

## Assessment of the effects of municipal wastewater on the heavy metal pollution of water and sediment in Arak Mighan Lake, Iran

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### Abstract

Heavy metal concentrations in surface water, wastewater and sediments collected from Mighan Lake in Markazi province, Iran were analyzed, and assessment contamination were evaluated according to the water and sediment quality guidelines. The results showed that the average concentrations of heavy metals in water were ranked as: Ni > Cr > Pb > Fe > Cu > Zn. In comparison with results the background values, the Mighan Lake was polluted by Ni, Cr, Pb and Fe, Most of metals might be mainly from industrial effluent and wastewater around the lake. In the sediments in the Mighan Lake, Fe, Ni, Pb and Cr showed a decreasing trend while Cu and Zn present an increasing trend. The increase of Cu and Zn might due to industrial activity around of Mighan Lake. The analysis of assessment contamination based on quality guidelines suggested that heavy metals in most water and sediments from the Mighan Lake had high toxicity, with Ni, Cr and Pb in water and moderate toxicity, with Zn and Cu in sediment being the highest priority pollutant.

**Keywords:** Assessment contamination, Heavy metal, Sediment, Mighan Lake, Wastewater.

### 1-Introduction

Lakes are considered one of the most versatile ecosystems in the world, but they are more sensitive to environmental pollution and anthropogenic impacts (Forghani *et al.*, 2009). Heavy metals, such as arsenic (As), mercury (Hg), cadmium (Cd), lead (Pb), and chromium (Cr), are commonly detected in lakes (Anshumali-Ramanathan *et al.*, 2009; Onger *et al.*, 2009). Like other aquatic systems, heavy metals enter lakes through natural sources (e.g. weathering, erosion) and anthropogenic sources (e.g. mining, urban and industrial wastewater) (Zorer *et al.*, 2008; Alhas *et al.*, 2009; Nasehi *et al.*, 2013; Ghomi *et al.*, 2013). At present, heavy metal pollution has become a great environmental concern with their toxicity in the food chain (Li *et al.*, 2008; Yuan *et al.*, 2011; Sarfo *et al.*, 2013). Thus, heavy metals in lakes might ultimately have adverse biologic effects on human health through drinking water and consuming aquatic products (Huang *et al.*, 2009; Zhang *et al.*, 2009). For these reasons, it would be desirable and imperative to investigate

their distribution in lakes, which can provide valuable information of heavy metal pollution and help evaluate potential environmental risks (Bu-Olayan and Thomas, 2013; Kapungwe, 2013). Mighan Lake is the largest natural saline lake in Markazi province. The lake covers an area of 110 km<sup>2</sup>, with an average water depth of 1.5 m. It is mainly used for animals and birds. With rapid development of the local economy large amounts of wastewater from industrial, domestic and agricultural sources from Arak city in Markazi province, Iran was discharged into the lake. As a result, water environment in the Mighan Lake has deteriorated in terms of heavy metal pollution (Bolhasani *et al.*, 2010). A number of recent studies have discussed the distribution and sources of heavy metals in various aquatic systems (Yao *et al.*, 2009; Kurun *et al.*, 2010; Botsou *et al.*, 2011). The aims of this study were to: 1) investigate the contamination levels and distributions of heavy metals in surface water, wastewater and surface sediment from the Mighan Lake; 2)

compare their concentrations with quality standard; 3) evaluate the contamination of the metal concentrations based on sediment quality guidelines and 4) assessment of contamination source.

## 2– Materials and method

### 2.1–Sample collection

In Jun 2012, 13 water samples at 0.5 m below the surface water and wastewater close to the Mighan Lake were collected at sampling locations shown in Figure 1. All of these water samples were obtained using cleaned polyethylene bottles, which were washed with hydrochloric acid and then rinsed with distilled water. Subsequently, water samples were filtered through 0.45  $\mu\text{m}$  millipore filters, acidified to  $\text{pH}<2$  with 2 mL 6 N HCl and transported to the laboratory for analysis. During the sample collection, a global positioning system (GPS) was used to locate the sites. A sediment sample for metal analysis was recovered at the central part of the Mighan Lake using a static gravity corer. Total 6 sediment samples were obtained from the sampling site. All the samples were then placed into sealed polyethylene bags and transported to the laboratory for storage at  $-20^{\circ}\text{C}$  until analysis. In the laboratory, samples were air-dried at room temperature and sieved through a 100–mesh sieve. For heavy metal analysis, 0.1 g of dry sample was digested using 10 mL HCl–HNO<sub>3</sub>–HF. The solution was finally diluted to 25 mL with deionized distilled water.

