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Trace metal pollution in surface water and sediment of a Mountain River in an East African country

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ABSTRACT

The concentration and spatial distribution of trace metals Cr, Cd, Pb, Zn, Cu and Hg were measured in surface water and sediment of East Usambara Mountains (EUMs) River in Tanzania. Concentrations ranged from below detection limit (bdl) to 4.86 mg/l and bdl to 157.98 mg/kg dw in surface water and sediments, respectively. The levels of the metals in water were within the Tanzanian drinking water limit standards, but exceeded the World Health Organisation (WHO) drinking water limit guidelines. Concentrations of the metals in sediments in most of the locations were observed to be higher than the average concentration in the upper Earth’s crust. Wastes from artisanal gold mining and agricultural runoff were considered to be the major sources of the elevated metal concentrations in water and sediments of the study area. The extent of the trace metals pollution in surface water and sediments of EUMs rivers catchment implies that acute health consequences could be insignificant, but chronic adverse health effects due to prolonged pollution and long term exposure through food contamination, washings, animal keeping and fishing could be certain.

Introduction

Trace metals occur in aquatic systems, mainly from the weathering of bedrock in the catchment areas. The increasing trend of enhanced levels of these metals in the aquatic environment has attracted global attention due to its toxicity, abundance and persistence (Islam et al., 2014). Their degree of toxicity into the environment and living organisms generally depend on the extent of exploitation, application and consequent mobilization into the air, water and soil (Lone *et al*., 2008). Many trace metals are lipophilic in nature due to formation of conjugates with organic molecules, thus makes them easily taken up by living organisms and eventually lead to bio-concentration and magnification (Gray, 2002). In living organisms some of the trace metals such as Cd, Cr, Pb and Hg are reported to be associated with various health effects like kidney failure, liver damage, cholesterol metabolism effects, cancer and fetus effects (Islam *et al*. 2007; Singh & Taneja, 2009). The impacts range from abnormality, deformity, growth and reproduction interference (Velma *et al*. 2009). The Mina mata Bay in southern Japan during 1950 and 1968 is one of the well-known examples of metal effects, whereby Hg contamination in fish is associated with hearing and speech impairments, mental disturbance and kidney failure on humans.

Huge amount of chemicals especially trace metals have been released into rivers worldwide due to global population growth, intensive domestic activities and expanding industrial and agricultural production (Su et al., 2013). In Africa, rivers even in protected areas have been associated with water quality problems because of human activities that pollute and degrade water bodies and leads to the increase levels of metals in the river water and sediments. In Tanzania, environmental problems mainly due to artisanal mining and agricultural activities on natural landscapes have been gradually increasing during the last ten years. Of particular concern, is East Usambara Mountains (EUMs) where illegal gold mining by artisanal miners have substantially degraded water resources, natural vegetation and biodiversity particularly in the Amani Nature Reserve (Doggart et al., 2004). The degradation is also contributed by agricultural activities, predominantly tea plantation and horticulture farming through irrigation and application of chemical fertilizers and pesticides. These activities have tarnished the Zigi River catchment area and pose serious threat to aquatic biodiversity and human health especially for community of Tanga city who obtained their drinking water supply from this river. Presently there is no data available that quantifies the elemental contaminants inputs due to illegal mining and agriculture activities in the area. This study was therefore conceived in order to provide baseline information for possible monitoring and alleviating the problem.

Materials and Methods

Description of the study area

The East Usambara Mountains (EUMs) form part of the Eastern Arc of remote mountain building blocks situated southeast and southward from Mt. Kilimanjaro in northern Tanzania. It covers an area of 1,300 km square between 4°48’5°13S and 38°32’38°48’E (Hamilton, 1989). The mountains are the catchment of Zigi River which is permanent throughout the year (Litterick, 1989). The river consists of two tributaries, Zigi which flows east-north direction and Muzi which flows southwards. The Zigi-Muzi tributaries converge at Mabayani and then flow out of the Usambara near Lanzoni onward into the Indian Ocean (Litterick, 1989). The upper catchments consist mainly of metamorphic rocks of the Usagaran System that have comprehensively magmatized of pyroxene and hornblende granulites and gneisses. Top soils chiefly holds moderate amounts of organic matter (Litterick, 1989). The lower EUMs and Zigi catchment contain mainly sedimentary succession of rocks from Karroo to Quaternary that

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form a sequence of sandstones and shale's. The upper catchment of Zigi consists of dense forest integrated with tea plantations and traditional food crops cultivation. The down slope consist mainly of grazing land, dry savanna bush and coconut palms that stretch along the coastal strip. The rainfall varies from November to December as short rains and March to June is long rains (Bruen, 1989). The mean maximum daytime temperatures are approximately 4-5°C and the mean minimum temperatures 2-3°C (Hamilton, 1989).

