Assessment of heavy metal status of sediment and water in Mainefhi and Toker drinking-water reservoirs of Asmara City, Eritrea

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ABSTRACT

In the present study sediment and water samples collected from Mainefhi and Toker reservoirs supplying drinking water to Asmara city (Eritrea) were analyzed for the concentration levels of the heavy metals Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn using Flame Atomic Absorption Spectrometry. The mean concentration levels of heavy metals in the water of Mainefhi reservoir: Cd 0.0035 mg/l, Cr 0.028 mg/l, Cu 0.004 mg/l, Fe 0.242 mg/l, Mn 0.065 mg/l, Ni 0.011 mg/l and Zn 0.016 mg/l. Similarly, the concentration levels for the water of Toker reservoirs: Cd 0.001 mg/l, Cr 0.017 mg/l, Cu 0.011 mg/l, Fe 0.165 mg/l, Mn 0.096 mg/l, Ni 0.007 mg/l. Zn 0.016 mg/l. All the mean concentration values of the heavy metals, were below the WHO guideline limits for drinking water, excluding Cd in Mainefhi reservoir, which is of higher value than WHO value. Co and Pb were not detected in the water samples of both reservoirs. The mean concentrations of the heavy metals in the sediment of Mainefhi were Co 0.790 mg/l, Cr 14.607 mg/l, Cu 8.167 mg/l, Fe 11,421.9 mg/l (1.142%), Mn 87.254 mg/l, Ni 4.539 mg/l, Pb 1.752 mg/l and Zn 5.769 mg/l. Analogously, the levels of heavy metals in Toker sediment Cd 0.005 mg/l, Co 33.860 mg/l, Cr 252.598 mg/l, Cu 83.396 mg/l, Fe 87,502.6 mg/l (8.752%), Mn 1455.746 mg/l, Ni 87.806 mg/l, Pb 5.005 mg/l and Zn 88.936 mg/l. The mean concentration levels of Mainefhi sediment were below the consensus-sediment quality guidelines of NOAA. Moreover, Cr, Fe, Mn, and Ni were above the consensus-sediment quality guidelines of NOAA for Toker sediment. Four different methods of assessment of the accumulation and contamination status of the sediment of heavy metals were applied. The Geo-accumulation index (I-geo) and Contamination factor (CF) results indicated that Mainefhi sediment was unpolluted (I-geo ≤ 0 , CF<1), while the Toker sediment was moderately polluted for Co, Cr, Cu, Fe and Mn (I-geo 0-1, CF1-3). The contamination degree (CD) calculations also showed that Mainefhi sediment was not contaminated while Toker sediment was in the 'moderate contamination degree' (CD 6-12). Enrichment factor (EF) values for sediments of both Mainefhi and Toker reservoirs (EF < 2) indicated there was little or no influence by anthropogenic input in regards to the measured heavy metals. Strong positive correlation exists between the mean concentration of the heavy metals in the sediment and water of both reservoirs. A Pearson's correlation result gave r = 0.97, n = 9, p < 0.01 for Mainefhi reservoir and r = 0.86, n = 9, p < 0.01 for Toker reservoir.

Keywords: Sediments, water reservoirs, geo-accummulation index, enrichment factor, contamination factor, contamination degree

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INTRODUCTION

Water reservoirs play an important role in the livelihood of human populations especially in the developing countries. They can serve as sources of domestic water supply, for irrigation, fishing activities and hydropower generation. However, the pollution of these aquatic environments has become a worldwide problem and concern in recent years because of the toxic effects of the pollutants on the aquatic organisms (Macfarlane and Burchett, 2000). Reservoirs are being polluted by indiscriminate disposal of sewage, industrial wastes as well as by agricultural and other human activities. Among the various environmental pollutants of such water bodies, heavy metals are of particular concern, due to their potential toxic effects, their persistence and their ability to bio-accumulate in the ecosystem (Censi et al, 2006).