### 2.2– Sample analysis

Concentrations of Pb, Cd, Ni, Cr, Cu and Zn in all samples were determined using an inductively coupled plasma mass spectrometry (ICP–MS). For quality control, procedural blanks and duplicates were run for 6 samples. The standard deviations were below 10% for all elements. The detection limit for individual metals was 0.5 to 5 ng/L for a water

sample and 0.005 to 1  $\mu\text{g/g}$  for a sediment sample.

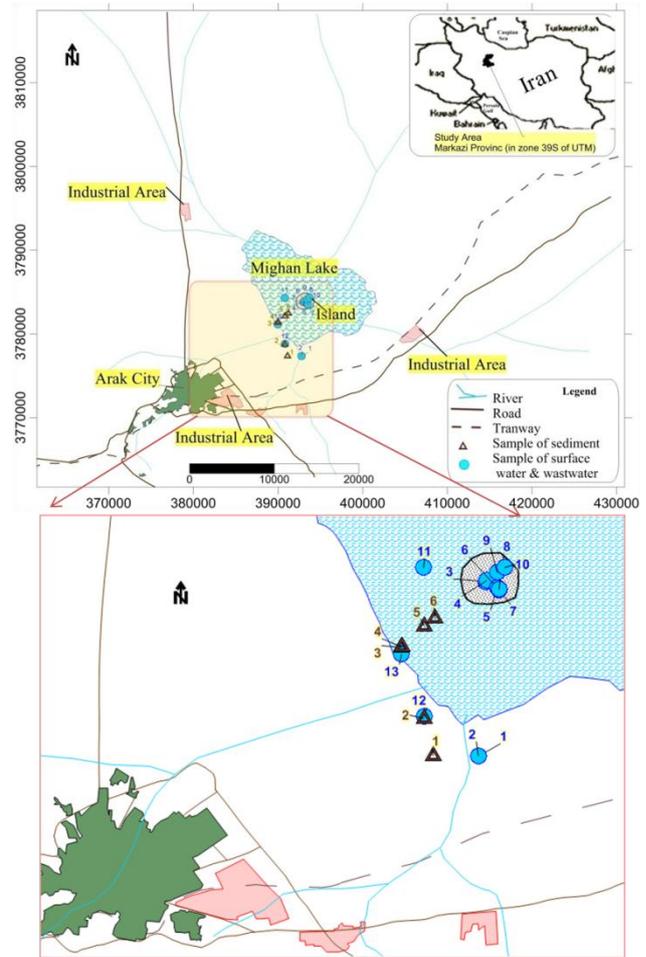


Figure 1) Location map of Mighan Lake and sampling sites of surface water, wastewater and sediment.

### 2.3–Geoaccumulation index (Igeo)

The geoaccumulation index allows estimation of contamination comparing premining and recent element concentrations. The geoaccumulation index is computed from the following Equation:

$$I_{geo} = \log_2 \left( \frac{C_n}{1.5B_n} \right) \quad \text{Eq. 1}$$

In the present work, geoaccumulation index was computed from the equation modified by Loska *et al.* (2004), where  $C_n$  is the measured concentration of the element in the surface water and wastewater samples and  $B_n$  is the permissible limit of WHO standard (Ogundiran and Afolabi, 2008). Müller divided the

geoaccumulation index into seven classes. They are as follows (Müller, 1969):

- $I_{geo} \leq 0$ : practically uncontaminated
- $0 < I_{geo} < 1$ : uncontaminated to moderately contaminated
- $1 < I_{geo} < 2$ : moderately contaminated
- $2 < I_{geo} < 3$ : moderately to heavily contaminated
- $3 < I_{geo} < 4$ : heavily contaminated
- $4 < I_{geo} < 5$ : heavily to extremely contaminated
- $5 \leq I_{geo}$ : extremely contaminated