Sample collection and preparation

A total of 28 samples of water and sediments were concurrently collected during the rainy season in June 2013. Samples were collected from fourteen different stations (S1-S14) from upstream to downstream of the river (Figure 1). The stations were marked using a hand held Geographical Positioning System (GPS), these include Mlesa [S1(S0457997 E9432530) S2(S0457834 E9432509) S3(S0457822 E9432709) S4(S0457429 E9432052) and S5(S0457785 E9432999)], Kwemkuyu [S6(S0460239 E9437929)], Makumba [S7(S0460779 E9436281)], Kisiwani [S8(S0462698 E9436741)], Lunguza [S9(S0465904 E9434374) and S10(S0465989 E9434273)], Mungubu K [S11(S0466269 E9435958)], Mambari L [S12(S0469745 E9447747)] and Lanzoni [S13(S0476329 E9446144) and S14(S0477847 E9445564)]. Water samples were collected by grab technique in 500 mL plastic bottles. After sampling, samples were measured for physico-chemical parameters temperature, pH, TDS and EC using hand-held portable water quality monitor and later filtered and preserved by concentrated HNO_3. The water samples were analysed immediately upon arrival to the laboratory. The river bed sediment samples were collected at a depth of 0-5 cm using a stainless steel scoop. They were wrapped in aluminium foil and labeled according to site name and number, then placed in the polyethylene bags and transported to the laboratory where they were kept at 20°C waiting for analyses.

Sample preparation and instrumental analysis

Water samples, previous filtered and preserved with HNO_3 were directly analysed for trace metals without any further treatment. Sediment samples for trace metals were oven dried at 105°C for 24 h, grinded and sieved by 0.18 mm sieve. 0.5 g of the sieved sample was extracted in a glass tube with 2 mL of concentrated hydrochloric acid-nitric acid mixture (1:3) in a hot plate (60 min at 95°C) and allowed to cool to room temperature. The extract was brought into the final volume of 10 mL with deionised water and left to settle overnight. The supernatant was filtered and analysed for trace metals.

Trace metals determination was performed by Perkin Elmer Analyst 100 Atomic Absorption Spectrometer with Perkin Elmer HGA 850 Graphite Furnace and Perkin Elmer AS 800 Auto-sampler made in Germany. The instrument detection limit is 0.01 ppm (0.1 mg/kg), slit width 0.70 nm and elements wavelength of 228.8, 357.9, 324.8, 283.3 and 213.9 nm for Cd, Cr, Cu, Pb and Zn, respectively. Standard solutions were prepared using high purity standard reagents. These solutions together with blanks were used in the process of analysis to ensure the precision and accuracy of the results. For analytical quality assurance, after every five samples readings, standards were run to make sure that the margin of error is within 5%.

Pollution load index (PLI) and contamination factor (CF)

To assess the sediment quality, an integrated approach of pollution load index of the six metals was calculated according to Suresh et al. (2011). The PLI is defined as the nth root of the multiplications of the contamination factor of metals (CF).

$$PLI = (CF_1 \times CF_2 \times CF_3 \times . . . \times CF_n)^{1/n}$$

where CF_{metals} is the ratio between the content of each metal to the background values in sediment, $CF_{metals} = C_{metal}/C_{background}$. The background values used to compare the metal concentrations are those of unpolluted sediments based on the world surface rock average. However, since the typical background values of these elements in the EUMs are hardly established the average upper earth's crust values are utilized as rough estimations (Charkravarty and Patgiri, 2009). The approach of using earth's crust values as background estimates in different environments where actual background levels are unknown has been widely used to study the sources and contamination of metals in different riverine, estuarine and coastal environments (Zhang et al., 2009). The PLI value of zero indicates perfection, a value of one indicates the presence of only baseline level of pollutants and values above one would indicate progressive deterioration of the site and estuarine quality (Tomilson et al., 1980). The PLI gave an assessment of the overall toxicity status of the sample and also it is a result of the contribution of the six metals. The ratio of the measured concentration to natural abundance of a given metal had been proposed as the index contamination factor (CF) being classified into four grades for monitoring the pollution of one single metal over a period of time (Turekian and Wedepohl, 1961; Loska et al., 1997): low degree (CF<1), moderate degree (1≤CF< 3), considerable degree (3≤CF<6), and very high degree (CF≥6). Thus the CF values can monitor the enrichment of one given metal in sediments over a period of time.

