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Investigating the Residue of Diazinon, Chlorpyrifos, and Dichlorvos in Urban Drinking Water Supply Sources and Determining the Water Quality Index in Tiran-Karvan in 2020

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ABSTRACT

Introduction: Contamination of water sources with toxins is an environmental problem and a serious threat to the health of societies. This study aimed to ascertain the remaining levels of toxins in urban drinking water networks and compute the water quality index.

Materials and Methods: To examine the residual concentration of diazinon, chlorpyrifos, and dichlorvos in urban drinking water distribution networks, 35 underground and surface water sources were sampled in Tiran-o-Karvan in Isfahan province, Iran. GC-ECD device was used to analyze the samples.

Results: Diazinon was not observed in any of the sampling seasons. The maximum concentration of chlorpyrifos and dichlorvos was 61 and 100 ppb, respectively. Although chlorpyrifos was seen more in the hot season and dichlorvos in the cold season, the mean concentration of these toxins in different seasons was lower than the guidelines of Iran and the World Health Organization (WHO). The mean concentration of nitrate, and hardness, in wet and dry seasons was 46.23 and 46.42, and 343.7 and 338.8 mg/l, respectively.

Conclusion: The mean and standard deviation of quality index of Iran's water resources (IRWQI) in wet and dry seasons were 52.16 ± 45.3 and 50.69 ± 15.8, respectively. The residual concentration of toxins in some water sources of this city exceeded the guidelines of Iran and the WHO in summer and spring.

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Introduction

Access to clean water and food is necessary for the survival of humans, animals, plants, and other living organisms and plays a determining role in their quality of life [1]. Therefore, food, and especially water, should not contain unwanted pollutants, microorganisms, or harmful chemicals [2, 3]. Studies have shown that factors such as rapid population growth, the development of industries and agriculture, and lack of proper management have diminished the quantity and quality of available water resources [4].

Groundwater and surface waters are important sources of water supply systems. Treated water sources are distributed through pipelines or in mineral water bottles. In Iran, people generally have access to tap water [5]. Water is a passive carrier of various pollutants, which causes many complications for human health. Note that low-quality drinking water causes 80% of all diseases.

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Thus, access to clean and unpolluted water is essential to a healthy lifestyle [6, 7]. Drinking water may become contaminated due to natural and human activities. Wastewater from agricultural, industrial, and mining activities are human sources of pollutants such as toxins and pesticides [7].

The contamination of water resources with pesticides is an environmental issue resulting from agricultural expansion, which is a consequence of population growth that leads to increased pesticide usage. Agricultural pesticides can infiltrate water sources by direct washing or irrigation. Precipitation can also occur on fields and contribute to introducing pesticides into surface water before their breakdown. In addition, herbicides can infiltrate groundwater by permeating through layers of soil. Occasionally, certain pesticides can enter the atmosphere and then pollute surface water and soil via precipitation [8, 9].

The entry of pesticides into drinking water sources and networks can harm human and environmental well-being due to their robust resilience to ecological variables, ability to dissolve in water, and high toxicity to living species. The manifestation of their adverse effects is contingent upon the chemical's nature, period of usage, duration of exposure, concentration of intake, and human toxicity threshold. The entry of pesticides into the body brings acute and chronic effects such as abdominal pain, headache, skin and digestive diseases, respiratory diseases, nervous disorders, cancer, etc. [9, 10].

Many pesticides with different chemical compositions used worldwide are classified as herbicides, insecticides, fungicides, acaricides, nematicides, etc., according to their usage. The most common are herbicides, which constitute approximately 80% of all pesticide use. In terms of chemical composition, pesticides belong to organochlorine, organophosphorus (diazinon, chlorpyrifos, and dichlorvos), organonitrogen or carbamate, and pyrethroid groups. Among the given pesticides, organophosphorus compounds are so diverse that they account for about 40% of the world pesticides [11, 12].

