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# The Effect of Industrial Development on Health Hazard Risk Posed by Drinking Water Containing Heavy Metals with HRA<sub>EPA</sub> Index, Case Study; Semnan Province

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#### Abstract

**Background:** Developmental activities especially the establishment of more than 20 industrial units in Semnan province caused the officers worries about increased concentration of heavy metals in drinking water supplies and probably increasing of health risk at consumers. The aim of the study was to investigate the effect of Semnan province industrial development on the health risk level caused by drinking waters heavy metals with HRA<sub>EPA</sub> index.

**Methods:** For this descriptive-analytical study, sampling from 17 wells was perfumed at first. Then the heavy metals concentrations were measured at laboratory with Standard methods (Arsenic, Cadmium and Mercury with Atomic Absorption GTA-95- Lead and Chromium with Atomic Absorption Spectrophotometry). Meantime, HRA<sub>EPA</sub> index was calculated with three step process of health risk assessment. Statistical analysis was performed by Repeated Measure One sample T-test and One-Way ANNOVA methods. Finally, zoning map of heavy metals in Semnan province has been prepared with GIS.

**Results:** Results showed that the levels of all heavy metals are at national, WHO and EU standard ranges. HRA<sub>EPA</sub> indexes was 4.48/10-4, 4.36/10-4 and 4.46/10-4 for 2001-2011, 2012-2015 and 2015 period, respectively. Also, the highest and lowest HRA<sub>EPA</sub> index was for 2001-2011 and 2015, respectively.

**Conclusions:** The study showed that the heavy metals concentrations in groundwater resources werelower than threshold toxic level. However with the industrial development started at Semnan province, water resources quality must be protected by law enforcement and tight supervision on industrial and mining-excavation activities.

Keywords: Heavy metals, Health risk, Semnan, Industrial development.

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# Introduction

Development activities to fulfill human needs have negatively influenced natural resources and the environment.<sup>1,2</sup> Concentrations of heavy metals, a group of pollutants, in water resources have significantly increased during recent years.<sup>3</sup> More than 50 elements are categorized as heavy metals; 17 of them are very common and toxic, with the potential to cause many serious and adverse effects on humans, living species, and the environment.<sup>4,5</sup> These toxic elements include lead, mercury, arsenic, and cadmium.<sup>5</sup> Of the several ways by which heavy metals enter drinking water, waste water, sewage, and industrial waste are some of the most common ones.<sup>6</sup> These metals are naturally present in the earth layers and in surface and ground waters. If their amount increases above the natural baseline, it will pose a serious problem, considering their stability, poor degradability, and ability for bioaccumulation in organisms.<sup>7,8</sup> These elements form bonds with other molecules, such as oxygen, sulfur, and nitrogen, which are common constituents of enzymes and proteins in the body and thereby interrupt the normal functions of such substances.<sup>9</sup> The main issue that makes heavy metals a serious problem is the inability of the human body to metabolize such elements. In fact, after entering the body, heavy metals are not excreted; instead, they accumulate in body tissues, such as fat, muscle, bones, and joints, and cause several diseases and abnormalities.<sup>10,11</sup>

To date, many studies have been conducted to determine the concentrations of heavy metals in water resources, food chain, and soil. However, few of them aimed to investigate the potential health hazard posed by such elements. In Iran, only one study has been conducted to assess this issue; in that study, the concentration of heavy metals was monitored over a 6month period, which is too short a time to be able to assess the actual risk of these elements, considering their cumulative nature. The results of the study revealed that the average concentration of these elements in drinking water in Hamadan province, Iran, is above the level recommended by the standards.<sup>12</sup>

Savari et al. assessed the possibility of heavy metals entering drinking water through the corrosion of the drinking water pipeline network; they reported the concentrations of lead, cadmium, chromium, iron, and manganese as 8.48, 0.97, 3180, 168, and 30.6  $\mu$ g/L, respectively.<sup>13</sup> It was reported by Miranzade et al.<sup>14</sup> that the concentrations of heavy metals in the drinking water network of Kashan, Iran, was not above national and international standards and did not pose any risk to consumers. Samir et al. assessed the concentrations of heavy metals in the North Delta Lake and in the animals living in that region and concluded that the concentrations of heavy metals in water, fish tissues, and deposits on the floor of the lake were higher than those recommended by international standard bodies.<sup>15</sup>

