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Original Article

Determination of Radionuclide Concentrations in Tea Samples Cultivated in Guilan Province, Iran

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Abstract

Introduction
Foodstuffs are known to contain natural and artificial radionuclides. Determination of radionuclide concentration is of great significance for the protection of human health. The main objective of the present study was the quantification of radionuclides in tea samples, cultivated in Guilan Province in North of Iran.

Materials and Methods
The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in 18 tea samples were measured, using a gamma spectrometry system. In addition, radium equivalent index (Ra_{eq}) and radiation hazard index (HI) were calculated. ANOVA test was used for the statistical analysis of the data

Results
The concentration of ^{137}Cs was below the minimum detectable activity (MDA). The concentrations of ^{226}Ra and ^{232}Th ranged from < MDA to 0.042 and < MDA to 0.037 Bq/kg respectively. The mean concentration of ^{40}K was 410±15 Bq/kg. Based on the findings, the concentration of ^{40}K was significantly higher than other radionuclides ($P<0.01$). Also, the mean Ra_{eq} value was estimated at 31.8±1.2 Bq/kg, and HI in the samples ranged from 0.075 to 0.093.

Conclusion
According to the findings, the activity level of radionuclides in tea samples was found to be within the acceptable range and therefore, non-threatening to public health.

Keywords: Radionuclide, Tea, Gamma Spectrometry

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1. Introduction

Sources of radioactivity in the environment have both natural and artificial origins. The main natural radioactive sources of ionizing radiation include ^{40}K, ^{232}Th, ^{238}U, and their decay series. Artificial radionuclides such as ^{137}Cs may be released into the environment as a result of human activities including energy production, nuclear weapon testing, and nuclear accidents [1-3].

Based on a report by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), exposure to natural radionuclides accounts for nearly 70% of the population radiation exposure. Radiation doses of radionuclides are threatening to human health. In fact, environmental radionuclides may be transferred to the human body through food, water, and air particles, inducing internal radiation doses [4].

Foodstuffs are known to contain natural and artificial radionuclides, which could be accumulated in certain parts of the body such as the lungs, kidney, liver, and skeletal tissues after ingestion. According to previous studies, at least one-eighth of the mean annual effective dose radiated by natural sources is caused by the consumption of foods and beverages [1, 5, 6].

Black tea is one of the popular beverages in the world, prepared from the leaves of a shrub known as *Camellia sinensis*. Recently, black tea has gained particular attention for its health benefits, and studies have revealed its biological activities. The polyphenolic compounds of black tea have anti-inflammatory, anti-oxidant, and anti-cancer properties, as well as metabolic regulatory effects [7, 8].

Environmental radionuclides are concentrated in tea leaves and consumption of black tea as a beverage may contribute to an internal radiation dose [9]. Measurement of the concentrations of radionuclides present in black tea permits the assessment of the dose received by the intake of this beverage. Radiation levels of the samples can be determined by a gamma spectrometer, equipped with a high-purity germanium (HPGe) detector and a multi-channel analyzer [10, 11].

Guilan is one of the Northern provinces of Iran, with thousands of hectares of land cultivated for tea. Although the concentrations of lead, cadmium, and arsenic in black tea samples, cultivated in North of Iran, have been reported in the literature [12], the concentrations of natural and artificial radionuclides remain undetermined.

The main objective of this study was to quantify the presence of natural and artificial radionuclides (i.e., ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in tea samples cultivated in Guilan and to estimate the potential biological hazards by calculating the radium equivalent index (Ra_{eq}) and hazard index (HI) for consumers. The obtained data could be used as the baseline for future reference. For this purpose, 18 samples of black tea were obtained from Guilan tea factories. Then, the radionuclide levels were measured via HPGe gamma spectrometry, and then, the results were analyzed.

