

Determination of radioactivity levels of soil samples and the excess of lifetime cancer risk in Rize province, Turkey

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ABSTRACT

Background: Turkey, especially the northern part of it, was one of the countries which were contaminated by the Chernobyl accident. Rize is a city located in the Northeastern district of Turkey which was heavily influenced by the Chernobyl nuclear accident. **Materials and Methods:** In this study, the activity concentrations of natural (²²⁶Ra, ²³²Th, ⁴⁰K) and artificial (¹³⁷Cs) were measurements in soil samples collected from 132 different points in Rize province of Turkey using gamma spectrometry with a high-purity germanium detector. **Results:** The average activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were found to be 85.75±11.77, 51.08±9.42, and 771.57±37.65 Bq/kg in soil samples, respectively. In order to evaluate the radiological hazard of the natural radioactivity, radium equivalent activity (Ra_{eq}), representative level index (I_{vr}), the external hazard index (H_{ex}), the total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess life time cancer risk (ELCR) have been calculated and compared with the internationally approved values. **Conclusion:** The outdoor air absorbed dose rates (D) due to terrestrial gamma rays for soil have been calculated because of agricultural area and living in the surrounding. It is important to determine background radiation level in order to evaluate the health hazards. Annual effective gamma doses and the lifetime risks of cancer were higher than the world's average. Moreover compared to the World's average, the lifetime risk of cancer doubled for most of the localities.

Keywords: Radioactivity, soil, lifetime cancer risk, gamma dose.

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INTRODUCTION

Human beings are exposed to background radiation that stems both from natural and man-made sources. Natural background radiation, which is equivalent to 2.4 mSv per person, makes up approximately 80% of the total radiation dose a person is exposed in a year ⁽¹⁾. Soil radionuclide activity concentration is one of the main determinants of the natural background radiation. Volcanic geographic structures as well as rocks that are rich in phosphate, granite and salt contain natural radionuclides like ²³⁸U, ²³²Th and ⁴⁰K. When rocks are disintegrated through natural

processes, radionuclides are carried to the soil by rain and flows ⁽²⁾.

The natural radioactivity in soil comes from ²²⁶Ra, ²³²Th and from natural ⁴⁰K. Some other terrestrial radionuclides, including those of the ²³⁵U series, ⁸⁷Rb, ¹³⁸La, ¹⁴⁷Sm and ¹⁷⁶Lu exist in nature but at such low levels that their contributions to the dose in the humans are small. Artificial radionuclides can also be present such as ¹³⁷Cs, resulting from fallout from weapons testing. ¹³⁷Cs is a fission product which is formed through nuclear tests and accidents. The deposition of ¹³⁷Cs in soil is important since its half-life is 30.2 years and it has a gamma emission of 661 keV ⁽³⁾. The radiological

implication of these radionuclides is due to the gamma-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters. Therefore, the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population (4). Natural environmental radioactivity and the associated external exposure due to gamma radiation depends primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world (5).

Turkey, especially the northern part of it, was one of the countries which were contaminated by the Chernobyl accident (6). Rize is a city located in the Northeastern district of Turkey which was heavily influenced by the Chernobyl nuclear accident. The radioactive plume from the accident reached Turkey by 5 May 1986, substantially contaminating various regions and ecosystems of the country. During the emergency, Cekmece Nuclear Research and Training Center (CANEM) performed an analysis of various substances. In their report, it has been noted that the surface soil ^{137}Cs activity concentration of the eastern part of the Black Sea mountains was around 4000–4500 Bq/kg at the 0.5cm soil in the year 1988 (7).

It is critical to evaluate soil radioactivity in order to understand background radiation

concentrations. Measuring terrestrial gamma dose rates is also essential since gamma radiation provides information concerning excess lifetime cancer risks. Yet in Turkey, there are only a limited number of studies which evaluate soil radioactivity and terrestrial gamma dose rates (8-25).

The aim of this study is to determine natural (^{226}Ra , ^{232}Th , ^{40}K) and artificial (^{137}Cs) radioactivity levels in soil collected from different points in Rize province of Turkey. Also, the average radium equivalent activity (R_{eq}), representative level index (I_{yr}), the external hazard index (H_{ex}), the total absorbed dose rate (D), the annual effective dose equivalent (AEDE) and excess life time cancer risk (ELCR) which will be defined later have been calculated and compared with the results in literature. The results of this study will provide background data on the natural and artificial radioactive isotopes and environmental pollution.

