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Co-Contamination of Arsenic and Other Trace Elements (Hg, Pb, Al, Fe, Cr, Ni, and Cd) in the Rafsanjan Plain Alluvial Aquifer SE of Iran and Arsenic Risk Assessment

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Abstract

Assessing the concentration of trace elements in aquifers is increasingly subjected to study in Iran due to the lack of groundwater resources. This study was undertaken with the objective of determining trace elements in the alluvial aquifer located in the southern part of the Rafsanjan plain, Kerman province, Iran. The total of 73 groundwater samples from individual water wells were analyzed by atomic absorption spectroscopy (AAS). Results showed that the levels of As, Hg, Pb, Al, Fe, Cr, Ni, and Cd elements were above the World Health Organization standards for drinking-water in some parts of the plain. Thus, statistical data analyses and spatial distribution interpretation were performed to identify the main sources of the pollution. A health risk assessment model derived from US environmental protection agency was applied to calculate the cumulative exposure to As as well as toxic and carcinogenic risks caused by drinking contaminated raw groundwater. Results show that residents of some part of region may suffer from significant adverse toxic health impacts and are exposed to drinking water with As concentrations.

Keywords

Arsenic, Health Risk, Trace Elements, Rafsanjan Plain, Groundwater Pollution

1. Introduction

Arsenic (As) is a toxic metalloid with ubiquitous distribution into the environment ([1] [2] [3]). This element originates either from anthropogenic and geogenic sources [4] and has been classified as Group 1 human carcinogen by the International Agency for Research on cancer [5]. There is no evidence that arsenic is essential in human bodies. In contrast, chronic oral consumption of arsenic is considered to cause an adverse impact on human beings, known as "arsenicosis" or "arsenic poisoning disease". Arsenicosis can cause skin lesions, pigmentation of the skin, and the development of hard patches of skin on the palm of the hands and soles of the feet. Arsenic poisoning finally leads to skin, bladder, kidney, and lung cancers, as well as diseases of blood vessels of the legs and feet. Diabetes, high blood pressure, and reproductive disorders may also be the side effects of chronic arsenic exposure ([6] [7] [8]).

Groundwater is the basic sources of water for millions of people dwelling in arid and semi-arid regions like the Rafsanjan plain, Kerman province, Iran. The quality of groundwater exerts a strong influence upon human health, industries, and agriculture. Therefore, researchers did many studies in this regard around the world. For the first time in Iran, Mosaferi [9] reported arsenic contamination in water resources of Kordestan province. After that several investigations have done to give us a better view of heavy metal pollution throughout the country ([7] [8] [10] [11] [12]).

The objectives of the present study were: 1) to determine the distribution of toxic trace elements in ground water of the Rafsanjan plain and recognize the sources of pollution, 2) to assess non-carcinogenic and carcinogenic risks among the populations exposed to arsenic through groundwater drinking pathways.

2. Study Area

The Rafsanjan plain is located at a latitude between 30˚05' to 30˚45' and a longitude between 55˚45' to 56˚24' in the southern part of the Central Iran. This study is focused on the southern part of the plain (**Figure 1**). Geologically, the Rafsanjan plain belongs to the Central Iran Zone. The Sarcheshmeh Mountains which form the southern border of the plain, are composed of volcanic rocks (**Figure 2**). The study area consists of Plio-Quaternary alluvials which decrease in size towards the north. The alluvial aquifer of southern Rafsanjan plain is mainly composed of gravels with some sands and clays. Clay contents increase towards the north [3]. This region has dry climate with yearly average precipitation of about 108 mm which mainly falls during December–March period. Because of the hydroclimatic conditions, there is no considerable surface flow in this area [13].

The main parts of the plain which include alluvial fans and salt flats are considered as barren lands or poor pastures. Pistachio orchards, croplands and residential areas are the main land use units of the plain. Many water wells have been drilled to develop pistachio orchards [14].

Several seasonal and intermittent streams flow from the northern slopes of Sarcheshmeh Mountains into the plain (**Figure 3**). Among them Shour, Givdery

