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Investigation of Gorgan City groundwater contamination with Emphasis on Heavy Metals

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Abstract

This study was conducted to evaluate the quality of groundwater resources in Gorgan using heavy metal indices. For this purpose, the chemical analysis results of 63 drinking water wells were used in the two seasons of spring and autumn. Concentration of four heavy metals (arsenic, lead, iron, and manganese) in groundwater was measured by ICP in the Golestan Provincial Water and Wastewater Department laboratory. To evaluate the groundwater contamination by heavy metals, Heavy Metal Pollution Index (HPI), Degree of Contamination (Cd), and Heavy Metal Evaluation Index (HEI), were used. Results revealed that the concentration of heavy metals did not change much in spring and autumn. Also, the results of paired t-test showed that there is no significant difference between heavy metal concentrations in spring and autumn. Heavy metal distribution map shows that average value of heavy metals in the western part of the aquifer is increasing from south to north. The highest concentration of these metals in residential areas with high population density and the old texture of the city increased dramatically. Results revealed that the average values of HPI, Cd, and HEI index in the spring and fall of are less than the critical amount of contamination for drinking water. Although, Gorgan groundwater is currently not contaminated in terms of metal indices, but essential operations should be taken for the proper disposal of municipal wastewater and industrial and agricultural effluents. It is also recommended to monitor the groundwater resources of study area periodically in terms of heavy metal concentrations.

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Keywords: Groundwater, Heavy Metals, Contamination Indices.

1. Introduction

Excessive use of natural resources and excessive production of waste in advanced societies often threatens groundwater resources and contaminates it. Numerous factors, including climate, soil characteristics, groundwater circulation from stone types, area topography, saline water penetration in coastal areas, human activity on the ground, and so on affect water quality. (Selvam ET AL., 2015). Investigating the quality of groundwater resources is a very important environmental issue that should be taken into consideration by water and soil and environmental experts. Therefore, before using these water resources in drinking, agriculture, industry sectors their qualitative characteristics must be identified.

In recent decades, attention to heavy metals accumulation in the environment has been the focus of their toxicity and sustainability (Upadhyay et al., 2006). What has made groundwater pollution to heavy metals become one of the most important environmental issues in the present world is the high toxicity of these metals even at low concentrations (Kumar et al., 2012). Heavy metals are one of the environmental contaminants with carcinogenic compounds that reflect the presence of natural or human contamination in water and the environment (Akoto et al., 2008). Heavy metals in colloidal phases, particles, and insoluble with natural origin (mainly due to soil and rock weathering, environmental erosion, forest fires, and volcanic eruptions) or human (such as discharging urban, industrial, and agricultural waste urban runoff, etc.) are found in water (Marcvecchio et al., 2007; Yeh et al., 2020). Generally, neurological disorders, types of cancers, and in acute cases of death are the results of heavy metal entry effects into the human body. Exposure to toxic heavy metals such as mercury, cadmium, arsenic, chromium, tallium, and lead even at low concentrations will impair normal body function. However, some metals such as zinc, iron, and manganese are required in small amounts for metabolic activities of the body. However, these elements at high concentrations can have negative effects on human health. Manganese prevents children's intellectual development and iron causes genetic and metabolic diseases (Nejatijahromi et al., 2018).

Pollution indices are useful tools for water sector managers, environmental managers, and decision makers to take action in these areas, which are affected by all the parameters applicable to qualitative indices. The spatial analysis of heavy metals using these indices can be very useful in detecting and quantifying the water quality trend (Mishra et al., 2017).

So far, many research in Iran has been conducted to study groundwater quality using water quality contamination indices, some examples are mentioned below. Hosseinpour et al. (2014) evaluated the quality of groundwater resources around the Khorasan Steel Complex Company. Although, the results of this study revealed that the average amount of HPI index of heavy metals was much lower than the threshold. But if the appropriate environmental approaches are not adopted over time and with the increase in the concentration of these elements in groundwater, environmental problems will arise in the stuy area. In a study was conducted by Bayati et al. (2020), the water quality of the Choghakhor lagoon was evaluated using Mi, HPI, HEI and CD indices. Their results revealed that the water quality of Choghakhor lagoon based on MI, HPI, and HEI indices

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is in the category of danger threshold, contamination-free, and low contamination, respectively. Also, based on the Cd index, groundwater has low pollution.

