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Investigation and assessment of heavy metals in surface sediments of Ganjiang River, China

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Abstract

Surface sediment samples were collected from four sampling areas (Ganzhou nonferrous metal processing zones (GZ), Jian city sewage discharge areas (JA), Non city areas (NC) and Zhangshu middle-city areas (ZS)) to investigate the degree and chemical partitioning of heavy metals. Samples were analyzed for Cu, Zn, Pb, Cd, Cr, As and Ni using sequential extraction scheme. Based on geo-statistics analyses, the results showed maximum values of Cu (131.84 μg g^{-1}), Zn (290.68 μg g^{-1}), Cd (99.57 μg g^{-1}), Cr (53.75 μg g^{-1}), As (25.58 μg g^{-1}) and Ni (64.75 μg g^{-1}) were in sediments dated at GZ areas, while ZS areas had the highest concentrations of Pb (128.51 μg g^{-1}). Almost all metal values were higher than their corresponding background values in the following order: Ni<Cr<Zn<Pb<As<Cu<Cd. Meanwhile, sediments showed that heavy metals were predominantly occured in Fe-Mn oxide, residual, organic, exchangeable and carbonate fractions respectively. It was found that concentration in carbonate fractions were generally low for most of the studied metals. The geo-accumulation index and sediment quality values were used to determine the potential risk of heavy metal contamination. The results of geo-accumulation index indicated that contamination degree ranged from uncontaminated to extremely contaminated degrees. When metal partitioning characteristics were also considered, over 60% of metals showed potential bioavailability and toxic effects were probable.

Key words

Ganjiang River, heavy metals, partitioning, risk assessment, sediments

Introduction

The term `heavy metal' should probably be reserved for those elements with atomic mass ≥ 200. In practice, the term has come to embrace any metal, exposure to which constitutes a potential hazard (Baldwin and Marshall, 1999). Heavy meals are one of the serious pollutants in our natural environment because of their toxicity, persistence and bio accumulation potential (Duruibe *et al.*, 2007). Sediments are the main sink for pollutants and have been recognized as important indicators of water contamination (Varol and Sen, 2012). Analysis of pollutants in water is not conclusive owing to water discharge fluctuation and short resident time. The same remains true for suspended matter. The study of sediments play an important role for their longer residence time, and the role is called “the record of history”. Spatial distribution and concentration of heavy metals in sediments are affected by both natural environmental and anthropogenic factors (Bai *et al.,* 2011). Natural environmental factors include benthic agitation, flow changes and natural erosion etc. The anthropogenic sources of heavy metals in sediments include mining, smelting, waste disposal, urban effluent, vehicle exhausts, sewage sludge, pesticides, fertilizers application etc. (Zhang *et al.,* 2012). Man, through industrialization and technology, has developed the capacity to alter these natural interactions to the extent that the aquatic life therein have been threatened to a devastating extent. For this reason, bottom river sediments are not only the sink of heavy metals, but also potential secondary sources of heavy metals in aquatic system (Bing *et al.,* 2013).

The mobility, biological availability and toxicity of trace metals generally depend on the distribution of heavy metals in various chemical phases (Usero *et al.,* 1998). Thus, knowing the partition of total metal concentrations into non-residual and

residual fractions or phases and mobility of heavy metals is very important for evaluating the phytoremediation efficiency (Zhang *et al.*, 2013). Sometimes, partitioning of heavy metals is used in sedimentary geochemistry to provide information regarding the binding sites of metals as well as their source and pathways by which they have been transported to the aquatic environment. Using sequential extraction procedures, it is possible to identify a number of different fractions: "exchangeable", "carbonates", "Fe/Mn oxides", "organic/sulphidic" and "residual" (Tessier *et al.*, 1984). The non-residual, including exchangeable, carbonate, reducible and oxidizable, is considered to be mobile or environmentally reactive fraction in respect of geological and chemical processes. The more soluble forms of metals are considered to be potentially more bio-available than less soluble forms (Koukina and Vetrov, 2013). In contrast, the residual also called lithogenous or detrital fraction is usually considered to be immobile or environmentally unreactive. It presents metals that are transported in solid form. It is unlikely for residual metals to be released into solution through dissolution and remain fixed in sediments within matrix of silicates and other detrital minerals (Wang *et al.,* 2012).