## 2.4–Contamination degree

The quality of water surface and wastewater can be assessed with the use or calculation of environmental factors and indices, which include a wide range of parameters. Such factors may become valuable tool for the assessment of environmental condition of an area. According to Backman *et al.* (1998), contamination index ( $C_d$ ) may be considered as such if the measured concentration of parameters and the upper permissible levels of a contaminant are taken into account. According to Backman *et al.* (1998), contamination index is defined as Eq. 2 and Eq. 3:

$$C_d = \sum C_f \quad \text{Eq. 2}$$

$$C_f = \frac{CA}{CN} \quad \text{Eq. 3}$$

Where  $C_d$  is contamination degree;  $C_f$  is contamination factor, CA is analytical value of the component and CN is upper permissible concentration of the component according to WHO (2011). Hakanson (1980) suggested four classes of  $C_f$  to evaluate the metal contamination levels as follows (Loska *et al.*, 2004): low ( $C_f < 1$ ), moderate ( $1 \leq C_f < 3$ ), considerable ( $3 \leq C_f < 6$ ), and very high ( $6 \leq C_f$ ) contamination. The degree of contamination ( $C_d$ ) is the sum of contamination factors for all of the elements. Contamination degree ( $C_d$ ) is calculated individually for each water sample, as a sum of the contaminant

factors of single component that exceed the maximum contaminant levels (Ramos *et al.*, 2004). Hence, contamination degree summarized the combinational effects of several quality parameters that may have harmful consequences to health/the environment. The value scale for contamination degree consists of 4 ranges as (Edet and Offiong, 2002):

- $C_d < n$ : low degree of contamination
- $n < C_d < 2n$ : moderate degree of contamination
- $2n < C_d < 3n$ : high degree of contamination
- $C_d > 3n$ : very high degree of contamination

Where  $n$  is the number of contaminants involved in the  $C_d$  determination.

## 2.5–Statistical analysis

To identify relationship among metal data in surface water and wastewater and their possible sources, cluster analysis (CA) were performed using statistics software package Statistica Version 8 for windows. CA is the most common multivariate statistical methods used in environmental studies (Lambrakis *et al.*, 2004; Liu *et al.*, 2003; Love *et al.*, 2004). CA classifies a set of observations into two or more mutually exclusive unknown groups based on combination of internal variables. CA often group individual parameters and variables (Colby, 1993). The purpose of CA is to discover a system of organizing observations where a number of groups/variables share observed properties. A dendrogram is the most commonly used method of summarizing hierarchical clustering. In the current study, CA was used to evaluate the sources similarities metal data in water and sediments.

## 3–Results and discussion

### 3.1–Characterization of surface water and wastewater

#### 3.1.1–Hydrochemical data

The level of the detected constituents in water and wastewater samples (Table 1 and Fig. 2) shows that in all the sampling locations, Zn and Cu are low relative to the WHO standard limit. Cr, Pb, Ni and Fe were found to be above the maximum permissible limit of standard in all locations.

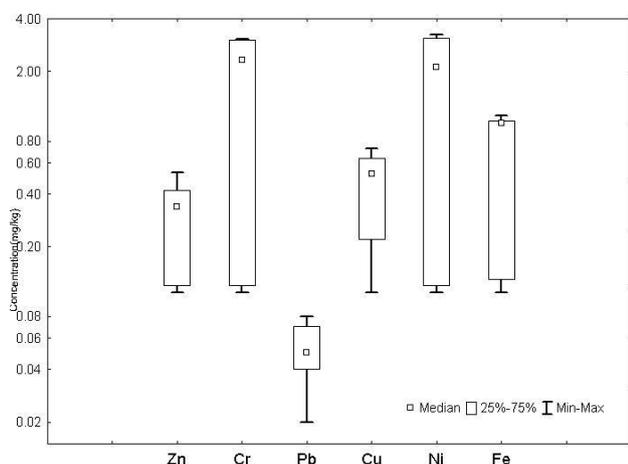


Figure 2) Concentrations for constituents in surface water and wastewater samples in Mighan Lake.

