Evaluation and risk Assessment of potentially toxic trace metals in water, sediment and fish from the City of Addis Ababa, Central Ethiopia

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ABSTRACT

Because of fast urban expansion and increased industrial activities, large quantities of solid and liquid wastes contaminated by trace metals are released into the nearby water bodies situated in Addis Ababa. Greater Akaki River (GAR) and Little Akai River (LAR), which join at Aba Samuel Reservoir, are the main rivers draining the city. Eleven (11) water, 11 sediment and 18 fish samples were taken and tested for the level of trace metals content, distribution, pollution level, bioaccumulation, possible sources of pollutants and associated ecological risk. Trace metals were tested using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). USEPA guideline values, geo-accumulation index, contamination factor, and pollution load index were used to evaluate contamination levels and eco-toxicity. The results indicated that the mean concentration of Cr, Fe, Mn and Ni in water samples and Cd, Pb, Mn, Ni and Zn in sediment samples exceeded their respective background values. Besides, the levels of Cr, Fe, Mn, Ni and Pb in the muscle of *Clarias gariepinus* were above the permissible limits for human consumption. Furthermore, ecological risk assessments using sediment samples revealed the widespread pollution by Cd and Pb and moderate pollution by Mn, Ni and Zn. A comparison with similar studies revealed that sediments from the study area had the highest average value of Pb. Generally, the study indicated that Akaki River catchment and Aba Samuel Reservoir were found to be moderately to strongly contaminated by heavy metals and thus posed high ecological risks.

kEY words*: Addis Ababa; bioaccumulation; Greater Akaki River; Akaki River; muscle; water pollution.*

1. INTRODUCTION

Contamination of the aquatic environment by trace metals in excess of the natural loads has become a problem of increasing concern. Large quantities of trace metals are discharged into the environment because of anthropogenic activities such as urbanization, industrialization and extension of irrigation and other agricultural practices. The situation is particularly alarming in developing countries, where most rivers, lakes and reservoirs are receiving untreated wastes because of poor setup of environmental sustainability (Mwanamoki et al., 2014). The most vulnerable river-reservoir systems are those crossing large cities and densely populated areas, and nearby industrial establishments (Yousaf *et al*., 2016). Trace metals are among the conservative pollutants that are not subject to degradation process and are permanent additions to aquatic ecosystems (Igwe & Abia, 2006). As a result, higher levels of trace metals are found in water, soil, sediment and biota. The toxic effects of trace metals occur because of bioaccumulation and biomagnification of the elements in the tissues of living organisms such as fish irrespective of the extent of exposure (Mulu& Mehari, 2013).

Water, sediment and fish are often exposed to toxic substances such as trace metals. Thus, quantification of trace metals in water, sediment and fish is extremely important (Kebede et al., 2012). Sediments provide important information about the long-term trends (Kebede et al., 2012). Fish are notorious for their ability to concentrate trace metals in their muscles (Olowu et al., 2010). Fishing has been the main sources of protein supply for many Ethiopians particularly for those who are residing in the vicinity of major water bodies. In Ethiopia, catfish is the second most commonly consumed type of fish species and it represents over 20 % of the total fish consumption in the country (Gashaw & Matthias, 2014).Previous studies have shown that African catfish has the ability to accumulate trace metals and its applicability as bioindicator (Ermias et al., 2014). In the current study site, Aba Samuel Reservoir, a significant percentage of *Clarias gariepinus* is captured and consumed by the local dwellers.

Addis Ababa city, estimated to have approximately 5 million population, hosts a large number of industries whose untreated wastewaters are discharged into the small river network. The primary sources of trace metals pollution in the river system include wastes from metal finishing industries (iron and still processing), tannery operations, textile industries, domestic, agrochemicals and leachates from landfills and contaminated sites (Nigatu et al., 2011)

Previous studies on trace metals in different media are hardly sufficient in the catchment area. Therefore, the objective of this work was to determine the occurrence, distribution and ecological risk of trace metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in water, sediments and fish samples from Akaki River catchment and Aba Samuel Reservoir, central Ethiopia.