Environmental and health effects of heavy metals in sediments depend on mobility and bioavailability of metals, which are function of their partitioning with sediments (Guven and Akinci, 2008). Hence, not only the determinant of metal concentrations but also identification of binding sites and phase associations is essential for assessing environmental quality. Thus, to provide reliable information on the forms of association of heavy metals regarding their availability levels and hence potential mobility to food and water sources, a sequential extraction procedure has been largely relied upon to partition heavy metals, such that their chemical reactivity can be elucidated (Nemati *et al.,* 2011).

Poyang Lake, China's largest freshwater lake, receives water from five rivers. Ganjiang River, which is one of the five river catchments and account s for about 45% water of Poyang Lake, located in Jiangxi province, flows north through Lake Poyang to enter the Yangtze River after a course of 815 km. There are more than 300 branches with a total area of 81600 km^2 (Fig. 1). At the same time, this area has large population, and the aquatic ecosystem is challenged by the expanding demands of land use due to local economic development. Therefore, it is necessary to investigate spatial distribution of heavy metals in sediments from the Ganjiang River and assess the risk caused by heavy metals the in order to protect corresponding aquatic ecosystem. The present study aimed at investigating the heavy metal (Cu, Zn, Pb, Cd, Cr, As and Ni) contamination of four areas by calculating index of geo-accumulation (I_{geo}) and comparing with sediment quality values (SQVs) and examing chemical partitioning of heavy metals retained in sediments by using sequential extraction techniques.

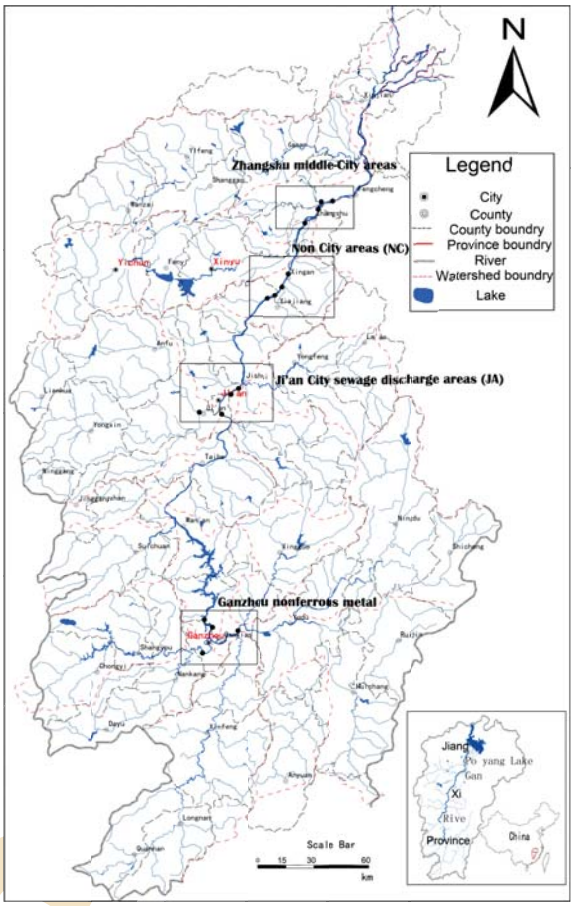

Fig. 1 : Map showing the research area and the geographical location of the sampling sites

Materials and Methods

According the hydrologic, topographic and pollution characteristics, Ganjiang River was subdivided into three sections namely (Jin *et al.,* 2011). Respectively, upper stream of the river was from the source to Ganzhou City with altitudes ranging between 600 m to 1800 m. From Ganzhou City to Xingan county, the river entered the middle stream where both river sides was covered by low hills with altitudes ranging between 200 m to 600 m. Down from Xingan county was the lower reach with man-made banks to prevent the flood flowing downward. The river in this areas were characterized by flat water and the river bed was formed by alluvial plain (Poyang Lake plain) which was heavily affected by the Poyang Lake hydrological processes. Four special sites were selected to represent the whole river: Ganzhou nonferrous metal processing zones (GZ), Jian City sewage discharge areas (JA), non City areas (NC) and Zhangshu middle-City areas (ZS).

The sediment core samples were collected from 4 sites

(GZ, JA, NC and ZS) using a Hydro-Bios stainless-steel grab sampler and wrapped in polyethylene plastic bags during November 2011. All samples were taken from both top horizon (0-20cm deep from the surface) and sub-horizon (20-40cm deep from the surface). At each site, four samples were taken to reflect heavy metal pollution. Samples were left to dry in Petri-dishes at room temperature, ground in a mortar and pestle to grain size, and then dried again at 105 ºC prior to laboratory analyses.