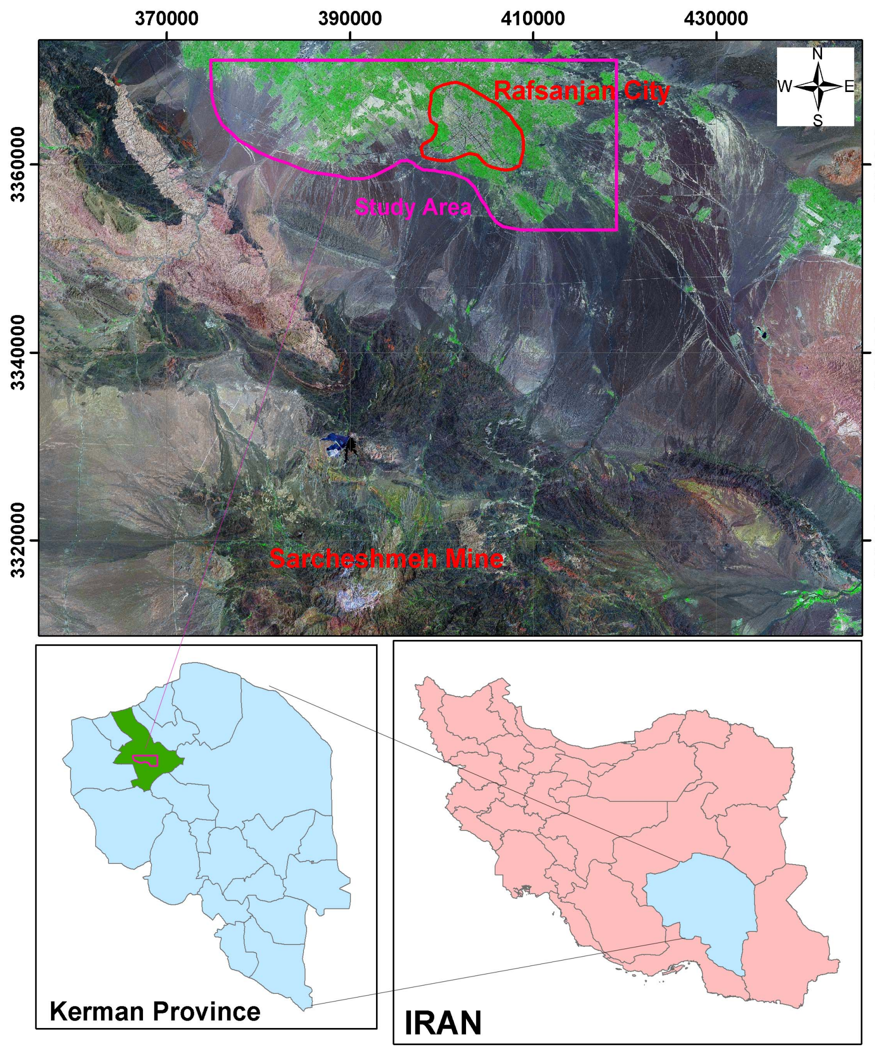

Figure 1. Schematic map of the study area.

and Qalandary streams are of more importance. They exert strong influence on replenishment of the alluvial aquifer of the plain via extensive alluvial fans [15].

Hydrogeologically point of view, the alluvial aquifer of southern Rafsanjan plain is mainly composed of gravels with some sands and clays. Clay contents increase towards the north. Hydrogeological investigations indicate an unconfined aquifer [16] (**Figure 4**). Groundwater flows from south and southeastern part into the northeastern salt flats. Clays, silts and evaporates of Miocene age are considered as the base of the aquifer [17].

There is different land use in the region. The main parts of the plain which include alluvial fans and salt flats are considered as barren lands or poor pastures. Pistachio Orchards, croplands and residential areas are the main land use units of the plain [18].

Figure 5 presents the location of mining and industrial area .It is obvious that mining and mineral processing industries were mainly developed due to the presence of numerous copper mines in the area.

