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ORIGINAL ARTICLE

Contamination, toxicity and risk assessment of heavy metals and metalloids in sediments of Shahid Rajaie Dam, Sefidrood and Shirinrood Rivers, Iran

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Abstract In this paper, the heavy metals and metalloids (As, Cd, Co, Cr, Ni, Cu, Pb and Zn) levels were studied to investigate their contamination and ecological effects in surface sediments of Shahid Rajaie Dam Lake and two major upstream rivers. Contamination factor and modified degree of contamination values indicate a very low to considerable contamination effects for the studied sediments while pollution load index indicates no pollution (except five stations). The comparison of selected elements concentrations with sediment quality guidelines reveal that the average concentration of As, Cr and Ni in the present sediments is higher than threshold effect level. In addition, Ni shows higher concentration than probable effect level and effect range low values. These sediments based on PELQ and ERMQ calculations, for Cr, As, Cd, Zn, Cu, Ni and Pb are slightly toxic. The result of enrichment factor evaluation similarity to principal component analysis indicates that the main source of Pb, Cd, and Zn for 15.38–42.3 percentages of samples is anthropogenic. PC1 agrees with the measured enrichment factor suggesting a geogenic source for Cu, Mn, As, Cr, Al, Co, Ni and Sc in the sediment samples. Higher positive loadings of P with Ni and Co for a small number of sediment probably indicate amplified concentration due to anthropogenic sources such as application of phosphorus fertilizers in the agricultural lands. High positive loading of Pb, Zn and Cd with organic carbon (OC) and clay reveals a significant role of OC and clay in the dissolution and distribution of Pb, Zn and Cd in the dam's lake sediments. In summary, the present study has provides a practical baseline data for the long-term monitoring of heavy metals pollution in the study area.

Keywords Sediment contamination · Pollution load index · Ecological risk assessment · Shahid Rajaie Dam · Iran

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Introduction

The pollution of natural environment by heavy metals and metalloids is a topic of much discussion recently (Barakat et al. 2012; Xiao et al. 2013; Fujita et al. 2014; Kukrer et al. 2014). The issue of toxic elements pollution in water and sediment of lakes and rivers has received much more attention from many environmental researchers over the past few decades. The rapid development of industry and agriculture has resulted in increasing pollution by potentially toxic elements, which are harmful to the natural environment due to their toxicity, persistence and bioaccumulation (Uluturhan and Kucuksezgin 2007; Varol and Sen 2012). Inputs of pollutants, particularly of toxic metals and metalloids to the aquatic environment are increasingly worrying (Peng et al. 2009). Metals such as copper, iron, chromium and nickel are essential since they play an important role in biological systems, whereas cadmium and lead are non-essential metals, as they are toxic, even in trace amounts (Fernandes et al. 2008). Elements such as mercury, cadmium, arsenic, lead and chromium are dangerous to human health (Ouyang et al. 2002; Hogan 2010).

Pollutants concentrations in aquatic ecosystems are usually monitored by measuring their concentrations in water, sediments and biota (Kang et al. 2001; Zhang et al. 2002). Sediments can reflect the quality of water system, and can be used to detect the insoluble contaminants in water. Their capacity to accumulate contaminants is an important factor to assess the environmental impact on aquatic ecosystems (Silva and Rezende 2002; Joksimovic et al. 2011; Nasehi et al. 2013). Depending on the environmental conditions, heavy metals and metalloids tend to adsorb from water of fine particles and usually move thereafter with the sediments, and can affect the organisms and food chain if toxic levels are reached, resulting in health risk (Saha and Zaman 2013).

Agricultural lands cover a large part of the river basins and application of mineral fertilizers, manure and pesticides release toxic metals and metalloids in the ecosystem (Kabata-Pendias and Mukherjee 2007).

The Tajan River is one of the major rivers for water supply in north of Iran and Shahid Rajaie Dam was constructed on it at 1997 (Fig. 1). The demand for water in the domestic, industrial, and agricultural sectors is steadily increasing due to developments, population growth as well as the demand for a sufficient food supply. Considering the potential for direct discharge of organic and inorganic contaminants into the Shahid Rajaie dam reservoir and upstream (Sefidrood and Shirinrood Rivers); no attempt has been made for monitoring the content of heavy metals and metalloids in sediments of Dam Lake, Sefidrood and Shirinrood Rivers. Catchment of the study area is used for food and agricultural industries and tourism. A major road with high traffic load, especially, in the weekend, passes from the vicinity of Shahid Rajaie Dam and Shirinrood Rivers in the upstream. The Shahid Rajaie Dam receives discharges from agricultural land wastewater (approximately 1500 ha, paddy field), traffic related pollutants and domestic and livestock wastewater. The other issue is lack of waste management in the neighboring cities and villages, using many small unengineered landfills and direct swage discharge to upstream rivers.

Therefore, the main goals of present study are (1) determination of the distribution and concentrations of Cd, Cu, Cr, Zn, Ni, Pb, As and Co in the surface sediments of Shahid Rajaie Dam and upstream; (2) evaluating degree of contamination and pollution load index (PLI); (3) study the ecological effects using sediment quality guidelines and