Nejatijahromi et al. (2018) evaluated the quality of groundwater resources of the Varamin aquifer for the drinking section using indexes of heavy metal pollution such as HEI and HPI. The results showed that the pollution of Varamin aquifer is not hazardous to heavy metals in most parts, but in some areas, clear changes in the concentration of some metals are observed. With the continuation of pollution sources in Varamin plain, we can expect that in the long run, the risk of groundwater pollution relative to heavy metals will continue.

Selvam et al., (2015), evaluated groundwater pollution in Tamilado, India, using the HPI (HPI), Heavy Metal Evaluation Index (HEI) and the degree of contamination (D_C). For this purpose, they measured concentration of nine heavy metals (Al, Cr, Fe, Cu, Mn, Ni, Zn, Cd and Pb) in thirty -six groundwater samples during the summer of 2013. Initial results of this study revealed that groundwater is contaminated with manganese, copper, lead, chromium, and cadmium. Also, based on the results of groundwater contamination indices, most groundwater samples have moderate to high contamination.

The source of heavy metals in groundwater, as well as the risk of human health pollution around the Auto Landfill site in the Komasi area of Ghana were evaluated by Boateng et al. (2019). The results of this study revealed that the average concentration of lead, iron, cadmium and chrome is higher than the World Health Organization (WHO) for drinking water. The results of contamination indices indicated that the groundwater has been contaminated to heavy metals. The results of the multiplier analysis also showed that lithology and human factors are potential sources of groundwater pollution to heavy metals in the study area.

Another research has been conducted by Atangana and Oberholster (2021) on the assessment of the quality of surface and groundwater in the Olive River Basin in South Africa using heavy metal contamination indices. This study was aimed to determine the changes of surface and groundwater on heavy metals contamination. Results of Heavy Metal Pollution Index based on selected heavy metals (Cd, Zn, Ni, Mn, Cu, Fe, Cr, Al and Pb) showed that more than 75 % of the region did not have good water quality, while less than 35 % is appropriate. In addition, this study shows that infants, children, and adults who consume contaminated water are exposed to significant amounts of metals.

Measuring the concentration of heavy metals of groundwater resources is of great importance due to the concentration of water quality in terms of concentration of these metals and the possible contamination of these resources, which requires proper and periodic research. Therefore, in the present study, the concentration of heavy metals, including lead, arsenic, iron and manganese, was investigated with the aim of monitoring spatial changes in groundwater supplies in Gorgan. It was also evaluated using statistical techniques, geochemical methods and contamination indices of groundwater resources compared to heavy metal contamination.

2. Materials and methods

2.1. Study area

Gorgan city with a longitude of 54.26 and a latitude of 36.50, an altitude of 160 meters above sea level and an approximate area of 1615 square kilometers is the center of Golestan province. (Fig. 1). This city is located near Semnan province, Aq-Qala and Turkmen cities, Aliabad and Kordkoy. Based on 2016 census, Gorgan has a population of nearly half a million people. The city has a temperate Caspian or Mediterranean climate. Although the rainfall in this city is less than the western cities of Mazandaran province. However, rainfall in this city is less than the western cities of Mazandaran province Ziarat, Qarahsoo, Kafshgiri rivers are the most important rivers in this area that flow into the Caspian Sea. The studied aquifer is mostly influenced by sedimentation of Ziarat River.