Statistical analysis

The data were statistically analysed using Mocrosoft Excel 2003. The means and standard deviations of the metal concentrations in water and sediments were calculated at 95% confidence level.

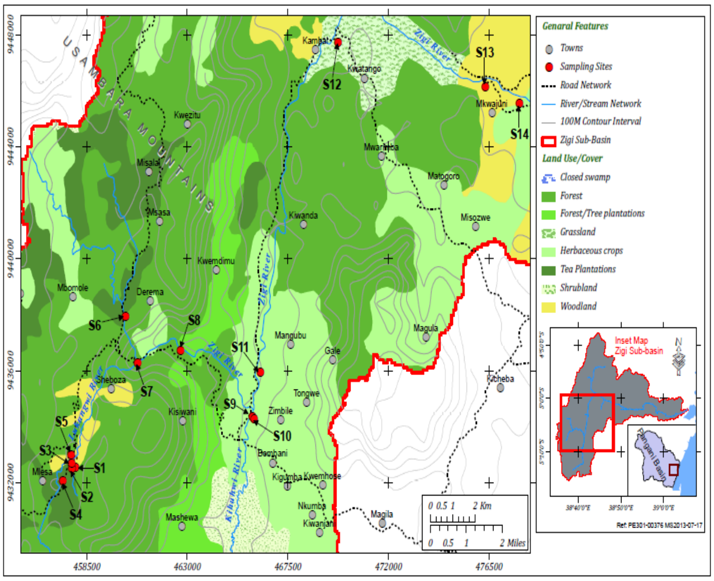

Figure 1. Map of EUMs showing site descriptions and sampling stations

Results and Discussion

Physicochemical properties and metal concentrations in surface water

Physicochemical parameters are important factors that influence the natural processes in rivers water. Besides affecting each other, deviation of these parameters from the normal values is an indication of pollution. In this study physicochemical parameters temperature, pH, electrical conductivity (EC) and total dissolved solids (TDS) were determined, results of which are presented in Figure 2. The figure shows that temperature levels were in the range of 19-25°C.

This reflects the temperature levels of the studied sites. Temperature of water is important as it affects dissolution of oxygen, pH and salt dissolubility in water (Florescu et al., 2011). The pHs in all the sites were in range of 7.5-8.5 which is normal for natural surface water, being optimum for most organisms. These levels are also within guidance values for drinking water in Tanzania and as per WHO (TBS, 2003; WHO, 2006). Neutral pH suggests that the water is not susceptible to trace metals pollution originating from geological materials and rocks (Sampson et al., 2012). This implies that deviation of trace metal concentrations reported in this study may be due other sources such as agricultural runoff, mineral from rain water runoff, mining wastes or other discharges from human settlements. Electrical conductivity in natural waters is the normalized measure of the water's ability to conduct electric current, and is an indication of the physical presence of dissolved chemicals in water. EC can provide a clear view of the concentration of metal ions in water. Increase conductivity can be the results of released trace metals ions in the water (Florescu et al., 2011). EC values reported in this study were in the range of 45-100 mS/cm and TDS values in the range of 25-50 mg/l. The guidance values for EC and TDS are 1000 mS/cm and 1000 mg/l, respectively, signifying that the surface waters investigated in this study are generally fresh.

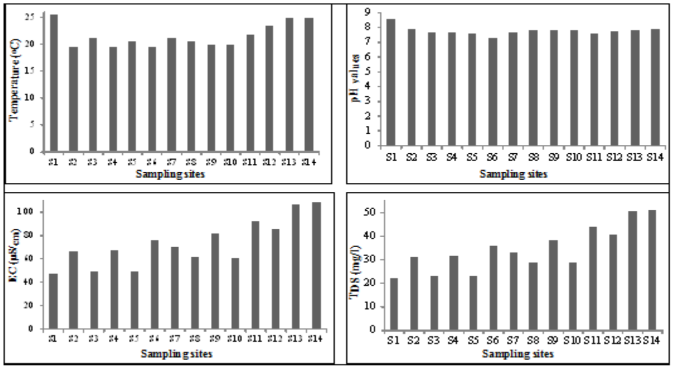

Figure 2. Levels of the physicochemical properties measured in surface water samples of EUMs