Table 1) Heavy metal data of surface water and wastewater samples (concentration is mg/kg).

| Sample No.       | Zn   | Cr   | Pb   | Cu   | Ni   | Fe   |
|------------------|------|------|------|------|------|------|
| 1                | 0.11 | 0.11 | 0.02 | 0.11 | 0.12 | 0.11 |
| 2                | 0.11 | 0.12 | 0.07 | 0.12 | 0.11 | 0.12 |
| 3                | 0.21 | 3.02 | 0.05 | 0.73 | 3.21 | 1.12 |
| 4                | 0.53 | 3.01 | 0.06 | 0.71 | 3.23 | 1.02 |
| 5                | 0.42 | 3.04 | 0.04 | 0.61 | 2.21 | 1.01 |
| 6                | 0.51 | 3.03 | 0.03 | 0.64 | 3.11 | 1.04 |
| 7                | 0.42 | 3.01 | 0.05 | 0.62 | 3.21 | 1.05 |
| 8                | 0.42 | 2.56 | 0.07 | 0.72 | 3.05 | 1.01 |
| 9                | 0.34 | 2.23 | 0.08 | 0.52 | 2.06 | 1.03 |
| 10               | 0.43 | 2.33 | 0.08 | 0.41 | 2.11 | 1.11 |
| 11               | 0.12 | 0.11 | 0.06 | 0.22 | 0.11 | 0.13 |
| 12               | 0.13 | 0.12 | 0.05 | 0.11 | 0.12 | 0.12 |
| 13               | 0.11 | 0.96 | 0.04 | 0.23 | 1.11 | 1.01 |
| Max.             | 0.53 | 3.04 | 0.08 | 0.73 | 3.23 | 1.12 |
| Min.             | 0.11 | 0.11 | 0.02 | 0.11 | 0.11 | 0.11 |
| Mean             | 0.30 | 1.82 | 0.05 | 0.44 | 1.83 | 0.76 |
| Reference Value* | 3    | 0.05 | 0.01 | 2    | 0.02 | 0.3  |

Figure 2 and Table 1 present the statistics of elemental constituents in water and wastewater of the study area. The result shows that the mean concentration of the examined parameters Zn, Cr, Pb, Cu, Ni and Fe included 0.30, 1.82, 0.05, 0.44, 1.83 and 0.76 mg/kg respectively.

Among the examined heavy metal constituents Cr, Pb, Ni and Fe have the highest mean while Zn and Cu remained the least. Further, the World Health Organization (WHO) standard limit for drinking water quality adopted to adjudge the suitability of surface water and waste water for drinking in the study area showed that only the parameters examined of Cr, Pb, Ni and Fe was found to be above the maximum permissible limit for drinking water standard. High Cr, Pb, Ni level noticed in water samples is characteristic of water in the around the industrial area which is due to the local wastewater treatment plant.

### 3.1.2–Geoaccumulation index (Igeo)

Contamination assessment showed that geoaccumulation index of Zn and Cu in all locations is practically uncontaminated (Fig. 3).

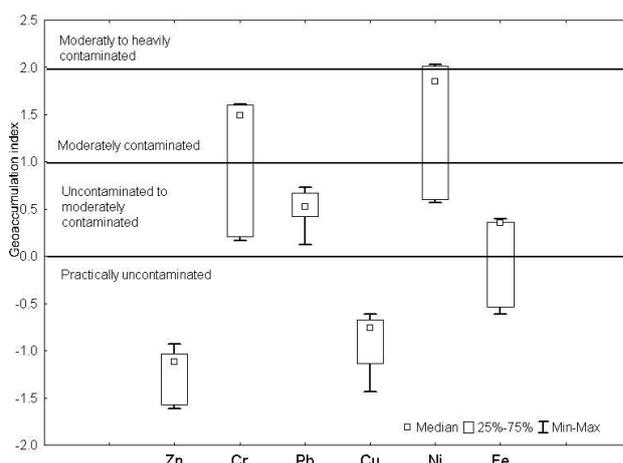


Figure 3) Geoaccumulation index for constituents in surface water and wastewater samples in Mighan Lake.