The situation in various geographical regions of the world is different. In recent years, there has been an increasing interest in determining the concentrations of heavy metals in drinking water networks and many studies have been conducted in this regard, for example, those by Gabarkidan et al.<sup>16</sup>, Chen et al.<sup>17</sup>, Mohammed et al.<sup>18</sup>, and Cao et al.<sup>19</sup>Gabarkidan et al. evaluated the health risk related to heavy metals in soil, rice, and vegetables grown in the vicinity of

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Gibfel River in the north of Ethiopia. They reported that the concentrations of these hazardous elements were higher than the limits recommended by the relative standards.<sup>16</sup> Chen et al. conducted their study on vegetables irrigated with waste water. They concluded that the concentrations of heavy metals, such as lead and cadmium, were higher than the permissible limits and the concentration of mercury was comparable to what was proposed by the relative standards.<sup>17</sup>

Considering the recent industrial development in Semnan province of Iran, and the establishment of more than 30 chemical plants in the past four years, many concerns have been raised about the quality of the water resources in this region; accordingly we set this investigation to evaluate changes in the heavy metals concentrations over past years. Moreover, the annual health hazard risk posed by the drinking water polluted with these elements was assessed using a new index known as HRA<sub>EPA</sub>, in several time intervals.

### **Materials and Methods**

The present study was conducted in Semnan province, Iran. The number of people in this province is estimated to be 631218 individuals. The area of the province is 97491 sq. km and is located to the east of Tehran, about 220 km away. Figure 1 demonstrates the location of the province. The latitude of the area is 51 to 57 degrees and the longitude of the area is 34 to 37 degrees. The height of the region is 1630 m above sea level. The northern neighbors of the province are Mazandaran and Golestan provinces, the southern neighbor is Esfahan province, the eastern neighbors are KhorasanRazavi and Jonobi, and finally the western neighbors of this province are Tehran and Qom. Moreover, the province is located in the southern vicinity of the Alborz Mountains and its height is reduced moving from the north to the south, which ends in the Kavir Desert.<sup>20</sup>

The main resources of the province drinking water are 41 wells, which are located in various areas of the province <sup>20</sup>. In the present study, we used the data available on the concentrations of heavy metals in drinking water for the past 10 years. In addition, we measured the concentrations of these elements in ground water resources by sampling from homes, which used these resources for one year. These new measurements were taken owing to several reasons; to validate the results of previous measurements and to assess the changes in the concentrations of heavy metals considering the establishment of several new plants in the area. We conducted the sampling from the waters of all the wells seasonally. According to the standards recommended by the standard method book (2008), the all-sampling apparatus was made of plastic and glass polyethylene. Before being used, all the sampling apparatus was cleanly washed in three steps with a diluted detergent, sulfuric acid, and deionized water. In the next step, all apparatus was exposed to a heat of 180°C for 1 h to dry. As mentioned previously, all water samples were gathered from home drinking water. First, the faucet was removed, the pipe was physically cleaned, and finally, after 2 min, the sampling process was performed. The samples were carefully and quickly transported to the laboratory of the Water and Sewage Company of Semnan province and the concentrations of heavy metals in the samples were evaluated according the standard method. The concentrations of arsenic, cadmium, and mercury were determined using the Atomic Absorption Spectroscopy method with the graphite furnace GTA-95, and lead and zinc were quantified using the Atomic Absorption Spectroscopy. The results of this step are later used for the health hazard risk assessment. It is worth noting that the quantities of copper, nickel, and zinc that were reported as low as negligible by the measurements conducted in previous years were proven by several sampling trials, so we made a decision to exclude these three elements from the study.

The results obtained from the field sampling over years were analyzed using Statistical Package for the Social Sciences software package. One-way analysis of variance and the repeated measure test were some statistical tests applied in the present study for comparing the mean of metals concentrations and their health hazard risk over the years <sup>21</sup>. Moreover, the normality of the variables (the concentration of heavy metals) was analyzed using Kolmogorov-Smirnov test and for comparing the mean of concentrations to the national and international standards<sup>22</sup> the One-way sample T-test was employed. In Table 1, the permissible limits for the concentration of heavy metals in drinking water recommended by national and international bodies, are presented.

