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Potentially toxic elements and persistent organic pollutants in water and fish at Shahid Rajaei Dam, north of Iran

A. Shakeri · R. Shakeri · B. Mehrabi

Abstract The purpose of this study was to assess selected potentially toxic elements and persistent organic pollutants concentrations in the Shahid Rajaei Dam reservoir as well as their accumulation in barbel and *Leuciscus cephalus* fish species. The concentration of arsenic, mercury, nickel, chromium and cadmium in all water samples is less than WHO drinking water standard. Chromium shows a higher concentration than WHO standard in both fish species, while nickel and mercury content in barbel fish is higher than WHO standard. The result of principle component analyses indicates an increased elemental concentration due to application of phosphorus fertilizers in the agricultural lands, especially paddy field, ending up to the Tajan River and Shahid Rajaei Dam reservoir. Ni, Co, Mn, Pb and Cr display a quasi-independent behavior within the groups (PC1, PC2 and PC3) reflecting contribution of both geogenic and anthropogenic sources. The concentrations of persistent organic pollutants in all water samples, except M-5 sample, are less than WHO and EPA drinking water standards, while persistent organic pollutants (detected in both fish species) indicate higher contents than those of EPA standard values. Based on carcinogenic health risk value, maximum allowable fish consumption for arsenic is two meals per month. The result of noncarcinogenic health risk of mercury indicates one and three meals consumption per month for barbel and *L. cephalus* fishes, respectively. The calculation of maximum allowable fish consumption based on carcinogenic health for dieldrin, $\sum$HCH,

heptachlor epoxide, $\sum$DDT and $\sum$PCBs reveals monthly meals limitation of both fish species grown in Shahid Rajaei Dam.

Keywords Health risk assessment · Inorganic and organic pollutants · Maximum allowable fish consumption · Organochlorine pesticides · Polychlorinated biphenyls · Shahid Rajaei Dam

Introduction

The pollution of the aquatic environment with organic and inorganic pollutants has become a worldwide problem in recent years, as they are indestructible and most of them have toxic effects on organisms (Tanabe et al. 1994; MacFarlane and Burchett 2000; Malik et al. 2009; Dai et al. 2011). The major organic pollutants are polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), and inorganic one is potentially toxic elements (PTEs). These pollutants are ubiquitous contaminants in different environments (Martin et al. 2003; Fu and Wu 2005, 2006). Aquatic environments are important biotopes that should be controlled sensitively from unsafe industrial, agricultural and other anthropogenic activities (Mohiuddin et al. 2011; Zeng et al. 2013; Chakraborty and Owens 2014). PCBs are a group of chlorinated hydrocarbons widely used in industry with many different applications. However, their physical and chemical stability and also their lipophilic character make them serious environmental pollutants, which are highly persistent and tend to accumulate in soils, sediments, tissues and aquatic biota (Lana et al. 2008; Eqani et al. 2011). OCPs are organic compounds that to a varying degree resist photolytic, biological and chemical degradation. They are renowned for their persistence and

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bioaccumulation characteristics. Dieldrin and DDT, as well as the persistent organochlorine compounds of their decomposition, are still present in the environment, despite the ban on their use which was introduced in most countries in the 1970s (Cleemann et al. 2000; Feng et al. 2003; Eqani et al. 2011; Pawelczyk 2013). In fact, many of these compounds are now classified as the so-called persistent organic pollutants (POPs), because they can be recycled through food chains and produce a significant magnification of the original concentration at the end of the chain (Doong et al. 2002; Dai et al. 2011; Sharma et al. 2014). POPs are of international concern owing to their negative impact to both wildlife and human being due to lipophilic properties (Sanpera et al. 2002; Eqani et al. 2011). Agricultural lands cover a large part of the river basin and even in some areas in the world; these lands are located very close to the river. The use of PCBs and OCPs in the agricultural land has led to the accumulation of these compounds in the environment. Toxic elements enter in the environment from variety of sources (Dalai and Ishiga 2013). Main anthropogenic sources of heavy metal contamination are mining, disposal of domestic waste, as well as metal chelates from different industries and indiscriminate use of heavy metal-containing fertilizer and pesticides in agricultural fields (Nouri et al. 2008; Atar et al. 2012; Gupta et al. 2013). These pollutants migrate to surface and ground waters. They may also affect organisms directly by accumulating in their body or indirectly by transferring to the next species in food chain (Vander Oost et al. 2003; Sayg and Yigit 2012). Pollutant concentrations in aquatic ecosystems are usually monitored by measuring their concentrations in water, sediments and biota (Zhang et al. 2002; Dai et al. 2011; Eqani et al. 2011). Different groups of organisms can be used as a bioindicator of environmental change, though fish is usually considered as a good indicator of aquatic environmental changes and ecosystem health, especially in toxic pollution of waters (Yim et al. 2005; Moiseenko et al. 2008; Zrncic et al. 2013).

The Tajan River is one of the major rivers for water supply in north of Iran. The demand for water in the domestic, industrial and agricultural sectors of the Tajan River region is steadily increasing due to development, population growth as well as the demand for a sufficient food supply. For this purpose, Shahid Rajaei Dam was constructed on the Tajan River in 1997. This dam has been subjected to anthropogenic inputs of pollutants. Despite the potential for direct discharge of organic and inorganic contaminants into the Shahid Rajaei Dam reservoir, no measurement has been taken for monitoring the content and composition of POPs and trace metals in dam and Tajan River, especially in upstream. The Shahid Rajaei Dam receives discharges from agricultural land wastewater (approximately 1,800 ha, paddy field) and domestic and livestock wastewater. The present work is the first attempt for evaluating the PTEs and POPs contamination in the Shahid Rajaei Dam. Therefore, the concentrations of PTEs, OCPs and PCBs in water during two periods (November 2012 and September 2013) and fish in November 2012 were analyzed and compared. The main goals of present study are: (1) to determine the geochemical composition of the Shahid Rajaei Dam lake water, (2) to determine concentrations of persistent organic pollutants (OCPs and PCBs) and selected metals in water and their accumulation in fish, (3) to evaluate their hazard quotient (HQ), and (4) to calculate monthly fish consumption limits for carcinogenic and noncarcinogenic health of each contaminant.

Materials and methods

Study area

Shahid Rajaei 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 1,244 Km^2. It is constructed on the Tajan River, and its reservoir is fed by Shirinrood and Sefidrood rivers (in the confluence of these rivers, Tajan River arise). 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 and Sekuya 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 in the region in terms of lithology are mainly limestone, dolomitic limestone, sandstone, marl and shale (Fig. 1).