MATERIALS AND METHODS

Study area

Rize is a province of north-east Turkey, on the eastern Black Sea coast (figure 1). Rize stands between the latitudes of $40^{\circ}\text{-}20'$ and $41^{\circ}\text{-}20'$ N and the longitudes of $40^{\circ}\text{-}22'$ and $41^{\circ}\text{-}28'$ E. Rize has a catchment area of 3920 km². It is on the

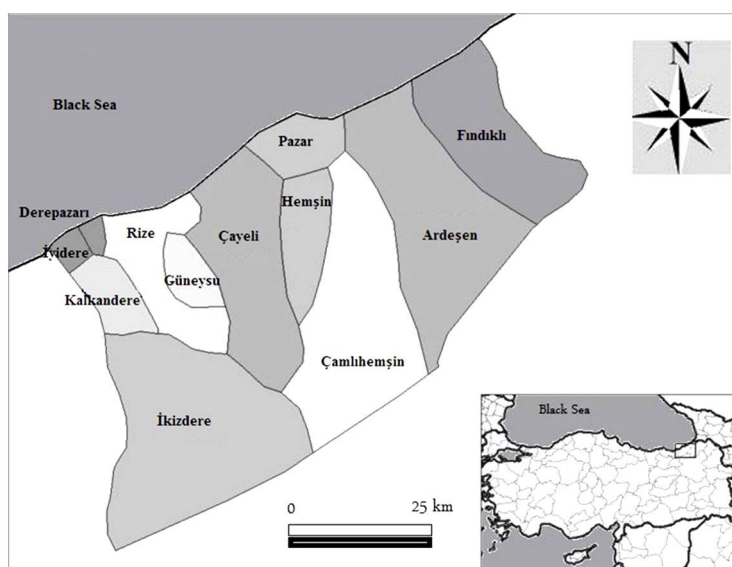


Figure 1. Location of sampling sites indicating the Rize Province, Turkey.

north side of the range of mountains that run along the Black Sea coast. Overlooking the sea this is the wettest corner of Turkey and Rize is the country's largest producer of tea. Summers are cool (July average 22°C), winters are warm (January average 7°C) and it is wet all year round. It has twelve district areas. The population of Rize is 361353 ⁽²⁶⁾.

Sampling and sample preparation

Soil samples were collected from 12 locations of the study area during the year 2010. In all a total of 132 samples were analyzed. After clearing the ground of stones, pebbles, vegetation and roots, 1–2 kg of material from the first 10 cm of topsoil was placed in labeled polythene bags and then transferred to the laboratory. The samples were dried at 60 °C for 48 h, grained, passed through 2 mm sieves. The dried samples then were homogenized and weighed and transferred into uncontaminated empty cylindrical plastic containers of uniform size. The samples were weighed and stored for a minimum period of one month to allow daughter products to come into radioactive secular equilibrium with their parents ²²⁶Ra and ²³²Th and then were counted for 50.000–100.000 s depending on the concentration of the radionuclides.

Experimental method for γ - spectroscopy

The radiation levels of samples were analyzed using gamma spectrometry, which was equipped with a 55% efficiency high purity germanium (HPGe) detector and a multi-channel analyzer. The gamma spectra were analyzed by using the ORTEC Maestro 32 data acquisition and analysis system. The detector had coaxial closed-facing geometry with the following specifications: resolution full width half maximum (FWHM) at 122 keV ⁵⁷Co was 1.00 keV and at 1.33MeV ⁶⁰Co was 1.90 keV. The detector was shielded by a cylindrical lead shield, which had average thickness of 10 cm in order to achieve a background level as low as possible.

Efficiency of the detector was determined with a ¹⁵²Eu source (Amersham Company, UK) of known activity. ¹⁵²Eu source have been widely

used for calibration and efficiency determination due to their large range of energies (122, 244, 344, 411, 443, 779, 964, 1112 and 1408 keV) with emission probabilities of 3-29 % ^(27, 28). An ideal measuring geometry of cylindrical source (homogeneously distributed activity with constant volume and distance) was placed coaxially with the detector for the efficiency determination and the same procedure applied for the sample measurements.

Soil samples were placed symmetrically on top of the detector and measured for a period of 100.000 s. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks. From the net area of a certain peak, the activity concentrations in the samples were obtained using equation 1:

$$C(\text{Bq/kg}) = \frac{C_n}{\epsilon P_\gamma M_s} \quad (1)$$

where C is the activity concentration of the radionuclide in the sample given in Bq/kg, C_n is the count rate under the corresponding peak, ϵ is the detector efficiency at the specific γ -ray energy, P _{γ} is the absolute transition probability of the specific γ -ray, and M_s is the mass of the sample (kg).

For the measurement of ²²⁶Ra activity concentration, the γ -ray energies of 295.21 and 351.92 keV of ²¹⁴Pb, 609.31 keV of ²¹⁴Bi were used. The activity concentration of ²³²Th was determined at the γ -ray energies 911.07 keV and 969.11 keV of ²²⁸Ac, ⁴⁰K and ¹³⁷Cs were measured directly from the 1460.8 keV and 661.66 keV peak energies, respectively ^(29,30). ⁴⁰K activity determined from the 1460.7 keV emission gamma-lines and ¹³⁷Cs activity determined from the 661.1 keV emission gamma-lines.