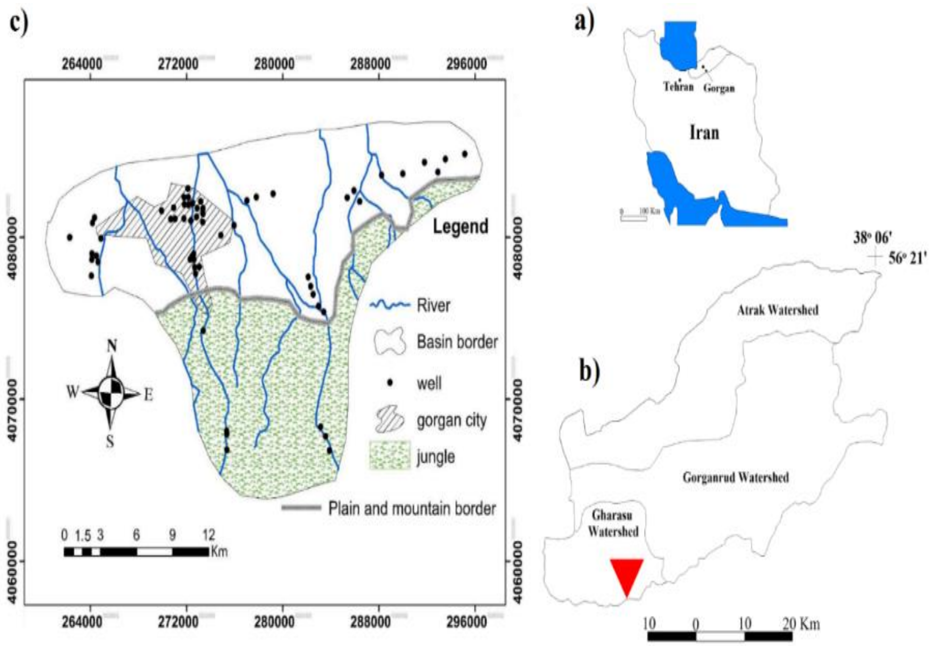

Figure 1. Location of the study area in the country (a), Golestan province (b) and the border of the area(c)

2.2. Indices of heavy metal pollution

In this study, the concentration of heavy metals (including lead, arsenic, iron, and manganese) of 63 Gorgan groundwater supplies in two seasons of spring and autumn 2018 was collected from Golestan Province Water and Wastewater Department. The location of the sampling wells is shown in Figure 1. In this research, Heavy Metal Pollution Index (HPI), and Degree of Contamination(Cd), were used to evaluate groundwater contamination.

Heavy metal pollution index (HPI) determines the effect of heavy metals on human health (Equation 1). This index is calculated and evaluated based on the weight assigned to each selective parameter. The selected weight has the desired amount between zero and one, and Its choice depends on individual qualitative and observational considerations. In Equation (1), W_i Unit weight is related to the parameter of i which is considered as the inverse ratio of the standard allowable value (maximum allowable drinking water limit) for each heavy metal (S_i) ($W_i = 1 / S_i$). In the present study, I_i (the ideal value) of the parameter of i is defined according to the standard of the World Health Organization. Also, n is the number of parameters and Q_i is the sub-index of parameter i, which can be calculated from Equation (2).

$$\mathbf{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}$$
(1)

(2)
$$\mathbf{Q_i} = \sum_{i=1}^{n} \frac{|M_i - I_i|}{(S_i - I_i)} * \mathbf{100}$$

In the Equation (2), M_i relationship indicates the measured value of the heavy element in parameter of I. The numerical value of 100 was introduced as the critical limit of the pollution index (Majhi and Biswal, 2016).
The degree of pollution index (Cd) includes the combined effects of several water quality parameters that these parameters are considered as harmful elements for water in the drinking water sector (Prasanna et al., 2012) which can be calculated based on the following equation:

$$\mathbf{Cd} = \sum_{i=1}^{n} \mathbf{Cfi}$$
(3)

$$\mathbf{Cfi} = \frac{M_i}{S_i} - \mathbf{1} \qquad (4)$$

In Equation 3 and 4 CFi indicates the ith element pollution factor
In general, the numerical values of Cd can be divided into three categories of low contamination (Cd<1), moderate pollution (Cd = 1 to 3) and high contamination (Cd>3).
Heavy Metal Evaluation Index (HEI) shows the overall quality of water according to heavy metals (Edet and Offiong, 2002; Prasanna et al., 2012). This index can be calculated from the following Equation.