Sampling and analysis

For water quality assessment, nineteen samples were collected from the surface waters including nine sites along the Shirinrood (Sh-1 to Sh-9) and four sites along Sefidrood (S-1 to S-4) rivers in addition to five samples from Lake Dam (M-1 to M-5) and one sample from a mineral spring (Sp) during two periods (November 2012 and September 2013). Location of the sampling points is shown in Fig. 2. Each water sample was collected in a 1.5-l polyethylene bottle. The bottles were thoroughly washed with dilute hydrochloric acid and then with distilled water in the laboratory. In field, each bottle was filled and emptied twice with the water to be sampled before sampling. Water samples were filtered using 0.45-µm-pore-size filter papers in order to separate particulate matter. The filtered samples
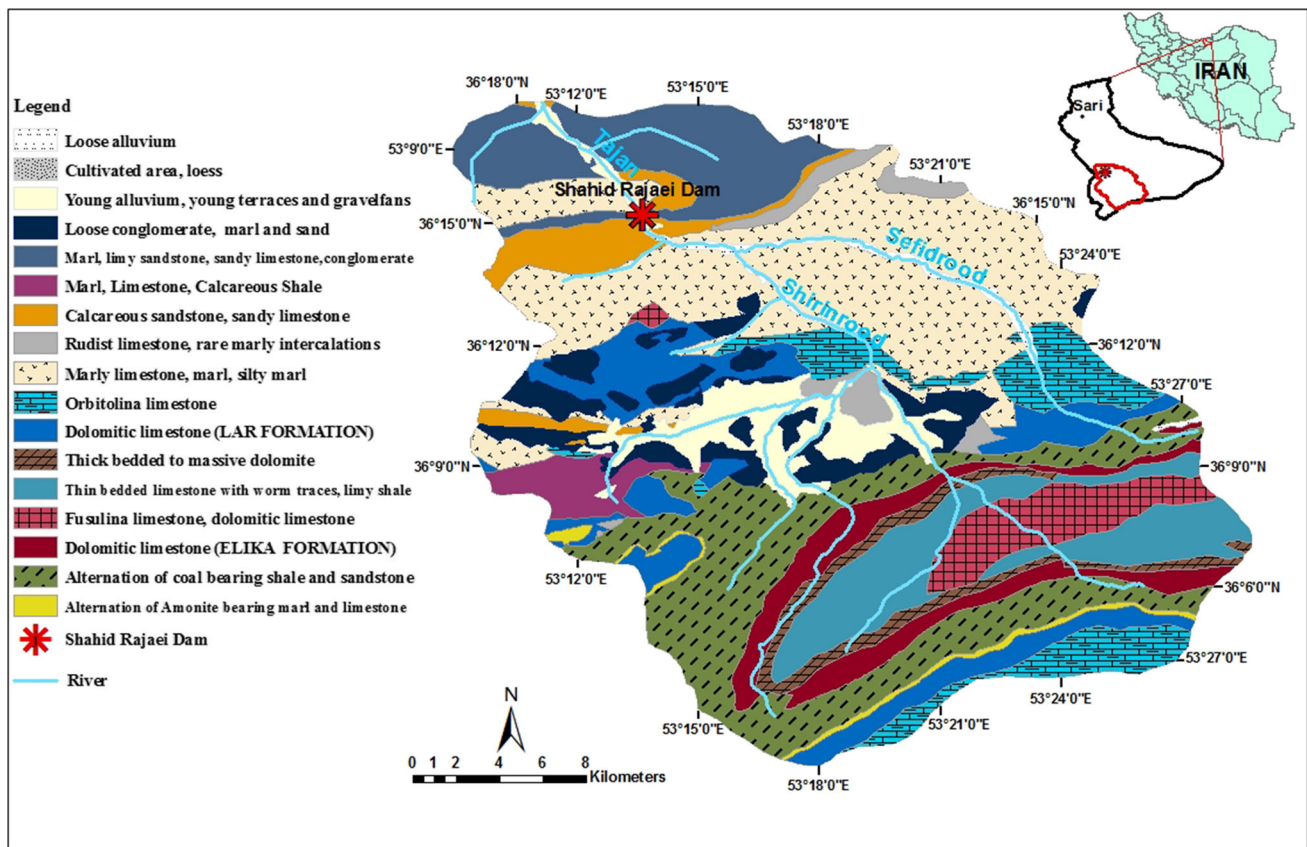
Fig. 1 Geological map of the study area

were then split into two subsamples: one was acidified with HNO_3 for dissolved element measurement and the second one unacidified for the determination of dissolved anion. The samples were kept at 4 °C prior to analysis. Electrical conductivity (EC) and pH were measured in the field during sampling using portable measuring devices (Eutech instruments, PCD650). The concentrations of calcium, potassium, magnesium and sodium were measured using standard titration, and bicarbonate, sulfate and chloride ions were measured using ion chromatography techniques (in Mazandaran regional water laboratory). Trace elements and PTEs were analyzed by ICP-OES/ICP-MS in West lab, Australia. Certified reference materials, duplicates and reagent blanks were analyzed with each batch of samples for measuring accuracy, precision and baseline contamination, respectively. For measuring OCPs and PCBs, water samples at nine stations were collected in a 1-l dark glass bottle and sent to the Kimia Shangarf Pars laboratory for gas chromatography (GC) analysis. Fish samples, including two species barbel and *L. cephalus* of Cyprinidae family, were collected from the Lake Dam. The fish samples were washed with deionized water, packed in polyethylene bags and kept at −20 °C, then transported on ice to the Kimia Shangarf Pars laboratory. In laboratory, 10 g of whole fish

weight was prepared according to the EPA-821-08-021 (for PCBs) and EPA-821-R-00-017 (for OCPs) methods. POPs were analyzed by gas chromatography, and selected PTEs (As, Cd, Cr, Ni and Hg) were analyzed by atomic absorption spectrometry.