2. MATERIALS AND METHODS

2.1. Study area and Sampling Stations

Addis Ababa is one of the fast expanding cities in Ethiopia. It is the country's commercial, manufacturing and cultural center. Addis Ababa has uncontrolled urbanization, industrialization, and poor sanitation resulting from waste disposal from different sources. The Great Akaki River (GAR) (locally known as Tiliku Akaki River) and Little Akaki River (LAR) (locally known as Tinishu Akaki River), with their tributaries, drain the city from north to south. These river systems are the dumping sites of solid and liquid wastes and thus, the environment of the city is threatened by severe pollution of toxic chemicals (Samuel et al., 2007). The confluence point of the two river systems is Aba Samuel Reservoir, 37 km south-west of Addis Ababa. It was the first hydropower station in Ethiopia to have been built in 1939, and was left unrepaired since 1970s (Eshetu et al., 2004). It was rebuilt and came to life again in 2016. The local people around the Reservoir use the river system for irrigation, drinking water for cattle, washing clothes, fishing and other domestic needs without strict water quality monitoring (Samuel et al., 2007).

2.2. Sample Collection and Pretreatment

Eleven (11) water and sediment samples were collected in January, 2017. 500 mL water and 500 g sediment samples were collected from the following sampling stations: GAR (S1- Entoto Kidanemihiret Monastery (control site 1), S2-Tirunesh

Beijing Hospital, S3-below Akaki Town); LAR (S4- Geferesa Reservoir (control site 2), S5- Lafto Bridge, S6- Jugan Kebele); Aba Samuel Reservoir (S7-below the confluence point of GAR and LAR, S8- Reservoir at the midpoint, S9-Reservoir

Sample code	Site description	Sample code	Site description
S1	GAR at Entoto Kidanemihiret Monastery	S7	Below the meeting and LAR

above the Dam) and downstream (S10-50 meter from the Reservoir and S11-1000 meter from the Reservoir) (Fig. 1). The samples were stored on ice-cooled container and transported to the laboratory, where they were filtered with Whatman 42 filter paper and preserved by 69% HNO_3 until the pH is less than 2. The filtered samples were stored in a refrigerator until further pretreatment and analyses were carried out. On the other hand, sediment samples were air dried at ambient temperature in a shade, grounded, homogenized, sub-sampled and passed through a stainless steel sieve of 45 µm and stored for further treatment. The sample codes shown below are described below and shown in Figure 1with sample codes.

Sample code	Site description	Sample code	Site description
S1	GAR at Entoto, Kidanemihiret Monastery	S7	Below the meeting point of GAR & LAR
S2	GAR at Tirunesh Beijing Hospital	S8	Aba Samuel Reservoir at the midpoint
S3	GAR below Akaki town	S9	Aba Samuel Reservoir above the Dam
S4	LAR above Gefersa Reservoir	S10	Aba Samuel Reservoir below the Dam
S5	LAR at Lafto Bridge	S11	Aba Samuel Reservoir 1 km downstream
S6	LAR at Jugan kebele		