Organochlorine, organophosphorus, and carbamates pesticides are particularly important due to their properties such as cumulative properties, carcinogenicity, inhibition of acetylcholinesterase activity, mutagenicity, and effects on the nervous system [9, 10]. Therefore, it is necessary to prevent the entry of Pesticides into water sources, and the measurement of Pesticides residue in water is necessary for human health and environmental protection. The most important step in preventing contamination of water sources with residual Pesticides is to determine their residual concentration accurately and compare the value with standard [13]. This study aimed to determine the residual concentration of diazinon, chlorpyrifos, and dichlorvos and calculate the water quality index in the studied area.

Materials and Methods

This descriptive-analytical cross-sectional study was conducted in Tiran-o-Karvan in Isfahan province, Iran. The sampling stations are depicted in Figure 1.

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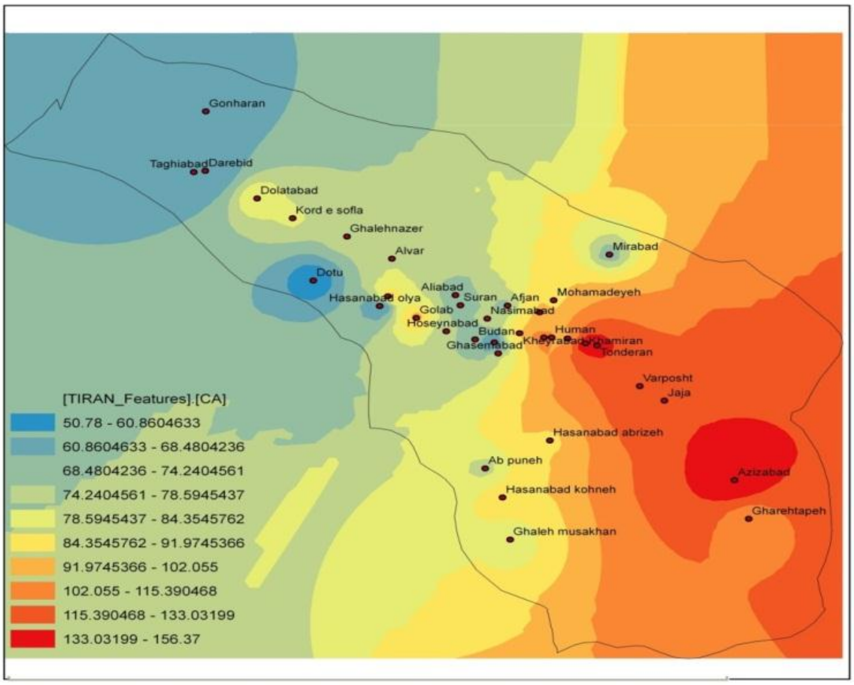

Figure 1: Sampling stations

Sampling

Based on the city drinking water supply sources, the total number of sampling stations in each sampling stage was 35 stations (33 samples from underground water sources and two samples from surface water). The drinking water supply sources of this city are provided by surface and underground water. One of the surface water sources is Zayanderud Dam, which supplies drinking water of Tiran, Rezvanshahr, Asgaran (partly), and several villages.

The second surface water station is Cheshme Morghab, located in the city. The water of several villages comes from underground water sources. As a result, all sources of drinking water in the city were identified and selected for sampling. Since water from several sources is mixed in some distribution networks, the water supply sources of these networks were sampled separately. A total of 245 samples were taken from the water sources of this city.

Sampling for testing the toxins was performed using dark glass containers capped with aluminum foil. To prepare these containers, they were first washed with a detergent and hot water, and then they were placed in a 5% nitric acid solution for a day. Next, the containers were placed in the oven for 4 hours. Finally, after cooling down, these containers were used for sampling the toxins. The samples were transferred to the laboratory on ice. A 1.5-L capped plastic container was used to test chemical parameters, and glass containers were used for microbial tests. The number of samples is presented in Table 1.

Table 1: The number of samples taken from water sources in different seasons

Row	Test	Number of samples			
		Winter	**Spring**	**Summer**	**Autumn**
1	Chemical parameters	35	Not measured	35	Not measured
2	Faecal coliforms	35	Not measured	35	Not measured
3	Pesticides	Not measured	35	35	35

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Determining the hardness of water samples

Total hardness: First, 25 ml of the sample was poured into a 250-ml Erlenmeyer flask. Then, 2 ml of ammonium buffer solution was added to it, and then, an appropriate amount of Eriochrome Black T reagent was added to it, and it was tittered with the 0.01 molar EDTA standard solution while the Erlenmeyer flask was shaken. A change in color from red to blue indicated the end of the titration. The samples for whose titration more than 15 ml of the standard EDTA solution was used were diluted with distilled water to the volume of the sample, and this test was repeated twice for each sample [14].

