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Evaluation of Radioactivity Levels and Lifetime Risk Associated with Drinking Water of Ardabil Province

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ABSTRACT

The ingestion and intake of healthful and high-quality water are crucial aspects of human existence. Therefore, it is imperative to closely monitor the contamination of drinking water, including potential exposure to radioactive elements, in order to assess the level of radiation that individuals may be subjected to through water consumption. The purpose of this paper is to measure the levels of gross beta and alpha activities in water samples obtained from various sources in Ardabil. Using liquid scintillation methodology, the gross beta and alpha activity concentrations of 25 water samples were scrutinized and measured to determine their radiological health. The findings of this analysis revealed that the gross alpha activity concentration ranged from 30 $mBqL^{-1}$ to 180 $mBqL^{-1}$, with an average of 71.6 $mBqL^{-1}$. Similarly, the gross beta activity concentration in the samples varied from 49 $mBqL^{-1}$ to 210 $mBqL^{-1}$, with an average of 114.4 $mBqL^{-1}$. Furthermore, there was a moderate positive correlation of 0.49 between alpha concentration and beta concentration. The annual effective dose ranged from 0.036 to 0.104 mSv, with an average of 0.072 mSv. The variation in risk ranges from 2.02E-04 to 7.91E-04, with an average of 4.01E-04. These outcomes demonstrate that the water samples analyzed are free from any radiological hazards and are deemed safe for consumption.

Keywords: Lifetime risk; Annual effective dose; Gross beta; Gross alpha.

1. Introductions

The consumption of water is of utmost significance in sustaining human life. Ensuring access to safe drinking water is a crucial aspect that relates to health and progress at national, regional, and local levels. It is considered both a fundamental human right and a vital component of a comprehensive health protection policy [1]. Consequently, the

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surveillance and assessment of any discharge of radioelements into drinking water carry substantial importance from a biological standpoint, aiming to protect human and environmental well-being [2].

Radioactivity found in drinking water primarily comes from radionuclides in the natural decay series of ^{238}U and ^{232}Th, as well as potassium (K) elements. Certain radionuclides within these categories can quickly dissolve in water. The composition of the resulting solution depends the geological and mineralogical components of the rock, the duration of groundwater residence in the bedrock and the ambient temperature of the area radioactive substances can also be present in drinking water from sources like mining and nuclear activities. When radioelements enter the human food chain and are ingested or inhaled, they cause internal exposure within the human body [3,4].

When assessing radioactivity, measuring gross alpha (α) and beta (β) activity concentrations in drinking water is an efficient initial screening parameter. Analyzing individual radioelements is time-consuming and costly, making it necessary to determine the gross α and β activity concentrations in drinking water sources, regardless of specific radionuclides present. According to guidelines from the World Health Organization (WHO), screening levels for gross α and β activity concentrations in drinking water are 500 and 1000 $mBqL^{-1}$, respectively [5].

If results are positive, concentrations of radionuclides like radium-226, radon-222, and uranium must be accurately measured and an operational plan must be implemented. Consequently, numerous investigations have been conducted worldwide to assess compliance with these limit values in drinking water across various countries.

So far, numerous studies have been conducted on the levels of radioactivity in various water samples collected from different countries. For example, research has been done on the natural radioactivity in drinking water in Albania [6], the gross alpha and beta radioactivity in the waters of the Gilan province [7], the natural radioactivity in drinking water in Hatay [8], the total alpha and beta radioactivity in Tehran city water [9], the radiation levels in groundwater of drinking water Vietnam [10], the radioactive substances in thermal springs water in Turkey [11], the total alpha and beta radioactivity in tap water from Jordan [12], the gross alpha and beta radioactivity in groundwater and surface water in Kaduna, Nigeria [13] and the total beta and alpha concentration in drinking water from Calabria, Italy [14] and India [15].