The HRA<sub>EPA</sub> index is regarded as one of the most suitable ways to approximate the health risk posed by the presence of heavy metals in drinking water. The index is promulgated by United States Environmental Protection Agency (USEPA).<sup>23</sup> Three steps are needed to calculate the index:

Step 1; calculating ADD<sub>d</sub>,

(1)  $ADD_d = DW \times C/BW$ 

Where  $ADD_d$  is the daily dose of a hazardous substance entering the body by drinking water, DW is the volume average of daily water in individual drinks, C is the concentration of the hazardous substance in water ( $\mu$ g/L), and BW is the body weight.

#### (2) Step 2; Risk = $ADD_d \times Ur$

Where  $ADD_d$  is the daily dose of the hazardous material, Ur is the unit of risk related to the concentration or the available dose of the hazardous material; Ur also is related to the adverse effect of the element on the human body.

(3) Step 3;  $\operatorname{Risk}_{\operatorname{sum}} = 1 - (1 - \operatorname{Risk}_1) \times (1 - \operatorname{Risk}_2) \times (1 - \operatorname{Risk}_n)$ 

In the final step, the total risk of all elements is calculated. In equation 3,  $Risk_{sum}$  is the total risk and  $Risk_n$  is the risk posed by element n.

In order to provide a map representing the location of each water well, the geographical coordinates of each well were determined using a GPS navigation device (Oregon, GPS 550, UTM format). After gathering the coordinates of wells, the final conversion of data was performed using the Arc GIS software, and the outputs were transformed into digital layers with the point format. In the next step, the qualitative tables of the wells' locations were updated in order to construct a map demonstrating the level of heavy metals concentrations in each well based on the total risk scores and HRA<sub>EPA</sub> index.

#### **Results**

The results of water sampling from the wells: After sampling from the water wells and analyzing the samples, the concentrations of heavy metals were quantified and the results are presented in Table 2.

The annual average of the heavy metals' concentrations from 2012 to 2015 is also presented in Table 3. As explained in the previous section, some data were directly gathered by the authors, and some were obtained from the archives of the Semnan province Water and Sewage Co.

The health hazard risk in terms of carcinogenicity calculated for each element identified in ground water resources of Semnan province: The results of the health hazard risk assessment (carcinogenic and non-carcinogenic) calculated for heavy metals in groundwater resources are presented in Table 5 and Figure 2.

The analysis of the results in GIS software: The coordinates of the wells investigated in the present study are shown in Table 6.

The concentrations of heavy metals in various wells in different time intervals, depicted using GIS, is exhibited in Figure 3.



Figure 1. The studied area

Table 1: The permissible concentration of heavy metals in drinking water recommended by national and international bodies.

Standard		Concentration (mg/L)					
Stanuaru	Pb	Cd	Hg	Cr	As		
National	0.05	0.05	0.05	0.05	0.05		
WHO	0.01	0.05	0.01	0.05	0.01		
European Union	0.01	0.01	0.05	0.05	0.05		

Table 2. The annual average concentrations of the heavy metals obtained by sampling from Semnan province
drinking water in 2015 (mean±SD).

uninking water in 2015			Concentration	(ug/L)		
Well Name	As	Pb	Cd	Hg	Cr	Total
Semnan	1.2±0.5	5.5±0.6	0.2±0.08	14.5±6	1±0.4	22.4
Shahroud	1.1±0.6	5.3±0.5	0.1±0.06	14.6±3.4	1.3±0.5	22.4
Garmsar	1.3±0.4	5.4±0.4	0.3±0.07	14.2±3.9	1±0.3	22.2
Damghan	1.4±0.6	5.6±0.5	0.2±0.06	14.1±3.8	1.8±0.4	23.1
Darjezin	1.4±0.6	5.3±0.4	0.1±0.04	14.8±3.8	1.9±0.5	23.5
Sorkheh	1.1±0.7	5.1±0.6	0.3±0.06	14.9±2.7	1.4±0.6	22.8
Shahmirzad	1±0.4	5.7±0.7	0.9±0.04	14.7±5.9	1.8±0.7	24.1
Ivanaki	1.3±0.4	5.8±0.3	0.8±0.07	14.2±4.9	1.7±0.8	23.8
Bastam	1.4±0.6	5.9±0.7	0.9±0.04	13.6±4.7	1.3±0.5	23.1
Aradan	1.8±0.6	4.9±0.6	0.8±0.06	13.7±5.2	1.7±0.8	22.9
Mojen	1.1±0.6	5.1±0.3	0.2±0.06	12.6±5	0.9±0.5	19.9
Kalatehkhij	1.2±0.5	5.4±0.5	0.4±0.05	13.7±4.5	1.4±0.6	22.1
Mahdishahr	1.1±0.4	5.8±0.4	0.2±0.07	14.2±3.5	1.7±0.6	23
Dibaj	1.4±0.6	5.9±0.8	0.4±0.03	14.1±3.6	1.8±0.7	23.6
Miami	3±0.7	6.5±0.4	0.6±0.04	13.8±5.2	1.3±0.4	25.2
Biarjmand	1.1±0.4	6.3±0.6	0.6±0.05	14.5±6	1.5±0.4	24
Amirieh	1.1±0.5	6.5±0.5	0.8±0.05	13.2±4	1.1±0.6	22.7
Average	1.35±0.54	5.6±0.53	0.458±0.056	14±4.51	1.38±0.55	22.8