Calcium hardness

First, 25 ml of the sample was poured into a 250-ml Erlenmeyer flask. Next, 2 ml of the sodium hydroxide was added to it. Subsequently, an appropriate amount of murexide reagent was added to it and titrated with EDTA standard solution while the Erlenmeyer flask was shaken. A color change from pink to purple indicated the end of the titration [14].

Phosphate determination

Four milliliter of ammonium molybdate was added to 100 ml of the sample, and it was shaken completely. Then, 10 drops of ascorbic acid were added to the solution, and its absorption value was read in the spectrophotometer after 10 minutes and before 12 minutes at a wavelength of 690 nm [14].

Nitrate determination

Samples were prepared using the standard American Public Health Association (APHA) method. After that, nitrate concentration was determined using spectrophotometric method and at the wavelength of 275 and 220 nm [14].

Measurement of toxins

The dispersive liquid-liquid microextraction (DMLLE) method was employed for the extraction of toxins. In this method, 5 mL of sample, 1 mL of acetonitrile solvent, and 20 mL of chlorobenzene were used. First, 5 mL of the sample was transferred to a 15-ml funnel with a screw cap. Then, 1 mL of acetonitrile and 20 μL of chlorobenzene were added to it. Next, it was placed in a centrifuge device at 5000 rpm for 5 minutes. Finally, 2 μL of the precipitate was taken and injected into the device. An Agilent Gas Chromatography equipped with a mass spectrometer and a DB5 capillary column (with a length of 60 m, an internal diameter of 0.25 mm, and a solid phase thickness of 250 μm) was used to test the toxins. The injection temperature was 240 °C in 2:1 split mode. The initial temperature of the oven was kept at 80 °C for 1 minute; then, the temperature was raised to 280 °C with a gradient of 30°/min and was finally kept at this temperature for 5 minutes [15].

Sodium adsorption ratio calculation

Calculations were made using Eq.1 to determine the amount of sodium absorption coefficient. All the units of measurement of the parameters in the sodium absorption coefficient equation were meq/l [16].

$$\boldsymbol{SAR} = \frac{\boldsymbol{Na}}{\sqrt{(\frac{\boldsymbol{Ca+Mg}}{\boldsymbol{2}})}} \qquad \text{Eq. 1}$$

Calculating the $IRWQI_{GC}$

The index of each parameter was calculated based on its measured data and by using the ranking curves in Iran water quality calculation guideline [17]. The numerical value of the quality index was obtained by Eq. 2.

$$IRWQI_{GC} = \left[\prod_{i=1}^{n} I_i^{wi}\right]^{\frac{1}{y}} \qquad \text{Eq. 2}$$

$$y = \sum_{i=1}^{n} w_i \qquad \text{Eq. 2}$$

Where, wi is the weight of the ith parameter, n is the number of parameters, and Ii is the value of the index for the ith parameter from the ranking curve.

The weight and value of each parameter for calculating IRWQI are presented in Table 2.

Results

Measured toxins

Based on the results, it was determined that the highest concentration of identified toxins was 100, 61, and 0.1, respectively, belonging to dichlorvos, chlorpyrifos, and diazinon (Table 3).

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Table 2: The weight and value of each parameter for calculating IRWQI

Row	Parameter	First rank	Second rank	Third rank	Fourth rank	Fifth rank	Sixth rank	Seventh rank
1	Nitrate	33	2					
2	Hardness	2	21	3	5	4		
3	EC		1	21	13			
4	Phosphate		4	3	4	15	9	
5	pH		1	10	8	11	5	
6	SAR		7	4	11	7	6	
7	F.C							35

Table 3: Comparison of the concentration of pesticides measured and their permissible limit in drinking in terms of micrograms per liter (ppb)

Pesticide	Min	Max	Maximum limit	
			National standard	WHO and EPA guidelines
Diazinon	N.D*	< 0. 1		0.1
Chlorpyrifos	N.D	61	30	30
Dichlorvos	N.D	100		

* Not detect

Concentration of physical and chemical parameters

This study measured the concentration of physical and chemical parameters of water samples. Based on the results, the highest nitrate concentration was found in the cold season and the lowest in the warm season (Table 4).