Ardabil is a city in a mountainous region of northwestern Iran (Fig. 1), located at approximately 38° 17' N latitude and 48° 18' E longitude. The province of Ardebil relies heavily on groundwater for its drinking water supply. This region is of significant importance compared to other provinces in Iran due to higher natural radiation levels such as Ramsar, Talash, Mahallat, and Qazvin, etc. Prolonged exposure to elevated levels of radioactivity in drinking water can increase the risk of health issues, including cancer. Therefore, it is crucial to determine the concentration of radioactivity in the drinking water of Ardebil and assess the annual effective dose rate and lifetime cancer risk for the population in this area.

Despite this importance, there is a lack of systematic information in the literature regarding the level of radioactivity in drinking water sources in Ardabil province. Thus, the main objective of this study is to determine the gross alpha (α) and beta (β) activity concentrations in the drinking water and estimate the resulting effective rate. Additionally, the assessment aims to calculate the excess lifetime cancer risk for the population of Ardabil province.

2. Material and methods

Ardabil Province is located in northwestern Iran, covering an area of approximately 17,953 square kilometers. It has an estimated population of 1.2 million people according to recent figures. The province is administratively divided into 11 counties with the city of Ardabil serving as the capital. Some of the main industries include food processing, textiles, and leather goods production which provide an important source of jobs. Agriculture and animal husbandry also form a significant part of the local economy, with crops such as wheat, barley, potatoes, tomatoes, and various vegetables commonly grown in the region. In addition to farming, there are mineral reserves of iron ore, mica, lead, and zinc present. The mountainous terrain and western climatic influences result in cool, wet winters and mild, dry summers.