Among the heavy metals, Cr, Ni, Pb, Cd, and As are regarded as toxic substances; Cr (VI), Ni and Cd are carcinogenic; As and Cd are teratogenic, and the health effects of Pb include neurological impairment and malfunctioning of the central nervous system (Nadal *et al.*, 2004). Although some heavy metals such as Fe, Mn, Co, Cu, and Zn are essential micronutrients for aquatic fauna and flora, they may be dangerous at high levels (Nadal *et al.*, 2004; Ochieng *et al.*, 2007). Thus, for organisms living in aquatic ecosystems, elevated essential and non-essential heavy metal contents may bring about a significant impact on their health, reproduction, and survival. Contaminants may also eventually pass through the food chain to humans and result in a wide range of adverse health effects.

Heavy metals in water reservoirs originate from both natural processes and anthropogenic sources. Contribution of natural processes like atmospheric inputs and geological weathering of rocks and soil, directly to surface waters, is usually the largest natural source. Relatively, anthropogenic sources are mainly from industrial processing, urban sewage and agricultural run-off (Rezaei and Sayadi, 2015).

Once heavy metals and other pollutants are discharged into water, they rapidly become associated with particulates and are incorporated in bottom sediments (Binning and Baird, 2001). Aquatic sediments are principally derived from weathering processes, or from terrestrial sources under high runoff storms and floods. Heavy metals are also present in aquatic sediments mainly as a result of chemical leaching of bed rocks (lithogenic or natural sources), as well as discharge of urban and industrial wastewaters from agricultural activities (anthropogenic sources). The occurrence of elevated levels of trace metals especially in the sediments can be a good indication of man-induced pollution where high levels of heavy metals can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Davies *et al.*, 1991).

The Contamination of sediments by heavy metals and other pollutants is one of the major threats to aquatic ecosystems and leads to serious environmental problems (Loizidou et al., 1992). Pollutants released to surface water accumulate to harmful levels in sediments (Chukwujindu et al., 2007). Like soils in the terrestrial system, sediments are important sinks for a range of substances including nutrients, hydrocarbons, pesticides and heavy metals (Baldwin and Howitt, 2007). Sediments act as both carriers and potential sources of contaminants in an aquatic environment. Contaminants are not necessarily fixed permanently by the sediments, under changing environmental conditions (temperature, pH, redox potential, salinity), they may be released to the water column by various processes of remobilization. Interactions commonly occur in the sediment-water interface (Chapman, 2000; Soares et al., 1999). This may lead to serious changes in the water quality of the reservoir. Moreover, distinguishing the anthropogenic inputs from the natural ones is the basis of environmental management (Wu et al., 2007). Heavy metals discharging into a lake from both natural and anthropogenic sources are distributed between bed sediments (as sink and source of heavy metals) and aqueous phases. Thus, in evaluating the pollution condition of a water body, investigation of both sediment and water should be considered (Lee et al., 2003).

Asmara reservoirs, Mainefhi and Toker, in addition to serving as drinking water sources, are also used for the development of freshwater fish breeding which are intended for the surrounding communities. Accordingly, the study and evaluation of heavy metals in the sediments and water of these reservoirs is necessary. This study is actually the first of its kind on these reservoirs. The main objective of the present study is, therefore, to determine the concentration levels of heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in the surface sediment and water of the two reservoirs, and to assess the pollution status of the sediment by the application of different methods of geochemical criteria computations. Moreover, the outcomes of this study may give a preliminary insight and indication of the mineralogical composition of the surrounding highlands around these reservoirs. Furthermore, the results obtained may be used by the authorities for the monitoring, management, and remediation of the quality and also as a baseline for further studies of these water reservoirs.

MATERIALS AND METHODS

Description of Sampling Site

The sediment and water samples were collected from Mainefhi and Toker drinking water reservoirs, which are located in the surroundings of Asmara city, the capital city of the state of Eritrea. Natural runoffs rain water, the primary source, from the mountains, hills and valleys around the reservoirs are directed to fall in these reservoirs. Asmara city is located at coordinates of 15.33°N, 38.93°E and an elevation of 2,325 meters above sea level in the central highlands of Eritrea (Figure 1). Asmara gets an average of about 500 millimeters of precipitation annually in the months of June to September (Ministry of Land Water and Environment, 2007). The geologists claimed that these highlands surroundings of Asmara city are rich in minerals.