Statistical analysis

Principal component analysis (PCA) is the most common multivariate statistical methods used in environmental studies (Kortatsi et al. 2009). It also helps in assigning source identity to each one of the PCs, as well as being a very powerful method of exploratory data processing and interpretation. PCA is widely used to reduce data (Loska and Wiechuya 2003) and to extract a small number of latent factors for analyzing relationships among the observed variables. The most common PCA type producing more interpretable components is the varimax rotation, which is applied in the current study. Factor loadings of >0.71 are typically regarded as excellent and <0.32 very poor for interpretation (Nowak 1998; Garcia et al. 2004). The number of significant principal components is selected on the basis of the Kaiser criterion with eigenvalue higher than 1 (Kaiser 1960).
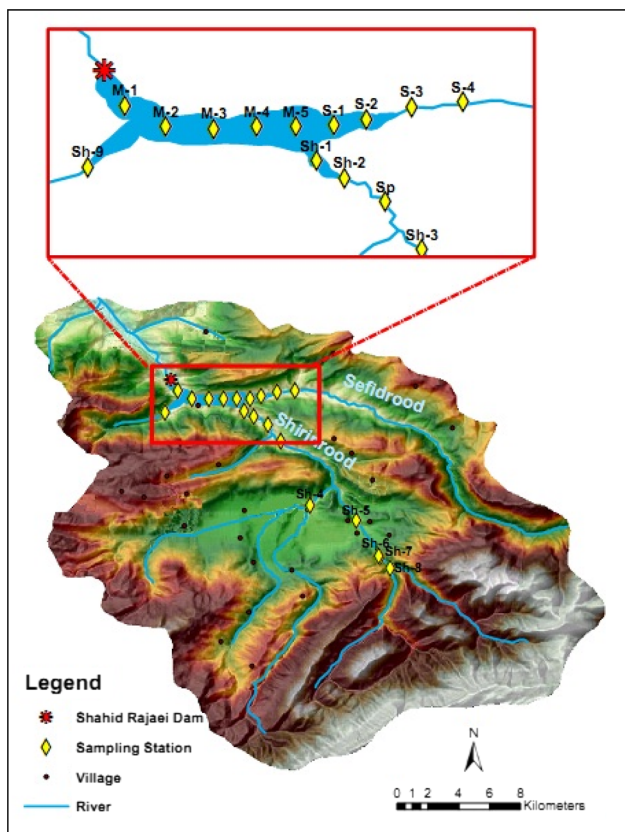
Fig. 2 Location of water sampling stations

Risk assessment

Hazard quotient (HQ)

To assess the public health risk of exposure to POPs and potentially toxic elements through fish consumption, the hazard quotient (HQ) is calculated by comparing the average daily doses (ADD) of a pollutant taken in, with the reference doses (RfD) (Vallero 2004; USEPA 1992). The HQ was calculated for each contaminant using Eq. (1).

$$HQ = \frac{ADD}{RfD} \tag{1}$$

The ADD exposure level (intake) is expressed in milligram per kilogram per day and is calculated by the Eq. (2).

$$ADD = \frac{C_m \cdot CR}{BW} \tag{2}$$

where: RfD = oral reference dose (mg/kg/day); ADD = average daily dose (mg/kg/day); BW = consumer body weight (70 kg); C_m = measured concentration of chemical contaminant m in fish (mg/kg); CR = mean daily fish consumption rate (0.05 kg/day)

The HQ has been defined as if it is <1.0, there is no significant risk or systemic toxicity and ratios higher than 1.0 representing a potential risk. There is a likelihood that adverse outcome can occur at these concentrations, but an HQ > 1 does not necessarily mean that adverse effects will occur (Vallero 2004).

Risk-based consumption limits

The USEPA approach is designed to manage health risks by providing risk-based consumption advice regarding contaminated fish (for example, one should limit consumption of a particular species to a specified number of meals per month or week). Two equations are used for deriving meals consumption limits for either carcinogenic or noncarcinogenic health effects. To calculate consumption limits for carcinogenic effects, it is necessary to specify an acceptable lifetime risk level (ARL). The appropriate risk level for a given population is determined by risk managers that calculate a risk level of 1 in 100,000 (10^{-5}).

Equation (3) is used for calculating an allowable daily consumption of contaminated fish based on a contaminant's carcinogenicity, expressed in kilograms of fish consumed per day (USEPA 2000).

$$CR_{lim} = \frac{ARL \cdot BW}{C_m \cdot CSF} \tag{3}$$

Equation (4) is used for calculating an allowable daily consumption of contaminated fish, based on a contaminant's noncarcinogenic health effects and is expressed in kilograms of fish per day.

$$CR_{lim} = \frac{Rfd \cdot BW}{C_m} \tag{4}$$

where:

CR_{lim} = maximum allowable fish consumption rate (kg/day); ARL = maximum acceptable individual lifetime risk level (unit-less); BW = consumer body weight (70 kg); C_m = measured concentration of chemical contaminant m in fish (mg/kg); CSF = cancer slope factor (mg/kg/day); RfD = oral reference dose (mg/kg/day)

Cancer slope factor and oral reference dose obtained from the USEPA Integrated Risk Information System for PTEs and POPs contaminants (http://www.epa.gov/iris/).

Equation (5) is used to convert daily consumption limits, in kilograms, to meals consumption limits over a given period (month) as a function of meals size (USEPA 2000).

$$CR_{mm} = \frac{CR_{lim} \cdot T_{ap}}{MS} \tag{5}$$

where:

CR_{mm} = maximum allowable fish consumption rate (meals/month); T_{ap} = time averaging period [365.25 day/ 12 months = 30.44 (day/months)]; MS = meal size (0.227 kg fish/meals)

Results and discussion

Major ion chemistry

Major ions concentration, EC, TDS and pH of collected samples are summarized in Table 1. The pH ranges from 7.4 to 8.1 with an average value of 7.9 in November 2012 and ranges from 7.1 to 8.1 with an average value of 7.8 in September 2013, which is reflecting the alkaline nature of the water samples. The EC and TDS values range from 307 to 1235 μs/cm and 304–945 mg/l with an average value of 462 μs/cm and 455 mg/l, respectively. The order of cation chemistry for 16 water samples in November (Sh-1, Sh-2, Sh-3, Sh-5, Sh-6, Sh-7, Sh-8, S-1 to S-4, M-1 to M-5) is $Ca^{++} > Mg^{++} > Na^+ > K^+$, while for Sh-4 and Sh-9 samples is $Ca^{++} > Na^+ > Mg^{++} > K^+$ and for the Sp mineral spring is $Na^+ > Ca^{++} > Mg^{++} > K^+$. The order of cation chemistry for water samples, except Sh-1 ($Ca^{++} > Na^+ > Mg^{++} > K^+$) and Sh-4 ($Ca^{++} > Mg^{++} > Na^+ > K^+$), in September is similar to those of November. The sodium concentration ranges from 4.6 to 103.5 mg/l with an average of 16.3 mg/l in November and 4.6–142.6 mg/l with an average of 21.9 mg/l in September (Table 1). Sodium concentration for Sp mineral spring is higher than WHO and EPA standard values. Calcium and magnesium concentrations in water vary from 44 to 190 and 10.8 to 26.4 mg/l with average concentrations of 65 and 17.2 mg/l, respectively. Potassium concentration varies between 1.56 and 4.29 mg/l with an average of 1.96 mg/l (Table 1). The order of anion concentration for all water samples except Sh-4, Sh-9 and Sp samples in November and S-1 to S-4 and Sp in September is $HCO_3^- > SO_4^- > Cl^-$. In general, bicarbonate and sulfate are the dominant anions ranging from 140.3 to 366.0 mg/l and 4.8 to 307.2 mg/l with average concentrations of 206.5 and 61.7 mg/l, respectively. According to the Piper diagram, all water samples are $Ca–HCO_3–SO_4$ type, while Sp mineral spring water is $Na–HCO_3–Cl$ type (Fig. 3).