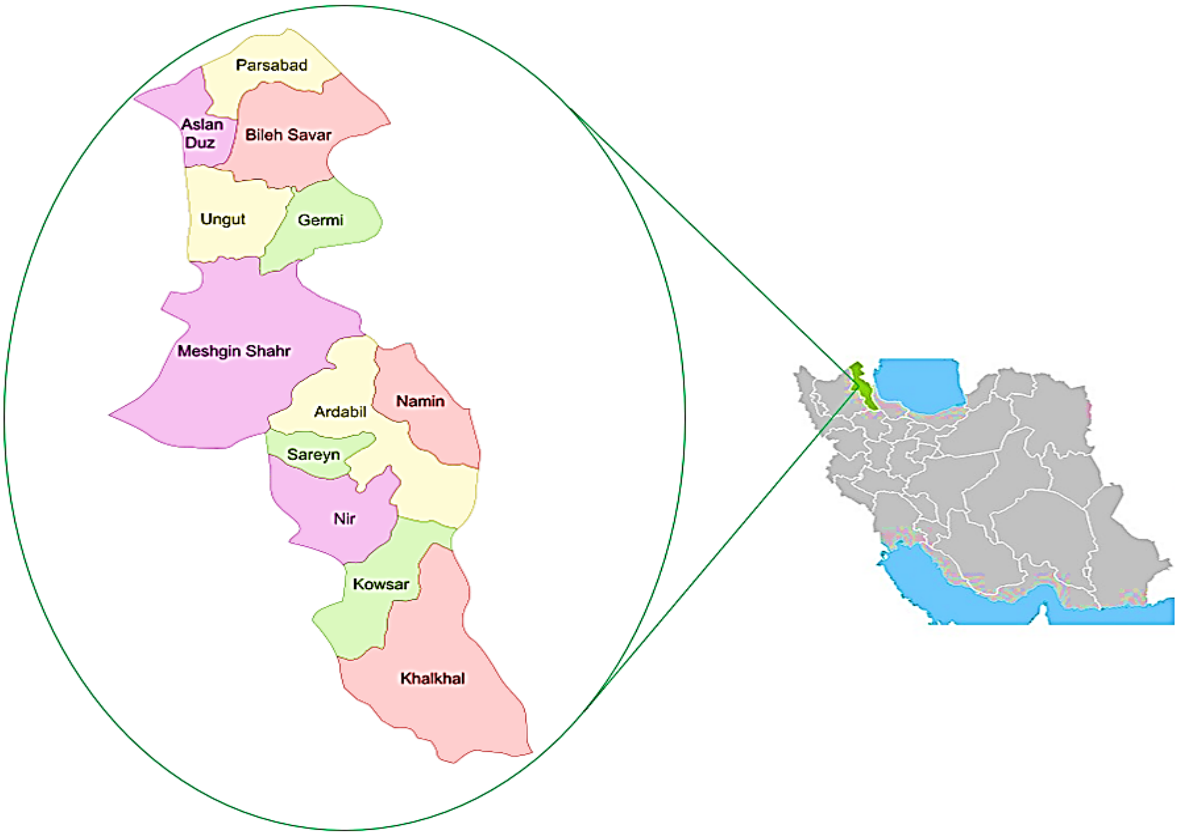

Fig. 1. Geographical context for Ardabil province, Iran: The province lies at coordinates 38°26′N 48°06′E within the country's borders.

2.1. Minimum detectable activity

The Minimum Detectable Activity (MDA) refers to the lowest level of radioactivity that can be detected when analyzing a sample using a detection system. The MDA is influenced by several factors, including counting time, sample size, counting efficiency, and background radiation. Increasing the counting time or sample size can lead to an increase in MDA. In the case of the Liquid Scintillation Counting (LSC) detection system, the following equation can be employed to calculate the MDA:

$$MDA = L_d / (\varepsilon \times V \times T \times 60) \quad (1)$$

Here, V represents the sample volume, T denotes the duration of the measurements in minutes, and ε represents the counting efficiency. The parameter L_d is defined as:

$$L_d = 2.71 + 4.65\sqrt{(B \times T)} \quad (2)$$

In Eq. (2), B corresponds to the background radiation measured in counts per minute.

2.2 Methods for sample preparation

According to ISO 11704 standard [16], 250 milliliters of each prepared sample were transferred to a glass container. To prevent sample deposition on the container walls, the accumulation of organic materials, and variations in ion states, a dilute solution of nitric acid (HNO_3) was used to acidify the samples to a pH of 2.5. The glass container was then placed on a hot plate aid in dying. The heating temperature was carefully controlled to stay below 80 °C to avoid the evaporation of alpha and beta emitters, which could result in inaccurate readings.

During the heating process, each sample was stirred using a magnetic stirring capsule to ensure homogeneity and efficient heat transfer. The resulting compound residue was then dissolved by adding 0.1 M HNO_3, producing a solution. To obtain an appropriate sample volume, the samples were diluted using double-distilled water. Following dilution, the samples were transferred to 20 mL vials. To create the final sample, 10 mL of HighSafe3 scintillation cocktail (from PerkinElmer Inc) was added to each vial, bringing the total volume to 20 mL. Subsequently, the prepared samples were moved to a Liquid Scintillation Counting (LSC) system for counting and analysis.

2.3 Methods for efficiency calibration

To determine the calibration efficiency for gross beta and alpha counts, a standard solution containing of pure beta and alpha emitting materials with known activities was used. Each material was measured separately. The efficiency calibration was established under optimal conditions by utilizing the appropriate standard solution for the gross alpha/beta detector efficiency:

$$\varepsilon = (N_S - B) / (A_{Standard} \times V \times T \times 60) \quad (3)$$

Here, ε represents the gross efficiency, N_S denotes the alpha or beta count, B indicates the background count, $A_{Standard}$ represents the pre-identified activity of the standards, and V corresponds to the volume of the standard solution.

Finally, the radioactivity concentrations of gross beta or alpha in a given quantity were determined using Eq. (4) [17]:

$$A = N_{net} / (\varepsilon \times V \times 60) \quad (4)$$

In this equation, N_{net} denotes the net count, A represents the actual counting rate, ε denotes the detector efficiency, and V signifies the sample volume in liters. The conversion from decay per minute (dpm) to decay per second (dps) was achieved by employing a coefficient of 60 in the aforementioned equation.

The regions of interest (ROIs) for alpha and beta counting in the alpha and beta MCAs should be set to optimize the figure of merit (E^2/B) while ensuring that all radionuclides of concern are included in each respective region. For an unquenched sample, a ROI ranging from 400 keV to 700 keV would capture most alpha emitting radionuclides of concern. Depending on the quenching properties of the samples, the ROI should be adjusted to include the radionuclides of concern, while minimizing the background count rate. For the mixture used in this testing method, an ROI from 50 keV to 400 keV generally accounts for the alpha-emitting radionuclides of interest. For beta-emitting radionuclides, an ROI from 2 to 2000 keV is typically used. A low energy threshold of 2 keV is generally enough to eliminate luminescence and low energy interference.