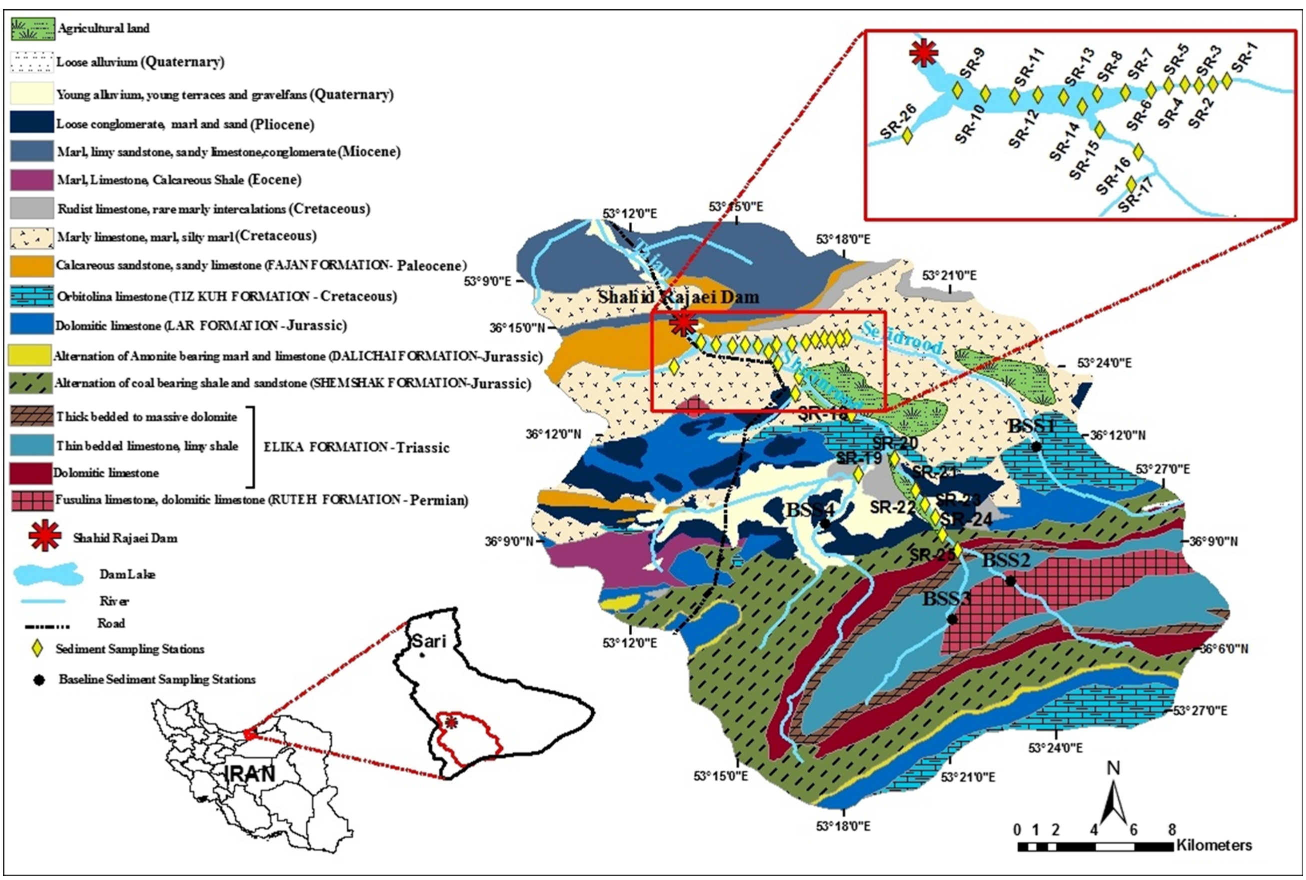

Fig. 1 Geological map of the study area and sample location

potential ecological risk and (4) identifying the anthropogenic or natural sources of elements.

Materials and methods

Study area and sample collection

Shahid Rajaie Dam is located in 40 km south of the Sari City, in the northern part of Iran (Fig. 1) with 160 million cubic meters capacity and approximate catchment of 1244 km^2. It is constructed on Tajan River. It was designed to provide irrigation, drinking, and industrial water in the region. The main activities in this area are agriculture, crop irrigation, and dairy activities. The main human settlements are in upstream including Ferim, Afrachal, Ali-Abad, Sekuya, Damad Kola villages with a total of more than 10,000 habitants. Average temperature in the basin is 12 °C and average annual precipitation is about 650 mm. Geological formations from the oldest to youngest are Ruteh, Elika, Shemshak, Dalichai, Lar, Tizkuh and Fajan formations. The main lithologic units include marl, limestone, dolomitic limestone, shale and sandstone (Fig. 1). Figure 1 displays the location of sampling stations.

About 1 kg of sediment samples were collected in October 2012 from 26 sites of Shahid Rajaie Dam using a pre-cleaned stainless steel grab sampler (10 cm), Sefidrood and Shirinrood Rivers using plastic scoop (10 cm). Finding suitable unpolluted samples for background determinations proved to be difficult at the study area, which has a long history of agricultural practices and animal husbandry. However, four sediment samples believed to be unaffected by anthropogenic activity and representative of local lithological unites, were chosen as local baseline/background in the upstream (Fig. 1).

Analytical methods

The collected samples were immediately stored in polyethylene bags and air-dried in the laboratory at room temperature. Then, gravel and plant root were removed samples passed through a 2 mm stainless steel sieve. Grain size plays a significant role in determining the elemental concentrations in sediments (Szefer et al. 1996). Salomons and Forstner (1984) recommended a particle size fraction of <63 μm for analysis because they thought it was most nearly equivalent to materials carried in suspension, the most important system for transport of fine sediments.

The <2 mm fraction was ground in an agate mortar and pestle and passed through a 63 μm sieve. Physicochemical properties of the sediment samples were measured using standard analytical methods. Organic carbon content was determined using Gaudette et al. (1974) titration method. Sediment pH was determined through equilibration with a homogenized suspension of 10 g of sample with 50 ml of distilled water after shaking for 5 min, and 1 h pause using a calibrated ELE pH meter. To determine the concentration of metals and metalloids, complete dissolution of sediment samples (approximately 1 g of each) was carried out using a mixture of HF, HNO_3, $HClO_4$ and H_2O_2 in a Teflon beaker on sand bath at atmospheric pressure. The concentrations of As, Cd, Co, Cr, Ni, Cu, Pb, Sc, Al, Mn, Pb and Zn were measured by an accredited commercial laboratory (ZarAzma Laboratory, Iran) using ICP-MS methods (Agilent, 7700x, USA). Data quality was ensured through the use of internal duplicates, blanks, and HRM. The precision and accuracy of measurements are 95 and ±5 %, respectively.

Statistical analysis

Multivariate statistical approaches such as principal component analysis (PCA) have been used by researchers for deriving the significance of specific parameters that lead to sediment metals and metalloids enrichment (in the present study) among the data generated (Shirodkar et al. 2009; Shakeri and Moore 2010; Kukrer et al. 2014). PCA has been applied for determining the degree of pollution by heavy metals and metalloids from lithogenic action and anthropogenic sources (Sun et al. 2010). After the application of PCA, a varimax normalized rotation was applied to minimize the variances of the factor loadings across variables for each factor. These analyses were conducted by using the IBM/SPSS package. The number of significant principal components is selected based on the Kaiser criterion with eigenvalues higher than one (Kaiser 1960).

Pollution indicators and potential ecological risk

There are several methods to assess sediment quality and describe the contamination adverse effects (Ridgway and Shimmield 2002). The assessment of sediment contamination was carried out using indexes such as the enrichment factor (EF), contamination factor (C_f) and modified degree of contamination (mC_d), sediment quality guidelines (SQGs), PLI and risk index (RI).

To evaluate natural or anthropogenic sources of heavy metals and metalloids in sediment, the enrichment factor was calculated for sediment samples (Zhang et al. 2007) using conservative elements such as Al, Fe, Sc and Ti (Lee et al. 1998; Reimann and de Caritat 2000; Bergamaschi et al. 2002; Hernandez et al. 2003; Mishra et al. 2004; Abrahim and Parker 2008, Shakeri and Moore 2010) as reference elements.

The reference values were adopted from the baseline concentration of heavy metals and metalloids using following equation:

$$\mathrm{EF} = \frac{[\mathrm{M}]/[\mathrm{Sc}]_{\mathrm{sediment}}}{[\mathrm{M}]/[\mathrm{Sc}]_{\mathrm{baseline}}}$$

where [M] = total heavy metals and metalloids concentrations measured in sediment samples (mg/kg) and [Sc] = total concentration of scandium as the reference element (mg/kg). According to Hernandez et al. (2003), EF values between 0.5 and 2 can be considered in the range of natural variation, whereas ratios greater than 2 indicate some enrichment corresponding mainly to anthropogenic inputs. Also, five categories are recognized on the basis of enrichment factor (Sutherland 2000; Loska and Wiechuya 2003) (Table 1).

