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Carcinogenic and Non-carcinogenic Risk Assessment of Heavy Metals in Water Resources of North East of Iran in 2018

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ABSTRACT

Introduction: Contamination of water with heavy metals has turned into a health concern, particularly in the developing countries. In this study, concentration of heavy metals and associated carcinogenic and non-carcinogenic risk was investigated in water samples collected from Gonbad-e Kavus, a high-risk area for cancer.

Materials and Methods: Samples were collected from Gorgan River, Golestan reservoir and wells around villages with high prevalence in 2018.Samples were analysed through inductively coupled plasma mass spectrometry. After determining the concentration of heavy metals in water samples from different sources, health risk assessment was carried out according to the Environmental Protection Agency.

Results: Arsenic in samples 6-9 was higher than 10 µg/L, calcium and magnesium in sample 5 was higher than 200 mg/L and 150 mg/L respectively, and sodium in all samples was higher than 50 mg/L. According to the findings, these concentrations were higher than the maximum allowed limit in most water samples. Hazard quotient (HQ) in samples 8 and 9 were associated with arsenic and health risk in sample 1 was related to antimony. Furthermore, since all samples contained high amounts of lithium, water from this area better should not be consumed by children older than one year.

Conclusion: Given the high rate of arsenic contamination, consumption of water in the study area could be health threatening for all individuals and is not recommended for children. This highlights the need for taking immediate actions to review the water treatment process and ensure safety of the drinking water in this area.

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Introduction

Heavy metals are one of the most important environmental pollutants. Widespread water pollution is a common challenge in the developing countries, which may be result in physical and biological changes as well as accumulation of toxic and harmful substances in water[1-2]. Contamination reduces the quality of water, so that it cannot be consumed by living creatures anymore. Contrary to some degradable contaminants such as agricultural wastes, heavy metals including cadmium, lead, and arsenic are non-biodegradable and could be health threatening[3-5].

Presence of heavy metals in the drinking waterand food can have adverse health effects on humans. One of the important results of heavy metals' sustainability in the environment is the entry of metals in the food chain[6-8].Some heavy metals such as iron, manganese; cobalt, copper,

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and zinc are essential for plants and animals, but dangerous at high concentrations. However, certain heavy metals such as arsenic, cadmium, and lead are toxic even at very low concentrations[9-11].

Industrial wastewater, chemical fertilizers, solid waste leachate, and geological structures are known sources of heavy metals contamination in water systems[6,12]. Contamination of water with heavy metals can spread to different parts of the aquatic ecosystem, such as water, sediments, and plants. Groundwater is the main water source for drinking, agricultural, and industrial purposes. Almost one-third of the world's population uses groundwater to supply drinking water[13].

Several risk assessments studies were conducted on contamination of water sources with heavy metals. Lee et al. assessed bioavailability of arsenic, copper, lead, and zinc in soil and performed chemical analysis for groundwater and stream water samples from abandoned mine areas (Dukeum, Dongil, Dongjung, Myungbong and Songchun mine areas). High values of cancer risk for As (1.16×10^{-5}) were detected through soil ingestion pathways in the Songchun mine area and assessed through water exposure pathways in the all mines except Dukeum[14].In China, Wang et al.analysed several water quality indices and performed risk assessment of heavy metals including iron, copper, manganese, zinc, arsenic, chromium, mercury, lead, and cadmium in community water sources. The highest rate of contamination was observed in reservoirs and river water caused by chromium and arsenic[15].

In the northeast of Iran, Gonbad-e Kavusis a high-risk area for cancer. Northern Iran lies on the Asian belt with predominance of upper-gastrointestinal cancers. The Golestan population-based cancer registry (GPCR) was established in Golestan province. Overall, 19807 new cancer cases were registered during the study period (2004-2013) with an average of 1981 cases per annum as well as overall Age-Standardized Incidence Rates (ASR) of 175.0 and 142.4 in males and females, respectively[16] .