Table 3. Four-year average concentrations of heavy metals in groundwater resources in Semnan province between 2012 and 2015 (mean±SD).

Well Name			Concentration	n (μg/L)		
	As	Pb	Cd	Hg	Cr	Total
Semnan	1±0.2	4.5±0.2	0.2±0.04	11.2±1	1±0.1	17.9
Shahroud	0.9±0.4	3.3±0.1	0.1±0.01	12.1±2.3	0.8±0.2	17.2
Garmsar	1.1±0.1	3.5±0.2	0.2±0.02	12.1±2.1	0.8±0.1	17.7
Damghan	1.3±0.5	4.3±0.2	0.18±0.02	13.2±3.3	1.7±0.3	20.7
Darjezin	1.1±0.3	5.1±0.2	0.07±0.02	13.2±3.2	1.4±0.3	20.9
Sorkheh	1±0.6	5±0.3	0.3±0.03	13.1±1.9	1.2±0.5	20.6
Shahmirzad	1±0.2	5.4±0.3	0.9±0.02	14.7±4.6	1.8±0.5	23.8
Ivanaki	1.2±0.2	5.6±0.3	0.8±0.05	14.1±4.3	1.6±0.6	23.3
Bastam	1.2±0.5	5.6±0.2	0.9±0.02	13.4±4.4	1.1±0.4	22.2
Aradan	1.8±0.4	4.7±0.5	0.8±0.04	13.5±5.4	1.7±0.3	22.5
Mojen	1.1±0.3	5±0.1	0.2±0.03	12.2±4	0.9±0.04	19.4
Kalatehkhij	1.1±0.3	5.3±0.2	0.4±0.04	13.4±4.1	1.3±0.4	21.5
Mahdishahr	1.2±0.5	5.8±0.5	0.2±0.05	14.2±3.1	1.7±0.5	23.1
Dibaj	1.4±0.5	5.7±0.6	0.4±0.02	14±3.2	1.8±0.4	23.3
Miami	3±0.6	6.3±0.3	0.6±0.03	13.6±5.1	1.2±0.3	24.7
Biarjmand	1.1±0.3	6.4±0.5	0.6±0.04	14.8±5	1.5±0.5	24.4
Amirieh	1.1±0.6	6.4±0.4	0.8±0.04	13.4±5	1.1±0.7	22.8
Average	1.2±0.38	5.2±0.38	0.45±0.036	13.4±4.8	1.3±0.43	24.4

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Table 4.Ten-year average concentrations of heavy metals in groundwater resources in Semnan province between 2001 and 2011 (mean±SD).