Hakanson (1980) proposed an overall indicator of contamination based on integrating data for a series of seven specific potentially toxic elements and the organic pollutant polychlorinated biphenyl. This method is based on the calculation of a C_f for each pollutant. The individual contamination factors are calculated according to the following equation:

$$C_\mathrm{f}^i = \frac{M_x^i}{M_b^i}$$

where M_x and M_b, respectively, refer to the mean concentration of each metal in the sediment and the baseline or background value (concentration in unpolluted sediment). C_f^i is defined according to four categories as Table 2 (Liu et al. 2005).

Hakanson (1980) study suggests that the numeric sum of all the aforementioned contamination factors should express the overall degree of contamination in the sediment by using the following formula:

$$C_d = \sum_{i=1}^{n} C_\mathrm{f}^i$$

where C_f^i are the individual contamination factors for the selected element and n is the number of the C_fs examined for specific sediment. The degree of contamination defines the quality of the environment in the following way: $C_d < 8,\ 8 < C_d < 16,\ 16 < C_d < 32,\ C_d > 32$ indicates low, moderate, considerable, and very high degree of contamination accordingly.

Abrahim (2005) presented a modified and generalized form of the Hakanson (1980) equation for the calculation of the overall degree of contamination using following equation:

$$mC_d = \frac{\sum_{i=1}^{n} C_\mathrm{f}^i}{n}$$

where n is the number of analyzed elements, i the element and C_f the contamination factor.

For the classification of the sediments according to the mC_d gradations are proposed by Abrahim and Parker (2008) (Table 3).

Sediment quality guidelines (SQGs) have been used as an interpretive tools for assessing the biological significance of individual chemicals (Mucha et al. 2003). Two sets of guidelines are commonly used; the effects range low/median (ERL/ERM) and threshold/probable effect level (TEL/PEL). The low range values (ERL or TEL) have been estimated as the concentration of contaminants with a relatively low effect on biological communities.

Table 1 Classification of enrichment factor (Sutherland 2000; Loska and Wiechuya 2003)

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 2 Gradations of contamination factor (Liu et al. 2005)

$C_\mathrm{f}^i < 1$	Low contamination
$1 < C_\mathrm{f}^i < 3$	Moderate contamination
$3 < C_\mathrm{f}^i < 6$	Considerable contamination
$C_\mathrm{f}^i > 6$	Very high contamination

Table 3 Gradations of modified degree of contamination (Abrahim and Parker 2008)

$mC_d < 1.5$	Nill to very low degree of contamination
$1.5 \leq mC_d < 2$	Low degree of contamination
$2 \leq mC_d < 4$	Moderate degree of contamination
$4 \leq mC_d < 8$	High degree of contamination
$8 \leq mC_d < 16$	Very high degree of contamination
$16 \leq mC_d < 32$	Extremely high degree of contamination
$mC_d \geq 32$	Ultra high degree of contamination

Table 4 Degrees of contamination and potential ecological risk corresponding to RI and E_r^i values (Hakanson 1980)

Range	Degree
E_r^i	
<40	Low
40–80	Moderate
80–160	High
160–320	Very high
>320	Dangerous
RI	
<150	Low
150–300	Moderate
300–600	High
>600	Very high

Table 5 Total concentration and descriptive statistics of heavy metals and metalloids (mg/kg) along with some physico-chemical properties of sediment samples

Sample No.	Al	As	Cd	Co	Cr	Cu	Ni	Zn	Pb	Mn	P	Sc	pH	OC %	Sand %	Clay %	Silt %
Detection limit	100.0	0.1	0.1	1.0	1.0	1.0	1.0	1.0	1.0	5.0	10.0	0.5	–	–	–	–	–
Sefidrood																	
SR-1	59,047	9.2	0.50	15.0	74.0	26	39.0	81.0	14.0	729	585	9.3	7.19	1.08	24.2	16.1	59.7
SR-2	51,856	7.8	0.30	13.5	63.0	19	31.0	66.0	12.0	548	525	7.2	7.21	1.26	37.8	8.9	53.3
SR-3	48,940	7.5	0.20	15.2	60.7	16	39.5	70.4	25.3	484	599	7.2	7.97	1.12	23.5	15.6	60.9
SR-4	63,968	8.4	0.30	15.5	75.0	25	38.0	78.0	16.0	713	548	9.0	7.21	1.10	8.6	21.7	69.7
SR-5	67,021	9.0	0.40	16.3	72.0	29	42.0	89.0	17.0	779	565	9.5	7.30	1.05	6.6	28.9	64.5
SR-6	65,240	7.3	0.35	19.9	81.1	27	55.3	96.5	32.6	658	712	10.5	7.66	1.17	12.9	22.1	65.0
Dam's Lake																	
SR-7	68,927	7.0	0.50	16.8	74.0	30	42.0	88.0	17.0	712	605	9.7	7.16	1.75	12.6	21.7	65.7
SR-8	56,816	7.1	0.40	13.6	69.0	21	32.0	71.0	11.0	588	530	7.5	7.21	0.74	24.2	10.9	64.9
SR-9	54,790	7.4	0.34	17.2	65.2	19	45.4	78.2	18.7	541	669	8.1	7.98	1.29	16.6	18.2	65.2
SR-10	74,099	10.8	0.30	17.1	78.0	29	46.0	93.0	17.0	780	621	10.9	7.11	1.35	2.6	40.1	57.3
SR-11	43,068	5.7	0.80	12.0	58.0	20	28.0	143.0	65.0	722	495	6.5	6.79	1.84	14.6	32.9	52.5
SR-12	41,070	7.1	0.60	13.6	52.5	17	35.4	146.0	70.1	639	610	6.7	6.91	1.38	14.1	24.8	61.1
SR-13	52,301	8.2	0.70	14.0	67.0	24	33.0	148.0	74.0	841	565	8.0	7.02	1.98	0.6	66.9	32.5
SR-14	70,035	8.8	0.50	16.7	76.0	33	44.0	161.0	65.0	767	622	11.2	6.84	2.19	11.2	50.9	37.9
SR-15	65,300	7.5	0.40	19.6	79.7	30	57.3	169.7	71.6	683	744	11.6	7.04	1.49	13.6	40.5	46.0
Shirinrood																	
SR-16	59,946	8.4	0.50	15.1	72.0	29	41.0	140.0	110.0	851	609	10.0	7.75	3.13	28.4	19.3	52.3
SR-17	42,587	2.3	0.40	11.9	76.0	17	33.0	59.0	11.0	378	552	7.5	7.15	1.35	50.0	13.7	36.3
SR-18	38,620	7.0	0.20	17.3	79.9	11	42.3	61.4	18.6	352	683	6.9	8.12	1.45	30.7	24.5	44.9
SR-19	42,086	6.4	0.40	11.1	52.0	18	27.0	108.0	11.0	616	452	6.9	7.34	1.17	69.2	18.1	12.7
SR-20	14560	7.1	0.25	11.1	46.6	8	29.1	48.8	28.1	364	585	4.7	8.14	1.45	66.4	15.6	18.0
SR-21	53,706	9.5	0.50	16.1	80.0	27	37.0	122.0	22.0	705	567	9.0	7.28	0.68	48.0	24.5	27.5
SR-22	40,790	7.3	0.41	13.8	47.9	16	32.1	114.3	22.8	565	502	6.2	8.04	1.39	49.3	22.4	28.4
SR-23	52,694	6.9	0.30	15.5	73.0	26	32.0	102.0	21.0	744	564	8.0	7.20	0.72	80.0	15.1	4.9
SR-24	46,786	10.1	0.50	12.5	80.0	23	34.0	120.0	48.0	681	577	7.6	7.11	1.14	72.8	8.1	19.1
SR-25	47,820	5.1	0.38	17.2	61.9	24	41.7	107.0	31.4	672	688	7.7	7.93	1.48	59.1	20.7	20.3
SR-26	25,377	3.6	0.30	8.5	57.0	13	27.0	45.0	22.0	390	522	4.8	7.17	1.10	70.8	11.7	17.5
Mean	51,825	7.4	0.41	14.9	68.1	22	37.8	100.2	33.5	635	588	8.2	7.38	1.38	32.6	23.6	43.8
Minimum	14,560	2.3	0.20	8.5	46.6	8.4	27.0	45.0	11.0	352	452	4.7	6.79	0.68	0.6	8.1	4.9
Maximum	74,099	10.8	0.80	19.9	81.1	33	57.31	169.7	110	851	744	11.64	8.14	3.13	80.0	66.9	69.7
W.M.S[a]	72,000	8	0.17	14	72	33	52	95	19	–	–	–	–	–	–	–	–
Shale average[b]	80,000	13	0.30	19	90	45	68	95	20	–	–	–	–	–	–	–	–
Upper crust[c]	–	2	0.10	17	85	25	20	71	20	–	–	–	–	–	–	–	–
Baseline	48,494	7.4	0.3	14.8	80.0	22.1	34.9	81.0	18.0	–	–	–	–	–	–	–	–