Therefore, this study was aimed to determine the chemical properties and level of heavy metals in different water sources (surface and groundwater in high risk area, Fajr, Soltan Ali, and Aq Abad villages). Furthermore, the study targeted at assessing the risk factor, cancer and non-cancerous effects, as well as adverse threshold for men, women, and children.

Materials and Methods

The city of Gonbad-e-Kavus is located at 55° 18' longitude and 37° 17'min latitude in the northern and central parts of the Golestan Province (Figure 1). The soil of this area is mainly composed of volcanic plains. The geosciences of the Gonbad-e-KavusCity show that this area is marshy, habitable, abandoned, impassable, and there exists major faults.

Sampling was done based on hydro geological studies and from certain sampling stations suggested by the regional water authorities (Figure 2).

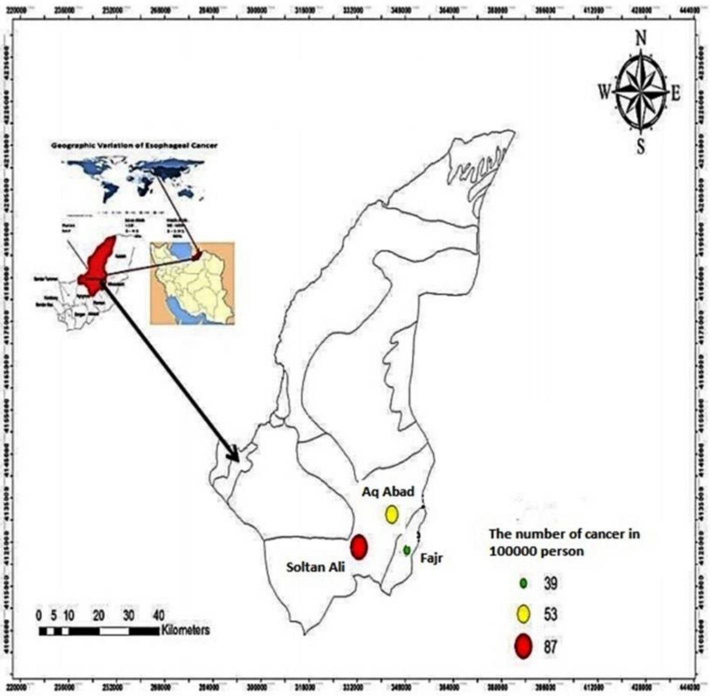

Figure 1: Location of the study area and epidemiology of cancer in Gonbad-e Kavus