The results of trace metal Cu, Cd, Cr, Pb, Zn and Hg concentrations in surface water of river Zigi are shown in Table 1. The results show that the trace metal concentrations from upstream to downstream (S1-S14) varied without any distinct patterns. This spatial variability reflect the impact of varying settings, lithogenic characteristics and land use practices spread over different sites along the river. The origins of these metals may therefore be attributed to the artisanal mining waste discharge and agricultural runoff, and to the small extent the nature of the study area. For instance application of phosphoric fertilizers is one of the major sources of Cd release into the environment (Mirlean and Roisenberg, 2006). The pathways of Cu contamination in waterways include the use of Cu based fungicides such as the Bordeaux mixture (Nogueirol et al., 2010). The most important anthropogenic sources of Hg pollution in aquatic systems are atmospheric deposition, urban discharges, agricultural runoffs, mining, combustion and industrial discharges (Wang et al., 2004). In this study, the trend of mean concentration of the metals in surface water was Cr>Pb>Hg>Zn>Cd>Cu.

The mean concentrations of Cu, Cd and Zn were lower than the limits for Tanzania drinking water standard (TBS, 2003). The mean concentrations of Cu and Zn were also within the WHO guideline limits for drinking water, but that of Cd was higher than the WHO guideline limits. Concentration of trace metals Cr, Pb and Zn higher than the maximum limits for Tanzanian standards and WHO guidelines were detected mostly from upstream and downstream of the river. Mercury was the least found trace metal with the highest concentration of 4.86 mg/l at S3. The metal has been reported to be part of amalgamation process during gold mining activities, production and pose a risk of water pollution (Tschakert and Singha, 2007). The abundance per individual metals in terms of occurrence from various sites was in the following order Cr>Cd>Pb>Zn>Cu>Hg.

Table 1 also shows concentrations of metals in individual sampling sites, which were observed to vary among locations. The concentrations of Cu in all the sites were within the permissible drinking water limit as per Tanzanian standard and WHO guideline. Concentrations of Cd at S5 and S7 were above Tanzanian standards (TZS) and WHO guidelines limits (TBS 2003; WHO 2006), while at S1, S3, S4, S5, S6, S7, S8, S11 and S14 were within TZS limits but higher than WHO limits. Concentration of Cr at S1, S2, S3, S4, S10, S11, S12, S13 and S14 were higher than TZS and WHO limits. Concentrations of Pb at S3, S4, S11, S13 and S14 and Hg at S3 were above the TZS and WHO limits.

Table 1. Trace metal concentration (mg/l) in water sample of EUMs and maximum allowable concentration (mg/l)

Sampling station	Cu	Cd	Cr	Pb	Zn	Hg
S1	bdl	0.02	0.38	bdl	bdl	bdl
S2	0.01	bdl	0.41	bdl	bdl	bdl
S3	bdl	0.03	0.37	0.20	bdl	4.86
S4	0.04	0.03	0.35	0.23	0.01	bdl
S5	0.01	0.05	bdl	0.30	0.02	bdl
S6	bdl	0.01	bdl	0.90	0.01	bdl
S7	bdl	0.07	bdl	bdl	bdl	bdl
S8	bdl	0.01	bdl	bdl	bdl	bdl
S9	0.01	bdl	bdl	bdl	bdl	bdl
S10	bdl	bdl	0.17	bdl	bdl	bdl
S11	bdl	0.03	0.45	0.30	0.16	bdl
S12	0.02	bdl	0.64	bdl	bdl	bdl
S13	0.04	bdl	0.45	0.18	0.03	bdl
S14	bdl	0.02	0.36	0.26	0.04	bdl
Mean ±SD	0.02±0.01	0.03±0.02	0.4±0.12	0.34±0.25	0.05±0.06	0.37±0.06
TZS	3.0	0.05	0.05	0.1	15.0	0.001
WHO	2.0	0.003	0.05	0.01	3.0	0.001

bdl=below detection limit. Highlighted values indicate levels above the TZS limits