Sample Collection

A total of 16 sediment and an equal number of water samples were collected from four sites within each of the two reservoirs, during the rainy (July and August) and the dry (February and March) months of 2014-2015. The sediment samples were collected by using clean plastic scoop and stored in polyethylene bags. The water samples were also collected in prewashed polyethylene bottles of 0.5 liter capacity. For preservation, the water samples were acidified in situ to pH < 2 with concentrated nitric acid (HNO₃) in the proportion of 1.5 ml of concentrated HNO₃ per 1 liter of sample water (APHA, 1998). The samples were then, carefully packed and transported to Khartoum where they were analyzed upon arrival.



Figure 1. Map of Eritrea showing the location of Asmara City

Preparation of Sediment Samples

The sediment sample was air dried, grounded using pestle and mortar and sieved with 2 mm sieve to obtain a fine powder. Conventional aqua regia digestion was performed in 250 ml Teflon beakers. A well-mixed sample of 0.5 g of the sieved sediment was digested in 12 ml of freshly prepared aqua-regia solution (1:3 HNO₃–HCl, v/v) on a hotplate for 3 hours at 110°C. The solution was then evaporated to near dryness. The sample was diluted with aqueous nitric acid (20 ml, 2%) and filtered through Whatman No. 42 paper into a 100 ml volumetric flask and then diluted to 100 ml with deionized distilled water (Chen and Ma, 2001). Every precaution was taken to avoid contamination during sampling, drying, grinding, sieving and storage.

Preparation of Water Samples

As the water samples were already acidified in situ with HNO_3 for preservation, the only procedure done for their preparation for analysis was to filter the samples with 0.45µm pore-size

filter papers into a 50 ml volumetric flask. Aqueous samples can be generally introduced for analysis directly and without any prior treatment (Bader and Zimmermann, 2012).

Determination of heavy metals by Atomic Absorption Spectrometer

Determination of metal concentrations present in the sediment and water samples was done by Flame Atomic Absorption Spectrometer (Model: *Shimadzu AA-7000*, Japan) using air-acetylene flame. Calibration line method was used for the quantification of selected metals and the digests were appropriately diluted whenever required (Radojevic and Bashkin, 1999). All measurements were made in triplicate. Similarly prepared blank solutions and standard reference materials were also analyzed alongside with the samples.

Data Analysis

Geochemical criteria, that is, Geo-accumulation index (*I-geo*) defined by Müller (1969), enrichment factor (*EF*) as defined by Rubio et al (2000), contamination factor (*Cf*) of heavy metals, and contamination degree (*Cd*) of sampling sites defined by Håkanson (1980) were determined as essential calculations for assessment of the sediment pollution status.

The Geo-accumulation index (*I-geo*), was calculated as follows (Mediola et. al, 2008):

$$I\text{-}geo = \log_2\left[C_n/1.5B_n\right] \tag{1}$$

Enrichment factor EF of heavy metals was calculated as the ratio of elemental concentration of sediment normalized to Fe (Muller, 1981):

$$EF = (C_n / Fe)$$
 sediment / (C_n / Fe) reference (2)

Contamination factor Cf was determined as (Håkanson, 1980):

$$Cf = C_n / B_n \tag{3}$$

The contamination degree (*Cd*) was defined as (Aksu et al., 1998):

The sum of all contamination factors (Cf) (4)

In the above formulas, C_n is the concentration of the examined element 'n' in the surface sediments, and B_n is the geochemical background concentration of metal 'n'. The world surface rock average (Martin and Maybeck, 1979) is used as background concentration reference for this study. The factor 1.5 in the *I-geo* formula (1) is incorporated to account for possible variation in the background data due to lithologic effect (Mediola et. al, 2008). The *EF* geochemical normalization 2, was obtained using *Fe* as the reference element and as a conservative tracer to differentiate natural from anthropogenic components, following the hypothesis that its content in the earth crust has not been troubled by anthropogenic activity and because natural sources (98%) greatly dominate its contribution (Tippie, 1984).

Statistical Analysis

The computer software "Statistical Program for Social Sciences" (SPSS, PASW Statistics 18, SPSS Inc.) was used for data analysis. Correlation matrices were determined with *Pearson product-moment correlation coefficient* computational software program in SPSS.