Well Name	Concentration (µg/L)					
	As	Pb	Cd	Hg	Cr	Total
Semnan	1.6±0.5	3.2±0.5	0.25±0.06	13.4±3	1.5±0.5	19.9
Shahroud	0.6±0.05	3.7±0.4	0.23±0.04	11.9±2	0.6±0.06	17
Garmsar	1.2±0.6	4.2±0.35	0.31±0.03	12.7±3.6	0.4±0.05	18.8
Damghan	1.1±0.5	3.6±0.45	0.21±0.05	14.5±2.4	1.2±0.3	20.6
Darjezin	1.2±0.5	3.9±0.5	0.4±0.06	13.1±2.8	1.1±0.35	19.7
Sorkheh	2±0.3	6±0.9	0.5±0.07	13.6±3.8	1.1±0.5	23.2
Shahmirzad	1.6±0.5	3.5±0.56	0.6±0.05	13.2±4.6	1.2±0.4	20.1
Ivanaki	1.1±0.5	4.2±0.39	0.6±0.04	13.1±3.4	1.2±0.6	20.2
Bastam	1.4±0.4	6.5±0.27	0.56±0.05	14.7±2.9	1.5±0.1	24.7
Aradan	1.3±0.28	4.2±0.19	0.76±0.02	12.9±2.4	1.4±0.3	20.6
Mojen	1.2±0.42	5.7±0.28	0.6±0.02	13.6±2.1	1.2±0.2	22.3
Kalatehkhij	1.1±0.5	4.6±0.47	0.7±0.05	12.1±5.6	1.5±0.4	20
Mahdishahr	1.2±0.4	5.2±0.31	0.6±0.03	14.8±4.7	1.4±0.35	23.3
Dibaj	1.2±0.21	5.3±0.23	0.9±0.01	15±4.1	1.6±0.59	24
Miami	2.9±0.54	5.8±0.56	0.9±0.06	14.7±3.5	1.6±0.26	25.9
Biarjmand	13±0.36	4.3±0.32	0.7±0.04	12.4±4.8	1.2±0.19	19.9
Amirieh	1.3±0.11	4.5±0.48	0.6±0.05	12.2±2.9	1.3±0.35	19.9
Average	1.37±0.38	4.61±0.38	0.55±0.026	13.4±3.9	1.23±0.32	21.2

#### Table 5. The results of the health hazard risk assessment in water ground resources over the years

Period	Element	C (mg/l)	ADDd (mg/kg)	UR <sub>cancer</sub> (kg-day/mg)	$Risk_{cancer}$	UR <sub>noncancer</sub> (kg-day/mg)	Risk <sub>noncancer</sub>
	As	0.00135	5.78×10 <sup>-5</sup>	1.5	8.67×10 <sup>-5</sup>	0.003	1.73×10 <sup>-7</sup>
	Cr	0.00138	5.91×10 <sup>-5</sup>	0.1	5.91×10 <sup>-6</sup>	0.005	2.95×10 <sup>-7</sup>
2015	Hg	0.0134	$5.74 \times 10^{-4}$	0.6	3.44×10 <sup>-4</sup>	0.0035	2.01×10 <sup>-6</sup>
2015	Cd	0.000458	1.96×10 <sup>-5</sup>	0.38	7.45×10 <sup>-6</sup>	0.0005	9.8×10 <sup>-9</sup>
	Pb	0.0056	0.00024	0.0085	2.04×10 <sup>-6</sup>	0.000785	1.88×10 <sup>-8</sup>
	Total health risk	-	-	-	4.46×10 <sup>-4</sup>	-	2.51×10 <sup>-6</sup>
	As	0.0012	5.14×10 <sup>-5</sup>	1.5	7.71×10 <sup>-5</sup>	0.003	1.54×10 <sup>-7</sup>
2012	Cr	0.0013	5.57×10 <sup>-5</sup>	0.1	5.57×10 <sup>-6</sup>	0.005	2.79×10 <sup>-7</sup>
2012 to	Hg	0.0134	$5.74 \times 10^{-4}$	0.6	3.44×10 <sup>-4</sup>	0.0035	2.01×10 <sup>-6</sup>
2015	Cd	0.00045	1.92×10 <sup>-5</sup>	0.38	7.29×10 <sup>-6</sup>	0.0005	9.6×10 <sup>-9</sup>
2015	Pb	0.0052	2.23×10 <sup>-4</sup>	0.0085	1.89×10 <sup>-6</sup>	0.000785	$1.75 \times 10^{-8}$
	Total health risk	-	-	-	4.36×10 <sup>-4</sup>	-	2.47×10 <sup>-6</sup>
	As	0.00137	5.87×10 <sup>-5</sup>	1.5	8.81×10 <sup>-5</sup>	0.003	1.76×10 <sup>-7</sup>
2001	Cr	0.00123	5.27×10 <sup>-5</sup>	0.1	5.27×10 <sup>-6</sup>	0.005	2.63×10 <sup>-7</sup>
2001 to	Hg	0.0134	$5.74 \times 10^{-4}$	0.6	3.44×10 <sup>-4</sup>	0.0035	2.01×10 <sup>-6</sup>
	Cd	0.00055	2.35×10 <sup>-5</sup>	0.38	8.93×10 <sup>-6</sup>	0.0005	1.18×10 <sup>-8</sup>
2011	Pb	0.00461	1.97×10 <sup>-4</sup>	0.0085	1.68×10 <sup>-6</sup>	0.000785	1.55×10 <sup>-8</sup>
	Total health risk	-	-	-	4.48×10 <sup>-4</sup>	-	2.48×10 <sup>-6</sup>