Phosphate concentration in unpolluted waters typically ranges from 0.01 to 0.1 mg/l (Samecka-Cymerman and Kempers 2001). The phosphate content for 89.5 % of water samples is higher than this range (EPA standard 0.03 mg/l). Phosphatic fertilizers used in agricultural lands in the Tajan basin may contribute to observe excess phosphate. In general, the mean concentration of NO_3^- and NO_2^- in the water samples is below the maximum allowable concentration (Table 1).

Inorganic and organic pollutants

Water

Concentration of selected trace elements and PTEs is summarized in Table 2. The average abundance order of trace elements contents for water samples in November is: Fe > Mn > Zn > Ni > Cr > Pb > Mo, As > Co > Cd and in September is: Fe > Mn > Zn > Cr > Ni > Mo > Pb > Co > As. The concentration of selected elements in water samples is as follows: Fe <10–290 μg/l, Mn 1.20–30.50 μg/l, Zn 0.90–17.87 μg/l, Ni 2.3–11.0 μg/l, Cr 1–6 μg/l, Pb 0.4–4.4 μg/l, Mo 0.30–1.89 μg/l, As <0.01–1.60 μg/l, Co <0.02–0.63 μg/l and Cd <0.05–1.01 μg/l. Concentrations of PTEs in all the water samples are less than WHO and EPA standard (Table 2).

The results of PCA for selected elements and major ions along with some physicochemical properties of water samples are presented in Table 3. Five factors are statistically representing the contributions influencing chemical composition of the water samples. These components describe 84.52 % of the total variance of the data. The first component (PC1) explains 30.41 % of the total variance, which is strongly and positively loading related to Na, Cl, K, NO₃, Ca, SO₄ and EC. Ni and Co display moderate positive loading, while B and HCO₃ show weak positive loading. The presence of Na, Cl, K, Ca, SO₄ and NO₃ indicates the geogenic origin. PC2 explains 18.96 % of the total variance and shows high positive loading on HCO₃, Mo and Cr, while Ni (0.59) and Pb (0.50) display moderate positive loading. High positive loading factor of bicarbonate in PC2 could be due to activity of plants root in aeration zone at the area with high rainfall in forest environment. Plant roots respire and produce CO_2 which will turn to carbonic acid. High and moderate positive loadings of HCO₃ with Mo, Cr, Ni and Pb reveal a significant role of bicarbonate ion in the dissolution and distribution of these elements in the surface water of the Tajan River and Shahid Rajaei Dam. PC3 has a high factor loading for Fe and PO₄, and moderate positive loading for Mn (0.68), Co (0.51), Cr (0.59) and pH (0.54). As and Ni have weak positive loading. Nickel, Co, Mn, Pb and Cr display a quasi-independent behavior within the groups (PC1, PC2 and PC3) reflecting contribution of both geogenic and anthropogenic sources. The higher loadings of PO₄ indicate amplified concentration due to application of phosphorus fertilizers as well as organophosphate pesticides in the agricultural lands, especially paddy field, ending up to the Tajan River and Shahid Rajaei Dam reservoir. High and moderate loadings of Fe and pH on PC3 indicate that iron content and pH play a significant role in dissolution and distribution of Cr, Ni and Co. PC4 has a high factor loading for Cd, Zn and significant positive loading for Pb, indicating a

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Table 1 Concentration of major ions (mg/l) along with some physicochemical properties of water

Sample no.	pH	EC	TDS	Ca^{++}	Mg^{++}	Na^+	K^+	HCO_3^-	Cl^-	NO_3^-	NO_2^-	SO_4^-	PO_4^-
November 2012													
S-1	8.1	500	315	58	18.0	11.5	1.56	152.5	17.75	3.1	0.013	86.4	0.14
S-2	8.1	500	315	58	18.0	11.5	1.56	152.5	17.75	3.1	0.018	86.4	0.14
S-3	8.1	541	341	70	16.8	9.2	1.95	140.3	17.75	0.9	0.036	115.2	0.19
S-4	8.1	505	318	56	19.2	11.5	1.56	164.7	17.75	3.5	0.069	76.8	0.10
M-1	7.8	514	324	62	16.8	11.5	1.95	152.5	17.75	1.8	0.019	91.2	0.05
M-2	7.7	514	324	64	14.4	13.8	1.95	195.2	24.85	3.1	0.041	48.0	0.05
M-3	7.9	513	323	62	16.8	11.5	1.95	152.5	17.75	2.6	0.046	91.2	0.03
M-4	7.9	517	326	66	15.6	13.8	1.95	158.6	24.85	2.6	0.044	76.8	0.09
M-5	8.0	508	320	60	16.8	11.5	1.56	158.6	17.75	2.6	0.042	81.6	0.06
Sh-1	8.0	515	324	62	16.8	11.5	1.95	158.6	17.75	3.1	0.029	86.4	0.13
Sh-2	7.8	747	471	110	15.6	11.5	2.73	201.3	21.30	4.8	0.007	158.4	0.13
Sh-3	8.0	518	326	64	15.6	11.5	1.95	164.7	21.30	2.6	0.045	76.8	0.02
Sh-4	7.8	1,235	778	190	16.8	27.6	4.29	262.3	49.70	4.9	0.001	307.2	0.05
Sh-5	7.9	492	310	66	15.6	4.6	1.56	201.3	10.65	2.2	0.003	52.8	0.15
Sh-6	8.0	484	305	66	14.4	4.6	1.56	213.5	10.65	1.8	0.001	38.4	0.07
Sh-7	7.9	485	306	66	14.4	4.6	1.56	213.5	10.65	2.2	0.001	38.4	0.11
Sh-8	7.8	482	304	64	15.6	4.6	1.56	213.5	7.10	1.8	0.002	43.2	0.15
Sh-9	7.9	502	316	62	10.8	20.7	1.56	231.8	28.40	2.2	0.004	9.6	0.08
Sp	7.4	983	619	60	26.4	103.5	3.51	366.0	113.60	1.8	0.008	19.2	0.02
September 2013													
S-1	7.8	316	501	54	19.2	13.8	1.56	237.9	24.85	2.6	0.0300	9.6	0.14
S-2	7.8	319	507	54	19.2	13.8	1.56	244.0	24.85	3.1	0.0278	4.8	0.11
S-3	7.8	322	511	54	20.4	13.8	1.95	244.0	24.85	2.6	0.0285	9.6	0.15
S-4	7.8	319	507	54	19.2	13.8	1.56	244.0	24.85	1.8	0.0285	4.8	0.13
M-1	7.9	313	497	54	16.8	16.1	1.56	183.0	28.40	3.1	0.0429	43.2	0.09
M-2	7.8	313	497	54	16.8	16.1	1.95	183.0	28.40	3.1	0.0451	43.2	0.04
M-3	7.8	313	497	54	16.8	16.1	1.95	183.0	28.40	1.8	0.0450	43.8	0.02
M-4	7.8	313	497	54	16.8	16.1	1.95	183	28.40	1.8	0.0453	44.5	0.07
M-5	7.8	315	500	54	16.8	16.1	1.95	183	28.40	1.8	0.0457	48.0	0.05
Sh-1	7.8	312	496	54	16.0	16.2	1.56	164.7	28.40	1.8	0.1248	57.6	0.07
Sh-2	7.8	317	503	54	18.0	16.2	1.56	179.9	24.85	2.2	0.0294	57.6	0.09
Sh-3	7.8	318	504	54	19.2	13.8	1.56	201.3	24.85	2.2	0.0291	57.6	0.03
Sh-4	7.1	309	490	56	21.6	4.6	1.56	262.3	10.65	2.2	0.0067	48.0	0.08
Sh-5	8.1	307	488	64	18.3	17.2	1.95	181.3	16.52	3.1	0.0079	41.7	0.12
Sh-6	8.0	316	501	54	18.7	16.9	1.95	185.2	15.31	2.6	0.0217	40.8	0.12
Sh-7	8.0	310	492	54	18.0	16.4	1.95	174.3	15.17	1.8	0.0183	42.5	0.09
Sh-8	7.5	559	887	126	19.2	18.4	3.12	353.8	28.40	3.5	0.0083	96.0	0.17
Sh-9	7.8	315	500	54	16.3	18.1	1.56	249.9	18.23	2.2	0.0092	56.7	0.13
Sp	7.9	595	945	44	10.8	142.6	3.51	353.8	113.60	1.3	0.0240	9.6	0.02
Average	7.9	462	455	65	17.2	19.1	1.96	206.5	26.38	2.5	0.0275	61.7	0.09
Maximum	8.1	1,235	945	190	26.4	142.6	4.29	366.0	113.60	4.9	0.1248	307.2	0.19
Minimum	7.1	307	304	44	10.8	4.6	1.56	140.3	7.10	0.9	0.0007	4.8	0.02
USEPA	6.5–8.5	–	500	–	–	30–60	–	–	250	10	1	250	0.03
WHO	6–8	–	1,500	200	150	50	12	–	250	50	3	250	–