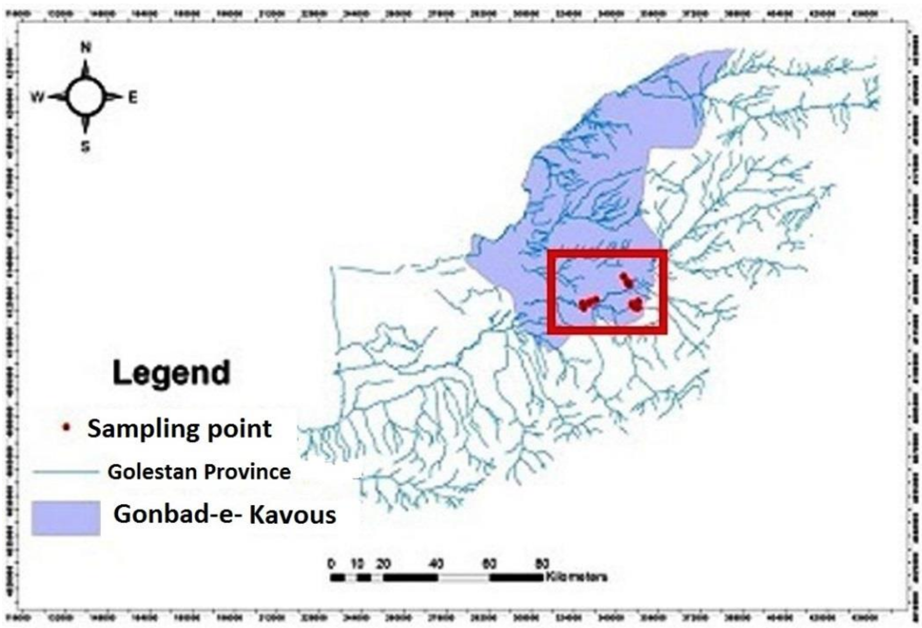

Figure 2: Location of water sampling stations in hydrographic network

In sampling, one sample was collected from well water located in Fajr village, three samples of drinking water were collected from all three villages(Fajr, Aq Abad, and Soltan Ali), and three samples of Gorgan River, which passes through these three villages were collected.The sample size for testing soluble/insoluble heavy metals was about 100 mL. The samples were stored immediately. Then, for preserve of sample, 1.5 mL of nitric acid (HNO_3) added to sample and the pH is brought to less than two. For samples with a high buffer capacity, the amount of acid was

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increased (5 mL for buffer or alkaline samples)[17]. The samples were analysed via inductively coupled plasma mass spectrometry (ICP-MS). Followed by obtaining the heavy metals' concentrations, health risk assessment was carried out according to the Environmental Protection Agency (EPA)[18].

According to the World Health Organization (WHO) and the International Chemical Safety Program, risk assessment is a process for estimating the risk to an organism, system or population[10]. In order to assess the risk of a Hazard quotient (HQ) < 1 risk, no significant toxicity should be observed and if HQ > 1, the probability of a potential hazard is calculated as follows:

HQ = ADD / RfdEq (1)

ADD (mg/kg-d) is equal to the mean daily dose and RfD (mg / kg-d) is equal to the reference dose(18).

ADD = (Cm.CR) / BW Eq(2)

Cm is the concentration of element measured in water and CR is the average daily water consumption (3 liters per day). The body weight (BW) of consumers is 78 kg for men, 65 kg for women, and 14.5 kg for children[19]. The water consumption limit in the area was calculated using the following equations:

CRlim = (RfD.BW) / Cm Eq(3)

CRlim = (ARL.BW) /Cm.CFS) Eq(4)

CRlim is the highest daily intake limit and CFS is the steady incidence of cancer. The amount of ARL is constant and equal to 10^{-5}.

Ethical Issue

This study was conducted with approval of Golestan University of Medical Sciences. Research Ethics Code was IR.GOUMS.REC.1398.034.

Results

In order to determine water contamination levels, the concentration of elements and heavy metals in water samples was compared with the existing standards set by the WHO[20] and the US Environmental Protection Agency[21] (Tables 1 and 2).

Table 1: Chemical parameters of the water quality in the study area compared with the global standards and guidelines

Sample	Water source	Cl(mg/L)	pH	TDS(mg/L)	Na(mg/L)	Mg(mg/L)	K(mg/L)	Ca(mg/L)
1	Groundwater	2.9	7.5	566	74	28.9	1.93	83.6
2	Groundwater	3.1	7.9	620	59.9	26	1.65	76.5
3	Groundwater	8	8.1	1103	77.9	38.6	2.66	96.6
4	Surface	33	7.48	3209	62.5	25.4	3.26	77.7
5	Surface	4.9	7.61	872	1300	328.9	9.82	202.5
6	Groundwater	53	7.21	4981	130.1	24.6	1.77	61.9
7	Groundwater	21	7.34	2826	224.1	27.9	2.05	52.6
8	Surface	2.5	7.7	533	464.1	111.2	5.78	115.4
9	Surface	3.1	7.76	564	228.6	80.5	8.14	66.2
Mean	-	14.61	7.62	1697.1	291.24	76.8	4.11	92.55
SD	-	17.81	0.27	1599.08	400.06	99.27	3.06	45.29
US.EPA	-	250	6.5-8.5	500	60-90			
WHO	-	250	6.5	1500	50	150	12	200