After freeze drying and homogenizing, a certain volume of sediment of each sample (about 0.5 g) was digested twice with 5 ml of HCl: HNO_3: $HClO_4$ 3:1:1 by MLS-1200 MEG high performance microwave digestion, performed as recommended by the manufacturer, for analyses of total concentration and other samples were used for analyses heavy metal partitioning following the method (Tessier *et al.,* 1979).

All the glass vessels used were cleansed with 15% HNO_3, followed by repeated rinsing using de-ionized water before use. Concentration of metals in solutions were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES). Accuracy of analytic determination was established using reference material of GSD-9 and GSD-11, supplied by the Chinese Academy of Geological Sciences. For each batch of 24 samples analyzed, two method blanks and two spiked blanks were processed. The relative standard deviation (RSD) of replicate analysis was < 4.8 %. Regression coefficient of calibration standards of different heavy metal were > 0.999.

The Geo-accumulation index (I_{geo}) is a common criterion to assess heavy metal pollution, and evaluate metal contamination in sediments (Müller, 1969). I_{geo} was calculated by computing the base 2 logarithm of the measured total concentration of the metal over its background concentration using the formula:

$$I_{geo} = \log_2 (C_n/1.5\ B_n)$$

Where C_n is the measured concentration of heavy metal in sediments and B_n is geochemical background concentration of the metal. Factor 1.5 was used as background value of lithological variability.

Results and Discussion

Concentration of heavy metals, including Cu, Zn, Pb, Cd, Cr, As and Ni for Ganzhou nonferrous metal processing zones (GZ), Jian City sewage discharge areas (JA), non City areas (NC) and Zhangshu middle-City areas (ZS) are presented in Table 1. Range (and mean values) of heavy metal concentrations (mg kg^{-1}) in surface sediments of the study area is: Cu, 11.79~131.18 (48.34); Zn, 30.92~290.68 (130.30); Pb, 12.55~128.51 (61.88); Cd, 0.57~99.57 (17.26); Cr, 4.22~53.75 (18.28); As, 0.33~25.58 (7.56); Ni, 12.23~64.75 (31.99). The data obtained from four areas of the Ganjiang River indicate that there was considerable spatial variability between areas within the reach. Highest value of metals was generally found in mud flats being in surface sediments located close to mining industry areas and heavily polluted industrial areas in GZ and ZS. This spatial variability was due to different proximities to contaminant sources and is discussed in detail elsewhere (Wang *et al.,* 2012). The significantly spatial variability of heavy metals indicate great impact of heavy metals by anthropogenic activities, i.e., mining activities, petroleum industry and domestic sewage runoff. It is widely recognized that water ecosystems can are contaminated by trace metals from diverse sources. However, anthropogenic activities such as mining and industrial processing of ores and metals still remain the principal cause of increased amount of heavy metals dumped in to water (DeGregori *et al.,*1996).

As seen in Table 1, the highest value of all heavy metals, excluding Pb, were found at GZ, where large amount of wastewater is produced during mining and beneficiating operations within mounts of nonferrous metal processing enterprises. Increased input of metals in GZ began during 1950s and then continued throughout the end 20th century as a result of increased mining industries and population growth. Highest concentration Pb metal are found in sediments of ZS sampling areas which is heavily polluted by the pharmaceutical and salt chemical processes. However, heavy metal concentration in NC area, where there are several small countries and towns, did not show high metal pollution due to minimum industrial, agricultural and domestic pollution.

Table 1 : Concentration of metals observed in sediments collected from Ganzhou nonferrous metal processing zones (GZ), Jian city sewage discharge areas (JA), Non city areas (NC) and Zhangshu middle-city areas (ZS)

	GZ (μg•g^{-1})		JA (μg•g^{-1})		NC (μg•g^{-1})		ZS (μg•g^{-1})	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Cu	95.94	64.75~131.18	35.23	26.98~39.72	17.94	11.79~28.26	43.60	36.69~53.69
Zn	232.24	181.00~290.68	106.34	94.29~112.37	50.55	30.92~86.83	132.27	127.46~136.38
Pb	84.79	53.84~116.14	27.53	20.61~33.73	21.97	12.55~31.69	113.61	103.52~128.51
Cd	57.48	3.32~99.57	3.98	3.47~5.21	2.94	0.57~5.63	4.50	3.74~5.46
Cr	40.48	20.25~53.75	8.63	6.99~10.62	5.34	4.22~7.12	18.83	16.98~20.43
As	18.09	1.62~25.58	2.94	2.45~3.25	1.16	0.33~1.62	8.02	6.42~10.00
Ni	47.11	29.71~64.75	27.42	25.76~29.08	21.11	12.23~33.39	30.79	26.38~37.73

The background values of heavy metals in Poyang Lake (Table 2) indicated that concentration of heavy metal in surface sediments of this study, excluding Cr, were all higher than their corresponding background values. This was especially true for Cd and Cu, which were 23.02 and 10.18 times of their background values, respectively. Compared with the corresponding background values in sediments from the Ganjiang River, degree of enrichment of 7 heavy metals increased in the following order Ni<Cr<Zn<Pb<As<Cu<Cd.