2. Materials and Methods

1.1. Study area and sample collection

This study was carried out from May to June 2015 on tea samples, cultivated in Guilan Province in North of Iran. This area is located between latitudes of 36°34′ and 38°27′ N and longitudes of 48°53′ and 50°34′ E (Figure 1). Guilan with a total area of 14,042 km^2 has a humid temperate climate with adequate annual precipitation [13]. In this study, 18 black tea samples were collected from Lahijan, Langroud, Amlash, Roudsar, Shaft, and Fuman cities, which have the major tea gardens in the province (three samples from each city).

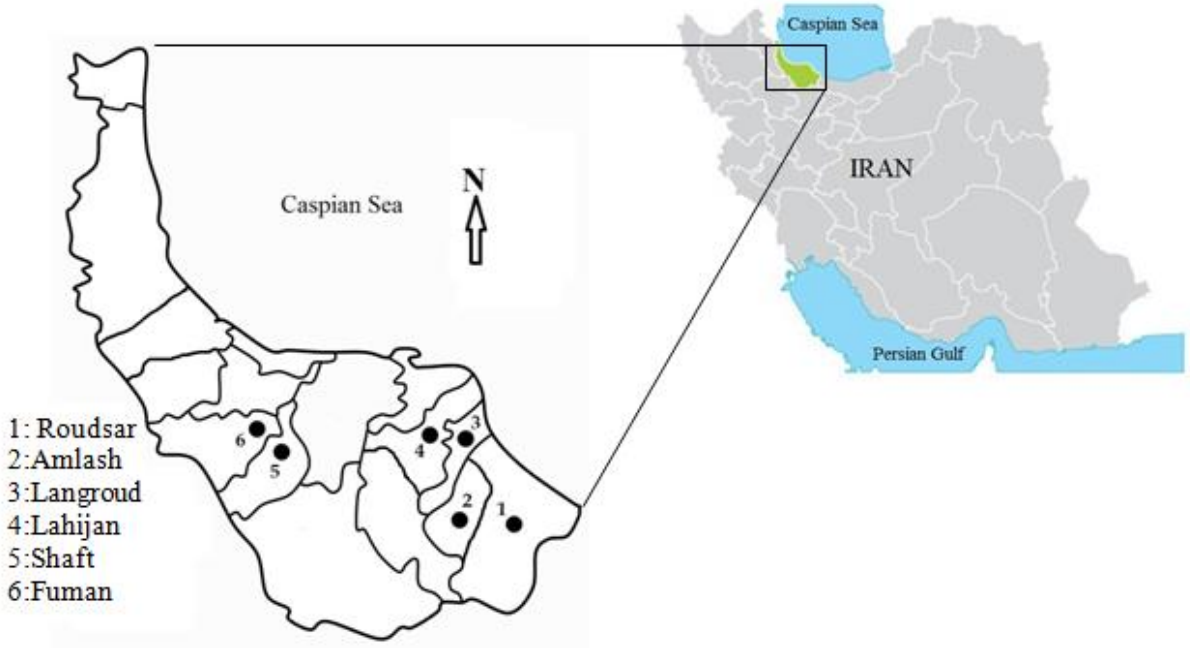

Figure 1. Location of Guilan province and the sampling regions

1.2. Radioactivity measurements

All the tea samples were homogenized, weighed, and transferred into uncontaminated cylindrical plastic containers of uniform size (Marinelli beaker). Each sample (500 g) was packed in a Marinelli beaker and sealed for 30 days at room temperature (22-25°C) before the analysis to allow secular equilibrium between thorium and uranium and their respective decay products [14].

Gamma radiation levels of the samples were measured, using a gamma spectrometry system (Model No. S100; Canberra, Austria). This system was equipped with an HPGe detector with a relative efficiency of 40%. The detector was shielded by 10 cm of lead on all sides and cadmium-copper in the inner sides to reduce background radiation.

