

## Dissolved Heavy Metals in the Reservoir of Shahid Rajaei Dam, Sari, Iran

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**ABSTRACT:** Concentrations of heavy metals (Ni, Co, Zn, Cd, Pb and Cu) in the reservoir of Shahid Rajaei Dam, one of the main water reservoirs in north part of Iran, were determined using a differential pulse polarography (Metrohm 797 VA) and compared with the national and international specified maximum contaminant levels (MCL) for different purposes of water use. The results showed that concentrations of Cd and Pb in the reservoir exceeded different MCLs and thus they could pose health problems for regional residents. Averaged concentrations of heavy metals exhibited a decreasing order of: Zn>Pb>Cd>Cu>Ni>Co. The spatial distribution and seasonal variation implied the significant effect of anthropogenic sources on concentration of heavy metals. These metals mainly originate from the fertilizers and pesticides which are used in upstream farm lands of the flood plain; however the natural sources should not be neglected. The results would help in water resource management of Shahid Rajaei Dam.

**Key words:** Dissolved heavy metals, Spatial distribution, Seasonal variation, Reservoir, Dam, Iran

### INTRODUCTION

Community development and growth of industrial and agricultural activities have lead to increase in aquatic contamination such as heavy metals and nutrients (Nriagu and Pacyna 1988, Donohue, *et al.* 2005, Palma, *et al.* 2010, Li and Zhang 2011, Prasanna, *et al.* 2012). Heavy metals are among the main contaminants of water resources because of their toxicity, bio-accumulative and persistent nature in various parts of aquatic life (Nurnberg 1982, Lasheen 1988, Diagonanolin, *et al.* 2004, Nordberg, *et al.* 2007, Li, Li and Zhang 2011, Grellier, *et al.* 2013). These metals originate from natural or anthropogenic sources and enter the aquatic ecosystem by atmospheric deposition, geologic weathering, agricultural activities and residential and industrial products (Dawson and Mackline 1998, Audry, *et al.* 2004, Donahue, Allen and Schindler 2006, Tuna, *et al.* 2007, Zamani, Yaftian and Parizanganeh 2012, Iqbal, Shah and Akhter 2013). Dam reservoirs are very important water supply resources for agricultural, industrial and domestic applications. Therefore, water assessment of dam reservoirs is crucial. In this regard, assessment of spatial and temporal variations of heavy metals' concentration can help to identify the sources of these contaminations and subsequently, a better

water quality management of the upstream basins is achieved. Reservoir of Shahid Rajaei Dam (36°14' - 36°15' N, 53°13' - 53°19' E), one of the main reservoirs in north part of Iran, was built in 1990s for agricultural purposes in Mazandaran province (Fig. 1). Water surface area of the reservoir is 428 hectares in normal water level and the total reservoir volume is 164 million cubic meters. It has a drainage area of about 1248 km<sup>2</sup> including the Sefid Rud and Shirin Rud Rivers' basins. Despite the preliminary aim of Shahid Rajaei Dam project, now this reservoir is utilized for fishery and boating purposes. Recently, this reservoir has been considered for drinking water supply of Sari, the provincial capital of Mazandaran province. So, regarding the extensive usage of this reservoir, water quality assessment including the evaluation of dissolved heavy metals' distribution is of utmost importance. In the present study the concentrations of dissolved heavy metals (Ni, Co, Zn, Cd, Pb and Cu) were quantified and compared with several maximum contaminant levels (MCL) suggested by the world organizations and other similar studies. Also, the variations of dissolved heavy metals were investigated through the depth of the reservoir in four seasons.

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Ultimately, the results of this research are important in resource management especially for drinking water supply of Sari.