Calculation of IRWQI

According to Figure 2, the highest value of IRWQI (i.e., 90) was obtained in stations 6 and 35, and the lowest value (i.e., 19) was obtained in station 27. The mean and standard deviation of IRWQI in wet and dry seasons were 52.16 ± 45.3 and 50.69 ± 15.8, respectively.

Table 4: Concentration of physicochemical and microbial parameters of water samples

Parameters	Season			
	Winter		Summer	
	Max	Min	Max	Min
Nitrate (mg/l)	85	22.3	77	5.4
Phosphate(mg/l)	0.099	0	0.099	0
Total hardness (mg/l $caco_3$)	1120	160	1120	160
Ca (mg/l $caco_3$)	640	60	640	62
Mg (mg/l $caco_3$)	560	52	564	20
Na (mg/l)	175	10	178	8
pH	7.9	7.2	7.8	7.2
Ec (mS)	1865	331	1900	320
SAR	3.09	0.14	3.04	0.12
Total coliform	N.D*	N.D*	N.D*	N.D*

* Not detect

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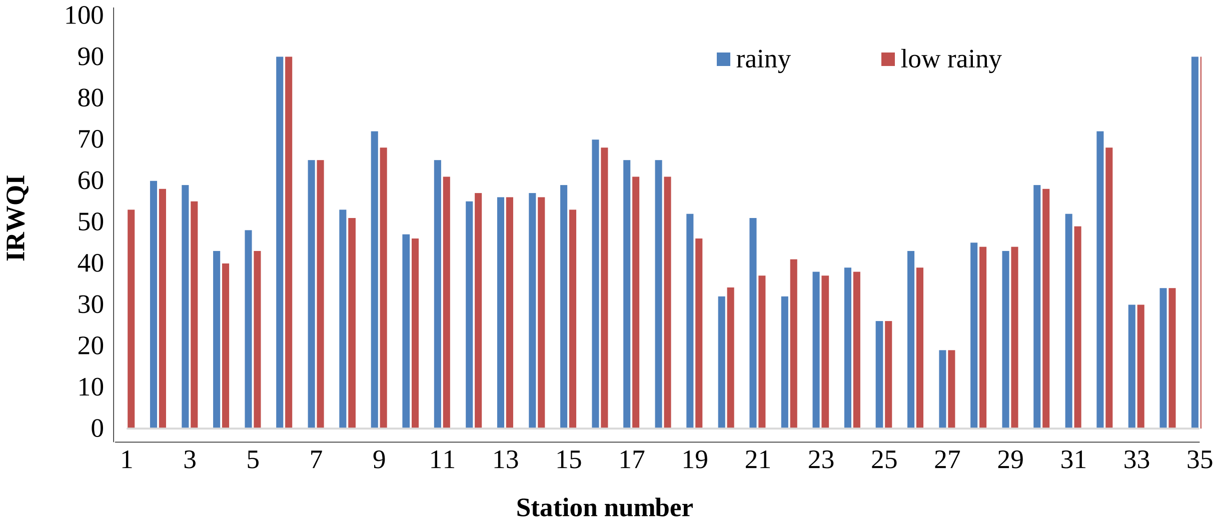

Figure 2: Water quality index in winter and summer seasons

Discussion

According to a study in 2019, about 96% of Iranian cities have access to safe water supply systems [18]. However, there is still a possibility of drinking water contamination with toxic compounds. The present study aimed to determine the chemical and microbial quality of drinking water sources and to investigate the presence of pesticide residues in water sources of Tiran-o-Karvan in Isfahan province.