common source. PC5 reveals high factor loading for As and B.

Considering extent of agricultural land in the study area, for determining the concentration of OCPs and PCBs, batch of three samples from the Shirinrood River (Sh-1, Sh-2 and Sh-3), Sefidrood River (S-1, S-2 and S-4) and the Lake Dam (M-1, M-3 and M-5) was collected and analyzed (in total nine samples). The OCPs and PCBs concentrations of the water samples are shown in Table 4. The total concentrations of OCPs in water is in range of <LoD (Limit of Detection) to 0.73 µg/l. The highest concentration was observed at M-5 station. The highest

concentration of organochlorine pesticides were: 0.09 µg/l (beta-HCH and gamma-chlordane), 0.17 µg/l (gamma-HCH), 0.01 µg/l (endrin and heptachlor epoxide), 0.03 µg/l (endrin ketone and alpha-endosulfane), 0.05 µg/l (dieldrin), 0.04 µg/l (alpha-chlordane), 0.12 µg/l (beta-endosulfane), 0.07 µg/l (endosulfane) and 0.1 µg/l (4,4′-DDE and 4,4′-DDD). Dieldrin concentration at M-5 sample (0.05 µg/l) is higher than WHO standard (0.03 µg/l). In other samples, the concentrations of OCPs are lower than WHO standard (2011). Total PCBs concentrations in water samples are in range of <LoD to 0.95 µg/l. The highest concentration of PCBs was observed at M-5 sample (0.95 µg/l), which is higher than EPA standard for drinking water (0.5 µg/l).

Fish

Mean concentrations of As, Ni, Cr, Hg and Cd in barbel and *L. cephalus* fish species are shown in Fig. 4. Cadmium (0.018 mg/kg) and As (0.035 mg/kg) show the lowest concentration, while Ni (1.44 mg/kg) and Cr (0.91 mg/kg) hold the highest mean concentrations in barbel and *L. cephalus* fish species, respectively. The average abundance order of toxic elements contents in barbel fish is Ni > Cr > Hg > As > Cd, while in *L. cephalus* fish is Cr > Hg > Ni > Cd > As (Fig. 4). The order in barbel fish is similar to the water samples (Ni > Cr > As > Cd > Hg), except for Hg. Mercury concentrations were very low in the water samples (<limit of detection), but concentrations in the fish samples were relatively high,

suggesting a particularly high bioaccumulation tendency compared with the other investigated trace elements. For human health protection, guidelines of PTEs presence in fish tissue have been set by different international organizations such as USEPA and WHO standards (Fig. 4). Concentration of analyzed selected PTEs in barbel and *L. cephalus* fish species in comparison with WHO standard (1989) contents is as follow: Cadmium indicates lower concentration than the WHO standard in two fish species. Nickel and mercury concentration in barbel fish is higher than the WHO standard. Chromium in barbel and *L. cephalus* fish species reveals higher concentrations than the WHO standards (0.15 mg/kg).

The concentrations of OCPs and PCBs in fish samples are summarized in Table 5. The highest concentrations of OCPs are 45 µg/kg (beta-HCH), 71 µg/kg (delta-HCH), 20 µg/kg (heptachlor), 10 µg/kg (heptachlor epoxide), 518 µg/kg (dieldrin), 44 µg/kg (4,4′-DDE), 8 µg/kg (4,4′-DDD), 216 µg/kg (4,4′-DDT) and 17 µg/kg (methoxychlor).