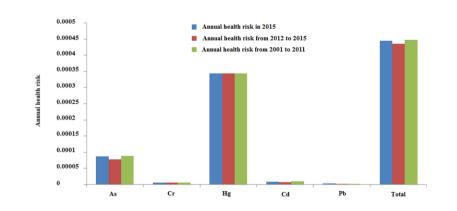


Figure 2. The comparison of the health hazard risk posed by different heavy metals over the years

Table 6.The	coordinates of	f the wells in	the GIS

Well Name	E	N
Semnan	53 <sup>0</sup> 23'34.01''	35 <sup>0</sup> 36'39.53''
Shahroud	54 <sup>0</sup> 57'35.79''	36 <sup>°</sup> 28'26.10''
Garmsar	52 <sup>°</sup> 24'49.70''	35 <sup>0</sup> 17'50.26''
Damghan	54 <sup>0</sup> 20'25.28''	36 <sup>0</sup> 08'42.08''
Mahdishahr	53 <sup>0</sup> 21'46.81''	35 <sup>°</sup> 44'54.57''
Sorkheh	53 <sup>0</sup> 12'48.07''	35 <sup>°</sup> 28'12.44''
Shahmirzad	53 <sup>0</sup> 18'47.03''	35 <sup>°</sup> 46'51.79''
Ivanaki	51 <sup>0</sup> 52'30.82''	35 <sup>°</sup> 22'36.54''
Bastam	54 <sup>0</sup> 59'12.90''	36 <sup>0</sup> 30'12.66''
Aradan	52 <sup>0</sup> 29'34.07''	35 <sup>°</sup> 16'07.06''
Mojen	54 <sup>0</sup> 38'18.42''	36 <sup>°</sup> 29'18.02''
Kalatehkhij	55 <sup>°</sup> 18'42.19''	36 <sup>°</sup> 42'09.24''
Darjezin	53 <sup>0</sup> 19'31.94''	35 <sup>0</sup> 39'58.22''
Dibaj	54 <sup>0</sup> 13'42.89''	36 <sup>°</sup> 25'25.04''
Miami	55 <sup>°</sup> 38'53.91''	36 <sup>°</sup> 24'11.23''
Biarjmand	55 <sup>0</sup> 54'38.25''	36 <sup>°</sup> 06'35.44''
Amirieh	54 <sup>0</sup> 08'35.16''	35 <sup>0</sup> 01'22.06''

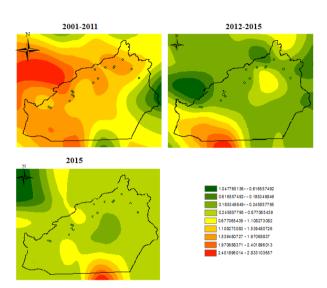


Figure 3. The layout of heavy metals concentrations in the investigated wells over the time

#### Discussion

Tables 2 to 4 represent the mean value of heavy metals concentrations from 2001 to 2015. As is evident from these tables, the heavy metal concentrations in all ground water resources did not cross the permissible limits recommended by national and international bodies demonstrated in Table 1. The reason for this is not exactly clear; however, it can be mainly due to the high underground depth of these wells, which provides enough time and natural filters for preventing the industrial pollutants from entering ground water resources. In addition, layers of the earth's crust in these geographical regions do not contain natural heavy metals and, more importantly, natural heavy metals are located far from the ground water resources. The heavy metals concentrations in the 17 wells investigated in the present study are also presented in these tables. According to these data, one can observe that the concentrations in all cities were nearly equal (0.022 µg/L), and also that their changes had been negligible over the years. The highest and lowest concentrations were founded in Miami