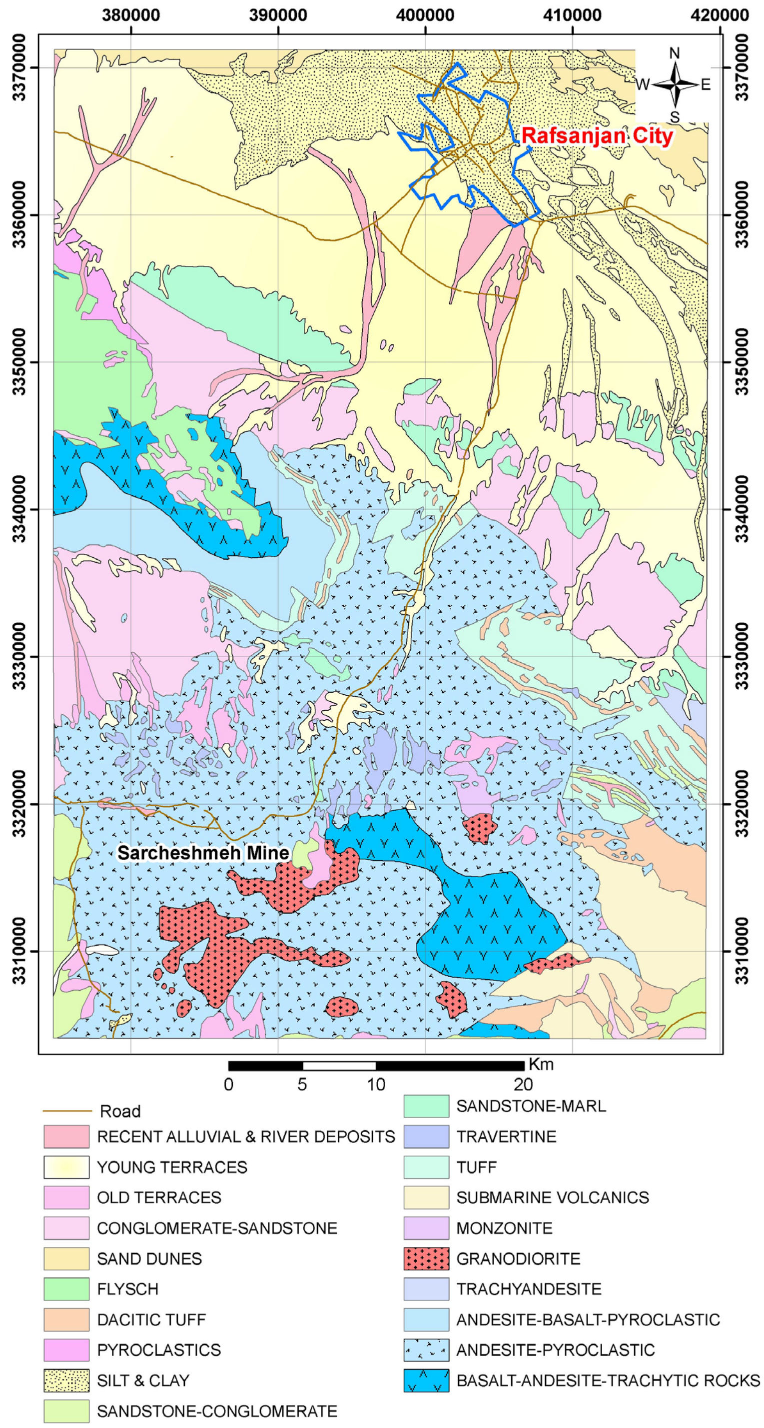

Figure 2. Geological map of the study area.

3. Materials and Methods

3.1. Sampling, Field Measurements and Laboratory Analyses

Groundwater samples were collected from 73 water wells during November to December 2015. Temperature, EC, Eh and pH were measured in situ using a WTW Multi150i multimeter. Water samples were collected in acid-washed

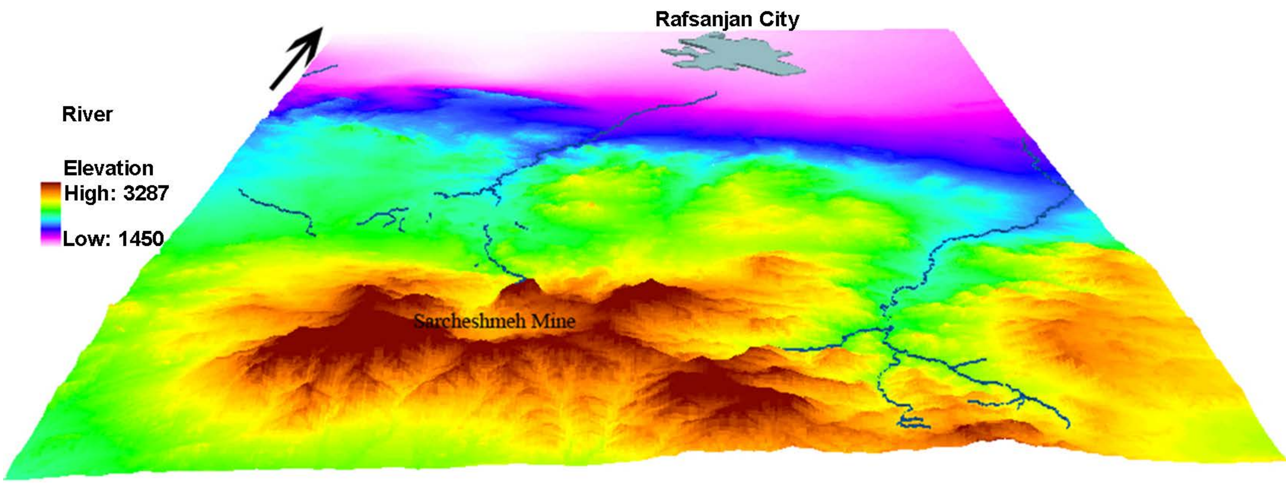

Figure 3. DEM model of the study area.