$$\mathbf{HEI} = \sum_{i=1}^{n} \frac{M_i}{S_i}$$
(5)

As stated in Equations 1 and 2, M_i and S_i represent the measured values and the maximum permissible concentrations of the ith heavy metal, respectively.
The high concentrations of heavy metal measured compared to the amount of Si indicate the inadequate and bad water quality in the drinking sector. Based on this index, this water cannot be used if the concentration of each metal is greater than the maximum permissible value in the standards (that is, the M_i / S_i ratio is greater than one), this water cannot be used for drinking purposes. Therefore, the value of one is considered as the threshold of the risk of contamination based on the HEI index.

2.3. Statistical Analysis

To investigate the changes in physicochemical parameters of selected wells in the two periods (spring and autumn), paired t-test was used at a probability level of 0.05 in the Minitab software environment. Prior to this test, the hypothesis of normality of the data was tested using the Anderson Darling test. In the t-test, if the calculated p value is smaller than 0.05, with the probability of 95 % H0's assumption, ie the mean of the two populations, is rejected and the assumption of H1 or significant differences is accepted (Shirazi, 2016).

3. Results and discussion

Statistical values for heavy metals in water resources related to drinking water wells are given in Table (1). Based on the average of the obtained results, there is not much difference between the spring and autumn values of the parameters. Also, the amounts of heavy metals examined are lower than the WHO standard values.

Table 1. Statistical characteristics of heavy metals data(in milligrams per liter)

Parameter	Min		Max		Mean		WHO standard 2011
	Fall	Spring	Fall	Spring	Fall	Spring	
Pb	0.005	0.005	0.015	0.015	0.008	0.008	0.01
As	0.005	0.005	0.011	0.011	0.007	0.007	0.01
Fe	0.02	0.02	0.2	0.2	.08	0.09	3
Mn	0.01	0.02	0.04	0.04	0.03	0.03	0.5

Figure 2 shows that the spatial distribution of As, Pb, Fe and Mn in the two seasons of spring and autumn. Results revealed that most of the changes in these parameters are related to the western part of the study area in the north-south direction, where the old texture of Gorgan City and the areas with the highest population density. Except for manganese, the changes in heavy metals in the spring are similar to those in the fall.

Variations in arsenic and lead concentration in spring and autumn ranging from 5 μg in the eastern part of the study range to 11 and 15 μg in the western part of the study range (Fig.2 A and B). Based on the distribution map of chemical parameters of arsenic and lead, it can be concluded that both follow the same pattern.hence, it can be concluded that these two parameters are likely to have a single origin. As shown in Figure 2, the amount of iron in groundwater of the study area did not change significantly in spring and autumn. The range of changes for this parameter is between 0.019 to 0.21 mg / l.

Variations of manganese in the groundwater of the study area in spring ranged from 0.02 to 0.04 and in autumn from 0.01 to 0.04 mg / ml, which has not changed much in the two seasons.

As shown in Figure 2, manganese distribution pattern in the two season is completely different. So that in spring the maximum value of the mentioned parameter can be observed in the western half, while the most changes in autumn are related to the eastern half. Most of the changes in this parameter are related to the western half of the study area, in accordance with the old texture of Gorgan. However, the amount of abnormalities in the spring is far greater than the fall season.