2.4 Evaluation of effective radiation dose

The calculation of effective dose for adults resulting from the ingestion of both beta and alpha emitting materials present in drinking water was performed using equation [18,19]:

$$\text{effective dose (mSv/y)} = A \times M \times CF \quad (5)$$

In this equation, A represents the gross beta or alpha activity concentration, M denotes the annual water intake per individual (L/y), and CF stands for the conversion factor of ingestion dose for gross alpha or beta (mSv/Bq). The main α-emitting radionuclides to determine CF values were assumed to be U-238, U-234, Th-230, Ra-226, Po-210 and Th-232. Also, the main β-emitting radionuclides to determine CF values were assumed to be Pb-210 and Ra-228. The average values of CF for main α- and β-emitting radionuclides are equal to 3.4×10^{-4} mSv Bq^{-1} and 4.6×10^{-4} mSv Bq^{-1} respectively.

It is important to note that annual water consumption can vary depending on various factors, such as outdoor temperature. For the purposes of this study, the adopted value for annual water consumption by adults was 730 liters [19].

3. Results and discussion

One of the crucial elements in measuring low levels of radioactivity is determining the minimum amount that can be reliably detected. In other words, the success of a method is often measured by its ability to accurately detect low levels of radioactivity. The minimum detectable activity (MDA) values were computed using Eq. 1 for gross alpha and beta measurements. For a duration of 240 minutes, the MDA values for blank sample measurements were 0.024 Bq L^{-1} and 0.034 Bq L^{-1} for alpha and beta, respectively. The results of the gross beta and alpha activity concentrations in water samples are presented in Table 1. The measurements for gross beta activity ranged from 30 to 180 mBq L^{-1}, with an average of 71.6 mBq L^{-1}, while for gross alpha activity, the measurements ranged from 49 to 210 mBq L^{-1}, with an average of 114.4 mBq L^{-1}.

Table 1 shows that the gross beta activities are higher than the gross alpha activities. Moreover, all gross alpha concentration values in Table 1 are below the recommended upper limit of 0.5 BqL^{-1}. Similarly, all gross beta concentration values are below the suggested upper limit of 1 BqL^{-1}. Table 2 provides basic descriptive statistics, for both gross beta and alpha radioactivity concentrations in the 25 water samples, including standard deviation, standard error, mean, maximum, minimum, kurtosis, and skewness, for both gross beta and alpha radioactivity concentrations in the 25 water samples (Table 2).

Table 1. The gross beta and alpha radioactivity concentration of water sources (n=3).

Code Number	pH	Alpha Concentration (BqL^{-1})	Beta Concentration (BqL^{-1})
A-P-DW-1	7.1	0.18	0.21
A-P-DW-2	7.6	0.07	0.16
A-P-DW-3	7.3	0.05	0.15
A-P-DW-4	7.5	0.05	0.16
A-P-DW-5	7.2	0.09	0.05
A-P-DW-6	7.4	0.05	0.13
A-P-DW-7	7.7	0.06	0.07
A-P-DW-8	7.2	0.03	0.12
A-P-DW-9	7.1	0.07	0.05
A-P-DW-10	7.3	0.10	0.14
A-P-DW-11	7.5	0.09	0.13
A-P-DW-12	7.6	0.06	0.06
A-P-DW-13	7.4	0.15	0.13
A-P-DW-14	7.2	0.06	0.11
A-P-DW-15	7.1	0.08	0.15
A-P-DW-16	7.7	0.10	0.06
A-P-DW-17	7.3	0.05	0.06
A-P-DW-18	7.5	0.14	0.15
A-P-DW-19	7.6	0.03	0.11
A-P-DW-20	7.4	0.07	0.16
A-P-DW-21	7.2	0.06	0.18
A-P-DW-22	7.1	0.03	0.06
A-P-DW-23	7.3	0.04	0.08
A-P-DW-24	7.5	0.03	0.07
A-P-DW-25	7.6	0.05	0.11

Table 2. Statistical analysis of gross beta and alpha radioactivity measurements from 25 water sample.