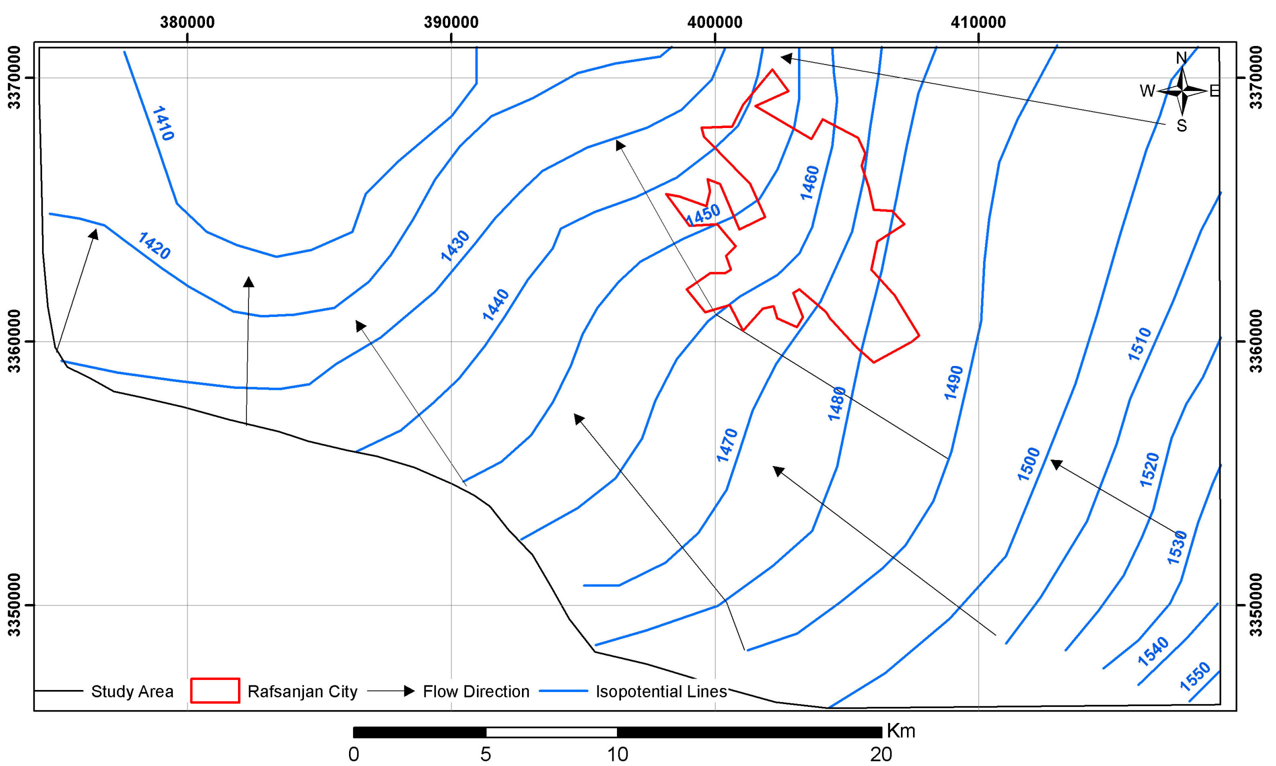

Figure 4. Isopiestic map of the study area.

polyethylene bottles for analyzing dissolved As and other trace elements. Samples filtered in situ using 0.2 μm pore-size Whatman CA filters. The samples were acidified with Merck Ultrapur HNO_3 (1:100 v/v) and stored at 4˚C. Another subsample was left unacidified for anion determinations. Ion chromatography was performed to determine major anion and cation concentrations. Trace elements and arsenic concentrations were measured using a flame atomic absorption spectroscopy (AAS) in Central laboratory of Graduate University of Advanced Technology of Kerman, Iran.