RESULTS AND DISCUSSION

Mean concentration levels of the heavy metal contents of water and sediment samples taken from Mainefhi and Toker reservoirs are present in Table 1 together with their respective international safe level guideline values. The mean concentration values of heavy metals in the water samples were all below the WHO (2011) maximum contamination level (MCL) guideline values with the exception of Cd in Mainefhi reservoir that was slightly higher. The concentration levels were Cd 0.0035 mg/l, Cr 0.028 mg/l, Cu 0.004 mg/l, Fe 0.242 mg/l, Mn 0.065 mg/l, Ni 0.011 mg/l and Zn 0.016 mg/l for Mainefhi reservoir and Cd 0.001 mg/l, Cr 0.017 mg/l, Cu 0.011 mg/l, Fe 0.165 mg/l, Mn 0.096 mg/l, Ni 0.007 mg/l. Zn 0.016 mg/l for Toker reservoir. Cadmium (Cd) and lead (Pb) were not detected in the water samples of both reserviors.

Table 1: Mean concentrations of heavy metals (ppm) in the sediment and water samples of
Mainefhi and Toker reservoirs with WHO and NOAA Guidelines

Source	Cd	Со	Cr	Cu	Fe ^a	Mn	Ni	Pb	Zn
MN Sed.		0.790	14.61	8.17	1.14	87.25	4.54	1.75	5.77
MN Wat.	0.004		0.028	0.004	0.242	0.065	0.011		0.016
TK Sed.	0.005	33.860	252.598	83.396	8.752	1455.746	87.806	5.005	88.923
TK Wat.	0.001		0.017	0.011	0.165	0.096	0.007		0.016
TEC*	0.99	50 ^b	43.3	31.6	2 ^b	460 ^b	22.7	35.8	121.0
PEC*	4.90	NG	111.0	149.0	4 ^c	1100 ^c	48.6	128.0	459.0
WHO MCL	0.003	NA	0.05	2.0	NG	0.5	0.02	0.01	3.0

MN Sed. = Mainefhi reservoir sediment; MN Wat. = Mainefhi reservoir water; TK Sed. = Toker reservoir sediment; TK Wat. = Toker reservoir water; TEC = Threshold Effect Concentration; PEC = Probable Effect Concentration; NA = Not available; NG = Not given.

* TEC and PEC Source is National Oceanic and Atmospheric Administration (NOAA, 2009)

 \mathbf{a} = For Fe the concentration values are calculated as percentages of iron in the reservoir sediments.

 \mathbf{b} = Lowest effect level (LEL) concentration (For Co, only LEL is given as 50+ ppm) (NOAA, 2009)

 \mathbf{c} = Serious effect level (SEL) concentration. (NOAA, 2009)

Similarly, the mean concentration values of heavy metals in the sediments of Mainefhi were Co 0.790 mg/l, Cr 14.607 mg/l, Cu 8.167 mg/l, Fe 11,421.9 mg/l (1.142%), Mn 87.254 mg/l, Ni 4.539 mg/l, Pb 1.752 mg/l and Zn 5.769 mg/l, whereas the levels in Toker sediments were Cd 0.005 mg/l, Co 33.860 mg/l, Cr 252.598 mg/l, Cu 83.396 mg/l, Fe 87,502.6 mg/l (8.752%), Mn 1455.746 mg/l, Ni 87.806 mg/l, Pb 5.005 mg/l and Zn 88.936 mg/l. Comparison between mean concentrations of heavy metals of both reservoirs with *Mann-Whitney 2-tailed Test* showed that

concentration levels for the metals at Toker reservoirs significantly exceeded (p < 0.01) those of Mainefhi, with the exception of Pb (p=0.450).