Delta-HCH, heptachlor, heptachlor epoxide, 4,4′-DDE, 4,4′-DDD and 4,4′-DDT indicate higher concentrations than those of EPA standard values (de Vlaming 2008; Davis et al. 2010) in both barbel and *L. cephalus* fish species (Table 5). Barbel fish has higher concentration of beta-HCH and dieldrin, while *L. cephalus* has higher concentration of methoxychlor compared with EPA guideline, respectively. The highest concentrations of PCBs for barbel and *L. cephalus* species were 17 (PCB28) and 45 (PCB28) µg/kg, respectively (Table 5). PCB28,
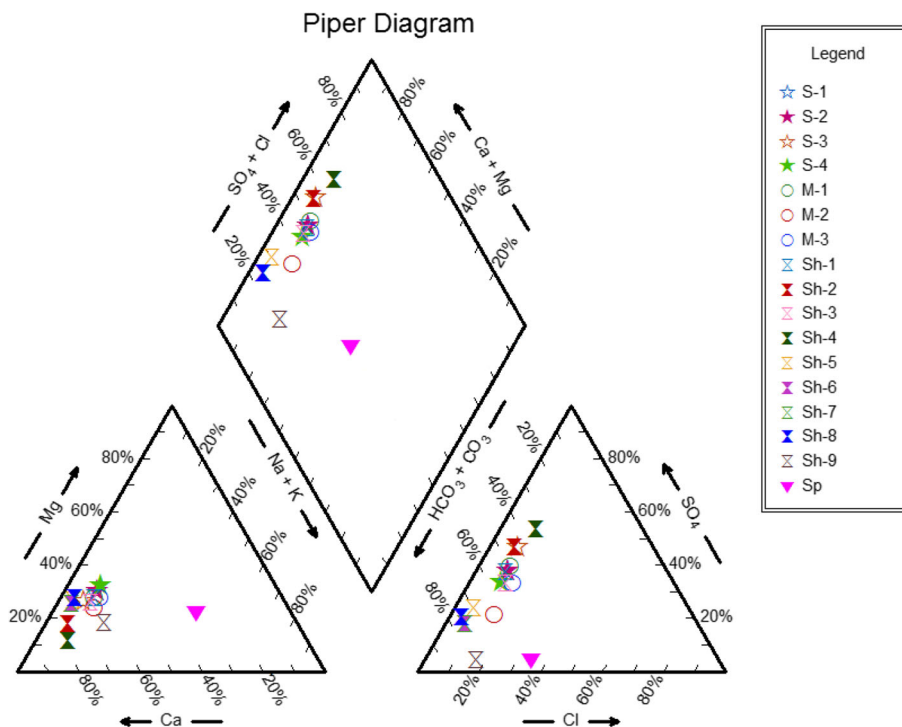
Fig. 3 Piper diagram of the water samples

Piper Diagram

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Table 2 Concentration of selected trace elements (µg/l) in water samples

Sample no.	As	Fe	Cr	Zn	Pb	Mo	Mn	Co	Cd	Hg	Ni
November 2012											
S-1	0.01	60	4	4.08	3.2	1.48	6.81	0.20	<0.05	<0.1	3.6
S-2	0.02	65	2	2.47	0.8	1.29	4.65	0.19	<0.05	<0.1	3.7
S-3	0.03	80	1	1.68	0.9	1.23	28.10	0.16	<0.05	<0.1	4.1
S-4	0.02	70	3	3.93	2.5	1.15	9.30	0.20	<0.05	<0.1	4.5
M-1	<0.01	<10	1	1.20	0.4	1.62	4.90	0.20	<0.05	<0.1	2.3
M-2	0.02	<10	2	1.42	0.5	1.41	9.70	0.21	<0.05	<0.1	3.7
M-3	0.04	<10	1	1.62	0.7	1.55	1.20	0.25	<0.05	<0.1	3.2
M-4	0.03	15	1	1.58	0.8	1.48	4.40	0.22	<0.05	<0.1	2.6
M-5	0.03	60	2	1.50	0.5	1.56	3.70	0.24	<0.05	<0.1	2.8
Sh-1	0.05	60	3	17.87	3.6	1.55	4.80	0.21	<0.05	<0.1	3.9
Sh-2	0.04	50	2	3.06	3.4	1.47	16.90	0.19	<0.05	<0.1	5.4
Sh-3	<0.01	45	1	1.48	0.5	1.47	4.50	0.20	<0.05	<0.1	2.8
Sh-4	0.03	90	2	2.37	1.4	1.89	18.20	0.22	<0.05	<0.1	8.7
Sh-5	0.03	80	1	1.34	0.7	1.32	8.40	0.17	<0.05	<0.1	3.9
Sh-6	0.02	65	1	1.75	1.1	1.45	3.40	0.19	<0.05	<0.1	4.1
Sh-7	0.03	70	1	1.23	0.8	1.53	4.10	0.21	<0.05	<0.1	4.3
Sh-8	0.02	70	3	2.39	1.4	1.85	2.30	0.38	<0.05	<0.1	4.1
Sh-9	0.01	85	2	2.04	1.0	1.12	6.45	0.25	<0.05	<0.1	5.9
Sp	0.04	55	2	2.30	1.8	0.64	1.80	0.20	<0.05	<0.1	3.5
September 2013											
S-1	1.20	60	2	2.10	0.7	0.80	8.81	0.51	0.15	<0.1	4.2
S-2	1.20	80	1	1.50	0.5	0.80	6.65	0.58	0.14	<0.1	3.8
S-3	1.10	290	4	8.70	2.6	0.50	30.50	0.52	0.31	<0.1	5.6
S-4	1.20	140	3	14.50	3.0	0.80	11.30	0.53	0.22	<0.1	5.9
M-1	0.90	<10	1	1.50	0.5	0.80	6.93	0.20	0.14	<0.1	3.3
M-2	1.00	<10	1	10.30	1.5	0.80	11.70	0.16	0.76	<0.1	4.2
M-3	0.80	<10	1	1.00	0.4	0.80	2.96	0.35	0.13	<0.1	3.4
M-4	1.00	30	1	17.50	3.6	0.70	6.40	0.10	1.01	<0.1	3.6
M-5	<0.01	80	1	6.90	1.1	0.70	5.72	<0.02	0.14	<0.1	3.8
Sh-1	1.20	80	1	1.70	0.9	0.80	6.86	0.37	0.26	<0.1	4.0
Sh-2	0.60	100	2	9.80	2.0	1.10	20.40	0.46	0.23	<0.1	6.5
Sh-3	0.50	60	1	1.70	0.7	0.90	6.72	0.23	0.09	<0.1	3.6
Sh-4	0.50	290	3	6.40	3.1	1.40	22.50	0.63	0.31	<0.1	11.0
Sh-5	0.50	190	4	4.60	3.4	1.20	10.60	0.09	0.15	<0.1	5.2
Sh-6	<0.01	140	4	8.80	2.0	1.30	5.84	0.61	0.16	<0.1	5.4
Sh-7	0.70	140	5	8.40	2.6	1.40	6.09	0.27	<0.05	<0.1	5.9
Sh-8	<0.01	120	2	8.90	4.4	1.40	4.14	<0.02	0.12	<0.1	5.3
Sh-9	1.60	220	6	4.60	2.6	1.00	8.88	0.33	0.23	<0.1	7.1
Sp	<0.01	80	1	0.90	0.4	0.30	3.12	0.14	0.11	<0.1	4.9
Average	0.45	97.50	2.08	4.61	1.6	1.17	8.68	0.28	0.26	<0.1	4.6
Maximum	1.60	290	6	17.87	4.4	1.89	30.50	0.63	1.01	<0.1	11.0
Minimum	<0.01	<10	1	0.90	0.4	0.30	1.20	<0.02	<0.05	<0.1	2.3
WHO (2011)	10	300	50	300	10.0	70	500	–	3.00	1	20.0
USEPA (2012)	10	300	100	–	15.0	–	–	–	5.00	2	–