Although heavy metal concentration studied were slightly lower than that reported in Raohe, another important river located in Poyang Lake Region, which was famous for its heavy metal pollution, affected enormous mining activities (Zeng *et al.*, 2011), sediment heavy metals from Ganjiang River were in relatively severe pollution degree (Table 3) as compared with the published data of other river located in China, especially Cu, Zn, Cd and As (Liu *et al.*, 2002; Zhang *et al.*, 2009).

Degree of heavy metal pollution in sediments can be assessed by determinating Geo-accumulation Index (I_{geo}). Contamination level of heavy metals can be classified into seven grades based on I_{geo} values (class 0: $I_{geo} \leq 0$, uncontaminated (UC); class 1: $I_{geo} \leq 1$, uncontaminated to moderately contaminated (UMC); class 2: $I_{geo} \leq 2$, moderately contaminated (MC); class 3: $I_{geo} \leq 3$, moderately to strongly contaminated (MS); class 4: $I_{geo} \leq 4$, strongly contaminated (SC); class 5: $I_{geo} \leq 5$, strongly to extremely contaminated (SEC); class 6: $I_{geo} > 5$, extremely contaminated (EC) (Förstner *et al.,* 1993; Kumar and Edward, 2009).

Table 4 shows that the range of I_{geo} values measured at different sites varied significantly. Ganjiang River was heavily polluted with Cu, with mean I_{geo} value of 2.34. According to these extremes of I_{geo} values of Cu, sediment samples of GZ could be considered as strongly contaminated. I_{geo} values ≤ 1 (uncontaminated to moderately contaminated) for Zn and AS account for 75%, the remaining 25% are moderately contaminated (≤ 2). Although I_{geo} values ≤ 1 (uncontaminated to moderately contaminated) for Pb account for 50%, I_{geo} values of the remaining 50% were more than 1.8. The I_{geo} values of Cd fell into three classes namely 1, 2 and 5 indicating varying sediment quality and local contamination. Especially GZ sampling areas of Cd fell in the range or class 5, suggesting strongly to extremely contaminated. I_{geo} values for Cr showed that all sampling areas fell in the uncontaminated class ($I_{geo} \leq 0$), which may be described as uncontaminated with respect to these heavy metals.

Furthermore, from Table 4, it could be found that the GZ sampling areas, where large amount of wastewater was produced during mining, beneficiating operations at mounts of Copper Mine factories and came down through branches, had the largest I_{geo} values for all measured metals, which fell into class 4, 2, 3, 5 and 3 respectively. There were not much differences of I_{geo} values for Cu, Zn and As between GA and ZS. However, I_{geo} values of Pb at ZS sampling areas were quite higher than JA. It is known that the pharmaceutical and light industries are the pillar industries for local economy since the 80th of the last century, lot of waste water containing Pb is directly discharged without treatment. The highest I_{geo} value of Cd was 1.80, which was found at the JA sampling areas due to the presence of historic anthropogenic activities, such as industrial wastewater, riverine discharge and domestic sewage (Li *et al.*, 2008). The

Table 2 : Background values and Multiple values in sediments along study area (μg g^{-1})

	Cu	Zn	Pb	Cd	Cr	As	Ni
Mean values	48.34	130.30	61.88	17.26	18.28	7.56	31.99
Background[a]	4.75	45.75	12.5	0.75	29.65	1.37	-
Multiple	10.18	2.85	4.95	23.02	0.62	5.52	-

Wt: wigh, Background[a] : the back ground values of heavy metal in sediments from the Poyang Lake(Gong *et al.*, 2006)

Table 3 : Comparison of heavy metal concentrations in study area with those in other regions (μg g^{-1})