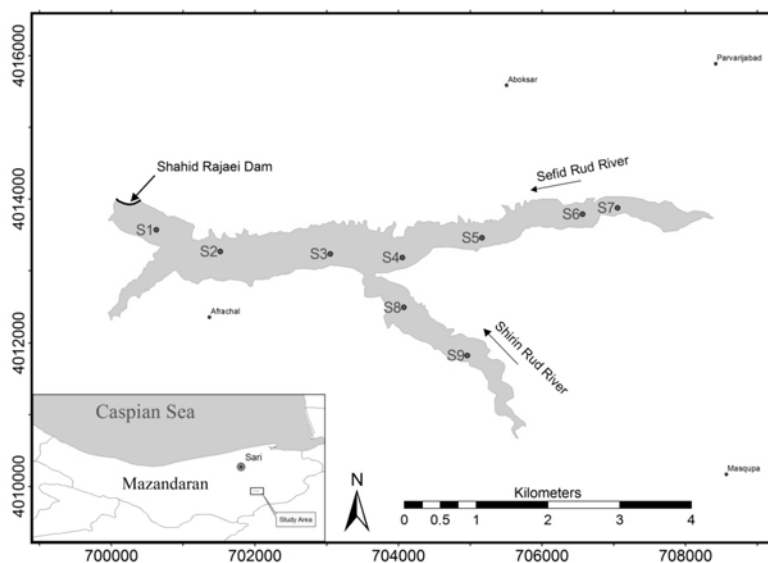
**MATERIALS & METHODS**

With studying topologic and hydrologic characteristics of the reservoir, nine sampling stations were positioned in Shahid Rajaei Dam reservoir, Iran (Fig. 1). Three stations (S1, S2 and S3) were located in the main part of the reservoir, starting from the Dam body to the junction point of Sefid Rud and Shirin Rud Rivers zones, four stations (S4, S5, S6 and S7) in Sefid Rud River zone and two stations (S8 and S9) in Shirin Rud River zone. Water samples were collected from September 2012 to August 2013 (September 2012, and January, April, and August 2013). Generally, in each station water samples were collected from surface up to 30 m under the water surface level with 5 m intervals

and up to 50 m under the water surface level with 10 m intervals (except in September 2012 which water samples were collected from surface, 5 m, 10 m and up to 50 m under the water surface level with 10 m intervals). So, a maximum of 9 water samples were collected from each station. However, some times the samplings were not implemented in some stations or some depths because of the low water level in the reservoir, and a total of 184 water samples were taken for the analysis (Table 1). Also, several water samples were taken from Sefid Rud and Shirin Rud Rivers at each sampling time. Water samples were collected in 1000 mL poly ethylene bottles and immediately transported to the laboratory and kept under 4°C to be ready for the analysis. Water samples were analyzed for the presence of Ni, Co, Zn, Cd, Pb and Cu ions using a differential pulse polarography (Metrohm 797 VA). Dissolved air was removed from the solutions by

**Table 1. Number of samples at each station**

Station	Sampling Date			
	Sep 2012	Jan 2013	Apr 2013	Aug 2013
S1	6	9	9	9
S2	7	9	9	9
S3	7	9	9	9
S4	5	7	9	6
S5	-	7	7	3
S6	-	-	3	-
S7	-	-	2	-
S8	5	7	8	4
S9	-	5	5	-



**Fig. 1. Location of Shahid Rajaei Dam reservoir, Iran, and nine sampling stations (S1, S2 and S3 in main part of reservoir, S4, S5, S6 and S7 in Sefid Rud River zone and S8 and S9 in Shirin Rud River zone)**

degassing with N<sub>2</sub> gas (99.999%) for 5-10 minutes prior to each run. The polarography parameters are listed in Table 2. The prepared standard method in DIN 38406 Part 16 describes the determination of Zn, Cd, Pb, Cu, Ni and Co ions in drinking, ground and surface waters. These metals can be determined in the following ranges: 1.0, 0.1, 0.1, 1.0, 0.1, and 0.1 µg/L respectively (Metrohm, Application Bulletin No. 231/2e). The digested samples were analyzed by standard addition in triplicate and the

average concentrations of metal ions were reported in µg/L. Repetitious relative standard deviation of less than 5% was accepted in all determinations. A detailed description of the method is given in Metrohm application bulletin No 231/2e. The data analyses were performed using PASW Statistics 18. Statistical methods including summary statistics, correlation analysis, and significance tests were employed in the data analyses. A probability

**Table 2. Instrument operating parameters for the analysis of the investigated heavy metals**

Parameters	Heavy metals	
	Ni and Co <sup>1</sup>	Zn, Cd, Pb and Cu <sup>2</sup>
Working electrode	HMDE	HMDE
Drop size	4	4
Stirrer speed	2000 rpm	2000 rpm
Mode	DP	DP
Purge time	300 s	300 s
Deposition potential	-0.7 V	-1.15V
Deposition time	90 s	90 s
Equilibrium time	10 s	10 s
Pulse amplitude	50 mV	50 mV
Start potential	-0.8 V	-1.15 V
End potential	-1.25 V	0.05 V
Voltage step	4 mV	6 mV
Voltage step time	0.3 s	0.1 s
Sweep rate	13 mV/s	60 mV/s
Peak potential	-1.13, -0.97V	-0.10, -0.98, -0.56, -0.38 V

<sup>1</sup>10 mL sample solution + 100 µL dimethylglyoxime solution (0.1 M) + 0.5 ml NH<sub>4</sub>Cl pH 9.5, <sup>2</sup>10 mL sample solution + 1 mL ammonium acetate buffer pH 4.6.