Table 3 Principal component analysis for variables in water samples

Component	Rotated component matrix				
	PC1	PC2	PC3	PC4	PC5
EC	0.97	0.22	0.08	0.02	−0.01
K	0.97	0.13	−0.01	0.07	0.01
SO$_4$	0.97	−0.10	0.14	−0.06	−0.11
Ca	0.94	0.31	0.09	0.05	−0.07
Cl	0.83	0.07	−0.04	0.14	0.50
NO$_3$	0.78	−0.06	−0.20	−0.03	0.07
Na	0.72	0.04	−0.07	0.06	0.64
Ni	0.67	0.59	0.38	0.10	0.06
HCO$_3$	0.38	0.89	−0.03	0.13	−0.01
Mo	0.20	0.84	−0.10	−0.09	−0.41
NO$_2$	−0.09	−0.78	−0.16	0.27	0.21
Mg	0.27	−0.76	0.15	−0.19	−0.41
Cr	−0.15	0.70	0.59	0.02	0.13
Fe	0.17	0.22	0.86	0.05	0.03
PO$_4$	−0.24	−0.02	0.76	0.09	−0.30
Mn	0.54	−0.03	0.68	0.22	0.05
pH	−0.28	−0.48	0.54	−0.35	0.08
Co	0.42	−0.11	0.51	−0.39	0.14
Zn	0.03	0.02	0.12	0.88	−0.12
Cd	0.11	−0.22	−0.13	0.81	0.26
Pb	0.04	0.50	0.30	0.68	−0.24
As	−0.21	−0.27	0.33	0.01	0.74
B	0.29	−0.01	−0.29	−0.14	0.72
% of variance	30.41	18.96	14.23	10.48	10.45
Cumulative %	30.41	49.36	63.59	74.07	84.52

Rotation method: varimax with Kaiser normalization

PCB37 (except *L. cephalus*2 and barbel1 samples), PCB44 (except *L. cephalus*2 and barbel2 samples), PCB138, PCB153 and PCB180 (except L. *cephalus*1 sample) indicate higher concentrations than those of EPA standard values (de Vlaming 2008; Davis et al. 2010).

The result of hazard quotient for fish samples is presented in Table 6. The HQ for As, Cd, Ni and Cr except Hg in barbel and *L. cephalus* species is <1.0. The HQ value of dieldrin (OCP) in barbel fish and ∑PCBs in both fish species were >1.0. HQ for other POPs in two fish species is below unit (Table 6).

The results of oral R*f*D values, CSF values, allowable daily consumption and other values for As, Hg, Cd, Ni, Cr and POPs and their carcinogenic or noncarcinogenic health effects are summarized in Table 6. Based on CR$_{mm}$ value, maximum allowable barbel and *L. cephalus* fishes consumption for carcinogenic health of As is two meals per month (Approximately 0.5 kg). The result of noncarcinogenic health for Hg indicates one and three meals consumption per month for barbel and *L. cephalus* fishes, respectively (Table 6). Maximum allowable barbel fish consumption for carcinogenic health is calculated as: ∑HCH (0.5 meals/month), heptachlor epoxide (1 meal/month), ∑DDT (4 meals/month), ∑PCBs (1 meal/month). The calculated CR$_{mm}$ for dieldrin is indicating no consumption for barbel fish. Maximum allowable *L. cephalus* fish consumption for carcinogenic health is calculated as: ∑HCH (2 meals/month), heptachlor epoxide (1 meal/month), ∑DDT (1 meal/month). Monthly fish consumption limits for carcinogenic health ∑PCBs in *L. cephalus* is 0.5 meals per month (Approximately 110 g).

Table 4 Range of OCPs and PCBs concentration (μg/l) in the water samples

Compound	Concentration	Compound	Concentration
Alpha-HCH	<LoD	Gamma-Chlordane	<LoD—0.09
Beta-HCH	<LoD—0.09	Alpha-Chlordane	<LoD—0.04
Gamma-HCH	<LoD—0.17	Alpha-Endosulfane	<LoD—0.03
Delta-HCH	<LoD	Beta-Endosulfane	<LoD—0.12
Aldrin	<LoD	Endosulfane	<LoD—0.07
Dieldrin	<LoD—0.05	4,4′-DDE	<LoD—0.10
Endrin	<LoD—0.01	4,4′-DDD	<LoD—0.10
Endrin ketone	<LoD—0.03	4,4′-DDT	<LoD
Endrin aldehyde	<LoD	Methoxychlor	<LoD
Heptachlor	<LoD	∑OCPs	<LoD—0.73
Heptachlor epoxide	<LoD—0.01	∑PCBs	<LoD—0.95

LoD: limit of detection (<0.01 μg/l)

∑OCPs = Sum of (alpha-HCH, beta-HCH, gamma-HCH, delta-HCH, aldrin, dieldrin, endrin, endrin ketone, endrin aldehyde, heptachlor, heptachlor epoxide, gamma-chlordane, alpha-chlordane, endosulfane, alpha- endosulfane, beta-endosulfane, 4,4′-DDE, 4,4′-DDD, 4,4′-DDT and methoxychlor)

∑PCBs = Sum of 32 PCB congeners (IUPAC No. 8, 28, 37, 44, 49, 52, 60, 66, 70, 74, 77, 82, 87, 99, 101, 105, 114, 118, 126, 128, 138, 153, 156, 158, 166, 169, 170, 179, 180, 183, 187 and 189)
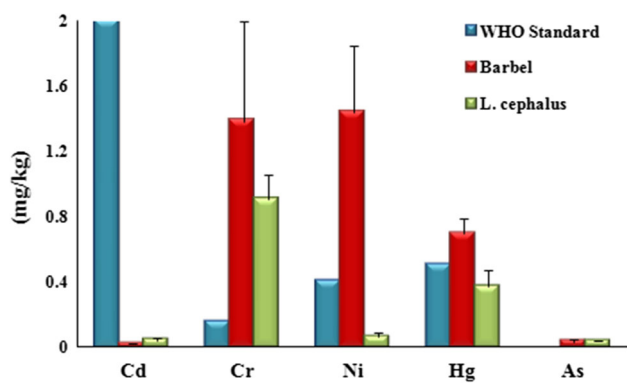
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Fig. 4 Mean concentration levels of potentially toxic elements in barbel and *L. cephalus* fish and WHO standard