Based on Table 3, in three sampling seasons, the concentration of diazinon was not detected or was below the detection limit of the device; as a result, the amount of this toxin in the city drinking water was lower than the standard defined by the World Health Organization (WHO) and the U.S. Environmental Protection Agency. The standard developed by the WHO and the U.S. Environmental Protection Agency is 20 µg/l [19]. In the study by Hasani et al. in 2010-2011 on the excavated drinking water wells of Shemiranat (Iran), the concentration of organophosphorus toxin diazinon was not detected in any of the studied wells, which is similar to the results of this study [20]. It seems that the pattern of using this toxin in the region (mostly in gardens and on fruit trees) has led to this phenomenon; the residual toxin on the foliage of trees is decomposed through physical factors such as sunlight and wind and does not have enough time to reach the surface of the soil, and does not enter the underground water sources. Diazinon is a relatively volatile organophosphorus toxin and insecticide, which is used in large quantities to kill flies and ticks, especially *Ornithodoros tholozani* ticks. This toxin alone is used abundantly in agricultural fields, and its residue is found in underground water and rivers [21]. Moreover, according to the drinking water standard of Iran, the concentration of chlorpyrifos toxin in water sources is 30 µg/l. Although the mean level of this toxin is lower than the standard in all seasons, it was above the standard in 5 stations in summer. The values in the table indicate the highest mean concentration of this toxin in summer in the analyzed samples. It seems that the most important reasons for this phenomenon include the existence of agricultural lands and the high consumption of this toxin in the region, precipitation in the previous months, and irrigation of agricultural lands. Khodadadi et al. investigated the residual organophosphorus and carbamate toxins in drinking water sources of Hamadan (Iran). They concluded that the presence of chlorpyrifos in the drinking water sources of this city is due to its high consumption in the previous months, precipitation in the previous months, and the abundance of agricultural lands in the region, which is similar to the results of the present study [22]. Chlorpyrifos is one of the most widely used organophosphorus pesticides, which is used

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to protect a wide range of products such as fruits, vegetables, grains, edible mushrooms, and cotton and to fight household and livestock pests [23, 24]. The excessive use of chlorpyrifos pollutes the air, underground water, rivers, lakes, and rainwater, and the resulting pollution can be found up to 24 km from the place of use [25]. The results of measuring the concentration of dichlorvos in spring and summer showed that, in most cases, the concentration of this toxin was below the detection limit of the device, and dichlorvos was not detected in 15% of the samples. The reason for the observation of dichlorvos in water resources of the city in the fall could be the presence of many greenhouses in the study area. Due to the widespread use of this toxin in greenhouses during the greenhouse cultivation seasons (mainly in late summer and fall), this toxin may have been released into the underground water sources of the city [19]. This toxin is widely utilized in greenhouses to fight pests, which is why it can enter the underground water of the region. Morovvati and Nematollahi investigated the concentration of pesticide residues in cucumber crops in greenhouses of Isfahan province. They found that 70% of the samples contaminated with illegal pesticide residues contained illegal residues of dichlorvos [26]. This finding indicates that dichlorvos is widely used in greenhouses. Khodadadi et al. examined the residue of organophosphorus and carbamate toxins in drinking water sources of Hamadan. They concluded that the concentration of diazinon in drinking water sources of this city was higher in the fall compared to other seasons, and its concentration had exceeded the standard limit in some cases. This result is similar to the present study in terms of the presence of toxin residues in the fall [22].

In other measured parameters, there was no significant difference between the concentration of samples of cold and warm seasons. According to the results of the current study, nitrate concentration was high and above the standards in several wells.

Pasban et al. investigated nitrate concentration in Bojnourd, Iran, drinking water wells. They found that the mean nitrate concentration in drinking water wells of this city was 54.9 mg/L. The mean nitrate concentration in wells within and outside the city showed a significant difference (P-value = 0.003), and the mean nitrate concentration in wells inside the city was higher [27]. Important factors can cause the concentration of nitrate to rise in the drinking water of this region. Improper use of nitrogen fertilizers in agriculture could be the main source of the increase in the nitrate ions in water resources of this region. Mohammad Zaheri et al. explored nitrate and nitrite pollution in underground water sources of Dehgolan (Iran) and concluded that after using fertilizer in agricultural lands, nitrate in underground water sources increased [28]. Other reasons could be the higher population density in the center of the studied area and factors that increase nitrate, such as improper waste disposal and the existence of more agricultural lands in the central part and around the villages. Hiyama and Babiker investigated the quality of underground water in Nasuno, Japan, using the geographic information system. They reported that the quality of underground water from north-west to south-east diminishes due to the decrease in water depth, increase in the entry of fertilizer from rice fields, and a rise in population density. They also concluded that the quality of underground water in the upper and lower parts is quite variable compared to the middle parts and showed that 30% of the samples has a concentration higher than 45 mg/l, which is similar to the results of the current study [29].