Location	Metal concentration							
	Cu	Zn	Pb	Cd	Cr	As	Ni	Reference
Ganjiang River, China	48.34	130.30	61.88	17.26	18.28	7.56	31.99	This study
Xinjiang West Estuary, China	25.00	87.00	3.00	3.00	8.05	na	na	Lu, 1994
Xinjiang West Estuary, China	100.11	217.08	53.79	4.32	na	na	na	Li *et al.,* 2008
Xinjiang East Estuary, China	290.30	212.80	117.50	2.50	22.14	na	na	Lu, 1994
Roahe Estuary, China	150.13	221.85	115.19	na	na	na	na	Zeng *et al.,* 2011
Nanjishang Reserve in Poyang Lake, China	45.59	105.6	103.4	0.413	na	na	na	Gong *et al.*, 2006
Zhujiang River, China	39.02	110.85	59.43	0.34	56.40	5.24	na	Liu *et al.,* 2002
Changjiang River, China	30.70	94.30	27.30	0.26	78.90	na	31.80	Zhang *et al.*, 2009

Table 4 : Geo-accumulation index (I_{geo}) of heavy metals in sediments

Site/Location	Cu	Zn	Pb	Cd	Cr	As	Ni
			GZ				
I_{geo} (mean values)	3.69	1.73	2.10	4.91	-0.24	2.55	-
I_{geo} class	4	2	3	5	0	3	-
			JA				
I_{geo} (mean values)	2.29	0.63	0.53	1.80	-2.39	0.51	-
I_{geo} class	3	1	1	2	0	1	-
			NC				
I_{geo} (mean values)	1.25	-0.55	0.15	0.98	-3.09	-1.06	-
I_{geo} class	2	0	1	1	0	0	-
			ZS				
I_{geo} (mean values)	2.13	0.42	1.80	1.24	-1.78	0.79	-
I_{geo} class	3	1	2	2	0	1	-

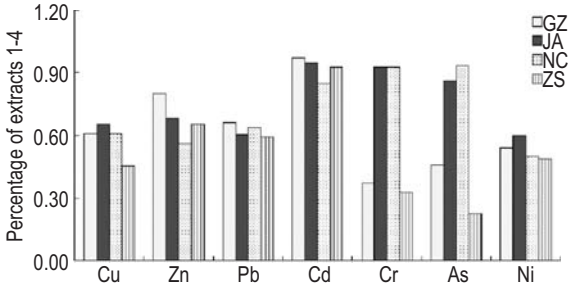

Fig. 2 : Metals extracted from fractions 1-4 as a percentage of total metal concentration

Table 5 : Screening Quick Reference Table for heavy metals in marine sediment ($\mu g \cdot g^{-1}$)

	TEL	ERL	PEL	ERM	AET
Cu	18.70	34.00	108.00	270.00	390.00 (MO)
Zn	124.00	150.00	271.00	410.00	410.00 (I)
Pb	30.24	46.70	112.00	218.00	400.00 (B)
Cd	0.68	1.20	4.21	9.60	3.00 (N)
Cr	52.30	81.00	160.00	370.00	62.00 (N)
As	7.24	8.20	41.60	70.00	35.00 (B)
Ni	15.90	20.90	42.80	51.60	110.00 (EL)

Bioassay endpoints: M - Microtos; O - Oyster larvae; I – Infaunal community impacts; B – Bivalve; N – Neanthes; E- Echinoderm larvae; L – Larval bioassay.

smallest I_{geo} values for all measured metals were calculated at NC sampling areas, where no massive industrial, agricultural and domestic pollution source was present. At this place, the I_{geo} values for Zn, Pb, Cd, Cr and As was > 1. Only Cu could be considered as moderately contaminated with a mean I_{geo} value of 1.25. On the basis of mean values of I_{geo}, degree of heavy metal pollution in the sediments was found in the following order: Cd>Cu>Pb>As>Zn> Cr.

Sediment quality value (SQV) evaluates the degree to which the sediment-associated chemical status might adversely affect aquatic organisms and were designed to assist help in the interpretation of sediment quality (Smith *et al.,* 1996). Here, screening quick reference table (SQUIRT) are introduced to assess whether heavy metals in sediments measured in Ganjiang River would breach these values. The guideline values used in SQUIRT are divided into five increasing categories of observable effects, which have been derived by using several different approaches. Sediment guidelines consist of A threshold effect level (TEL), effects range low (ERL), probable effects level (PEL) , a effects range median (ERM) and apparent effects level (AET) (Long *et al.,* 1995; Zheng *et al.,* 2008).