[a] Bowen (1979)

[b] Turekian and Wedepohl (1961)

[c] Rudnick and Fountain (1995)

Conversely, ERM and PEL values represent chemical concentrations above which adverse effects are likely to occur (Long and MacDonald 1998).

To obtain a more realistic measure of the sediments' toxicity, mean quotients were introduced according to the following equations:

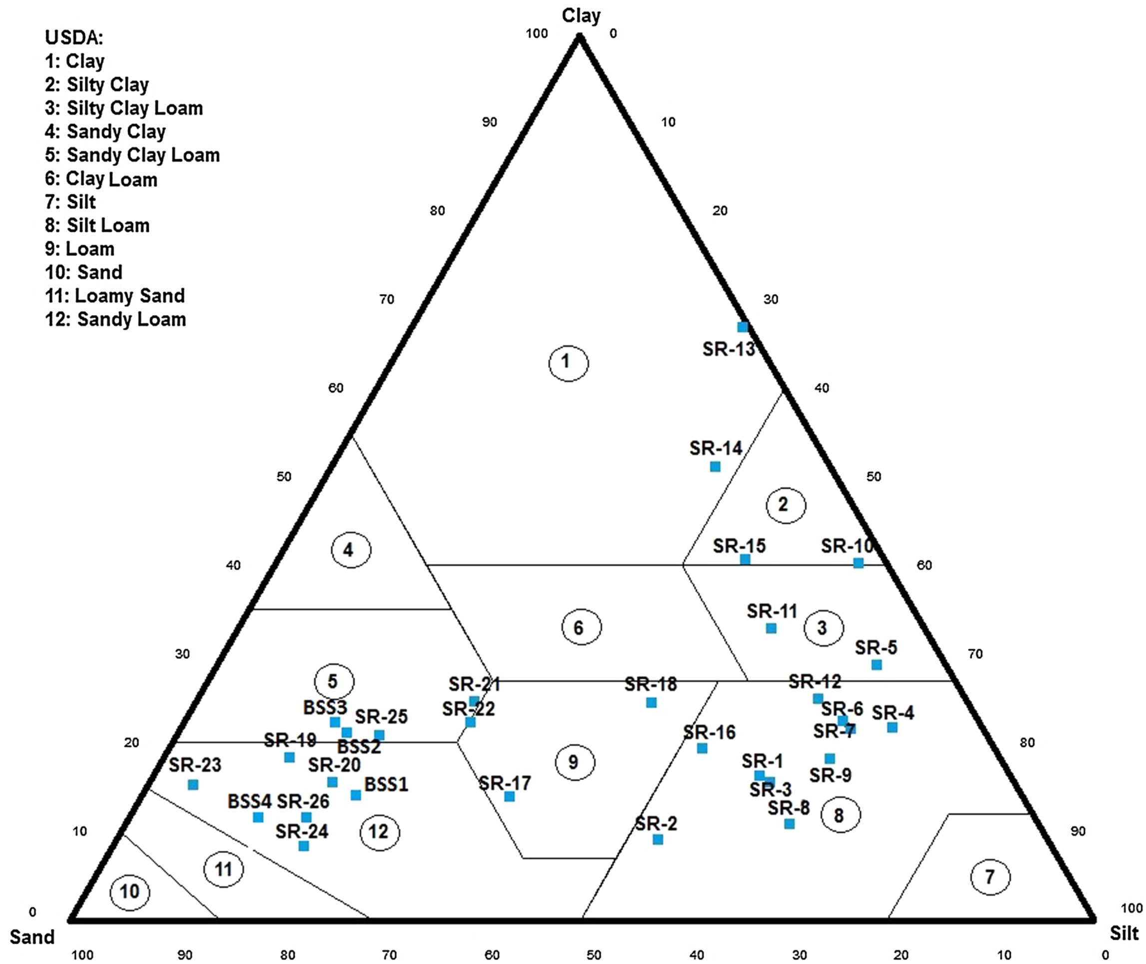

Fig. 2 Ternary diagram showing sediment texture

$$\mathrm{PELQ} = \frac{\sum_{i=1}^{n} M_i/\mathrm{PEL}_i}{n},$$

$$\mathrm{ERMQ} = \frac{\sum_{i=1}^{n} M_i/\mathrm{ERM}_i}{n}$$

where the PELQ and ERMQ factors are the average ratios between the heavy metal concentration in the sediment sample (M_i) and the related PEL and ERM values for the element i (PEL_i, ERM_i) and n is the number of metals. These factors describe the sediment contamination range as non-toxic (PELQ and ERMQ <0.1), slightly toxic ($0.1 < \mathrm{PELQ} < 1.5$ and $0.1 < \mathrm{ERMQ} < 0.5$), moderately toxic ($1.5 < \mathrm{PELQ} < 2.3$ and $0.5 < \mathrm{ERMQ} < 1.5$) and heavily toxic ($\mathrm{PELQ} > 2.3$ and $\mathrm{ERMQ} > 1.5$) (MacDonald et al. 2004; Leorri et al. 2008).