**Table 3. Heavy metal concentrations (average, minimum, maximum and standard deviation, n=184) in the Shahid Rajaei Dam Reservoir, Iran and guidelines (unit in µg/L)**

	Ni	Co	Zn	Cd	Pb	Cu
Mean	2.012	1.809	66.664	7.892	23.559	7.059
S.D.	1.080	1.750	39.087	6.129	12.991	9.618
Max	5.817	9.487	152.49	17.963	46.87	53.856
Min	0.158	0.278	9.617	0.249	4.034	0.940
Water quality Criteria (Drinking water quality)						
WHO <sup>a</sup>	70			3	10	2000
ISIRI <sup>b</sup>	70		3000 <sup>c</sup>	3	10	2000 (1000 <sup>c</sup> )
US EPA (MCLG) <sup>d</sup>				5	0.0	1300
US EPA (MCL) <sup>d</sup>				5	15	1300
Water quality Criteria (Protection of aquatic life) <sup>e</sup>						
China	100		100	5	100	10
USA	50		180	10	10	20
World average <sup>f</sup>	0.3	0.08	10	0.02	0.2	1.0
Danjiangkou Reservoir, China <sup>g</sup>	1.73	1.08	2.02	1.17	10.59	13.32
Mae Thang Reservoir <sup>h</sup>	0.3	0.15	2.92	0.01	0.03	0.46
Rawal Lake (mean of summer and winter data) <sup>i</sup>		107.5	18	15.5	192.5	13.5

Maximum contaminant level goal (MCLG), Maximum contaminant level (MCL)

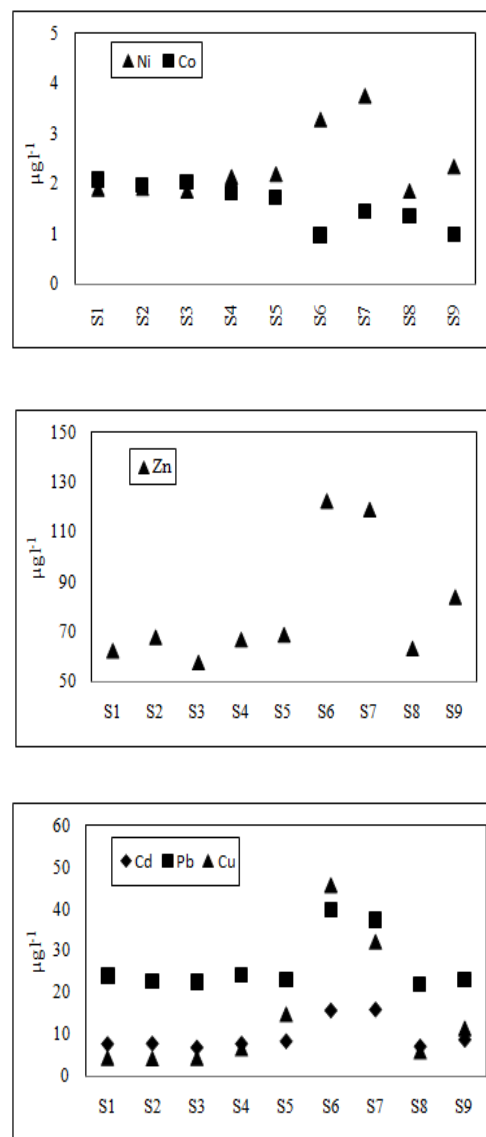
<sup>a</sup>(WHO 2011), <sup>b</sup>(ISIRI 2010), <sup>c</sup>Maximum desirable, <sup>d</sup>(U.S. Environmental Protection Agency 2012), <sup>e</sup>(Cheung, et al. 2003), <sup>f</sup>(Klavins, et al. 2000), <sup>g</sup>(Li, Xu, et al. 2008), <sup>h</sup>(Grellier, et al. 2013), <sup>i</sup>(Iqbal, Shah and Akhter 2013)

value of  $p < 0.01$  was considered as statistically significant in this research.