Conclusion

This article provides a basic data on inorganic and organic pollutants in water and their accumulation in two fish species from Shahid Rajaei Dam, North Iran. The results demonstrate that concentration of As, Cr, Ni, Co, Mo, Cd, Cu, Zn, Pb, Mn, Fe, Hg and POPs (except dieldrin and $\sum$PCBs contents for M-5 sample) in water are significantly lower than those of WHO drinking water standard. In contrast, Cr (in both fish), Ni and Hg (in barbel fish) and OCPs and PCBs (detected in the both fish species) indicate higher contents than WHO and EPA standard values, respectively, which suggests a particularly high bioaccumulation tendency in the fish samples.

Table 5 Organochlorine pesticides and polychlorinated biphenyls concentrations in the fish samples

POPs (µg/kg)	Barbel-1	Barbel-2	*Leuciscus cephalus*-1	*Leuciscus cephalus*-2	EPA (µg/kg)
OCPs					
Alpha-HCH	ND	ND	ND	ND	0.5
Beta-HCH	45	40	ND	ND	1
Delta-HCH	71	60	20	35	2
Gamma-HCH	ND	ND	ND	ND	0.5
Heptachlor	15	20	8	5	1
Heptachlor epoxide	10	7	3	5	1
Aldrin	ND	ND	ND	ND	1
Endrin	ND	ND	ND	ND	2
Dieldrin	480	518	ND	ND	0.5
4,4′-DDE	38	44	30	20	2
4,4′-DDD	8	5	6	4	1
4,4′-DDT	20	25	216	204	3
Alpha-endosulfane	ND	ND	ND	ND	2
Beta-endosulfane	ND	ND	ND	ND	5
Methoxychlor	ND	ND	12	17	3
PCBs					
PCB8	ND	ND	ND	ND	0.2
PCB28	17	15	45	40	0.2
PCB37	ND	5	10	ND	0.2
PCB44	4	ND	7	ND	0.2
PCB52	ND	ND	ND	ND	0.2
PCB87	ND	ND	ND	ND	0.2
PCB101	ND	ND	ND	ND	0.2
PCB118	ND	ND	ND	ND	0.2
PCB138	8	3	7	5	0.2
PCB153	6	3	8	2	0.2
PCB166	ND	ND	ND	ND	0.2
PCB179	ND	ND	ND	ND	0.2
PCB180	2	8	ND	6	0.2

Table 6 Monthly fish consumption limits for carcinogenic and noncarcinogenic health endpoints and other parameters of inorganic and organic pollutants

Fish species	Inorganic and organic pollutants		C_m	RfD	ADD	HQ	CSF	Noncancer		Cancer	
								CR_{lim}	CR_{mm}	CR_{lim}	CR_{mm}
Barbel	PTEs	As	0.0325	0.0003	2.3×10^{-5}	0.08	1.5	0.65	87	0.01	2
		Cd	0.018	0.001	1.3×10^{-5}	0.01	NA	3.89	521	–	–
		Hg	0.7	0.0001	5×10^{-4}	5.00	NA	0.01	1	–	–
		Ni	1.44	0.02	1.03×10^{-3}	0.05	NA	0.97	130	–	–
		Cr	1.39	0.02	9.9×10^{-4}	0.05	NA	1.01	135	–	–
	POPs	$\sum$HCH	0.108	8×10^{-4}	8×10^{-5}	0.10	1.6	0.52	70	0.0041	0.5
		Heptachlor epoxide	0.009	1.3×10^{-5}	6×10^{-6}	0.47	9.1	0.11	14	0.0090	1
		Dieldrin	0.499	5×10^{-5}	3.6×10^{-4}	7.13	16	0.01	1	0.0001	None
		$\sum$DDT	0.070	5×10^{-4}	5×10^{-5}	0.10	0.34	0.50	67	0.0294	4
		$\sum$PCBs	0.036	2×10^{-5}	2.5×10^{-5}	1.27	2	0.04	5	0.0099	1
Leuciscus cephalus	PTEs	As	0.035	0.0003	3×10^{-5}	0.08	1.5	0.60	80	0.01	2
		Cd	0.04	0.001	3×10^{-5}	0.03	NA	1.75	235	–	–
		Hg	0.37	0.0001	2.6×10^{-4}	2.64	NA	0.02	3	–	–
		Ni	0.065	0.02	4.6×10^{-5}	0.002	NA	21.54	2888	–	–
		Cr	0.91	0.02	6.5×10^{-4}	0.03	NA	1.54	206	–	–
	POPs	$\sum$HCH	0.028	8×10^{-4}	2×10^{-5}	0.02	1.6	2.04	273	0.0159	2
		Heptachlor epoxide	0.004	1.3×10^{-5}	3×10^{-6}	0.22	16	0.23	31	0.0109	1
		$\sum$DDT	0.240	5×10^{-4}	1.7×10^{-4}	0.34	0.34	0.15	20	0.0086	1
		$\sum$PCBs	0.065	2×10^{-5}	4.6×10^{-5}	2.32	2	0.02	3	0.0054	0.5

C_m = measured concentration of chemical contaminant m in a given species of fish (mg/kg); RfD = oral reference dose (mg/kg/day); ADD = average daily dose (mg/kg/day); CR_{lim} = maximum allowable fish consumption rate (kg/day); CR_{mm} = maximum allowable fish consumption rate (meals/month); CSF = cancer slope factor (mg/kg/day); NA = Not available in EPA's Integrated Risk Information System; None = No consumption recommended

The results of maximum allowable fish consumption based on carcinogenic health for POPs, As and Hg reveal limitation for consumption of both fish species in Shahid Rajaei Dam. The possible sources of the OCPs and PCBs compounds and selected toxic elements are possibly the effluents from residential and agricultural areas. The levels of these pollutants may cause health impact on the aquatic environment and accompanied life. Since POPs and toxic elements levels in tissues of barbel and *L. cephalus* fish track the pollution gradient, these species appears to be useful for biomonitoring of POPs and metal accumulation, reflecting local pollution conditions and incorporating local bioavailability of these pollutants. Extended monitoring studies are necessary to fully understand the behavior and toxicological effects of organic and inorganic pollutants to the aquatic environments in north of Iran.

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