For determining environmental quality of the sediment, an integrated pollution load index (PLI) was used, according to Suresh et al. (2011), using following equation:

$$\mathrm{PLI} = (Cf_1 \times Cf_2 \times \cdots \times Cf_n)^{1/n}$$

where n is the number of metals ($n = 8$ in this study) and C_f is the contamination factor.

Values of PLI > 1 imply that heavy metal pollution exists. Pollution load index <1 indicates no heavy metal pollution (Tomlinson et al. 1980).

The potential ecological risk index (RI) was used to evaluate the toxicity of metals in the sediment (Hakanson 1980). According to this method, the potential ecological risk factor (E_r^i) of single element and the potential ecological RI of multi-element can be computed by the following equation:

$$\mathrm{RI} = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} Cf^i \times T_r^i$$

where Cf^i is the contamination factor for the element ''i''; T_r^i is the toxic response factor for the given element of ''i'', which accounts for the toxic and the sensitivity requirements. The toxic response factors for Pb, Cd, Cr, Cu, Zn, As and Ni were 5, 30, 2, 5, 1, 10 and 5, respectively. The relationship between RI, E_r^i and pollution levels is given in Table 4.

Table 6 Principal component analysis for experimented variables in the sediment samples

Rotated component matrix			
	PC1	PC2	PC3
Al	0.94	0.10	0.14
Cu	0.91	0.30	0.01
Sc	0.91	0.22	0.25
Cr	0.77	−0.09	0.27
Mn	0.75	0.51	−0.24
As	0.72	0.09	0.02
Pb	0.02	0.93	0.07
Zn	0.34	0.83	−0.04
OC	−0.06	0.80	0.12
Cd	0.15	0.72	−0.49
Clay	0.31	0.70	0.10
P	0.25	0.17	0.90
Ni	0.67	0.14	0.70
pH	−0.42	−0.32	0.58
Co	0.68	0.08	0.71
% of variance	36.26	24.93	19.47
Cumulative %	36.26	61.18	80.65

Rotation Method: Varimax with Kaiser Normalization

Results and discussion

Descriptive statistics of selected heavy metals and metalloids, pH, organic carbon and percentage of sand, silt, and clay in sediments along with reference values of elements in average shale, world mean sediments (WMS) and upper continental crust (Turekian and Wedepohl 1961; Bowen 1979; Rudnick and Fountain 1995) are summarized in Table 5. The average abundance order of heavy metals and metalloids contents in sediment samples is Zn > Cr > Ni > Pb > Cu > Co > As > Cd. Comparison of mean concentration of the heavy metals and metalloids in sediments with the reference values reveals that Co, Cd, Pb and Zn have higher contents than the WMS. Cadmium, Pb and Zn indicating higher concentration than the average shale values and Co, Cd, Ni, As, Pb and Zn have higher contents than the upper continental crust values.

Table 5 shows values of sediment pH vary between 6.79 and 8.14. Average pH in sediment samples is 7.38. Organic carbon values are in range of 0.68 % (SR-21 station) to 3.13 % (SR-16 station). The dominant observed textural classes are silt loam, sandy loam, loam and clay, respectively (Fig. 2).

Principal component analysis

Principal component analysis was applied to reveal structure in data, which helps in finding relationship between sampling sites and elements. Results of factor analysis for selected elements (As, Cd, Co, Cr, Ni, Cu, Pb and Zn) along with Mn, Al, Sc, P, clay, pH and OC data at the sediment samples are tabulated in Table 6. Role of each sample on the factor analysis loadings is shown in Fig. 3. Table 6 represents three factors retained in the analysis and account for 80.65 % of variance in sediment samples. The first component, with 36.26 % of the total variance, has high and strong positive loadings of Al, Cu, Sc, Cr, Mn, As, Co and Ni. The strong association of these elements in sediments suggests a common source, showing a lithogenic control. Figure 3 indicates that the high loading of PC1 is corresponded to Sefidrood River (except SR-3), dam's lake (SR-7, SR-10 and SR-14) and Shirinrood River (SR-21, SR-23 and SR-24) samples. The second component,