	Alpha Concentration (Bq/L)	Beta Concentration (Bq/L)
Mean	0.07	0.11
Standard Error	0.01	0.01
Standard Deviation	0.04	0.05
Kurtosis	1.74	-0.98
Skewness	1.39	0.1
Minimum	0.03	0.05
Maximum	0.18	0.21

The gross alpha distribution is positively skewed (with a skewness of 1.39) and more peaked than normal (with a higher kurtosis of 1.74). In contrast, the gross beta distribution has very little skewness (close to zero at 0.10) and is flatter than normal (with a negative kurtosis of -0.98). These numerical measures of shape (skewness and kurtosis) provide insight into two separate probability distributions-alpha and beta. The gross alpha distribution exhibits more pronounced positive skewness and peaked-ness, while the gross beta distribution is closer to symmetrical with a flatter peak.

Fig. 2 illustrates the correlation between gross beta and alpha activity concentrations in the water samples. The correlation coefficient is a metric used to assess the relationship or dependence between two variables indicating how much changes in one variable align with changes in the other. With a correlation of 0.49, the measured levels of gross beta radiation moving up or down; end to correspond with similar movements in alpha radiation levels, showcasing a moderate linear association between the two. While not a perfect correlation, there seems to be some predictive capability between these variables.

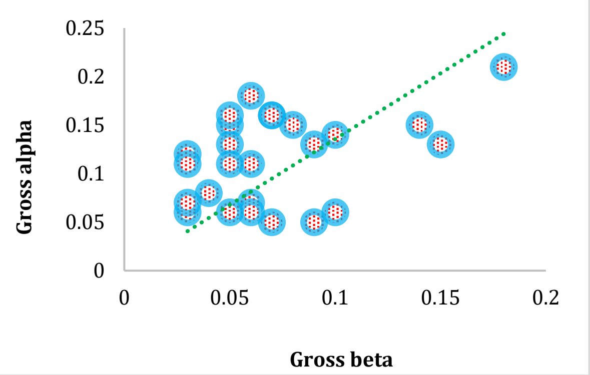

Fig. 2. The moderate correlation between the levels of gross beta and alpha radioactivity.

Table 3 presents the gross beta and alpha radioactivity concentrations measured in water samples from various countries. The reported values for gross alpha vary greatly, ranging from 1 in Serbia to 1700 in Catalonia. Similarly, the reported gross beta radioactivity concentrations range from 17 in Turkey to 2900 in Catalonia.

To evaluate the potential health risks for adults in society, we calculate the annual effective dose from radiation exposure through the consumption of water samples. The effective dose, measured in mSv, was determined by using the gross beta and alpha activity concentrations, dose coefficient, and annual water intake as outlined with Eq. 5. Table 4 presents the calculated effective dose values for the analyzed water samples. The annual effective dose varied from 0.036 to 0.104 mSv, with an average of 0.072 mSv.

Table 3. Comparison of the gross beta and alpha radioactivity concentrations results of this work and other study.

Origin	Gross alpha activity concentration (mBq L^{-1})	Gross beta activity concentration (mBq L^{-1})	References
Albani	39	220	Cfarku et al., 2014 [6]
Turkey	8 – 101	17 – 177	Turhan et al., 2019 [20]
Serbia	1 – 13	44 – 173	Janković et al., 2012 [21]
Mexico	< 11 – 601	211	Rangel et al., 2001 [22]
Catalonia	20 – 1700	40 – 2900	Ortega et al., 1996 [23]
Ardabil	30 – 180	49 – 210	This work

Table 4. Estimates of Annual effective dose and lifetime risk based on ingestion of gross beta and alpha radioactivity levels detected in water sample.