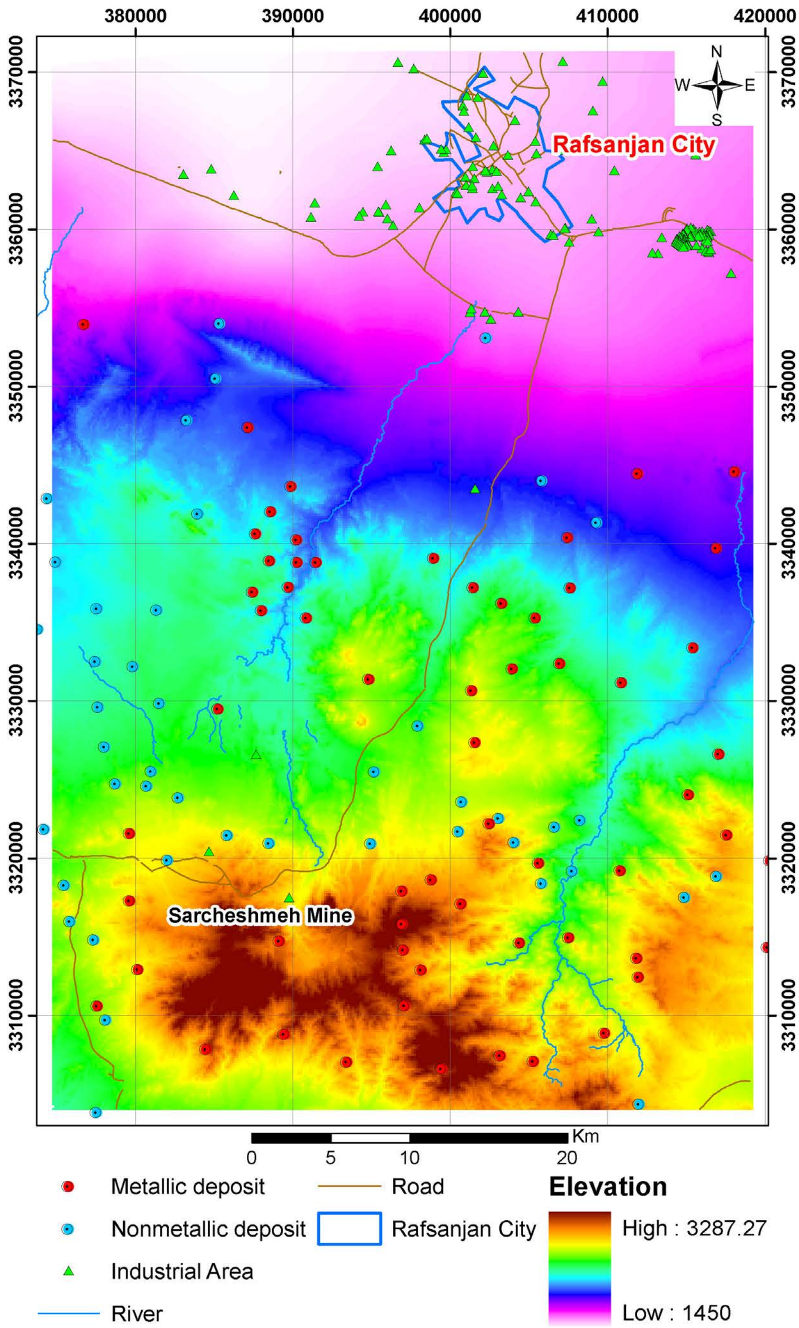

Figure 5. Mining and industrial map of the study area.

3.2. Health Risk Assessment Model

Heavy metal enters into human body through several pathways including food chain, dermal contact, and inhalation but in comparison to oral intake all others are negligible [19]. A health risk assessment model derived from the US EPA [20] was applied to compute the non-carcinogenic and carcinogenic effects of arsenic to individuals who consume groundwater as their drinking water source.

The Cumulative As exposure index (CAI) and (ADD) through water intake, was calculated according to the modified equations [21]:

$$CAI = Cw * IRw * EF * ED \quad (1)$$

$$ADD = (Cw * IRw * EF * ED)/(AT * BW) \quad (2)$$

where *CAI*: Cumulative As exposure index, *ADD*: Average daily dose from ingestion ($mg \cdot kg^{-1} \cdot d^{-1}$), *Cw*: Arsenic concentration in ground water ($mg \cdot L^{-1}$), *IRw*: Water ingestion rate ($L \cdot d^{-1}$), *EF*: Exposure frequency ($d \cdot y^{-1}$), *ED*: Exposure duration (year), *AT*: Average time/life expectancy (day), *BW*: Body weight (Kg).

To estimate the non-carcinogenic/chronic risk, *HQ* can be calculated by Equation (3) [22].

$$HQ = ADD/RfD \quad (3)$$

where *HQ*: Hazard Quotient (Toxic risk is considered occurring if $HQ > 1.00$), *RfD:* Oral reference dose, for As is equal to 3.04E−04 ($mg \cdot kg^{-1} \cdot d^{-1}$)

Carcinogenic risk is the probability of incidence of cancer from chemical exposure. It is calculated using Equation (4):

$$R = 1 - \exp(-SF * ADD) \quad (4)$$

where *SF* is the oral carcinogenic slope factor and for As, it is equal to 1.5 ($mg \cdot kg^{-1} \cdot d^{-1}$).

3.3. Spatial Distribution of Arsenic and other Trace Elements

Identification of contaminated areas is important to recognize the sources of pollution. We used ArcGIS (version 10.1) to prepare iso-concentration maps of As and other trace elements.

4. Results and Discussion

4.1. Chemistry of Groundwater in the Study Area

Physico-chemical parameters in sub-surface water of the study area are summarized in **Table 1**. Na values ranged from 146 - 730 mg/l, with mean values of 435 mg/l, Ca values in ranged from 17.2 - 234 mg/l, with mean values of 97.3 mg/l. Mg values ranged from 14.3 - 172.2 mg/l, with mean values of 52.7 mg/l, K values in ranged from 4.4 - 82 mg/l, with mean values of 13.29 mg/l. SO_4^{2-} values ranged from 71 - 770 mg/l, with mean values of 236.1 mg/l, HCO_3^- values in ranged from 48 - 380 mg/l, with mean values of 144.8 mg/l. Cl^- values in ranged from 84 - 1400 mg/l, with mean values of 550.1 mg/l. T values in ranged from 19 - 33 with mean values of 26.9. pH values ranged from 5.3 - 7.7, with mean values of 6.8. Furthermore 93%, 31%, 1%, 1%, 5% water samples showed higher concentration than their respective permissible limits by WHO [24] for Na, K, Mg, Ca, SO_4^{2-} respectively (**Table 1**). Evaporate formations such as gypsum and salt which located in this aquifer are responsible for pollution of these elements.