The mean concentration of Pb and Fe change from uncontaminated to moderately contaminated but Cr and Ni depict moderately contaminated. Based on Figure 3, the geoaccumulation index of Cr and Ni changes from uncontaminated to moderately contaminated in some of samples. Therefore, it is possible, there are three sources: Zn and Cu are of natural source; Pb and Fe are of natural and anthropogenic; Cr and Ni are of anthropogenic origin.

### 3.1.3–Contamination degree

The following terminology is adopted to describe the contamination degree ( $C_d$  values)

for seven substances indicating serious anthropogenic pollution:

- $C_d < 6$ : low degree of contamination
- $6 \leq C_d < 12$ : moderate degree of contamination
- $12 \leq C_d < 18$ : high degree of contamination
- $C_d \geq 18$ : very high degree of contamination

The computed contamination factor (Fig. 4 and Table 2) for the sampling shows that samples are very high contamination of Cr and Ni and are considerable contamination of Pb and Fe while, are low contamination of Zn and Cu.

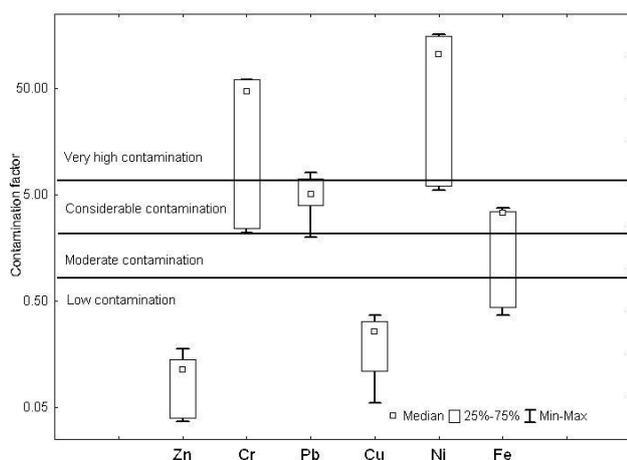


Figure 4) Contamination factor for constituents in surface water and wastewater samples in Mighan Lake.

Table 2) Contamination factors ( $C_f$ ) and contamination index ( $C_d$ ) in surface water and wastewater samples in Mighan Lake.

| Sampling No. | $C_f$ |      |    |      |     |      | $C_d$ | Contamination degree |
|--------------|-------|------|----|------|-----|------|-------|----------------------|
|              | Zn    | Cr   | Pb | Cu   | Ni  | Fe   |       |                      |
| 1            | 0.04  | 2.2  | 2  | 0.05 | 6   | 0.36 | 10    | Moderate             |
| 2            | 0.04  | 2.4  | 7  | 0.06 | 5.5 | 0.40 | 15    | High                 |
| 3            | 0.07  | 60.4 | 5  | 0.36 | 160 | 3.73 | 230   | Very high            |
| 4            | 0.17  | 60.2 | 6  | 0.35 | 161 | 3.40 | 231   | Very high            |
| 5            | 0.14  | 60.8 | 4  | 0.30 | 110 | 3.36 | 179   | Very high            |
| 6            | 0.17  | 60.6 | 3  | 0.32 | 155 | 3.46 | 223   | Very high            |
| 7            | 0.14  | 60.2 | 5  | 0.31 | 160 | 3.50 | 229   | Very high            |
| 8            | 0.14  | 51.2 | 7  | 0.36 | 152 | 3.36 | 214   | Very high            |
| 9            | 0.11  | 44.6 | 8  | 0.26 | 103 | 3.43 | 159   | Very high            |
| 10           | 0.14  | 46.6 | 8  | 0.21 | 105 | 3.70 | 164   | Very high            |
| 11           | 0.04  | 2.2  | 6  | 0.11 | 5.5 | 0.43 | 14    | High                 |
| 12           | 0.04  | 2.4  | 5  | 0.05 | 6   | 0.40 | 14    | High                 |
| 13           | 0.04  | 19.2 | 4  | 0.12 | 55  | 3.36 | 82    | Very high            |

Based on Cd values calculated for six elements (Zn, Cr, Pb, Cu, Ni and Fe) (Table 2), Mighan Lake generally presents contamination varying from a moderate degree of contamination to very high degree of contamination; the very

high and high degree of contamination belong to the most of samples in the surface water lake and wastewater of Arak city. In the study area, pollution can be related to wastewater of industrial activities.