In all the sampling stations in the two seasons, the amount of phosphate was below the standards, so it does not create any restrictions in terms of different uses, such as drinking, irrigation, and agriculture. According to Table 3, the amount of calcium varied from 60 mg/l to 640 mg/l. According to the Iranian standard, calcium concentrations of less than 75 mg/l in terms of calcium carbonate are desirable for drinking water, and its maximum allowed is 200 mg/l. Calcium hardness does not harm human health. In fact, hard water seems to be beneficial for the circulatory system [30]. The amount of magnesium

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in the city drinking water ranged from 60 mg/l to 562 mg/l, which is higher than the maximum allowed. Magnesium concentrations less than 50 mg/l are desirable for drinking water, and the maximum permitted is 150 mg/l. However, in many sources of drinking water supply, the amount of magnesium exceeds the standard values. Magnesium hardness, especially with sulfate ion, can cause diarrhea in people who are not used to it. In a study conducted in Ghana to assess the quality of drinking water from underground sources, the mean value of magnesium hardness was 122.79, and the mean value of calcium hardness was 55.28 mg/l of calcium carbonate [31]. The total hardness measured in two cold and warm seasons revealed that climate and atmospheric changes cannot affect the total hardness. It is possible to determine the mineral (inorganic) pollution of water relatively by measuring the value of E.C., which has a direct relationship with the amount of dissolved salts in water. E.C. values measured during the sampling period were 331 to 1865 millisiemens. It is believed that the limestone layers of the earth have increased these parameters in water resources of this city. The sodium adsorption ratio is a metric to evaluate the risk of sodium in irrigation water, which is determined by calculating the ratio of sodium to magnesium and calcium. Appropriate amounts of calcium and magnesium in irrigation make the texture of the irrigated soil granular and permeable and facilitate its efficiency; thus, the permeability of the soil decreases with an increase in the sodium ratio. This parameter is mostly used for agricultural purposes; for health purposes, electrical conductivity or total dissolved solids are mostly used. In fact, waters with SAR values of less than 3 have no restrictions for agricultural use.

In the wet season, most of the stations had a relatively good rank (descriptive equivalent). Besides, two sampling stations had a very good rank (descriptive equivalent). In the dry season, most of the stations had relatively good and relatively bad conditions based on the numerical index. The average IRWQI index was 52.45 in the wet season and 50.69 in the dry season. Water quality in the dry season decreased compared to the wet season (P-value = 0.006). According to the obtained mean values and comparison with the ranking table of the IRWQI, the state of water resources in the region was moderate. Therefore, if fundamental measures are not taken to prevent the entry of the pollutant into the underground water resources of the region, the quality index will decrease and be classified as poor. Edris Hoseinzadeh et al. investigated the water quality of Aydughmush Dam (Iran) by calculating the NSFWQI index. They concluded that water quality of this dam is within the good range and is suitable for various uses [32].

Conclusion

The results revealed that the residues of the studied toxins in all the samples were much lower than the maximum allowed by national and international guidelines (WHO and EPA). Although the mean concentration of pesticides in drinking water sources was lower than the standard, in some sources, especially in the western part of the region, the residual concentration of pesticides exceeded the standard limit, which can cause restrictions in terms of drinking uses in the future. As a result, it is necessary to take fundamental measures to prevent these pollutants from entering water resources.

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Conflict of interest

The authors declare that they have no conflict of interest.

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Code of ethics

The ethical code of the present study is IR.mui.rec.1395.3.394, which was carried out with

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Authors' contributions

Morad Mahmoudi baram: Investigation, Methodology; Karim Ebrahimpoor: Conceptualization, Supervision, Writing, Reviewing, Editing; Afshin Ebrahimpour: Methodology, Sofware, Formal analysis. All authors reviewed and approved the final manuscript.

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