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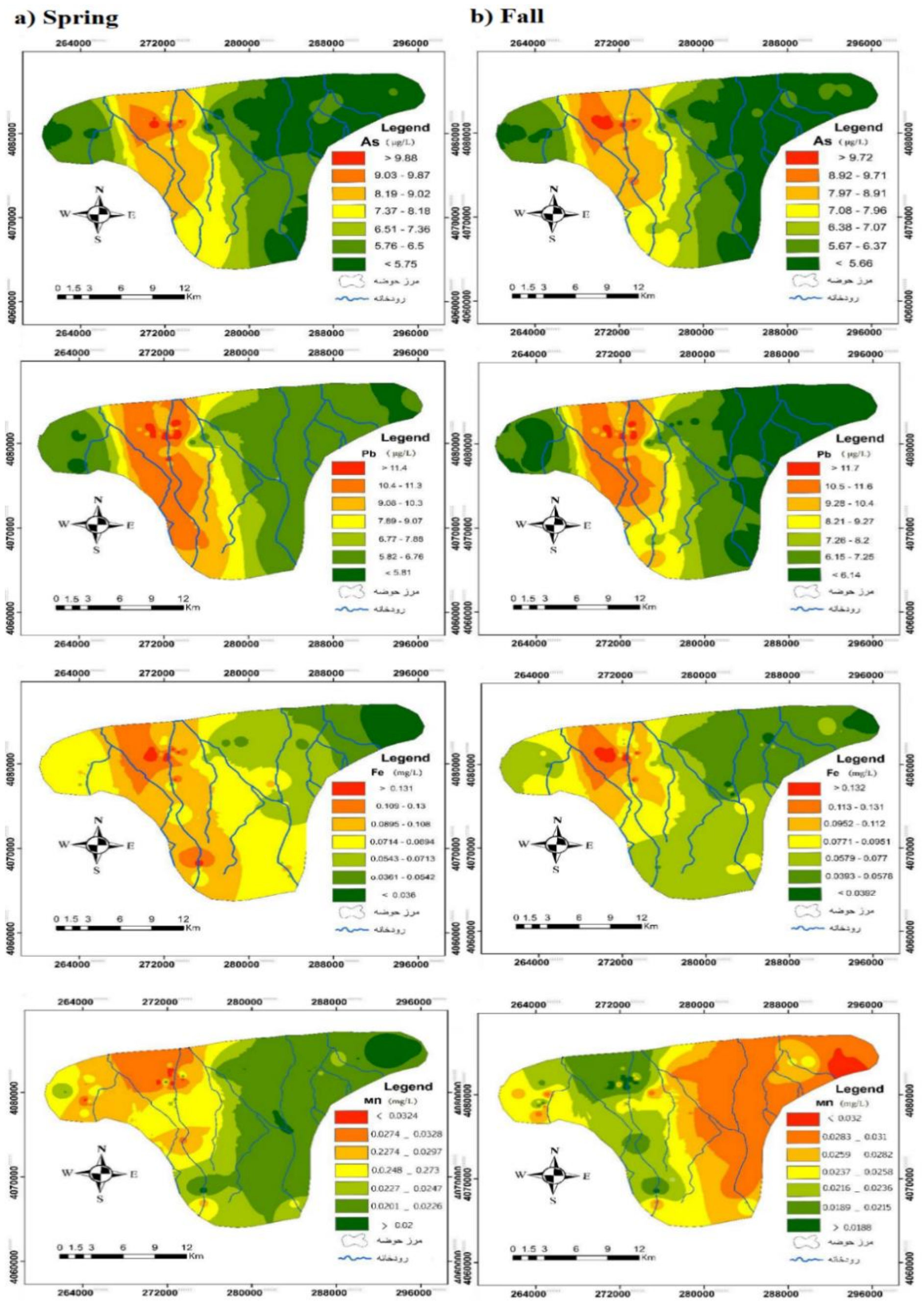

Figure 2. Spatial variation of As, Pb, Mn and Fe in spring and autumn in the study area in two season of spring and fall

To investigate the statistical differences between the measured heavy metals in the selected wells for two studied periods (spring and autumn), a pair t-test was used at a probability level of 0.05 in the Minitab software environment. Before performing paired t-test, the normality hypothesis of the data was checked at the probability level of 0.05. Table 2 shows the p-values of the paired t-test to investigate the significant differences in the chemical parameters of the selected wells in spring and autumn. The results of this study revealed that in all heavy metals, the calculated p-value is greater than 0.05. This indicates that a significant difference between the chemical parameters under is not accepted.

Table2: p-value of T-test of heavy metals in selected wells in spring and autumn

Factor	p-value
Pb	0.438
As	0.621
Mn	0.299
Fe	0.177

In this study, three heavy metals indices, pollution index, heavy metals evaluation index and metal index were used to determine the effect of heavy metals on human health. The values calculated for the heavy metal pollution index are presented in Table 3. Based on the HPI index classification, the groundwater of the studied aquifer in both spring and autumn seasons is free the contamination of heavy metals.