### 3.1.4–Cluster analysis

The heavy metal data concentrations (the variables) were chosen to calculate for similarities in the variable. Then hierarchical clustering by applying single method was performed on the data set. The CA results for heavy metal data studied are shown in Figure 5 as a dendrogram. Figure 5 displays three clusters: (1) Ni–Cr; (2) Fe; (3) Pb–Cu–Zn. It is observed, however, that clusters 2 and clusters 3 join together at a relatively higher level, possibly implying a common source. The results imply that some different anthropogenic activities occurred in the surface water and wastewater that illustrated as a dendrogram in Figure 5 from the study area. Cluster 1 is composed of the Ni and Cr was mainly derived from anthropogenic inputs.

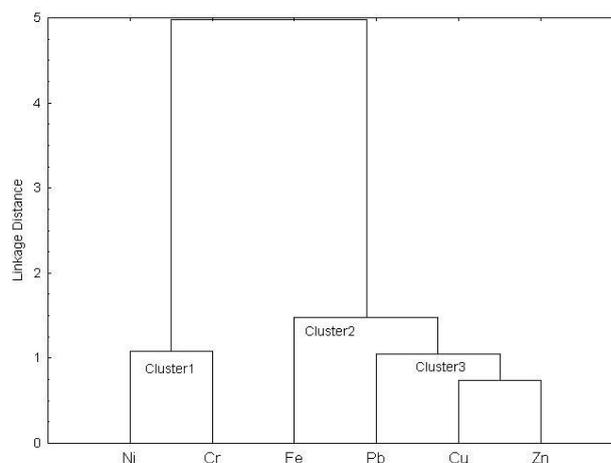


Figure 5) Dendrogram results single method of hierarchical cluster analysis for 6 variables.

Metal industrial activities (such as Aluminum Navard plant and Avangan plant) are the most important sources of heavy metals in wastewater (Table 3). Cluster 2 includes Fe was controlled by anthropogenic activity in the study area (such as Aluminum Navard plant, Avangan plant, Combain plant, Alemerol plant and Vagon Pars plant) (Table 3). Cluster 3 includes Pb, Cu and Zn appeared to be affected by both anthropogenic (wastewater of traffic activities, Aluminum Navard plant and Avangan plant)

and natural sources (especially Cu and Zn) (Solgi *et al.*, 2012; Kiham, 2013).

Table 3) Wastewater composition of metal plants in Arak city next to the Mighan Lake (concentration in mg/kg).

| Metal plant name* | Zn    | Cr   | Pb   | Cu   | Ni   | Fe   |
|-------------------|-------|------|------|------|------|------|
| Vagon-Pars        | 1.24  | 0.00 | 0.00 | 0.03 | 0.00 | 1.57 |
| Avangan           | 8.47  | 0.10 | 1.33 | 0.61 | 0.22 | 66   |
| Mashinsazi        | 0.03  | 0.00 | 0.05 | 0.00 | 0.00 | 0.32 |
| Combain           | 2.55  | 0.02 | 0.21 | 0.16 | 0.07 | 16   |
| Azarab            | 0.44  | 0.01 | 0.00 | 0.04 | 0.00 | 1.35 |
| Alemerol          | 0.86  | 0.02 | 0.05 | 0.04 | 0.05 | 6.14 |
| Al-Navard         | 12.81 | 0.27 | 0.57 | 0.90 | 0.42 | 117  |

### 3.2–Characterization of sediments

#### 3.2.1–Heavy metal concentrations of sediment samples

Summary of descriptive statistics such as minimum, mean, maximum for six elements used in this study are shown in Figure 6. Reference values of the studied metals (Caritat and Reimann, 2012; Taylor and McLennan, 1995) are also included Zn=52; Cr=35; Pb=17; Cu=14; Ni=19 and Fe=35000. Most of the elements have a wide range of variations of several magnitudes. This was evident for Zn, whose concentrations vary from 26 to 642 mg/kg with a mean of 174 mg/kg; for Cr from 15 to 80 with a mean of 29 mg/kg; for Pb from 8 to 17 with a mean of 12.43 mg/kg; for Cu from 10 to 33 with a mean of 18 mg/kg; for Ni from 14 to 19 with a mean of 17.43 mg/kg and for Fe from 3620 to 35000 with a mean of 12284 mg/kg (Fig. 6).