**RESULTS & DISCUSSION**

Heavy metals concentrations (in  $\mu\text{g/L}$ ) are presented in Table 3. Zn (66.664) is the dominant contributor, followed by Pb (23.559), Cd (7.892) and Cu (7.059). But a relatively lower concentration is observed for Ni (2.012). However, the lowest average level is found for Co (1.809). The metals exhibited the following decreasing concentration order:  $\text{Zn} > \text{Pb} > \text{Cd} > \text{Cu} > \text{Ni} > \text{Co}$ . Compared to the MCLs established by the World Health Organization (WHO 2011), Institute of Standards and Industrial Research of Iran (ISIRI 2010) and United States Environmental Protection Agency (U.S. EPA, 2012) for drinking water, the measured average concentrations of Cd and Pb are higher than the admissible values. Also, 59% and 69% of the total of 184 water samples exceeded WHO and ISIRI contamination limits for Cd and Pb, respectively whereas these percentages for US EPA standard are 56% and 63% for Cd and Pb, respectively. The concentrations of Cd and Pb are more than the Chinese and American criteria for protection of aquatic life value, respectively. Thus the main pollutants of heavy metals in the reservoir are Cd and Pb that may pose health risks to the regional residents and water receiving areas. By comparison with other studies, the concentrations of heavy metals in this study are higher than the world average, as well as those in Mae Thang reservoir, Thailand, and Danjiangkou reservoir, China (Cu is excluded). However, concentrations of heavy metals except for Zn in Rawal reservoir in Pakistan are higher.

The spatial distributions of dissolved heavy metals' concentrations are shown in Fig. 2. All heavy metals have rather uniform distributions in all stations except in S6 and S7 where concentrations are significantly higher. This is because the sampling in these stations were performed only in high concentration period of the whole reservoir (April 2013,



**Fig. 2. Spatial pattern of dissolved heavy metal concentrations (mean values) in the Shahid Rajaei Dam Reservoir, Iran**

**Table 4. Average entrance of heavy metals concentrations from Sefid Rud and Shirin Rud Rivers to the Shahid Rajaei Dam Reservoir in each season (unit in  $\mu\text{g/L}$ )**

River	Sampling Date	Ni	Co	Zn	Cd	Pb	Cu
Sefid Rud	Sep 2012	6.814	7.463	20.01	0.515	11.375	6.547
	Jan 2013	0.859	0.823	32.869	2.818	7.407	4.203
	Apr 2013	4.179	1.164	119.924	15.433	38.378	51.115
	Aug 2013	7.22	5.838	91.673	13.9	34.903	2.5
Shirin Rud	Sep 2012	3.76	7.076	19.303	0.549	10.73	7.418
	Jan 2013	1.263	0.892	23.922	1.883	8.014	3.816
	Apr 2013	1.87	2.059	131.183	16.2	40.295	47.8
	Aug 2013	1.471	3.619	78.139	11.78	30.661	1.435