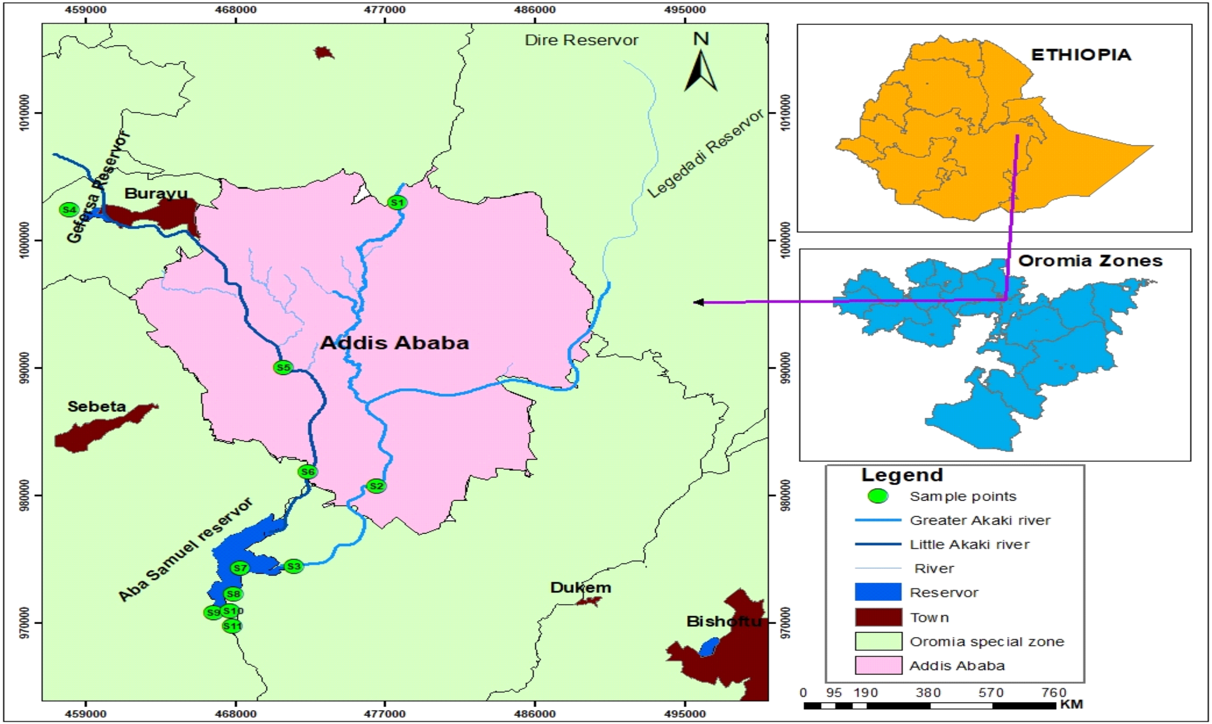

Figure 1. Sampling sites at the GAR, LAR, Aba Samuel Reservoir & downstream (S1-S11)

Eighteen fish samples were collected in January, 2017 from the Reservoir specifically at the Sites S8 and S9 (Fig. 1). This was done at the same locality and duration of time where the water and sediment samples were collected. Total length (in cm) and weight (in gm) were measured immediately after sampling. A sample from the back, dorso-lateral muscle, was removed from each fish using plastic knives. Three fish samples were pooled to form a composite sample of homogenized muscle tissue based on length. Samples were oven-dried at 105 ^oC for 24 h, powdered, homogenized using an agate mortar and pestle and stored in polyethylene bottles until the acid digestion procedure.

2.3. Sample preparation and Instrumental Analysis

Sediment samples (<45 μm) were digested using 20 ml of Mehlich 3 extractant (Mehlich, 1978). An inductively coupled plasma optical emission spectrometer, ICP-OES, (Arcos FHS2, Germany) was used for determining of trace metals in sediment samples. Calibration curves were prepared using 10, 20, 30, 40 and 50 mg/L of Fe and Mn; 0.04, 0.08, 0.12, 0.20, 0.40, 0.80, 1.20, 1.60, and 2.00 for Cu and Zn; 0.5, 1, 2, 3, 4, 5 for Cd, Ni and Pb and 1, 2, 4, 6, 8, 10 for Cr. In all cases, standard purity was >99.8%. The calibration curve showed linearity (r>0.995) by the detector response for the quantified elements.

Water and fish samples were digested using previously developed method by Ishak et al. (2015), with minor modifications (Ishak et al., 2015). Blanks were also prepared in a similar way. An inductively coupled plasma optical emission spectrometer, ICP-OES (Agilent 700 Series, USA), was used for determining trace metal concentrations in water and fish samples. Calibration curves were prepared using a series of 1, 2, 3, 5, 10 mg/L of mixed standards, containing Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn from 99.99% ICP grade standard. In addition, blank analyses were carried out to check interference from the laboratory. Individual reference standards of trace metals were used to identify and quantify the levels of samples. The same operational procedures were followed to analyze the samples as in the calibration routines. Quantification of the elements were recorded at the most sensitive emission wavelength of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn. USEPA sediment guidelines, geo-accumulation index (Igeo), contamination factor and pollution load index) were used to assess sediment contamination (Wang et al., 2014).