The concentration levels in the sediments of both reservoirs were also compared to the consensus-sediment quality guidelines (CSQGs) of NOAA (2009) to assess their status, because heavy metals in sediments can be secondary sources of pollution to the reservoir water, once environmental conditions are changed (Chen, *et al.* 1996). CSQGs include a threshold effect concentration (TEC) and a probable effect concentration (PEC) (Table1). Concentration of the contaminant in sediment below TEC means adverse biological effects are unlikely to occur (USEPA, 2000). Conversely, adverse biological effects are likely to occur if contaminant is above PEC level (Smith *et al.*, 1996). MacDonald, *et al.* (2000), noted that most of the TEC provide an accurate basis for predicting the absence of sediment toxicity, and most of the PECs, provide an accurate basis for predicting sediment toxicity. In this study, the mean concentration of all the heavy metals in the sediment samples of Mainefhi and that of Cd, Co, Pb and Zn in Toker are lower than the proposed TECs, indicating that there are no harmful effects from these metals. On the other hand, the concentrations of Cr, Fe, Mn, and Ni in Toker sediments exceeded PEC levels, which is an implication that they are likely to cause adverse effects on the aquatic organisms.

Nevertheless, it is important to mention in this context that especially the Toker reservoir catchment area includes the Embaderho locality, which is described as an area rich in minerals and a prospective mining site (Ministry of Energy and Mines, Eritrea, 2014). Moreover, anthropogenic pollution of the area is also unlikely as there are no industries and not much transport or traffic related activities around these reservoirs (Zerabruk, *et al.* 2015).

A Pearson product-moment correlation coefficient was computed to assess the relationship between the mean concentrations of the heavy metals in the sediment and those in the water of both the reservoirs. There was a strong positive correlation between the sediment and water concentration of heavy metals, with r = 0.97, n = 9, p < 0.01 for Mainefhi reservoir and r = 0.86, n = 9, p < 0.01 for Toker reservoir.

Geoaccumulation index (*Igeo*) of heavy metals in the sediments

Geo-accumulation index (*Igeo*) values are widely used to assess the degree of metal accumulation and contamination in terrestrial, aquatic and marine environments (Tijani et al, 2009). The geoaccumulation index scale, as defined by Muller (1969), consists of seven grades (0-6) ranging from unpolluted to extremely polluted (Table 2).

Igeo value	Class	Qualification of sediment
0	0	Unpolluted
0-1	1	From unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	From moderately polluted to strongly polluted
3-4	4	Strongly polluted
5-5	5	From strongly polluted to extremely polluted
>5	6	Extremely polluted

The results of the calculated *Igeo* in the sediment samples (Table 3) indicated that according to the Muller scale, the Mainefhi sediment was unpolluted, with values for all metals $Igeo \le 0$. On the other hand, the Toker sediment was 'from unpolluted to moderately polluted' or enriched for Co (0.175), Cr (0.904), Cu (0.305), Fe (0.343), and Mn (0.191), all having Igeo = 0-1. On the basis of the mean values of Igeo, the Toker sediment is enriched for metals in the following order: Cr > Fe > Cu > Mn > Co.

Table 3: Geoaccumulation index (Igeo) values of heavy metals in sediments of Mainefhi
and Toker reservoirs

Decompoin	Heavy metals in sediments and their Igeo values								
Reservoir	Cd	Со	Cr	Cu	Fe	Mn	Ni	Pb	Zn
MN Mean	0	-5.249	-3.208	-3.047	-2.594	-3.870	-4.490	-4.098	-4.626
TK Mean	-6.493	0.175	0.904	0.305	0.343	0.191	-0.216	-2.584	-0.680

Enrichment factor (EF) of the heavy metals in the sediments

A common approach to differentiate between the metal originating from anthropogenic activities and those from natural processes, and to assess the degree of anthropogenic influence is to calculate a normalized enrichment factor (EF) for the metal concentrations. EF is calculated by a comparison of each tested metal concentration with that of a reference metal (Muller, 1981). The normally used reference metals are Mn, Al and Fe (Liu et al., 2005). In this study iron was used as a conservative tracer to differentiate natural from anthropogenic components, following the hypothesis that its content in the earth crust has not been troubled by anthropogenic activity and it has been chosen as the element of normalization because natural sources (98%) greatly dominate its contribution (Tippie, 1984). According to Sutherland (2000), five contamination categories are defined based on the enrichment factor (Table 4). As the EF values increase, the contributions of the anthropogenic origins also increase (Sutherland, 2000).