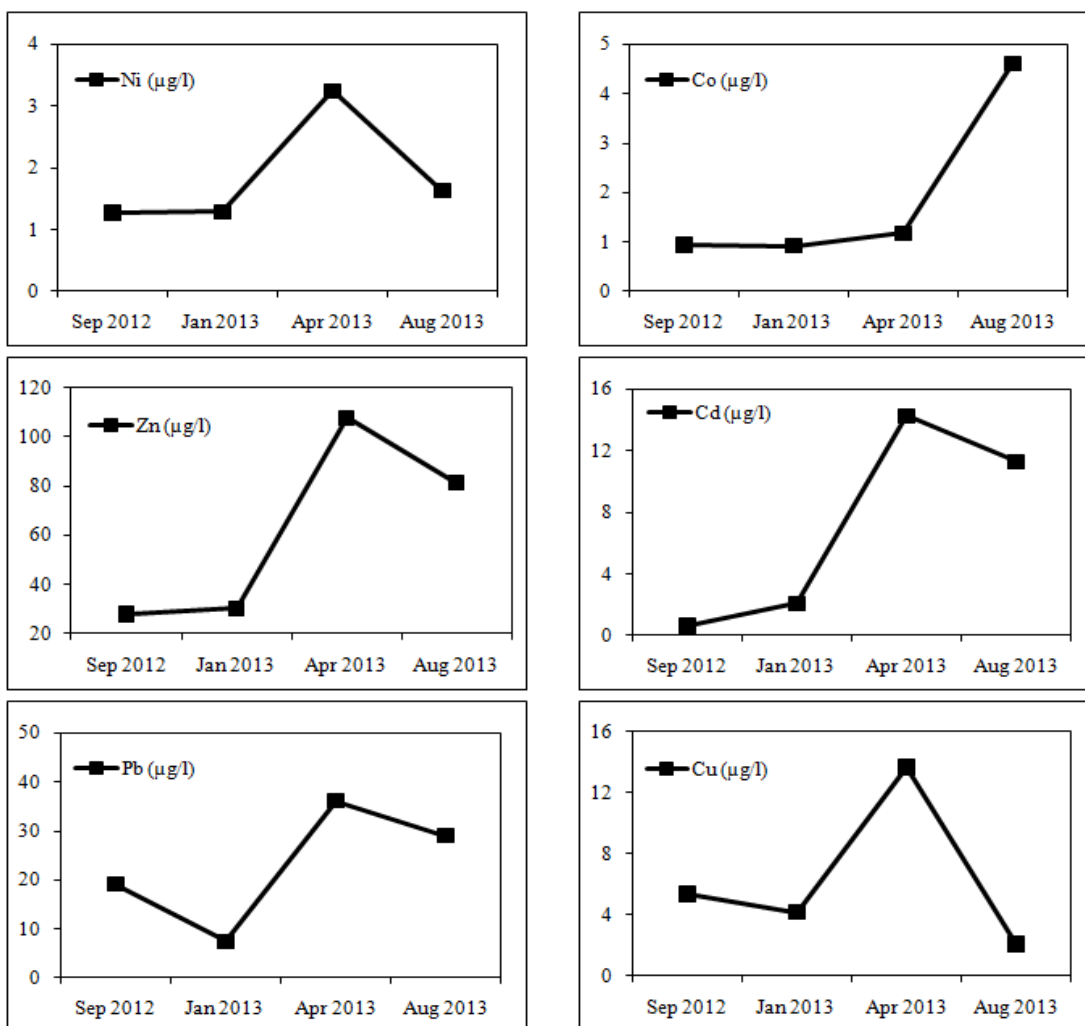


Fig. 3. seasonal variations of dissolved heavy metals' concentrations (mean value) in the Shahid Rajaei Dam Reservoir, Iran

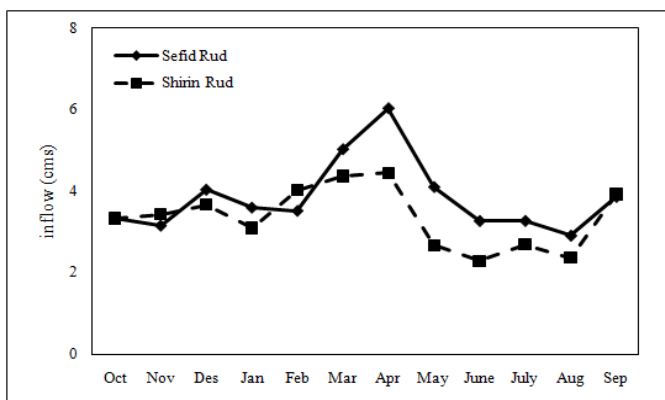


Fig. 4. seasonal variations of inflow (mean value) in the Shahid Rajaei Dam Reservoir, Iran

Fig. 3). In other sampling times the water surface level was lower than the bed elevation at these stations. This phenomenon was also partly seen in S9 located near Shirin Rud River entrance. Although a uniform distribution of Co concentration was seen in all stations. Zn, Cu and Pb contents showed large variations in the reservoir, from 57.731 (S3) to 122.255 (S6), 4.078 (S2) to 45.657 (S6) and 22.095 (S8) to 39.702 (S6) in  $\mu\text{g/L}$  respectively. Comparison of the average concentrations in Sefid Rud (S4 and S5) and Shirin Rud (S8 and S9) Rivers zones of the reservoir shows that the contents of heavy metals in Sefid Rud River zone are higher than Shirin Rud River zone, except for Zn. This can be caused by higher entrance of heavy metals from Sefid Rud River (Table 4) which can be attributed to the higher anthropogenic inputs due to most of the arable lands and natural inputs caused by existing of mineral streaks in upper stream of Sefid Rud River basin. Decreased heavy metal concentrations downstream of Sefid Rud (S5 to S4) and Shirin Rud (S9 to S8) River zones can be attributed to the dilution and sedimentation of particulate phase of heavy metals and decrease in water disturbance. This phenomenon was observed for all the investigated heavy metals except for Co in both zones and Pb in Sefid Rud River zone.