4.2. Distribution of Arsenic and other Trace Elements

Iso-concentration maps of As, Hg, Pb, Al, Fe, Cr, Ni, and Cd are presented in **Figure 6**.

Table 1. Mean, standard deviation, maximum, minimum and WHO [23] guidelines of the variables determined in the 73 groundwater samples.

Parameter	Mean	St dv	Max	Min	WHO
As (μg/l)	76.4	39.03	173	23.9	10
Cr (mg/l)	0.01	0.02	0.07	0	0.05
Pb (mg/l)	0.01	0.02	0.09	0	0.01
Fe (mg/l)	0.27	0.36	1.52	0	0.30
Al (mg/l)	0.19	0.17	0.84	0	0.20
Hg (μg/l)	6.69	2.69	11.88	1.09	6
Ni (μg/l)	71	70	445	5	70
Ca (mg/l)	97.33	47.02	234	17.2	200
Mg (mg/l)	52.77	31.45	172.2	14.3	150
K (mg/l)	13.29	12.55	82	4.40	12
Na (mg/l)	435.45	144.17	730	146	200
SO_4^{2-} (mg/l)	236.18	136.95	770	71	500
HCO_3^- (mg/l)	144.84	55.97	380	48	-
Cl^- (mg/l)	550.14	293.32	1400	84	-
T (˚C)	26.92	2.98	33	19	-
pH	6.83	0.58	7.70	5.30	-

Figure 6(a) displays the Iso-concentration map of arsenic. The concentration of this element in major parts of the plain exceeds its maximum permissible limitation (MPL), 10 μg/l [23]. The maximum observed value is 230 μg/l for the sample no. 19. The high level of arsenic in eastern parts is attributable to waste waters of the industrial park of the Rafsanjan. However, the higher background level of arsenic across the plain is ascribed to the widespread high-sulfidic volcanic and plutonic rocks of Sarcheshmeh mountains such as granodiorite, andesite and monzodiorite that contains Arsenopyrite (FeAsS) and pyrite (FeS_2) which are the main source of aquifer replenishment. The arsenic containing pesticides, however, may have an intensification role of pollution. Based on Figure 6(b), mercury shows a high concentration in main parts of the plain (MPL = 6 μg/l). The sample no. 15 has the maximum value of 11.88 μg/l. Copper mineral processing activities around Sarcheshmeh copper mineral is probably responsible of water pollution caused by this metal.

Considering 0.1 mg/l as the MPL of lead, its concentration is totally lower than this level (Figure 6(c)). The local increases of lead up to 0.06 mg/l in the north of the Rafsanjan may be attributed to a pollution caused by lead-bearing fuels consumed mainly in the city.

Figure 6(d) exhibits the Iso-concentration map of aluminum. Geochemical weathering of silicate minerals and some wastewaters of the industrial park of Rafsanjan caused concentration of aluminum higher than admissible value (MPL = 0.2 mg/l).