As shown in table 1, the mean concentrations (as mg/L) of the chemical parameters were as follows: Cl (14.61 ± 17.81), pH (7.62 ± 0.27), TDS (1607.1 ± 1599.1), Na (291.24 ± 400), Mg (76.8 ± 99.27), K (4.11 ± 3.06), and Ca (92.55 ± 45.29). The results indicated that the mean concentrations of the chemical parameters various area varied significantly. The mean concentrations of TDS and Na were higher than the permissible limit values.Calcium in sample 5 was higher than 200 mg/L, magnesium in sample 5 was higher than 150 mg/L, and sodium in all samples was higher than 50 mg/L, showing that these concentrations were higher than the maximum allowed limit in most water samples.

Table 2: Concentration (µg/L) of heavy metals in the water samples and its comparison with the global standards

Sample	Ba	Cs	Mo	Li	Rb	Sc	Se	Sb	Zn	As
1	87.44	< 0.1	0.49	17.30	< 1	2.36	2.15	1.60	93.61	2.75
2	61.22	< 0.1	0.36	12.73	< 1	2.12	2.77	1.57	20.04	2.88
3	89.77	< 0.1	2.84	19.56	1.63	2.75	2.66	1.02	< 1	2.47
4	93.10	< 0.1	0.48	12.13	< 1	1.38	2.36	0.84	< 1	3.18
5	51.29	0.28	9.47	86.68	1.55	< 1	7.47	< 1	< 1	7.29
6	55.47	0.29	3.91	19.60	1.28	3.02	14.82	3.02	< 1	12.87
7	94.77	0.36	2.31	25.07	1.55	3.42	13.13	3.42	488.09	13.4
8	70.20	< 0.1	0.08	40.20	1.20	3.20	8.61	3.20	< 1	18.2
9	105.05	0.49	2.79	39.36	1.94	1.08	10.49	1.08	3.17	18.17
Mean	78.70	0.36	2.53	30.29	1.53	2.42	7.16	1.97	151.23	9.02
SD	19.45	0.10	2.94	23.52	0.26	0.85	4.94	1.07	227.98	6.70
US.EPA	2000	-	-	700	-	40	5	6	5000	10
WHO	700	-	70	-	-	50	5	20	5000	10

As shown in table 2, the mean of heavy metals concentrations (as µg/L) were as follows: Ba (78.70 ± 19.45), Cs (0.36 ± 0.1), Mo (2.53 ± 2.94), Li (30.29 ± 23.52), Ro (1.53 ± 0.26), Sc (2.42 ± 0.85), Se (7.16 ± 4.94), Sb (1.97 ± 1.07), Zn (151.23 ± 227.98), and As (9.02 ± 6.7). The results indicated that the mean concentrations of the heavy metals varied significantly in various sampling areas. The mean concentrations of Se were higher than the permissible limit values.

Arsenic in samples 6-9 was higher than 10 µg/L, indicating that these concentrations were higher than the maximum allowed limit in most water samples.

Arsenic level exceeded themaximum allowable limit in water samples6 to 9 (Table 2). Concentrations of calcium and magnesium were also higher than the standard limit in several samples (Table 1). The concentration of sodium ion was higher than the global standard limit in most samples. In one sample, magnesium and calcium levels exceeded the global standard level. Results of the risk assessment for arsenic, lithium, and antimony are shown in tables 3 to 5.

Table 3: Arsenic risk assessment and maximum contaminant level for water resources in Gonbad-e Kavus, Iran