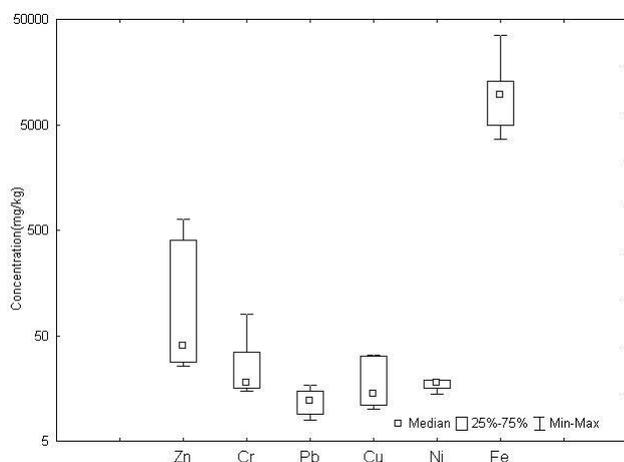


Figure 6) Concentration of metals (mg/kg) in sediment of Mighan Lake.

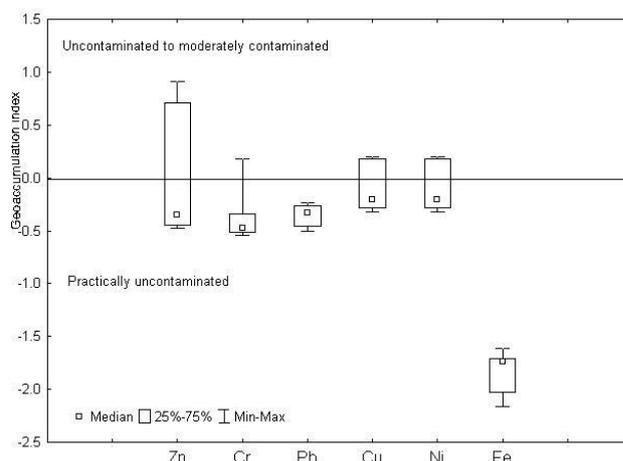


Figure 7) Geoaccumulation index for constituents in surface water and wastewater samples in Mighan Arak.

Cr, Pb and Fe concentrations in sediments of Mighan Lake are lower than values reported in the literature (Taylor and Mc Lennan, 1995), but the concentrations of Zn, Cu and Ni are higher than of upper continental crust. The anthropogenic sources of Zn, Cu and Ni are related to the metal industry activities around of Mighan Lake (Kabata–Pendias, 2000).

#### 3.2.2–Assessment of contamination

Assessment of contamination showed that Igeo of total elements are practically uncontaminated in most of locations (Fig. 7). In some of locations, Zn, Cu and Ni are uncontaminated to moderate contaminated. Therefore, it is possible that Cr, Pb and Fe are natural source and Zn, Cu and Ni are anthropogenic origin. The computed contamination factor (Fig.8 and Table 4) for the sampling shows that samples are low contamination for total of elements. While some of the samples show moderately to very high contamination for Zn and Cu (Fig. 8).

Table 3) Wastewater composition of metal plants in Arak city next to the Mighan Lake (concentration in mg/kg).

| Metal plant name* | Zn    | Cr   | Pb   | Cu   | Ni   | Fe   |
|-------------------|-------|------|------|------|------|------|
| Vagon-Pars        | 1.24  | 0.00 | 0.00 | 0.03 | 0.00 | 1.57 |
| Avangan           | 8.47  | 0.10 | 1.33 | 0.61 | 0.22 | 66   |
| Mashinsazi        | 0.03  | 0.00 | 0.05 | 0.00 | 0.00 | 0.32 |
| Combain           | 2.55  | 0.02 | 0.21 | 0.16 | 0.07 | 16   |
| Azarab            | 0.44  | 0.01 | 0.00 | 0.04 | 0.00 | 1.35 |
| Alemerol          | 0.86  | 0.02 | 0.05 | 0.04 | 0.05 | 6.14 |
| Al-Navard         | 12.81 | 0.27 | 0.57 | 0.90 | 0.42 | 117  |

Based on Table 4 sediments of Mighan Lake generally present contamination varying from a low degree of contamination to high degree of contamination; the high degree of contamination is belong to sample 1 and sample 2. Pollution can be related to a industrial activities (such as Aluminum Navard plant, Avangan plant, Combain plant, Alemerol plant and Vagon Pars plant).