Physico-chemical properties and metal concentrations in sediment

The physico-chemical parameters and trace metal concentration of sediments are presented in Table 2. The average pH in sediment was 5.91 which are slightly acidic. A wide range of values for metal concentrations was observed among the sampling stations. Factors such as salinity, geomorphological setup and land runoff might have played a crucial role in the variation of metals. The metal distribution was dominated by Cu, Pb and Zn which were recorded in all sampling stations, followed by Cd, Cr and Hg. The highest concentration level for all measured metals in sediments was that of Hg at S12 (157.98 mg/kg dw). The trace metals concentrations in sediments show a significant difference in relation to aquatic levels and distributions. Generally, there was a higher metal concentration in sediments than in river water samples. High levels in sediment than in water has to be expected because in water metals are absorbed onto suspended particles, and eventually settle to the bottom of the river in which sediments act as a 'sink' where chemicals tend to accumulate. In order to prevent or to minimize sediment pollution and contamination developed countries have set guidelines to form the basis for accepting sediment quality. Since Tanzania is yet to set sediment standard on the levels of trace metals in sediments of freshwater resources. The concentrations of environmentally significant metal contaminants quantified in EUMs sediments in this study were therefore compared with existing numerical sediment quality guideline (SQG) from other countries and regulatory agencies. Table 2 summarizes the Canadian Threshold Effect Level (TEL) and Probable Effect Level (PEL) sediment quality guidelines that were used for this purpose. In the reported study, Cu at S1, S3, S4, S5, S6, S7, S8, S9, S12 and S13 had concentrations above the TEL and (S3) above the PEL. Cd at S1, S2, S3, S4, S5, S6, S7, S8, S9, S11 and S12, and S13 had concentrations above the TEL. Concentrations of Pb at S1, S2, S7, S8, S10, S11, S13 and S14, and Zn at S3 were above the TEL. Source of Cu, Cd, Pb and Zn in the study sites could be from agricultural runoff and the metals being from pesticides, herbicides, fungicides and pesticides applied in tea plantations. Hg (S3, S8, S9 and S12) were excessively above the TEL and the PEL respectively. The extreme high concentration of Hg could probably originate from the artisanal gold mining activities particularly during the gold extraction process.

Table 3 summarises the concentration ranges and respective means of some trace metals quantified in the East Usambara Mountain river sediment samples. The concentrations are compared to those of the average earth's crust assumed to be the source of unpolluted river bedrock compositions (Martin and Meybeck, 1979). The typical background values in the study area are hardly established, thus even though natural metal background concentrations vary from place to place according to the composition of the bedrock and different geochemical characteristics of the specific area (Das and Krishaswami, 2007), in this study the upper Earth's crust values are only considered as very rough estimates. Table 3 also shows that metal concentrations in the EUMs sediments with exception to Cr are significantly exceeded the Earth's crust. Same trend is for metals in individual locations. However, as earlier pointed out this alone does not signify enrichment but rather a possibility of high actual natural background levels at the specific areas.

Table 2. Trace metal concentration (mg/kg-dw) and physicochemical parameters in sediment of EUMs, and standard metal limits (µg/g dw)

Sampling station	Cu	Cd	Cr	Pb	Zn	Hg	pH	EC (Ms/m)
S1	23.86	1.24	4.53	45.57	73.71	bdl	6.15	0.293
S2	14.28	0.23	1.97	48.71	22.59	bdl	4.99	0.367
S3	118.67	0.92	1.06	13.75	140.23	45.21	5.7	0.188
S4	57.86	0.36	1.91	10.45	72.94	bdl	6.27	0.396
S5	43.14	0.59	1.56	19.05	51.40	bdl	6.11	0.384
S6	32.32	0.57	1.73	21.07	44.45	bdl	5.91	0.347
S7	21.23	0.22	0.46	48.53	35.90	bdl	6.74	0.293
S8	23.63	1.13	0.80	26.34	35.67	52.20	5.5	0.479
S9	21.96	1.07	0.17	16.32	32.34	54.36	5.94	1.037
S10	17.53	bdl	bdl	39.50	35.21	bdl	5.67	0.456
S11	14.10	0.31	bdl	44.79	16.36	bdl	5.66	0.283
S12	59.36	0.44	1.94	24.75	47.00	157.98	6.5	0.548
S13	56.81	0.27	bdl	51.08	78.80	bdl	5.76	0.506
S14	17.15	bdl	bdl	51.07	28.90	5.60	5.8	0.436
CTEL	18.7	0.7	52.3	30.2	124	0.13	na	na
CPEL	108	4.2	160	112	271	0.7	na	na

bdl=below detection limit; CTEL- Canadian Threshold Effect Level; CPEL- Canadian Probable Effect Level; na – not applicable