EF value	Qualification of sediment
EF < 2	Deficiency to minimal enrichment
EF = 2-5	Moderate enrichment
EF = 5-20	Significant enrichment
EF = 20-40	Very high enrichment
EF > 40	Extremely high enrichment

Table 4: Classification of enrichment factor categories

The calculated *EF* values of heavy metals contents of the sediments of Mainefhi and Toker reservoirs (Figure 1), in descending order, are Fe (1.0) > Cu (0.741) > Cr (0.663) > Mn (0.419) > Pb (0.358) > Ni (0.273) > Zn (0.248) > Co (0.161) and similarly, Cr (1.498) > Fe (1.0) > Cu (0.989) > Mn (0.914) > Co (0.903) > Ni (0.273) > Zn (0.248) > Pb (0.358) > Cd (0.009), respectively. The calculated*EF*values for the heavy metals in the sediments of both reservoirs

indicate that there is little or no influence by anthropogenic input in regards to the measured heavy metals with EF < 2.



Figure 1: Enrichment Factor values of the heavy metals in sediments of Mainefhi and Toker reservoirs.

Contamination factor (CF) and contamination degree (Cd) of heavy metals in the sediments

In the present study the contamination factor (CF) and contamination degree (Cd) are used to assess the contamination status of sediments in the reservoirs. Håkanson (1980) described contamination factor (CF) as a ratio of the concentration of the element in samples to the preindustrial reference value for the element. In this study, the world surface rock average values (Martin and Meybek, 1979) are used for the calculation of CF as reference baselines. The degree of contamination (Cd) used in this study is also defined as the sum of all contaminant factor values of the heavy metals (Aksu et al., 1998). Both contamination factor (CF) and contamination degree (Cd) are classified in five categories (Table 5) according to the definition by (Håkanson, 1980).

 Table 5. Contamination factor (CF) and contamination degree (Cd) categories and terminologies

CF classes	CF and Cd terminologies	Cd classes
<i>CF</i> < 1	Low CF indicating low contamination / Low Cd	<i>Cd</i> < 6
$1 \le CF < 3$	Moderate CF/Cd	$6 \le Cd < 12$
$3 \le CF < 6$	Considerable CF / Cd	$12 \le Cd < 24$
$CF \ge 6$	Very high CF / Cd	$Cd \ge 24$

The calculated contamination factor (*CF*) results (Figure 2) indicated that Mainefhi sediment was unpolluted (CF<1), while the Toker sediment was moderately contaminated for Co (1.69), Cr

(2.81), Cu (1.85), Fe (1.90), Mn (1.71) and Ni (1.29), where the values are CF = 1-3. Accordingly, metal contamination levels were in order of Cr >Fe > Cu > Mn > Co > Ni in Toker sediment. Similarly, the contamination degree (*Cd*) calculation also showed that Mainefhi sediment was not contaminated (*Cd* < 1), while Toker sediment was in the 'moderate contamination degree' (*Cd* = 6-12).



Figure 2: Contamination factor (*CF*) and contamination degree (*CD*) of the heavy metals in sediments of Mainefhi and Toker reservoirs.

CONCLUSION

The present study shows that the heavy metals levels in both the water and sediments of the two reservoirs were within the values recommended in the WHO Guidelines for drinking water and Consensus-Based Sediment Quality Guidelines (CSQGs), with the exception of Cd in the water of Mainefhi reservoir, and Cr, Fe, Mn and Ni in Toker sediment which were higher than the respective guideline values. The geochemical parameters, *Igeo*, *EF*, *CF* and *Cd*, applied on the sediments of both reservoirs, also indicated there was no to moderate pollution and little or no anthropogenic input. These results, therefore, reinforce the belief that both reservoirs and their catchment areas are not influenced so far by anthropogenic polluting activities. Accordingly, the occurrence of some metals in the sediments being above the guidelines can be attributed, predominantly, to influx from the upper catchment areas of the Toker reservoir, which is described as an area rich in minerals and a prospective mining site.

It is noteworthy to mention that this study can serve as baseline information for further research to help in the monitoring of the safety and quality of the reservoirs for domestic water use as it is the first survey of its kind. It is recommended that regular water quality monitoring surveys are essential so as to enable the evaluation of possible build-up of heavy metals and other hazardous chemicals in the sediment and water of the reservoirs with the impending mining activities in the upper catchments.

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