2.4. Quality Control and Quality Assurance

All the glassware were thoroughly washed with detergent, soaked in 10% HNO_3 for 24 h and rinsed with de-ionized water. All reagents used were analytically graded. For sediment samples, certified reference material was employed to validate and evaluate the accuracy of the method. Blank analyses were carried out to check interference from the procedures used and contamination from the laboratory. Mean recovery rates of the 4 metals were: Zn, 100.52%; Fe, 106.69%; Mn, 118.52%; Cu, 74.14%.

2.5. Statistical Analysis

Statistical analyses of the results were carried out using Origin Pro (version 9.4, 2017) and Microsoft Excel 2007.

3. RESULTS AND DISCUSSION

3.1. Trace Metals in Water Samples

Cd and Pb were below the limit of instrumental detection (except at S7 and S8 for Pb). The levels of Cr and Fe were higher in LAR than that of GAR. The concentrations of the trace metals were as follows: 0.05 to 0.39 mg/L (Cr); nd (not detected) to 0.01 mg/L (Cu); 0.27 to 3.2 mg/L (Fe); 0.02 to 1.78 mg/L (Mn); 0.04 to 0.11 mg/L (Ni) and 0.17 to 0.31 mg/L (Zn). The mean concentrations in water samples were put in descending order as follows: Fe>Mn>Zn>Cr>Ni>Pb>Cu>Cd. Wide variations in the concentrations of trace elements were observed spatially along the course of the rivers and the reservoir. It was found that most pollutants (except cadmium and lead) increased in concentration irregularly form upstream (control areas) to downstream sampling stations. The levels of trace metals (Cr and Mn) from Aba Samuel Reservoir and Akaki River catchment exceeded the permissible levels of FAO's recommendation (Ayers & Westcot, 1994).

3.2. Trace Metals in Sediment Samples

The average concentrations of 8 selected trace metals in sediment samples from the Akaki River catchment and Aba Samuel Reservoir during the dry seasons and their standard deviations were presented in Table 1. Based on the elemental concentrations, the pattern in sediment was: Mn>Fe>Pb>Cr>Zn>Ni>Cu>Cd. The concentration ranges (mg/kg) of trace metals were shown as follows: 2.1-2.9 for Cd, 16.2-43.7 for Cr, 1.6-15.3 for Cu, 406.4-844.8 for Fe, 124.4-256.4 for Pb, 335.5-1319.2 for Mn, 15.6-36.2 for Ni and 4-110 for Zn. Toxic elements (such as Cd and Cu) showed minimum concentration while Mn and Fe manifested the highest concentration.

The spatial distribution of trace elements in the river and reservoir sediment depends on many factors including distance of the element sources to the reservoir, the chemical characteristics of the element and the hydrological conditions of the river and reservoir system (Zhang et al., 2013).

In order to better understand the distribution of trace metals, the study area was divided into four regions: GAR, LAR, reservoir and downstream to the reservoir. In this connection, LAR was found to be more contaminated than GAR (Samuel et al., 2007). Statistical analyses of the results ($p<0.05$) indicated that there were no significant spatial variations of the trace metal among the sampling stations except Pb. The concentration of Pb varied significantly from upstream to downstream area ($p=0.02$). It was assumed that pollutants from GAR and LAR eventually ended up at Aba Samuel reservoir. However, the levels of most trace metals investigated were lower at Aba Samuel Reservoir (S7, S8 and S9) than the upstream areas. The relatively lower concentration of sediment-bound trace metals in the reservoir might have been due to less accumulated metals in the sediment because the reservoir was rehabilitated in 2016.