The standard mix source (CERCA HM 395, France) was used for calibration and efficiency calculation of the gamma spectrometry system. The counting time for each sample was 72 hours, and the background radiation due to cosmic rays and environmental radionuclides was subtracted from each sample. The selected gamma peaks for the measurement of different radionuclides were 1461 keV for ^{40}K, 661 keV for ^{137}Cs, 609 keV for ^{226}Ra (^{214}Bi), and 583 keV for ^{232}Th (^{208}Tl).

The minimum detectable activity (MDA) was approximately 175, 21.8, 12.1, and 11.5 mBq/kg 0.175, 0.021, 0.012 and 0.011 Bq/kg for ^{40}K, ^{232}Th, ^{226}Ra, and ^{137}Cs, respectively. The activity concentrations in the samples were calculated using equation (1):

$$A_S = C_S / \varepsilon P_\gamma M_S \quad (1)$$

where A_S (Bq/kg) is the activity concentration of the sample, C_S is the sample count rate under the corresponding peak, ε denotes the efficiency of the detector at a specific gamma ray energy, P_γ is the absolute emission probability of the gamma ray, and M_s is the mass of the sample (kg) [11, 15].

Ra_{eq} is an index, used to describe the gamma output from different mixtures of ^{226}Ra, ^{232}Th, and ^{40}K by one quantity. Ra_{eq} for each sample was expressed in Bq/kg and calculated by equation (2):

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (2)$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively [1].

HI was used to estimate the level of gamma radiation hazards, associated with the natural radionuclides [16]. This index was measured using equation (3):

$$HI = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (3)$$

1.3. Statistical analysis

Data are presented as mean ± standard deviation (SD). After verifying the normality of variables, analysis of variance (ANOVA) was performed at a confidence interval of 95%. In all cases, P-value less than 0.05 was considered statistically significant. The results were analyzed, using SPSS version 17 (SPSS/PC Inc., Chicago, USA).

3. Results

The activity level of various radionuclides in 18 tea samples was measured via HPGe gamma spectrometry. To determine the concentrations of radionuclides in the samples, Equation 1 was used. By using this equation, the data were normalized for one kilogram of each sample (expressed as Bq/kg).

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the tea samples, collected from different parts of the studied areas, are presented in Table 1. The activity concentrations ranged between < MDA and 0.042 Bq/kg for ^{226}Ra, < MDA and 0.037 Bq/kg for ^{232}Th, 365 and 450 Bq/kg for ^{40}K, and < MDA for ^{137}Cs (< 0.011 Bq/kg).

The mean activity level of ^{40}K was 410±15 Bq/kg.

The average activity levels of ^{226}Ra, ^{232}Th, and ^{40}K were found to be <0.029, <0.031, and 410 ±15 Bq/kg in the tea samples, respectively. Based on the findings, the activity concentration of ^{40}K was significantly higher than other radionuclides in all the collected samples (P<0.01). Also, based on the ANOVA test results, the difference in the concentrations of ^{226}Ra and ^{232}Th was not significant in the tea samples.

Table 1. Activity concentrations (Bq/kg) of radionuclides in tea samples (mean±SD)

Study area	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Lahijan	<0.023	<MDA	430±18	<MDA*
Langroud	<0.019	<0.032	412±11	<MDA
Amlash	<MDA	<0.027	403±20	<MDA
Roudsar	<0.031	<0.028	391±21	<MDA
Fuman	<0.042	<MDA	426±14	<MDA
Shaft	<MDA	<0.037	421±13	<MDA
Activity rang	<MDA to 0.042	<MDA to 0.037	365 to 450	<MDA
Average	<0.029	<0.031	410±15	<MDA

*MDA: minimum detectable activity

Table 2. The radium equivalent (Raeq) (Bq/kg) and hazard index (HI) of tea samples

Study area	Raeq (mean±SD)	HI
Lahijan	33.13±1.38	0.089
Langroud	31.78±0.85	0.085
Amlash	31.06±1.54	0.083
Roudsar	30.17±1.61	0.081
Fuman	32.84±1.08	0.088
Shaft	32.46±1.03	0.085

Table 2 presents the calculated values of Ra_{eq} and HI for the tea samples. The results showed that Ra_{eq} ranged between 28.12 and 34.65 Bq/kg, and the average Ra_{eq} value was reported to be 31.8±1.2 Bq/kg. Moreover, the HI values varied from 0.075 to 0.093 in the samples.