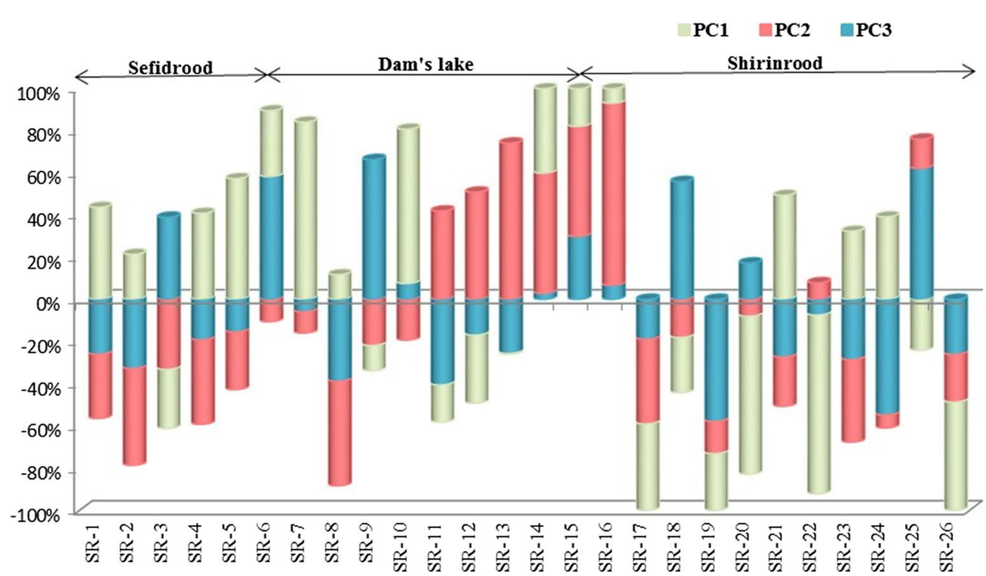

Fig. 3 Role of each samples on the factor analysis loadings

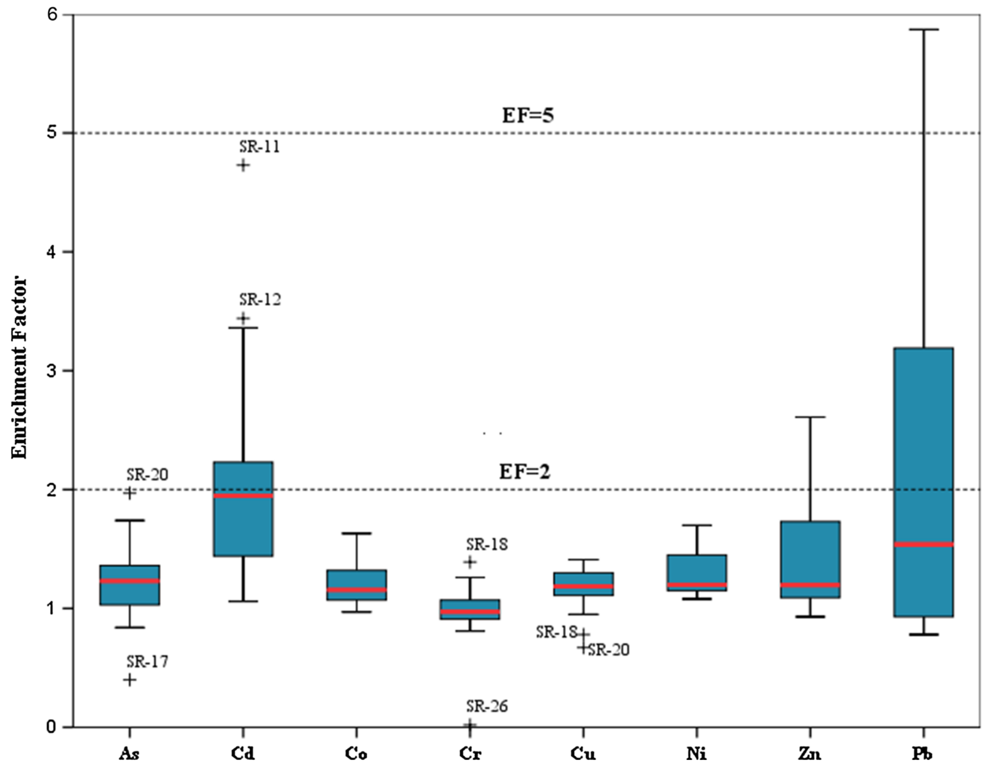

Fig. 4 *Box plot* of the enrichment factor of selected elements in the sediment samples. The *red line* is the median of data

explaining 24.93 % of the total variance, showed high positive loading of Pb, Zn, Cd, OC and clay. High positive loading of Pb, Zn and Cd with OC and clay (0.70 > PC3 > 0.93) reveals a significant role of OC and clay in the dissolution and distribution of these elements in the dam's lake sediments (Fig. 3). The third component with 19.47 % of the total variance has a high factor loading of P, Ni and Co. High positive loadings of P with Ni and Co for a small number of sediment samples (SR-3, SR-5, SR-9, SR-18 and SR-25) probably indicate amplified concentrations due to anthropogenic sources such as application of phosphorus fertilizers in the agricultural lands; especially paddy field, ending up to the Tajan River and Shahid Rajaie Dam reservoir. Gimeno-Garcia et al. (1996) examined the role of mineral fertilizers and pesticides application on heavy metals concentrations such as Ni and Co in paddy soils.

Enrichment Factor (EF)

Results of EF calculations based on local background for Shahid Rajaie Dam (SR-7 to SR-15), Sefidrood (SR-1 to SR-7) and Shirinrood (SR-16 to SR-26) rivers sediment samples can be categorized as follows (Fig. 4); deficient to low enriched As, Co, Ni, Cu and Cr, moderately enriched Cd, Zn and Pb for 15.38 to 42.3 % of samples and significant enriched Pb (SR-11, SR-12, SR-16). Mean EF order in sediment samples is Pb > Cd > Zn > Ni > As, Co > Cu > Cr. The results of EF evaluation indicate that Pb, Cd and Zn have more influence from anthropogenic sources such as paddy fields (Fig. 1), local unengineered landfills and sewage. Zinc and Pb concentration also in sediments of the study area can be affected by vehicles, especially to wearing out of tires and lead from gasoline, which is regarded as the main source of it in the environment (Callender and Karen 2000; Gallon et al. 2005; McKenzie et al. 2009; Fujiwara et al. 2011). Burning of wood/coal in cooking and fossil fuels for home heating by inhabitants of the upstream villages probably could be the other important sources of Pb contamination in the sediment samples. Also use of plastic containers, metal products (e.g., brass, bronze, castings, and galvanized metal) and cadmium batteries in the study area by local residents, farmers and tourism can release Pb, Zn and Cd in the surface sediment of Sefidrood and Shirinrood Rivers. In other similar environments in the world, markedly higher concentrations of potentially toxic elements were observed in the surface sediments by Kucuksezgin et al. 2008 (Gediz River), Fang et al. 2009 (East China Sea), Liu et al. 2012 (Nanfei River mouth), TavakolySany et al. 2013 (coastal sediments of Port Klang, Selangor, Malaysia), in which Zn, Pb and Cd exhibited moderately to severe enrichment factor.

Table 7 Contamination factors (C_f), degree of contamination (C_d) and modified degree of contamination (mC_d) of heavy metals and metalloids in the sediments

Sample No.	C_f								C_d	mC_d
	As	Cd	Co	Cr	Cu	Ni	Zn	Pb		
Sefidrood										
SR-1	0.92	1.7	0.9	0.9	0.7	0.8	1.0	0.8	7.7	1.0
SR-2	0.78	1.0	0.8	0.8	0.5	0.6	0.8	0.7	6.0	0.8
SR-3	0.75	0.7	0.9	0.8	0.5	0.8	0.9	1.4	6.6	0.8
SR-4	0.84	1.0	0.9	0.9	0.7	0.8	1.0	0.9	7.0	0.9
SR-5	0.90	1.3	1.0	0.9	0.8	0.8	1.1	0.9	7.8	1.0
SR-6	0.73	1.2	1.2	1.0	0.8	1.1	1.2	1.8	9.0	1.1
Dam's Lake										
SR-7	0.70	1.7	1.0	0.9	0.9	0.8	1.1	0.9	8.0	1.0
SR-8	0.71	1.3	0.8	0.9	0.6	0.6	0.9	0.6	6.4	0.8
SR-9	0.74	1.1	1.0	0.8	0.5	0.9	1.0	1.0	7.2	0.9
SR-10	1.08	1.0	1.0	1.0	0.8	0.9	1.1	0.9	7.9	1.0
SR-11	0.57	2.7	0.7	0.7	0.6	0.6	1.8	3.6	11.2	1.4
SR-12	0.71	2.0	0.8	0.7	0.5	0.7	1.8	3.9	11.1	1.4
SR-13	0.82	2.3	0.8	0.8	0.7	0.7	1.8	4.1	12.1	1.5
SR-14	0.88	1.7	1.0	1.0	0.9	0.9	2.0	3.6	11.9	1.5
SR-15	0.75	1.3	1.2	1.0	0.9	1.1	2.1	4.0	12.3	1.5
Shirinrood										
SR-16	0.84	1.7	0.9	0.9	0.8	0.8	1.7	6.1	13.8	1.7
SR-17	0.23	1.3	0.7	1.0	0.5	0.7	0.7	0.6	5.7	0.7
SR-18	0.70	0.7	1.0	1.0	0.3	0.8	0.8	1.0	6.3	0.8
SR-19	0.64	1.3	0.7	0.7	0.5	0.5	1.3	0.6	6.3	0.8
SR-20	0.71	0.8	0.7	0.6	0.2	0.6	0.6	1.6	5.8	0.7
SR-21	0.95	1.7	0.9	1.0	0.8	0.7	1.5	1.2	8.8	1.1
SR-22	0.73	1.4	0.8	0.6	0.5	0.6	1.4	1.3	7.3	0.9
SR-23	0.69	1.0	0.9	0.9	0.7	0.6	1.3	1.2	7.3	0.9
SR-24	1.01	1.7	0.7	1.0	0.7	0.7	1.5	2.7	9.9	1.2
SR-25	0.51	1.3	1.0	0.8	0.7	0.8	1.3	1.7	8.1	1.0
SR-26	0.36	1.0	0.5	0.7	0.4	0.5	0.6	1.2	5.3	0.7
Mean	0.74	1.4	0.9	0.9	0.6	0.8	1.2	1.9	8.3	1.0
Minimum	0.23	0.7	0.5	0.6	0.2	0.5	0.6	0.6	5.3	0.7
Maximum	1.08	2.7	1.2	1.0	0.9	1.1	2.1	6.1	13.8	1.7

Contamination factor (C_f), degree and modified degree of contamination (C_d and mC_d)

Table 7 shows the results of C_f, C_d and mC_d based on baseline in the sediment samples. The results of contamination factor reveal that Pb, Zn and Cd have moderate and As, Co, Cr, Ni and Cu have low contamination.