Sample	As (µg/L)	HQ			ADD			Carcinogenic effect			Non carcinogenic effect		
		Children	Male	Female	Children	Male	Female	Children	Male	Female	Children	Male	Female
1	2.75	0.0005	0.0001	0.0001	0.16	0.03	0.03	0.048	0.26	0.21	21.75	117	97.5
2	2.88	0.0005	0.0001	0.0001	0.16	0.03	0.03	0.048	0.26	0.21	21.75	117	97.5
3	2.47	0.0005	0.0001	0.0001	0.16	0.03	0.03	0.048	0.26	0.21	21.75	117	97.5
4	3.18	0.0006	0.0001	0.0001	0.16	0.03	0.03	0.032	0.17	0.14	14.5	78	97.5
5	7.29	0.001	0.0002	0.0003	0.33	0.06	0.1	0.013	0.07	0.061	6.21	33.42	65
6	12.87	0.002	0.0004	0.0006	0.66	0.13	0.2	0.008	0.043	0.036	3.62	19.5	16.25
7	13.14	0.002	0.0005	0.0006	0.66	0.16	0.2	0.007	0.04	0.03	3.34	18	15
8	18.2	0.003	0.0007	0.0008	1.25	0.26	0.26	0.005	0.028	0.024	2.41	13	10.8
9	18.17	0.003	0.0007	0.0008	1.25	0.26	0.26	0.005	0.028	0.024	2.41	13	10.8

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Table 4: Lithium risk assessment and maximum concentration level for water resources in Gonbad-e Kavus, Iran

Sample	Li(μg/L)	HQ			ADD			Non carcinogenic effect		
		Children	Male	Female	Children	Male	Female	Children	Male	Female
1	17.3	1.5	0.3	0.35	0.003	0.0006	0.0007	1.7	9.17	7.64
2	12.73	1	0.2	0.25	0.002	0.0004	0.0005	2.41	13	10.8
3	19.65	1.5	0.35	0.4	0.003	0.0007	0.0008	1.52	8.2	6.8
4	12.13	1	0.2	0.25	0.002	0.0004	0.0005	2.41	13	10.8
5	86.68	8.5	1.5	1.5	0.017	0.003	0.003	0.33	1.81	1.5
6	19.6	1.5	0.35	0.4	0.003	0.0007	0.0008	1.52	8.21	6.8
7	25.07	2.5	0.45	0.5	0.005	0.0009	0.001	1.16	6.24	5.2
8	40.2	4	0.5	0.5	0.008	0.001	0.001	0.72	3.9	3.25
9	39.36	4	0.5	0.9	0.008	0.001	0.0018	0.74	4	3.33

Table 5: Antimony risk assessment and maximum concentration level for water resources in Gonbad-e Kavus, Iran

Sample	Sb(μg/L)	HQ			ADD			Non carcinogenic effect		
		Children	Male	Female	Children	Male	Female	Children	Male	Female
1	1.6	0.75	0.25	0.25	0.0003	0.0001	0.0001	3.6	19.5	16.25
2	1.57	0.75	0.25	0.25	0.0003	0.0001	0.0001	3.8	20.8	17.33
3	1.02	0.5	0.17	0.1	0.0002	0.00007	0.00004	5.8	31.2	26
4	0.4	0.25	0.075	0.075	0.0001	0.00003	0.00003	7.25	39	32.5
5	< 0.1	0.025	0.075	0.01	0.00001	0.000003	0.000004	64.4	346	288.8
6	< 0.1	0.025	0.075	0.01	0.00001	0.000003	0.000004	64.4	346	288.8
7	0.69	0.25	0.05	0.05	0.0001	0.00002	0.00002	9.6	52	43.3
8	1.03	0.5	0.175	0.1	0.0002	0.00007	0.00004	5.8	31.2	26
9	2.08	1	0.25	0.25	0.0004	0.0001	0.0001	2.9	15.2	13

HQ in samples 8 and 9 was associated to arsenic and was related to antimony in sample 1. Given the lithium rate of above 1, using drinking water from this area is not recommended for all individuals, especially for children.

Discussion

Heavy metals constitute a very heterogeneous class of elements considerably varied in their chemical properties and biological functions. Increasing concerns exist about the potential deleterious impacts of these metals in many countries because they not only affect the productivity of ecosystems, but also impact on plants, animals, and human beings[22].