4. Discussion

Humans are exposed to both external and internal radiations from radionuclide sources [17, 18]. Internal exposure occurs through the uptake of radionuclides via inhalation or ingestion of contaminated foods and beverages. Ingestion exposure dose mostly results from ^{40}K, ^{137}Cs, ^{226}Ra, and ^{232}Th series [19, 20].

Chemical and physical processes following radionuclide radiation involve changes at molecular and cellular levels, which may lead to a wide range of health effects. In case damages occur in DNA molecules, hereditary disorders and cell death may ensue [21, 22]. Therefore, determination of radionuclide concentration in foods and beverages is of great significance for the protection of human health.

Considering the popularity of tea, following water in Iran, level of radionuclides in tea samples was measured in the present study. The results showed that the activity concentration in tea samples was <MDA to 0.042 0.029 Bq/kg for ^{226}Ra, <MDA to 0.037 0.031 Bq/kg for ^{232}Th, 410±15 365 to 450Bq/kg for ^{40}K and < MDA for ^{137}Cs. The values of Ra_{eq} and HI ranged from 28.12 to 34.65 Bq/kg and from 0.075 to 0.093, respectively.

The present findings were compared with the results reported in previous studies. In this regard, Harb et al. studied the concentration of radionuclides in tea samples obtained from Egyptian markets, using a gamma spectrometry system [23]. The average activity level of ^{232}Th, ^{226}Ra, ^{137}Cs, and ^{40}K were reported as 3.0±0.6, 3.1±0.8, 0.9±0.2, and 623±25 Bq/kg, respectively. Moreover, Korkmaz Gorur et al. measured the level of radionuclides in black tea samples from Turkish markets, using a gamma-ray spectrometer, equipped with an HPGe detector [15]. The activity concentrations of ^{232}Th, ^{226}Ra, ^{40}K, and ^{137}Cs were reported to be 3.2±0.6, 6.4±0.7, 445.6±17.8, and 42.0±1.4 Bq/kg, respectively.

In a previous study by Kilic et al. on tea samples from local Turkish markets [11], the mean activity levels of ^{232}Th, ^{238}U, ^{40}K, and ^{137}Cs were reported to be 2.7±1.0, 0.9±0.4, 501±42, and 45.04±2.6 Bq/kg, respectively. Also, the findings revealed that the average annual effective dose of ^{137}Cs as a result of tea consumption was not a public health hazard.

In consistence with previous studies, ^{40}K concentration in the present research was found to be significantly higher than other radionuclides. This radionuclide is a highly soluble element with a high transfer factor from soil to plant [6, 14]. The activity concentration of ^{137}Cs in the current study was significantly lower than the tea samples studied in Turkish markets. However, it should be noted that the Eastern Black Sea Region of Turkey was contaminated by the radioactive fallout from the Chernobyl accident, and therefore, ^{137}Cs entered the ecosystems [9, 15].

In the presents study, the concentrations of ^{232}Th and ^{226}Ra in all the selected tea samples were fairly lower than the values reported in the literature. Overall, if HI value is found to be less than one, there is no potential radiation hazard [16]. As presented in Table 2, the current findings indicated that the tea samples were free from radiation hazards. Also, the calculated values of Ra_{eq} were within the safety limits, recommended by the UNSCEAR 2000 Report [4].

5. Conclusion

In this study, the concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs radionuclides in tea samples cultivated in Guilan province were determined, using gamma spectrometry. The level of radionuclides was lower than the standard limits, established by international organizations. It was concluded that the

activity level of tea samples in the present study was non-threatening to public health.

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