Contamination factor in the Dam reservoir sediment samples (SR-11 to SR-16) indicates considerable contamination for Pb and moderate contamination for Cd and Zn (Table 7). Moderate degree of contamination ($8 < C_d < 16$) was mainly observed in SR-6, SR-11 to SR-16, SR-21, SR-24 and SR-25 stations (Table 7).

The applications of mC_d based on baseline indicate very low and low degree of contamination (0.7–1.7) in the sediment samples.

Assessment of sediment quality and potential ecological risk

The selected heavy metals and metalloids concentrations were compared with threshold effect level (TEL), probable effect level (PEL), effect range low (ERL) and effect range median (ERM) (Table 8). It is evident that the average concentration of As, Cr and Ni in the present sediments are higher than TEL. The average of Ni concentration is higher than PEL and

Table 8 Comparison of heavy metals and metalloids concentration with toxicological reference values (ERL, ERM, PEL and TEL) of the sediment samples

	Cr	Cu	Ni	Pb	Zn	As	Cd
Shahid Rajaie sediments	68.13	22.18	37.85	33.54	100.24	7.40	0.41
PEL[a]	90.00	197.00	36.00	91.30	315.00	17.00	3.53
Shahid Rajaie sediments/PEL	0.76	0.11	1.05	0.37	0.32	0.44	0.12
TEL[a]	37.30	35.70	18.00	35.00	123.00	5.90	0.60
Shahid Rajaie sediments/TEL	1.83	0.62	2.10	0.96	0.81	1.25	0.69
ERM[b]	370.00	270.00	51.60	218.00	410.00	70.00	9.60
Shahid Rajaie sediments/ERM	0.18	0.08	0.73	0.15	0.24	0.11	0.04
ERL[b]	81.00	34.00	20.90	46.70	150.00	8.20	1.20
Shahid Rajaie sediments/ERL	0.84	0.65	1.81	0.72	0.67	0.90	0.34

[a] MacDonald et al. (2000)

[b] NOAA (2009)

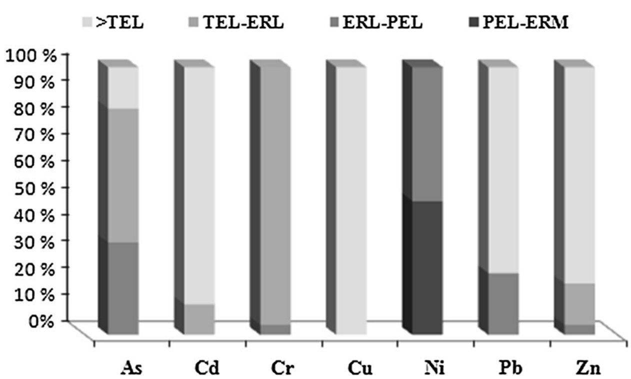

Fig. 5 Distribution of heavy metals and metalloids in all sediment samples with respect to effects range of median/low (ERM/ERL) and possible/threshold effect level (PEL/TEL) guidelines

ERL values. The results from the application of SQGs for the heavy metals and metalloids are shown in Fig. 5. These sediments based on PELQ and ERMQ calculations, are slightly toxic in Cr, As, Cd, Zn, Cu, Ni and Pb (Fig. 6).

Pollution load index (PLI) was calculated for every sampling site using the baseline elements data. The calculated PLI values of the present sediments are summarized in Table 9. The PLI values ranged from 0.61 to 1.33 with an average of 0.94. According to Tomlinson et al. (1980), values above 1 indicate progressive deterioration. Detected values for SR-6, SR-11 to SR-16, SR-21 and SR-24 stations is slightly exceeding 1 (Fig. 7). The results of evaluation on potential ecological risk factor (E_r^i) and potential ecological RI are summarized in Table 9. The order of potential ecological risk factor of the heavy metals and metalloids in sediments are Cd > Pb > As > Co > Ni > Cu > Cr > Zn. The E_r^i of Pb, Ni, Cr, Cu, Zn and As are lower than 40, which fit in low ecological risk criteria. Only the E_r^i values for Cd show moderate ecological risk (Hakanson 1980). Potential ecological RI is varied between 45.8 and 116.2. Average RI in the sediment samples is 72.3. All the sampling sites were at low risk level where the RI values were much lower than 150 (Hakanson 1980). The results indicated that there is a low potential ecological risk for selected elements in the sediments.

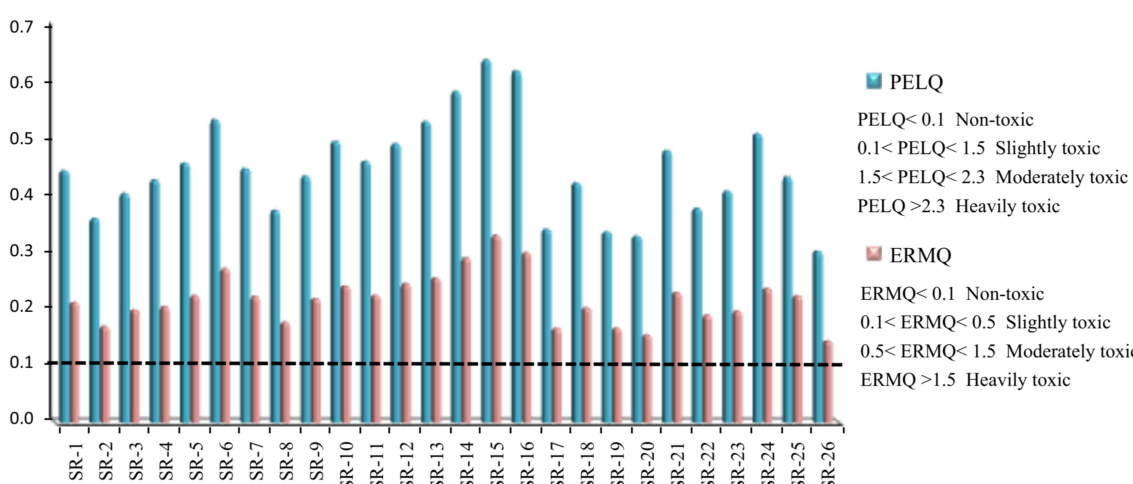

Fig. 6 PELQ and ERMQ diagram of Cd, As, Pb, Zn, Ni, Cr and Cu in the sediment samples