Table 1. Mean concentrations of trace metals (mean ± SD, mg/kg dry weight) in the sediment

Sample code.	Cd	Cr	Cu	Fe	Pb	Mn	Ni	Zn
S1	2.5±0.12	22.0±1.11	5.7±0.03	436.4±0.37	145.6±0.34	423.5±0.68	18.4±1.15	17.6±0.12
S2	2.6±0.16	28.4±1.19	3.6±0.01	844.8±2.93	134.1±5.34	1103.9±8.55	26.3±0.90	18.6±0.10
S3	2.6±0.14	23.1±1.23	3.2±0.01	557.0±1.21	133.3±4.68	576.9±2.47	20.7±0.98	29.1±0.17
S4	2.9±0.16	16.2±1.38	1.6±0.01	406.4±2.66	124.4±3.57	373.3±2.79	15.6±1.64	7.0±0.05
S5	2.7±0.16	26.9±1.24	15.3±0.09	579.3±1.19	170.8±5.04	669.6±3.66	26.2±1.18	59.4±0.25
S6	2.1±0.17	43.7±1.66	2.2±0.03	539.1±2.57	256.4±7.33	1319.2±10.51	36.2±1.37	5.2±0.04
S7	2.6±0.12	26.3±1.29	5.7±0.05	478.5±4.22	152.7±3.50	464.8±0.90	21.7±0.97	13.7±0.08
S8	2.6±0.17	27.7±1.45	5.2±0.01	492.1±2.48	128.5±2.80	591.7±3.17	22.9±1.62	4.0±0.02
S9	2.7±0.15	21.0±1.20	2.6±0.01	437.3±3.26	129.1±4.48	335.5±1.62	20.4±1.45	6.2±0.02
S10	2.5±0.17	31.4±1.35	4.0±0.19	818.4±3.59	145.4±1.41	1061.5±0.90	25.4±1.20	12.5±0.06
S11	2.8±0.13	32.1±1.23	7.4±0.02	539.9±3.83	171.2±1.31	522.0±3.11	26.3±1.40	110.0±0.55

Correlation analysis:

Pearson's correlation coefficients were computed in order to reveal the source(s) of pollution, distribution and similarity of behaviors of trace metals (Zhang et al., 2013). A significant positive correlation was observed for Pb with Cr (r=0.85), Mn with Cr (r=0.81), Mn with Fe (r=0.73), Mn with Pb (r=0.60), Ni with Cr (r=0.97), Ni with Pb (r=0.85) and Ni with Mn (r=0.83). This significant positive correlation suggests that the elements might have a common origin (Zhang et al., 2013).

Cluster analysis:

The dendrogram derived from the cluster analysis leaves two major clusters: (1) Cd, Cu, Cr, Ni, Zn, and Pb; and (2) Fe and Mn. In this regard, there are two distinct source factors: one that relates to the soil/geologic inputs (which may introduce Fe and Mn) and another that relates to a variety of anthropogenic activities in the catchment. Elements belonging to the same clusters are likely to have originated from common sources (Faisal et al., 2014).

Ecological risk assessment:

Based on the sediment quality guidelines (SQGs), the river and reservoir systems were found to be non-polluted with Cd and Cu, but non-to moderately polluted with Cr, Ni and Zn. At the same time, all of the sampling sites were heavily polluted with Pb.

Trace Metals in Fish Samples

Table 2 shows the mean concentration of trace metals in the muscles of *Clarias gariepinus* at Aba Samuel Reservoir. Considering guideline values by different authorities, the levels of Cr, Mn, Ni and Pb in the muscle of this fish were higher than the permissible levels. The rank of mean metal concentration in composite fish muscle was: Fe>Zn>Cr>Ni>Mn>Pb>Cu>Cd.In the present study, *Clarias gariepinus* muscle had mean Cr concentration ranging from 7.23 to 16.75 mg/kg. These measured values were much higher than the maximum permissible limit of 1 mg/kg by FAO (1983). The elevated Cr levels could be attributed to municipal and tannery wastes from upstream Addis Ababa city. The average concentration of Mn in the present study was 2.168 mg/kg which was above the WHO permissible limits of 1 mg/kg (Table 2) (Mokhtar et al., 2009). The higher amounts of Mn in this study might lead to dullness, weak muscle, headaches and insomnia (WHO, 2011). Mn in fish samples might have resulted from industrial effluents of iron and steel manufacturing. In the present study, the average concentration of Ni was 4.93 mg/kg which was above the maximum permissible limit of 1 mg/kg set by WHO (Mokhtar et al., 2009). Pb is classified as one of the most toxic trace metals. Level of Pb in the muscle of African catfish was above the maximum permissible limit of 1 mg/kg set by FAO/WHO, 2011.