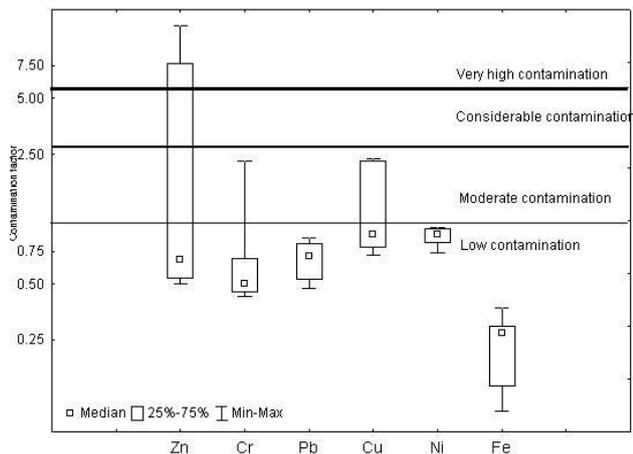


Figure 8) Contamination factor for constituents in surface water and wastewater samples in Mighan Arak.

### 3.2.3–Cluster analysis

Using single–linkage and Pearson's correlation coefficients cluster analysis (hierarchical cluster analysis) was carried out and the results are given in a dendrogram (Fig. 9).

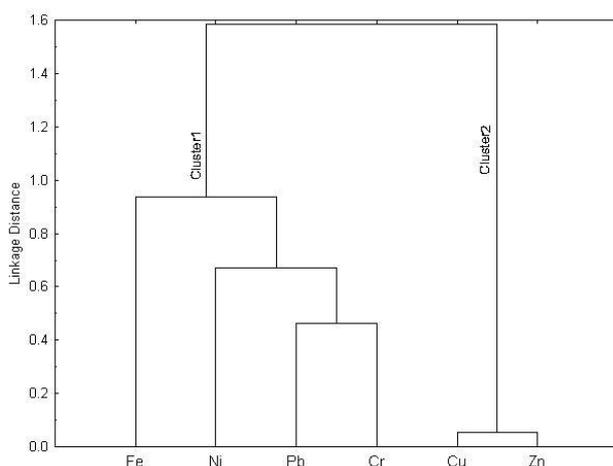


Figure 9) Dendrogram results of hierarchical cluster analysis for 6 variables.

Results of cluster analysis indicate that the elements comprise two groups. The first group is consisting of Fe, Ni, Pb and Cr. The second

group consisting of Cu and Zn. Cu and Zn source clearly represents the anthropogenic contribution, such as metal industrial activities (Ozsoy and Ornektekin, 2009; Song and Gao, 2009). These elements were enriched in comparison with crustal materials in some of the samples as shown in Figure 8. The cluster is usually showed from crustal sources or natural origins in sediments.

## 4–Conclusions

The study of the distribution of heavy metals in surface water, wastewater and the sediment from the Mighan Lake suggests that the lake is facing heavy metal pollution. In surface water and wastewater, abundances of heavy metals in water were ranked as: Ni> Cr> Pb> Fe> Cu> Zn. Some heavy metals exceeded background values, indicating that the Mighan Lake is polluted with some heavy metals. The spatial distribution of these metals indicated that they might be related to industrial effluent and wastewater around the lake. In the sediments, increase of Cu and Zn concentration reflected that this metal in the sediments might originate from the anthropogenic activities. Decrease in concentration of Cr, Pb, Ni and Fe was likely associated with the natural sources. Lake should be of high concern, and Cr, Ni in surface water and Cu, Zn in sediments were the highest priority pollutants.

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