Table 9 Potential ecological risk factors (Eri) and potential ecological risk indexes (RI) of heavy metals and metalloids in sediments from Shahid Rajaie Dam Lake and upstream

Sample No.	E_i								RI	PLI
	Cr	Cu	Ni	Pb	Zn	Co	As	Cd		
Sefidrood										
SR-1	1.9	3.7	3.9	3.9	1.0	4.4	9.2	50	78.0	0.93
SR-2	1.6	2.7	3.1	3.3	0.8	4.0	7.8	30	53.3	0.74
SR-3	1.5	2.3	4.0	7.0	0.9	4.5	7.5	20	47.6	0.78
SR-4	1.9	3.6	3.8	4.4	1.0	4.6	8.4	30	57.6	0.87
SR-5	1.8	4.1	4.2	4.7	1.1	4.8	9.0	40	69.8	0.96
SR-6	2.0	3.9	5.5	9.1	1.2	5.8	7.3	35	69.8	1.08
Dam's Lake										
SR-7	1.9	4.3	4.2	4.7	1.1	4.9	7.0	50	78.1	0.97
SR-8	1.7	3.0	3.2	3.1	0.9	4.0	7.1	40	63.0	0.77
SR-9	1.6	2.7	4.5	5.2	1.0	5.1	7.4	34	61.5	0.87
SR-10	2.0	4.1	4.6	4.7	1.1	5.0	10.8	30	62.4	0.98
SR-11	1.5	2.9	2.8	18.1	1.8	3.5	5.7	80	116.2	1.06
SR-12	1.3	2.4	3.5	19.5	1.8	4.0	7.1	60	99.7	1.08
SR-13	1.7	3.4	3.3	20.6	1.8	4.1	8.2	70	113.1	1.21
SR-14	1.9	4.7	4.4	18.1	2.0	4.9	8.8	50	94.8	1.3
SR-15	2.0	4.3	5.7	19.9	2.1	5.8	7.5	40	87.3	1.33
Shirinrood										
SR-16	1.8	4.1	4.1	30.6	1.7	4.4	8.4	50	105.2	1.3
SR-17	1.9	2.4	3.3	3.1	0.7	3.5	2.3	40	57.2	0.65
SR-18	2.0	1.5	4.2	5.2	0.8	5.1	7.0	20	45.8	0.75
SR-19	1.3	2.6	2.7	3.1	1.3	3.3	6.4	40	60.6	0.73
SR-20	1.2	1.2	2.9	7.8	0.6	3.3	7.1	25	49.0	0.65
SR-21	2.0	3.9	3.7	6.1	1.5	4.7	9.5	50	81.4	1.06
SR-22	1.2	2.3	3.2	6.3	1.4	4.1	7.3	41	66.8	0.84
SR-23	1.8	3.7	3.2	5.8	1.3	4.6	6.9	30	57.3	0.89
SR-24	2.0	3.3	3.4	13.3	1.5	3.7	10.1	50	87.3	1.1
SR-25	1.5	3.4	4.2	8.7	1.3	5.1	5.1	38	67.3	0.95
SR-26	1.4	1.9	2.7	6.1	0.6	2.5	3.6	30	48.7	0.61
Mean	1.7	3.2	3.8	9.3	1.2	4.4	7.4	41.3	72.3	0.94
Minimum	1.2	1.2	2.7	3.1	0.6	2.5	2.3	20.0	45.8	0.61
Maximum	2.0	4.7	5.7	30.6	2.1	5.8	10.8	80.0	116.2	1.33

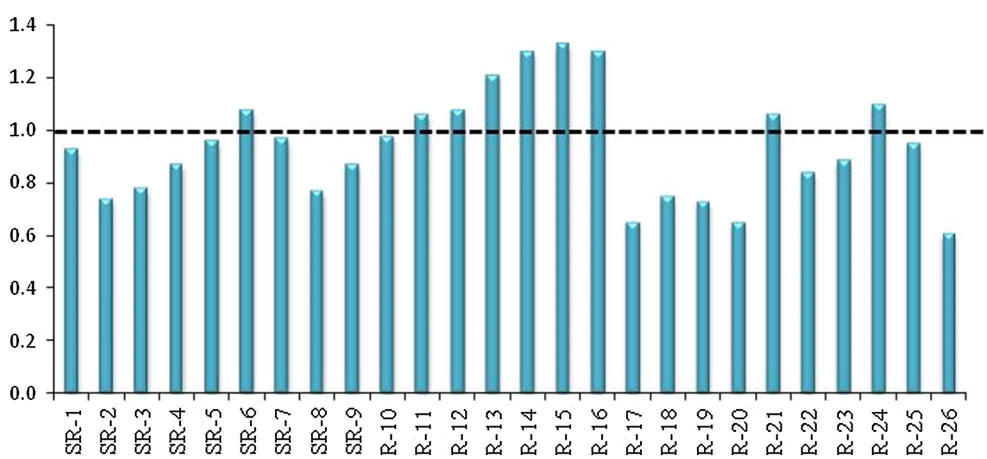

Fig. 7 Pollution load index estimated for Cd, As, Pb, Zn, Ni, Cr, Co and Cu in the sediment samples

Conclusion

In the present study, assessment of heavy metals and metalloids in surface sediments of Shahid Rajaie Dam, Sefidrood and Shirinrood Rivers were carried out using different environmental indices. Environmental factors PLI, *C*f, EF, C_d and mC_d and potential ecological risk show similar values for the levels of heavy metals and metalloids pollution in the sediment samples. The highest values of these factors for Cd, Zn and Pb in Shahid Rajaie Dam sediments, feed by Sefidrood and Shirinrood Rivers, are due to anthropogenic sources such as paddy fields, local unengineered landfills, sewage, vehicles, and burning of wood/coal and fossil fuels. Similar to EF, PCA data enabled us to indicate an elevated cadmium, lead and zinc concentrations result from inflow of contaminated water from upstream. PC1 is in agreement with EF showing a geogenic source for Cu, Mn, As, Cr, Al, Co, Ni and Sc in the sediment samples. Higher positive loadings of P with Ni and Co for a small number of sediment samples (SR-3, SR-5, SR-9, SR-18 and SR-25) probably is due to amplified concentration of phosphorus fertilizers in the agricultural lands. PC3 indicated that OC and clay contents control the distribution of Pb, Zn and Cd in the dam lake sediments. Although, the sediment contamination still is not a significant threat in the area, considering the increasing rate of agricultural activity and residential development in upstream of Shahid Rajaie Dam, contamination of sediment, especially by Cd, Zn and Pb would be plausible in the near future. Furthermore, since sediments play a significant role in the remobilization of contaminants in aquatic systems under favorable conditions, the sediment and water of Shahid Rajaie Dam require more monitoring to fully understand the behavior and ecological effects of toxic elements contamination in future.

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