1 Table 2. Trace metals in African catfish, *Clarias gariepinus* (mg/kg)

Pool number	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Pool 1	0.12±0.0004	9.02±0.0052	nd	62.42±0.0136	2.52±0.0004	4.06±0.0009	4.03±0.0099	19.79±0.0001
Pool 2	0.09±0.0002	7.23±0.0019	nd	42.78±0.0043	1.67±0.0002	3.81±0.0013	nd	4.88±0.0035
Pool 3	0.16±0.0005	10.84±0.0009	nd	66.26±0.0042	3.94±0.0001	4.94±0.0009	nd	20.60±0.0021
Pool 4	0.08±0.0003	7.99±0.0012	nd	42.30±0.0014	1.32±0.0002	3.3±0.0022	nd	7.21±0.0031
Pool 5	nd	16.75±0.0022	nd	95.6 ±0.0013	2.97±0.0006	7.7±0.0017	1.55±0.0042	14.03±0.0039
Pool 6	0.16±0.0006	12.14±0.0015	2.65±0.0013	89.55±0.0043	3.29±0.0005	5.74±0.0014	nd	38.77±0.0018
WHO 1989	**1**	-	**30**	**100**	**1**	**1**	**2**	**100**
FAO/WHO 2011	**0.2**	-	-	**43**	**5.5**	-	**1**	-
FAO 1983	-	1	-	-	-	--	-	-

Bioaccumulation of Trace Metals in Fish

As fish samples were collected from S8 and S9, the average levels of water and sediment samples at these sites were compared with the corresponding fish values. Bioaccumulation of trace metals in fish has been reported by many researchers (Ashraf et al., 2012; Dsikowitzky et al., 2013). Bioaccumulation of trace metals was evident from the present study in that the trace metal concentration was higher in fish than in water/sediment. This is in agreement with the findings of Reda and Ayu (2016). Iron concentrations were found to be high both in the water and fish tissue samples (Amanial and Adugna, 2016). However, cadmium concentration was found to be low in both water samples and fish tissue samples.

Table 3. Bioaccumulation of trace metals in fish

Element	Water (mg/L)	Sediment (mg/kg d.w)	Fish muscle (mg/kg d.w)
Cd	nd	2.7	0.12
Cr	0.07	24.5	10.66
Cu	0.01	3.9	2.65
Fe	1.90	464.7	66.50
Pb	0.02	128.3	2.79
Mn	1.19	463.6	2.62
Ni	0.05	21.7	4.93
Zn	0.19	5.1	17.55

4. CONCLUSIONS

In conclusion, most of the trace metals analyzed were available in measurable quantities in water, sediment and fish samples collected from Akaki river catchment and Aba Samuel Reservoir. It can be concluded that the catchment area had high influx of trace metals. Ecological risk assessment revealed the widespread pollution by Cd and Pb, and moderate pollution by, Mn, Ni, and Zn. The high level of trace metals in fish in comparison to water/sediment suggested that the fish were capable of accumulating the metals in their bodies from aquatic environment, which might affect human health as a result of fish consumption. Finally, safe disposal of domestic sewage and industrial effluents should be enforced in order to realize Acts of environmental protection. Setting national guideline for levels of trace elements in fish, and health risk assessments, are strongly recommended.

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CONFLICTS OF INTEREST

The authors declare that they have no competing interests.

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