Table 3. Concentration ranges (mg/kg dw) of some trace metals measured in EUMs sediments

Metal	**East Usambara Mountains sediments** (n=14)		**Average conc. in the upper Earth's Crust**
	Concentration range	Mean concentration ± SD	
Cu	14.10-118.67	37.28±28.68	25
Cd	bdl-1.24	0.61±0.38	0.09
Cr	bdl-4.53	1.61±1.21	35
Pb	10.49-51.08	32.93±15.41	20
Zn	16.36-140.23	51.11±32.04	71

Assessment of contamination levels in sediment

The degree of sediment contamination was scrutinized by the use pollution load index (PLI). Figure 3 summarise the calculated PLI, which range from 0.5 to 1.6, which confirm that some parts of sediment of the studied river were polluted (PLI>1). The figure shows that upper stations (S1, S3) and three lower stations (S12, S13, S14) had contamination index PLI>1, indicating that their polluted with trace metals. The PLI (Figure 3) and CF (Table 4) values indicate that Cu, Cd and Pb are the major contributors to the sediment pollution in the study area. The origins of these metals to the area might be due to the effect of agricultural and mining activities. The value of contamination factor (CF) for Cr and Zn showed low degree of contamination (CF<1), where as Cu and Pb showed moderate degree of contamination (1<CF<2). The value of contamination factor for Cd showed considerable degree of contamination (3<CF<6). Overall, the CF for all metals were in the descending order of Cd>Pb>Cu>Zn>Cr.

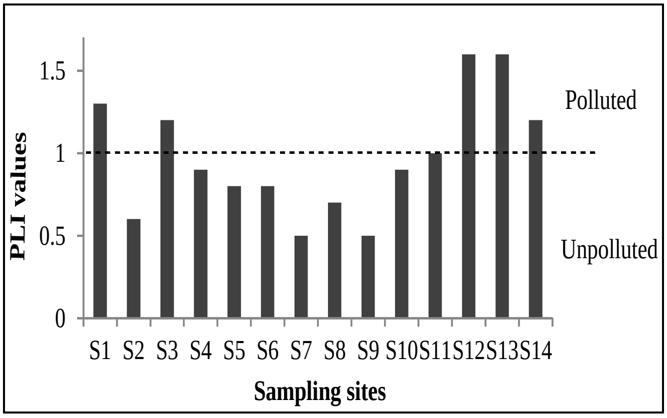

Figure 3. Pollution load index (PLI) value of trace metals in sediment of EUMs

Table 4. Contamination factor (CF) of trace metals in sediments of EUMs River

Sampling station	**Contamination factors**				
	Cu	**Cd**	**Cr**	**Pb**	**Zn**
S1	1.0	13.8	0.1	2.3	1.0
S2	0.6	2.6	0.1	2.4	0.3
S3	4.7	10.2	0.03	0.7	2.0
S4	2.3	4.0	0.1	0.5	1.0
S5	1.7	6.6	0.04	1.0	0.7
S6	1.3	6.3	0.05	1.1	0.6
S7	0.8	2.4	0.01	2.4	0.5
S8	0.9	12.6	0.02	1.3	0.5
S9	0.9	11.9	0.005	0.8	0.5
S10	0.7	-	-	2.0	0.5
S11	0.6	3.4	-	2.2	0.2
S12	2.4	4.9	0.1	1.2	0.7
S13	2.3	3	-	2.6	1.1
S14	0.7	-	-	2.6	0.4
Mean CF	1.5	5.9	0.04	1.7	0.7

Conclusions

The results of this study revealed that surface water and sediments in the studied sites of EUMs River are contaminated by trace metals. Concentrations of Cd, Cr and Pb in surface water for most of the sites generally exceeded the safe recommended values. This suggest that EUMs River is polluted by trace metals and even though acute adverse health effect could be insignificant, chronic effect due to prolonged pollution and long term exposure could be certain. The overall pollution load and contamination factors revealed that sediments in the study area were considerably polluted by Cu, Cd and Pb, and moderately by Zn and Cr. The study suggests that further analysis be extended aquatic organisms and plants in order to ascertain the ecological risks.

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