In this study, due to the normality of the data, Spearman method was used to determine the correlation coefficient between the data, at the confidence level of $\rho > 0.01$ and $r > 0.8$.Positive and strong correlation ($r = 0.9$) of lithium with strontium ($\rho = 0.01$) indicated a common original and geochemical behavior among these elements. These two elements are among the alkaline elements of the periodic table and are abundant in water. Positive and very strong correlation ($r > 0.8$) was found between scandium and antimony ($\rho > 0.01$). These two elements are not from the same geochemical source and do not show similar geochemical behaviors, only their ion radius is almost close to each other. A positive and very strong correlation ($r > 0.7$) was observed between selenium and cesium ($\rho > 0.01$), where cesium is a very active alkali metal that can compete with non-metal selenium. They do not have the same origin and geochemical behavior, also selenium has a very strong positive correlation ($r > 0.7$) with arsenic ($\rho > 0.05$), which indicates its common origin and geochemical behavior similar to these elements. These two elements are in a row of the periodic table and have the same electron arrangement and ionic radius. In most reports, arsenic was studied with selenium, perhaps due to the geochemical behavior of the two elements. Positive correlation ($r > 0.6$) of cesium with rubidium ($\rho > 0.05$) indicates their common origin and similar geochemical behavior because these

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two elements are in a group of the periodic table and both are alkali.

Given the carcinogenic effects of arsenic, consumption of water in Gonbad-e Kavus could be health threatening and is not recommended for children due to arsenic's non-carcinogenic effects on this age group. Similar to our findings, Bamuwamye et al. reported that concentrations of lead and arsenic in drinking water of Kampala region in Uganda exceeded the global limits[23]. Elumalaiet al. conducted a risk assessment of heavy metals in groundwater and used pollution indicators by multivariate statistical methods in South Africa[24]. They concluded that most of these waters are of good quality, but 15% are inappropriate for drinking. Moreover, hazard assessment in other heavy metals showed that groundwater was not suitable for drinking and the risk was high in all groups, including the older age group[24]. In the risk assessment of non-carcinogenic metals including lead, zinc, and chromium in drinking water sources in Hamedan (Iran), concentration of lead exceeded the WHO and Institute for Industrial Research Standardization limits in 46.41% of the drinking water samples. In addition, 39.02% of the samples contained a concentration higher than the EPA standard, while none of the samples had a sectional risk for children and adults. Although the level of chromium and zinc was lower than the standard limit, they showed risk in groups of less than one month and one to three months because of their high pathogenicity potential; so, the concentrations below the standard cannot be a guarantee of a lack of risk[25]. In a study by Lim HS et al., the concentration of cadmium and zinc in most water streams used for drinking water around the mining area was higher than the national limit but the HQ index. All heavy metals in drinking water were reported at the acceptablelevel[26]. Chromium, copper, cobalt, and manganese were permitted in the North Sea region of northern Pakistan in 2011 according to the EPA and WHO standards. Concentrations of cadmium, nickel, Lead, and zinc were 17%, 2%, 29%, and 6% higher than the normal level, respectively. The assessment of hazards quotient and risk indicator indicated that it is harmless to humans[27]. Momot et al. evaluated the health risk of metal in the middle of the Russian region; the risk assessment of metallic agents for carcinogenic diseases was 3.95×10^{-3} and 0.98×10^{3} for non-cardiovascular diseases[28]. This result can be justified by the geological conditions of the region, most of which are limestone and dolomite, and most water resources are salty.

We believe that both geogenic and anthropogenic factors are responsible for the contamination of water sources with various heavy metals in Gonbad-e-Kavus.

Conclusion

Analytical findings demonstrated that water resources of Gonbad-e-Kavushad high concentrations of As, Li, and Sb respectively. Given the hazards and health effects associated with arsenic, consumption of water in this area may be health threatening for all individuals and is not recommended for children. Further assessment of water quality indices and reconsideration of water treatment process are necessary to ensure safety of drinking water in the study area.

Due to the geochemical situation of the region and the high level of some elements such as arsenic, selenium, etc., water resources treatment as well as residents' education about the causes of cancer are recommended.

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Conflict of interest

The authors declare that there is no competing interest regarding publication of this study.

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