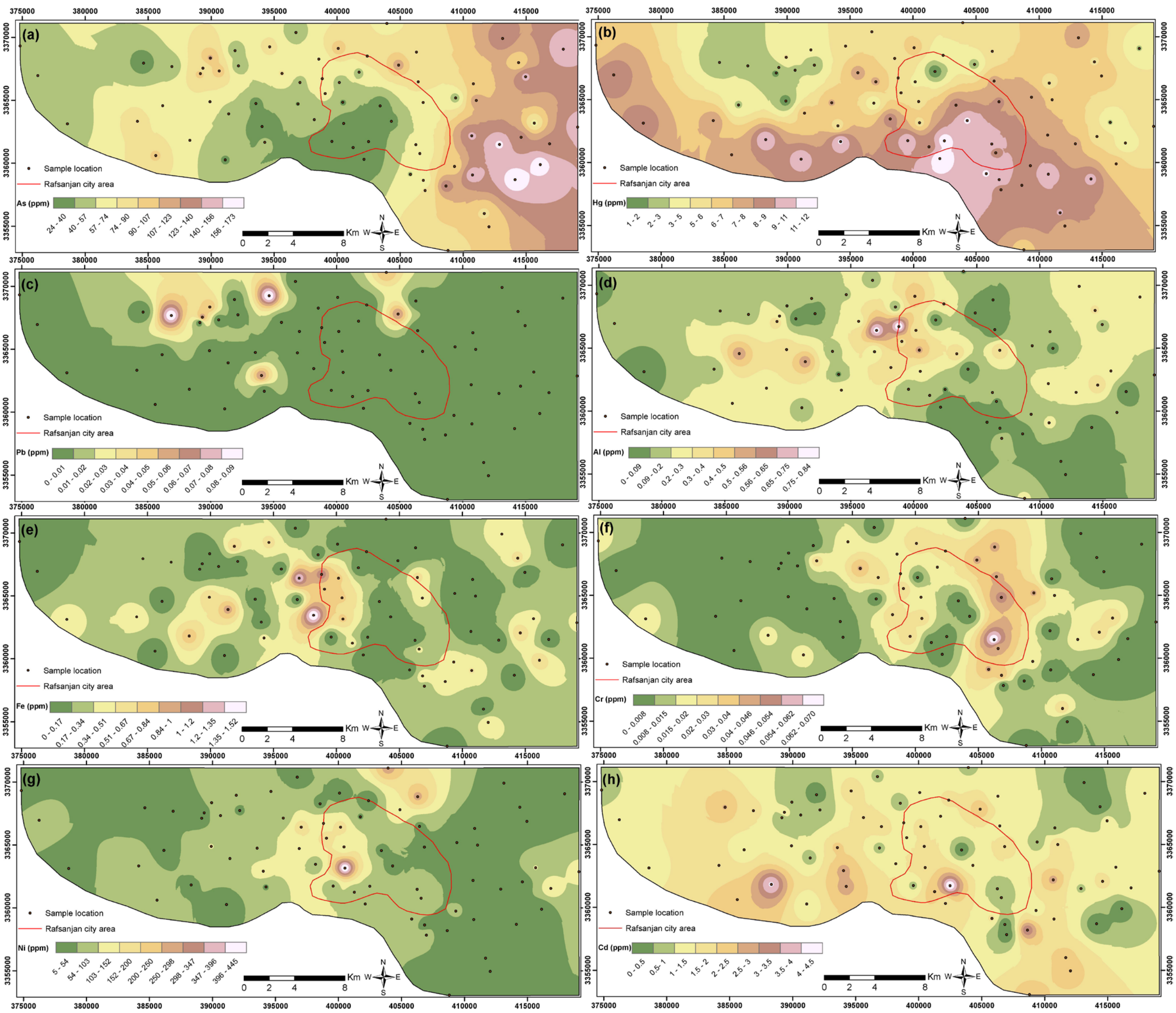

Figure 6. Iso-concentration maps of As, Hg, Pb, Al, Fe, Cr, Ni, and Cd.

The high Fe concentration in some parts (MPL = 0.3 mg/l) may imply iron containing fertilizers and silicate weathering as the main sources of this element in the water (Figure 6(e)).

In general, chromium level in studied groundwater samples is less than MPL value of 0.05 mg/l. Figure 6(f) demonstrates the variation in chromium values below the MPL.

High concentrations of nickel in the medial parts (MPL = 70 μg/l) of the plain are attributed to volcanic rock source and the high spots in northeast of the city may be attributed to the industrial wastewaters. Figure 6(g) exhibits the distribution of nickel in the study area.

The cadmium level in the study area is low (Figure 6(h)), except in the sample no. 21 that showed 4.06 μg/l (MPL = 3 μg/l). This sample located in the southwest of the plain, near a chemical plant. Wastewaters of the chemical plant could be responsible for the observed pollution.

4.3. Arsenic Risk Assessment

4.3.1. Hazard Identification

Groundwater is the main source of drinking water for rural inhabitants and border settlements of this region but another source has been provided for the residents who lives in the Rafsanjan city. As previously mentioned, concentration of arsenic in all water wells exceed MPL (10 μg/l) with an average amount of 76.40 μg/l in the study area. Risk assessment was conducted for 5 wells that are used as drinking water.

4.3.2. Cumulative As Exposure Index (*CAI*) and Average Daily Dose (*ADD*)

The cumulative AS exposure index (*CAI*) and average daily dose (*ADD*) can be calculated based on the results of the measurement of arsenic concentrations in the raw groundwater. Considering an average body weight of 70 kg for an adult with an average lifespan of 73.2 years in Iran and a water ingestion rate of 1.5 L/day of untreated groundwater rich in arsenic for an exposure duration of 35 years [24] (**Table 2**), a range of 1.41 g to 2.6 g is obtained for cumulative arsenic ingestion (*CAI*) of the Rafsanjan residents. Cumulative exposure (6.75 g) to arsenic was found to be lower than the threshold level for internal cancer caused by chronic exposure to arsenic in the groundwater for the residents. The lower cumulative ingestion of arsenic than the threshold level linked to internal cancer for most of the residents in the study area might be explained as the absence of the clearly demonstrated manifestations of arsenic-related cancer effects in the Rafsanjan region. However, more in-depth investigations of cancer incidence are required to confirm that there is a significant relationship of cancer incidence between exposed and the non-exposed populations in the Rafsanjan region. The average daily dose (*ADD*) was calculated using the same assumptions have been used to calculate *CAI*. The results indicated that residents who consume raw groundwater containing As concentrations between 73.92 μg/L and 135.93 μg/L ingest between 7.52×10^{-4} and 1.38×10^{-3} mg/kg of As per day (**Table 3**).