In the present study, marine sediment values for SQUIRT were introduced. Comparing total metal concentrations in surface sediments to PEL in Table 5, all the sediments tested would lie below PEL except for Cd and Ni in GZ sampling areas, Pb and Cd in ZS sampling areas. The average surfiacial Cd and Ni concentration at GZ was 57.48 and 47.11 $\mu g g^{-1}$ respectively, especially Cd concentration exceed ERM, indicating that any organism living within this sediment would potentially experience adverse biological effects. However, by comparing maximum total metal concentration in the sediments to SQUIRT guidelines in Table 5, sediments from GZ would lie above PEL for Cu, Zn, Pb, Cd and Ni, sediments from JA and NC would lie above PEL for Cd and the maximum concentration of Pb and Cd in ZS would lie above PEL, same for average values of Pb and Cd.

Partitioning data for sediment cores collected from GZ, JA, NC and ZS areas are given in Table 6. In this study, increased mobility was estimated by calculating the percentage of metals that are partitioned in extracts 1-4 (Fig. 2); this may also provide an indication of potential bioavailability. Analysis of the partitioning of metals in these sediments indicates that the percentage of metals collected from extracts 1-4 differs quite small between areas and this may indicate relative differences in

Table 6 : Partitioning of metals (Ganjiang River, μg g^{-1})

	Ext 1	Ext 2	Ext 3	Ext 4	Ext 5
			GZ		
Cu	38.05	5.23	12.53	2.34	37.81
Zn	89.71	8.98	52.22	34.54	46.78
Pb	21.48	2.89	27.62	4.08	28.72
Cd	45.06	1.33	6.67	2.84	1.60
Cr	1.07	0.83	10.51	2.78	25.30
As	1.70	0.55	5.04	0.99	9.81
Ni	11.74	1.31	6.20	6.17	21.69
			JA		
Cu	13.60	1.08	7.07	1.16	12.32
Zn	34.58	3.52	23.53	10.75	33.97
Pb	2.63	2.55	11.14	0.30	10.92
Cd	2.33	0.11	1.01	0.32	0.21
Cr	0.41	0.59	5.52	1.48	0.63
As	0.17	0.04	1.97	0.35	0.41
Ni	4.35	0.72	5.72	5.65	10.99
			NC		
Cu	5.83	0.40	4.21	0.42	7.09
Zn	12.35	1.57	8.36	6.13	22.15
Pb	2.13	1.01	10.55	0.23	8.04
Cd	1.59	0.03	0.65	0.23	0.44
Cr	0.19	0.39	3.99	0.38	0.40
As	0.01	0.02	0.96	0.11	0.07
Ni	2.80	0.65	2.39	4.76	10.52
			ZS		
Cu	8.24	2.40	8.54	0.91	24.16
Zn	51.07	4.51	21.62	9.16	45.71
Pb	4.98	2.67	58.89	0.35	46.34
Cd	2.41	0.07	1.09	0.30	0.32
Cr	0.25	0.22	4.42	1.23	12.53
As	0.06	0.03	1.37	0.34	6.26
Ni	3.75	0.89	5.19	5.94	16.56

bioavailability. Percentage "potential bioavailability" of partitioned fractions 1-4 for Zn, Pb, Cd, Cr and As in sediments collected from most sites are more than 60% of the total metals (Fig. 2).

By examining the concentration in the bioavailable fractions and comparing these values with the standards listed in SQUIRT, it is was observed that the concentration of Cd at all sampling areas were higher than ERL. The concentration in bioavailable fractions of Cd at JA sampling area was greater than ERL, Zn, Pb, Cr and As was less than TEL. Similar results were found at ZS, where the bioavailable fraction of Zn, Cr, As and Ni was less than TEL. However, the concentration in bioavailable fractions of Cu, Zn, Pb, Cd, AS and Ni at GZ sampling area greater than ERL. Long *et al.* (1995) it has been published that the incidence of effects increased about 10% for most trace metals when concentrations exceed ERL values but were lower than the effects range median (ERM). Furthermore, the greatest risk to biota appeared at GZ, where the concentration in bio available fractions of Cd was above PEL at which adverse effect were observed. It suggests that a full environmental risk assessment have been performed.

The results showed that heavy metal concentration showed significant spatial variation in surface sediments and most of the studied areas were heavily polluted characterized by high accumulation of Cd, Cu, As, Zn and Pb. Contamination assessment, based on Geo-accumulation Index (I_{geo}), showed that metals were found in the following uncontaminated to extremely contaminated degrees, and were in the decreasing order of : Cd>Cu>Pb>As>Zn>Cr.

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