Code Number	Annual effective dose-Alpha (mSv)	Annual effective dose-Beta (mSv)	Total calculated annual effective dose (mSv)	Lifetime risk
A-P-DW-1	3.68E-02	1.06E-01	1.43E-01	7.91E-04
A-P-DW-2	1.43E-02	8.06E-02	9.49E-02	5.27E-04
A-P-DW-3	1.02E-02	7.56E-02	8.58E-02	4.76E-04
A-P-DW-4	1.02E-02	8.06E-02	9.08E-02	5.04E-04
A-P-DW-5	1.84E-02	2.52E-02	4.36E-02	2.42E-04
A-P-DW-6	1.02E-02	6.55E-02	7.57E-02	4.20E-04
A-P-DW-7	1.23E-02	3.53E-02	4.75E-02	2.64E-04
A-P-DW-8	6.13E-03	6.04E-02	6.66E-02	3.69E-04
A-P-DW-9	1.43E-02	2.52E-02	3.95E-02	2.19E-04
A-P-DW-10	2.04E-02	7.05E-02	9.10E-02	5.05E-04
A-P-DW-11	1.84E-02	6.55E-02	8.39E-02	4.65E-04
A-P-DW-12	1.23E-02	3.02E-02	4.25E-02	2.36E-04
A-P-DW-13	3.07E-02	6.55E-02	9.61E-02	5.33E-04
A-P-DW-14	1.23E-02	5.54E-02	6.77E-02	3.75E-04
A-P-DW-15	1.64E-02	7.56E-02	9.19E-02	5.10E-04
A-P-DW-16	2.04E-02	3.02E-02	5.07E-02	2.81E-04
A-P-DW-17	1.02E-02	3.02E-02	4.04E-02	2.24E-04
A-P-DW-18	2.86E-02	7.56E-02	1.04E-01	5.78E-04
A-P-DW-19	6.13E-03	5.54E-02	6.15E-02	3.41E-04
A-P-DW-20	1.43E-02	8.06E-02	9.49E-02	5.27E-04
A-P-DW-21	1.23E-02	9.07E-02	1.03E-01	5.71E-04
A-P-DW-22	6.13E-03	3.02E-02	3.64E-02	2.02E-04
A-P-DW-23	8.18E-03	4.03E-02	4.85E-02	2.69E-04
A-P-DW-24	6.13E-03	3.53E-02	4.14E-02	2.30E-04
A-P-DW-25	1.02E-02	5.54E-02	6.56E-02	3.64E-04

Fig. 3 shows the total effective dose versus alpha concentration and beta concentration.

A Pearson correlation analysis was conducted to examine the relationships between the variables of total effective dose, alpha concentration, and beta concentration using the provided data (Fig. 3).

The analysis uncovered a very strong positive correlation between total annual effective dose and alpha concentration, with a Pearson's r value of 0.8854. This correlation was deemed highly statistically significant, supported by an extremely small p-value of 2.919E-08, well below the standard $\alpha = 0.05$ threshold. Similarly, a strong positive correlation was also observed between total annual effective dose and beta concentration ($r = 0.8731$, $p = 7.426E-08$).

A moderate positive correlation was observed between alpha concentration and beta concentration ($r = 0.4944$, $p = 0.0185$), though the association was not as strong as those with a total annual effective dose.