The seasonal variations of heavy metal'

concentrations are shown in Fig 3. Maximum concentrations of all heavy metals except Co were observed in April (start time for planting crops, particularly rice in the region). Also, concentrations of heavy metals except Cu in August were higher than September and January. The highest concentration of Co was observed in August. Because of the vast agricultural lands located in Sefid Rud and Shirin Rud Rivers' flood plains, the main source of heavy metal pollutants can be attributed to the agrochemicals, fertilizers, and pesticides which contain heavy metals. For example, Cd is found predominantly in phosphatic fertilizers because Cd is commonly present as an impurity in phosphatic rocks (Gimeno-Garcia, Andreu and Boluda 1996, Mann, Rate and Gilkes 2002, Otero, *et al.* 2005, Huang, *et al.* 2007). Also, superphosphate fertilizers contain Cd, Cu and Zn impurities and copper and iron sulphate are fertilizers containing Ni (Gimeno-Garcia, Andreu and Boluda 1996). Moreover, high values of Co and Pb in August can be attributed to the used pesticides in upstream farm lands (Gimeno-Garcia, Andreu and Boluda 1996, Markovic', *et al.* 2010). According to the flow data prepared by the Mazandaran Regional Water Company, the maximum inflow to the reservoir, which coincides with a period of high rainfall, occurs in April (Fig 4). Therefore, another reason of higher values of mentioned heavy metals in April can be rock weathering and

**Table 5. Correlation coefficient matrix of heavy metals in water samples in September (below the diagonal, n=30) and January (above the diagonal, n= 53)**

	Ni	Co	Zn	Cd	Pb	Cu
Ni	1	0.159	-0.002	-0.246	-0.152	-0.243
Co	0.135	1	-0.82	-0.129	0.006	-0.279
Zn	0.277	0.083	1	0.552	0.407	-0.006
Cd	0.069	0.138	0.614	1	0.644	0.25
Pb	0.161	0.226	0.597	0.638	1	0.009
Cu	0.062	0.022	0.552	0.582	0.301	1

**Table 6. Correlation coefficient matrix of heavy metals in water samples in April (below the diagonal, n=61) and August (above the diagonal, n= 40)**

	Ni	Co	Zn	Cd	Pb	Cu
Ni	1	0.497	0.04	-0.033	0.072	0.031
Co	0.599	1	-0.221	-0.208	-0.141	-0.088
Zn	0.183	0.046	1	0.742	0.73	0.464
Cd	0.174	0.028	0.763	1	0.92	0.425
Pb	0.184	0.032	0.668	0.906	1	0.487
Cu	-0.021	-0.057	0.407	0.397	0.37	1

transportation of sediments containing heavy metals due to higher values of rainfall in reservoir basin (McBride 1994).

A correlation analysis was also carried out in order to find out the appropriate relationships among the heavy metals as shown in Tables 5 and 6. A couple of strong correlations between Zn-Cd ( $R=0.614$ ) and Cd-Pb ( $R=0.638$ ) were observed in September, indicating a strong association between Cd with Zn and Pb in the water samples. It may share common sources with them such as herbicides (Gimeno-Garcia, Andreu and Boluda 1996). Some other significant correlations were also observed between Zn-Pb, Zn-Cu and Cd-Cu. A strong correlation between Cd-Pb ( $R=0.644$ ) and other significant correlations between Zn-Cd and Zn-Pb were observed in January. Because of agricultural inactivity at this time, the main sources of these heavy metals can be bedrock weathering (McBride 1994). Correlations observed between Zn, Cd and Pb were very strong in April and August. During these periods, the agricultural activities mainly planting rice in upstream lands of reservoir were superabundant. So at these times, the fertilizers and pesticides used in farm lands can be the main source of heavy metal pollutants such as Zn, Cd and Pb.

## CONCLUSION

The results indicated that Cd and Pb were the main pollutants in the Shahid Rajaei Dam Reservoir and it may pose health risks to residents and water receiving areas. Spatial distributions of heavy metals were rather uniform. Generally, the entered heavy metals from Sefid Rud River were partly higher than Shirin Rud River which may be due to higher anthropogenic and natural inputs to the upper streams. Seasonal variations of heavy metals indicated agricultural activities in upstream riverside lands of the reservoir. The sources of Ni, Zn, Cd and Cu could be mainly fertilizers and the main sources of Co and Pb could be pesticides. However, the natural sources such as bedrock weathering cannot be neglected. Further study is required to determine the exact sources of heavy metals along the upper streams.

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