4.3.3. Toxic and Carcinogenic Risks

The results of toxic and cancer risk indices calculated from the same parameters as *CAI* and *ADD* showed that border area of Rafsanjan city suffer toxic risk

Table 2. Exposure factors adopted in the risk assessment for the population of Rafsanjan plain.

Arsenic concentration (*Cw*)	Arsenic concentration in the raw groundwater sample in μg/l
Exposure rate (*IRw*)	1.5 l/day, average daily intake of water
Exposure frequency (*EF*)	365 days/year, the days of arsenic intake per year in evaluation period
Exposure duration (*ED*)	35 years, most of the wells are drilled in the period between 1975 and 1985, thus 35 years was assumed as mean exposure duration
Average exposure time (*AT*)	26,900 days (73.2 years),average lifetime in Iran
Body weight (*BW*)	Typical adult weight of 70 kg is considered

Table 3. Calculated *CAI*, *ADD*, *HQ* and *R* in 5 wells used as drinking water.

	Cw (μg/l)	*CAI* (g)	*ADD* (mg/kg-day)	*HQ*	*R*
Well no.16	74.23	1.42	7.55E−04	2.48	0.001131
Well no.17	85.99	1.65	8.75E−04	2.87	0.001312
Well no.18	94.78	1.82	9.64E−04	3.17	0.001445
Well no.72	135.93	2.60	1.38E−03	4.53	0.002068
Well no.73	73.92	1.41	7.52E−04	2.47	0.001128
Mean	92.97	1.78	9.93E−04	3.10	0.001488

indices (*HQ*) ranging from 2.47 to 4.53 (**Table 3**). The *HQ* index equal to 1.00 is suggesting that the residents of the region may suffer from significant adverse toxic health impacts. Furthermore, the cancer risk indices (*R*) ranged from 11 per 10,000 persons to 20 per 10,000 persons. The highest safe standard for cancer risk is 1 per 10,000 and the lowest safe standard is 1 per 1,000,000 [25]. Based on the results in **Table 3**, the residents in suburban area may be exposed to drinking water with As concentrations that could lead to cancer. However, no case of arsenicosis has not been reported in the Rafsanjan plain settlements yet. These results did not take into account the individual statuses of the residents such as the duration of exposure and the ingestion rate. To assess the health effects of chronic As exposure it would be necessary to accurately characterize residents' consumption patterns, namely to track changes in the sources of drinking water, duration of use and drinking rate [20]. The natural baseline concentrations of As in the groundwater defined during this study were used for As risk assessment. The results can be used as an essential component for future more in-depth epidemiological studies which will include exposure of the population to different As species. Here, it was assumed that raw groundwater was used untreated for drinking, which does not mean that certain changes did not take place. For example, As(III) oxidation and trapping of As on Fe precipitates in water distribution systems can be taken in to account. However, exposure to As in food cannot be ignored.

5. Conclusion

The iso-concentration maps of the heavy metals considered in this investigation exhibit higher than MPL levels of As, Hg, Cr, Pb, Cd, Al, Fe and Ni in some parts of southern portion of the alluvial aquifer of the Rafsanjan plain. Based on comparisons of anomalous levels of these elements with geological, hydrogeological, and land use maps of the area, the main sources of pollution include volcanic rocks of Sarcheshmeh mountains (As, Al, Fe, Ni), mineral processing in the Sarcheshmeh copper plant (Hg). Wastewaters of the Rafsanjan park of Industries (As, Al, Fe, Ni, Cd), urban wastewaters of the Rafsanjan city (Cr, Pb) as well as chemical pesticides consumed in pistachio gardens (As). Therefore, both anthropogenic and geogenic sources in variable proportions contribute to heavy metal pollution in the Rafsanjan plain. The concentration of As in all parts of the area is above the standard for drinking water, Which originates either from anthropogenic and geogenic sources. The results are in accordance with similar heavy metals pollution studies in other region of Iran metallogenic copper belt. Toxic and cancer risk assessment of arsenic showed that border area of the Rafsanjan city settlements suffer the toxic and cancer risk indices. However, cases of arsenicosis has not reported in this region yet, we had limitation in access to local medical data and also previous hydrochemical analysis. The results obtained are an essential component for future more in-depth epidemiological studies which will include exposure of the population to the different As species. We either suggest biogeochemical investigations on arsenic in Rafsanjan plain to determine the effects of this poisonous element on soil and other organisms such as plants and animals.

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