However, the average value of heavy metal pollution index in spring (12.42) is slightly higher than autumn (12.39). The lowest value of the above index (9.15) is related to wells no. 39 (spring) and 40 (autumn), respectively. Also, the highest amount of heavy metal pollution index in the two seasons is related to well no 22. As explained above, the HEI heavy metals evaluation index is calculated based on the ratio of the measured value of each element (heavy metal) to the maximum permissible concentration of those elements in the water. The calculated values of the HEI for both spring and autumn seasons are presented in Table 3.

Based on the values obtained for HEI, groundwater in the study area is not polluted with respect to heavy metals to low pollution to heavy metals. Similar to the HPI, the HEI also did not alter much in spring and autumn. Based on the results obtained for the metal index in spring, the highest value of the above index is for wells 32 and 39, the value of which is 0.3, and the lowest value is for well 9, the value of which is / 642. Also, in autumn, the maximum value for wells 32, 40, 54 and 55 is equal to 0.3 and the minimum value for well number 9 is about 0.642. The average of this index in both spring and autumn is equal to 0.36.

Cd pollution index includes the combined effects of several water quality parameters, which are considered as harmful elements for drinking water. (Prasnna et al., 2012). Based on the results are given in Table 3, the average pollution index in spring and autumn is 3.637 and 3.644, respectively. Hence, the studied water resources, in contrast with the other two indices, are polluted according to the Cd index. The highest and lowest values of Cd index in spring are related to wells No. 9 and 37, respectively. Also, in

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autumn season, the highest and lowest values of Cd index are related to wells No. 9 and 45, respectively.

Table 3- Heavy metal indices in spring and autumn

Season	Index	Average	Max.	Min.	Variance
Spring	HPI	12.42	19.05	9.15	7.914
	HEI	0.36	0.3	0.642	0.0095
	Cd	3.637	3.36	3.77	0.0095
Fall	HPI	12.39	19.05	9.15	8.131
	HEI	0.36	0.3	0.642	0.0097
	Cd	3.644	3.358	3.78	0.0097

3.1. Comparison of heavy metals concentrations with 1053 and WHO standards

In this study, the concentration of heavy metals of iron, manganese, lead, and arsenic was compared with standard WHO (2011) values (Table 4). The results of this comparison showed that the concentration of all heavy metals consisting iron, manganese, lead, and arsenic elements in both spring and autumn seasons is less than permissible level. Similar results were obtained from the comparison of heavy metals studied to the standard 1053.

Table 4- Comparison of heavy metal concentrations with 1053 and WHO standards

Elements	WHO standard	Standard 1053	Average concentration of elements (mg/l)	
			Fall	Spring
Fe	1	1	0.08	0.09
Mn	0.4	0.5	0.03	0.03
As	0.01	0.05	0.007	0.007
Pb	0.01	0.1	0.008	0.008

4. Conclusion

The study was conducted to evaluate the contamination of Gorgan City with the emphasis on heavy metals such as iron, manganese, arsenic, and lead. The comparison results of the statistical values of heavy metals showed that there is not much difference between the values of parameters in spring and autumn. Also, the amounts of heavy metals studied are less than the WHO and 1053 standard values.

Based on the distribution map of heavy metals, most of the changes in these parameters are related to the west part of the study area in the north-south direction located in the old texture of Gorgan and the areas with the highest population density. Also, except for manganese, the changes in heavy metals in the spring are similar to those in the fall. Based on the classification of heavy metal pollution index (HPI), heavy metal evaluation index (HEI) and degree of contamination index (Cd), the quality of groundwater in both spring and autumn was evaluated as good. Based on the increase in population of Gorgan city and the lack of a proper wastewater collection system, it is obvious that with the lack of proper management, the possibility of contamination of groundwater resources in this area in the future will not be far from expectation. Therefore, periodic and regular surveillance of Gorgan's groundwater resources is recommended.

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