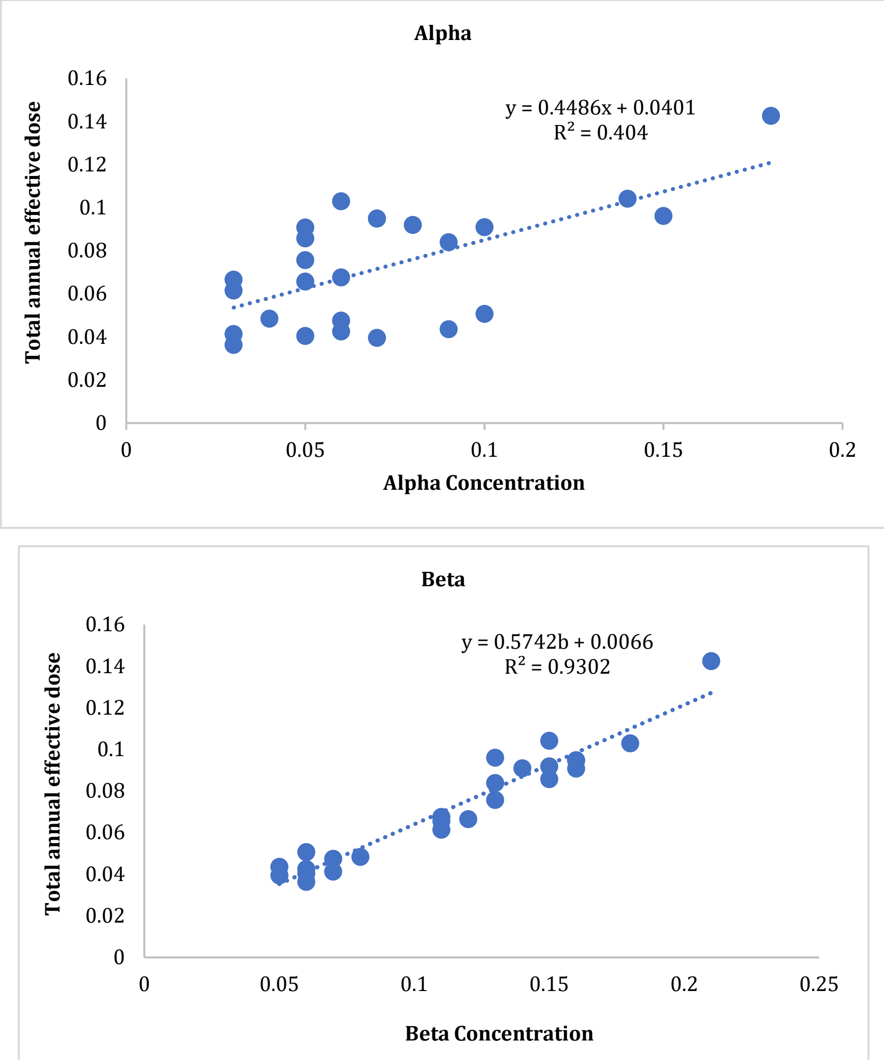

Fig. 3. The correlation between total annual effective dose and gross alpha and beta radioactivity levels.

Overall, the results suggest that higher levels of alpha and beta radiation exposure, as indicated by their respective concentrations, are highly predictive of greater total effective radiation doses accumulated. The nearly perfect correlation between total dose and each radionuclide concentration implies they are closely related. While alpha and beta concentrations were also somewhat correlated, their relationship was not as definitive based on these data. Overall, the analyses provide compelling statistical evidence for interdependencies between the radiation measures.

To meet national safety standards, it is crucial to assess all potential indicators related to water consumption, including the risk of radiation-induced health effects. The equation for lifetime risk (LifetimeRisk= risk factor* life expectancy*Annual Dose) takes into account the lifetime-mean dose, with a life expectancy of 76 years for Iran, and the risk factor, reported as 7.3×10^{-2} for radiation-induced stochastic health effects [22].

The variation in risk ranges from 2.02E-04 to 7.91E-04, with an average of 4.01E-04, which is slightly higher than the acceptable limit of 10^{-4}. In other words, based on these results, we can expect approximately 4 cases of disease for every 10,000 people exposed.

4. Conclusion

This research is the first comprehensive study on the radioactivity concentration in water of Ardabil and the associated health hazards, based on available evidence. Gross beta and alpha radioactivity measurements were used as the initial step in determining the water's radioactivity, as this technique offers a simple and cost-effective way to evaluate both beta and alpha levels in a relatively short time. The overall findings were significantly lower than the WHO's recommended reference level. Additionally, the average lifetime risk value is close to the appropriate 10^{-4} limit. These results suggest that Ardabil's water is radiologically safe and does not pose a significant public radiation threat. The data collected in this study can serve as the radiometric baseline values for water in this region, which can aid in establishing national guidelines for natural radioactive materials in drinking water (please note that the current conclusion is limited to this year and this research only).

5. Data availability statements

My manuscript has no associated data or the data will not be deposited. The author confirms that all data generated or analyzed during this study are included in this manuscript.

Conflict of interest

The authors declare no potential conflict of interest regarding the publication of this work.

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