(0.0259 µg/L) and Shahroud (0.0172 µg/L), respectively. The reason for higher concentrations of heavy metals in Miami in comparison to other cities can be attributed to the presence of these elements in the earth's crust layers in the southern part of the province. Moreover, the distance between ground water resources and industrial centers is high in Shahroud. Geologically, there are chromium and cadmium mines in the south-eastern parts of the province, so, before proceeding with the construction of any water well in these parts, an initial assessment should be performed on the risk posed by such mines on the health of the population who will drink the water sucked from these wells. Besides, the one sample t-test revealed that there was no significance in the heavy metals concentrations in different years and the permissible values recommended by national and international bodies, such as WHO and the European Union (P>0.05). In addition, the repeated measure analysis demonstrated that the differences in concentrations over the years had not been significant (P>0.05). These results are in agreement with the results obtained by Rajaeiet al.<sup>24</sup>, Ghanbari et al.<sup>25</sup>, and Shahriari et al.<sup>26</sup>, which is because of the geological similarities among the areas in which these studies have been conducted. However, these results differ from the data obtained by the study conducted by Nahidet al.<sup>27</sup>, which investigated the level of heavy metals in the drinking water of Tehran city. In that study, the concentration of lead was reported higher than permissible limits and the reason noted by authors was the entrance of lead through a corroded pipeline network.

The values of the HRA<sub>EPA</sub> index in various years are presented in Table 3. Accordingly, the health hazard risk posed by the heavy metals in drinking water was 4.48×10-4, 4.36×10-4, and 4.46×10-4 for time intervals 2001-2011, 2012-2015, and 2015, respectively. Moreover, the highest risk level was during the 2001-2011 time intervals, and the lowest one was in 2015. The results also demonstrate that the risk increased in 2015 compared to the 2011-2015 time interval; in contrast, the risk value decreased compared with the risk calculated for the 2001-2011 time interval. However, the statistical analysis revealed that there was no significant difference between the risks calculated for these three time intervals (P>0.05). In other words, the industrial development in recent years in this region has not resulted in a significant increase in the risk posed by these elements to people living in the region. The main reason for this is that the industrial centers are located far from the water resources and also, all these industries are mandated to use water treatment units for treating the produced sewages before they enter the environment. According to reports from the departments of the industrial centers of the province, all the industries located in the province are equipped with the wastewater treatment units. Moreover, all the industries located out of these industrial centers are regularly inspected by the province's department of environmental protection.

Rajaei et al. conducted a study to evaluate the risk posed by heavy metals in drinking water of AliAbadKatol city; they reported the risk as  $4.85 \times 10$ -4, which is similar to the risk scores obtained in the present study. Zinc was the only element the concentration of which, in that study, was different with what we found in the present study, which was because of the presence of natural zinc resources in that region 24. Momot et al. conducted the same study in middle Russia; they reported the HRA<sub>EPA</sub> index as 4.93×10-3, which is higher than the risk calculated for Semnan province.<sup>28</sup>

Two layouts are presented in Figure 3, which is related to the heavy metals concentrations in two different time intervals. As seen in the figure, the quality of the water in the ground water resources was very high, and there was no health risk threat to the public in this respect.

In the present study, the changes in concentrations of heavy metals in drinking water in Semnan province were measured during different years; in addition, the annual health risk posed by such elements was estimated using the new risk index known as HRA<sub>EPA</sub>. The results of the present study demonstrate that the heavy metals concentrations in the ground water resources are in the permissible range. In the other words, the concentrations of these elements in the drinking water are too low to cause any harm to the consumers. As a result, the health-hazard risk score obtained in the present study is much lower than that reported by other studies. There are several reasons that contribute to this, including the high depth of these resources beneath the earth's surface, mandatory use of waste-water treatment units by industries, and the absence of natural heavy metals resources in the area. However, considering the fact that the industrial development of the province has accelerated in recent years, in order to prevent the drinking water from being polluted it is recommended that the establishment of industrial centers in areas located in the vicinity of the ground water resources be prevented by the government. Besides, a quarterly monitoring program of water resources is of high importance because conducting such programs would help to identify any negative change in quality of the drinking water earlier, when taking preventive action is less expensive and more effective.

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#### **Conflict of Interest**

The authors declared that they have no conflict of interest.

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