Calculation of the radiological effects

The activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs measured in each of the soil samples. Radium equivalent activity is a widely used hazard index and it is calculated through the relation given by Beretka and Mathew ⁽³¹⁾. It is assumed that 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of

^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate

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

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

Another radiation hazard index called the representative level index, I_{yr} , is defined from the following formula^(32, 33):

$$I_{yr} = \frac{1}{150\text{Bq/kg}} A_{Ra} + \frac{1}{100\text{Bq/kg}} A_{Th} + \frac{1}{1500\text{Bq/kg}} A_K \quad (3)$$

where A_{Ra} , A_{Th} , and A_K have the same meaning as in equation 2.

The external hazard index, H_{ex} was calculated for the investigated samples using the model proposed by Krieger⁽³⁴⁾ assuming thick walls without windows and doors, where the external hazard index is given by

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (4)$$

where A_{Ra} , A_{Th} and A_K are the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Bq/kg, respectively. The calculated average external hazard index was found to be less than unity.

The absorbed dose rate, D (nGy/h) in air at 1 m above ground level due to the presence of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs in the soil samples at each site was calculated using the following equation 5⁽⁵⁾,

$$D = aC_{Ra} + bC_{Th} + cC_K + dC_{Cs} \quad (5)$$

where a is the dose rate per unit ^{226}Ra activity concentration (4.27×10^{-10} Gy/h/Bq/kg), C_{Ra} is the concentration of ^{226}Ra in the sample (Bq/kg), b is the dose rate per unit ^{232}Th activity concentration (6.62×10^{-10} Gy/h/ Bq/kg), C_{Th} is the concentration of ^{232}Th in the sample (Bq/kg), c is the dose rate per unit ^{40}K activity concentration (0.43×10^{-10} Gy/h/ Bq/kg), C_K is the concentration of ^{40}K in the sample (Bq/kg), d is the dose rate per unit ^{137}Cs activity concentration (0.30×10^{-10} Gy/h/ Bq/kg) and C_{Cs} is the concentration of ^{137}Cs in the sample (Bq/kg). The absorbed dose rate (nGy/h) in air at 1 m above the ground determined at each farm does not directly give the radiological hazard to which an individual is exposed.

The annual effective dose equivalent (AEDE) was calculated by using equation 6:

$$AEDE (\mu\text{Sv/y}) = D \times DCF \times OF \times T \quad (6)$$

where D is absorbed dose rate in air (nGy/h), DCF is dose conversion factor (0.7 Sv/Gy), OF is outdoor occupancy factor (0.2), T is the time (8760 h/y)⁽⁵⁾.

Excess life time cancer risk (ELCR) was calculated by using equation 7:

$$ELCR = AEDE \times DL \times RF \quad (7)$$

where DL is duration of life (70 years) and RF is risk factor (Sv^{-1}). For stochastic effects, ICRP 90 uses values of 0.05 for the public⁽²⁹⁾.

RESULTS AND DISCUSSION

The results of activity concentrations in the soil samples from twelve different sites are given in table 1 for the natural radionuclides of ^{226}Ra , ^{232}Th and ^{40}K and the artificial radionuclide of ^{137}Cs . Radium equivalent activity (Ra_{eq}), representative level index, external hazard index, absorbed dose rates, annual effective doses and the excess lifetime risks of cancer in soil samples are given in table 2.

The concentrations found in the present study ranged from 48.54 ± 6.30 (İyidere district) to 163.14 ± 21.29 (Kalkandere district) Bq/kg for ^{226}Ra , from 19.58 ± 4.01 (Hemşin district) to 125.53 ± 22.74 (Güneysu district) Bq/kg for ^{232}Th , from 302.40 ± 15.48 (Güneysu district) to 1159.51 ± 61.06 (Hemşin district) Bq/kg for ^{40}K . The average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were found to be 85.75 ± 11.77 , 51.08 ± 9.42 , and 771.57 ± 37.65 Bq/kg in soil samples, respectively. The activity of ^{40}K is seen to be higher than ^{226}Ra and ^{232}Th in all the selected soil samples. The world's mean values of ^{226}Ra , ^{232}Th and ^{40}K activity concentrations are 32, 45 and 420 Bq/kg, respectively⁽⁵⁾. The mean values of ^{226}Ra , ^{232}Th and ^{40}K are higher than the world's average values.

^{137}Cs does not exist in soil naturally. It is a product of fallout radioactivity. The ^{137}Cs might have been deposited in soil of study area, presumably as a result of the nuclear power

plant accident at Chernobyl on 26 April 1986. Moreover, measured ^{137}Cs activity concentrations can be attributed to the atmospheric nuclear weapon tests conducted by several countries. ^{137}Cs activities in soil samples varied from 75.80 ± 6.30 (İyidere district) to 481.81 ± 30.07 (Güneysu district) Bq/kg and average ^{137}Cs activity was found to be 236.38 ± 13.49 Bq/kg.

As shown in table 3, the radioactivity concentrations in soil samples were comparable

to other studies in various regions (8-25,35-40). Karadeniz et al., Tabat et al., Kiliç et al., Kam and Bozkurt, Celik et al. determined slightly lower activity concentrations of ^{226}Ra and ^{232}Th compared to this study (8,12,17,19,25). Merdanoğlu and Altınsoy et al., Orgun et al. and Abbaspour et al. determined a higher activity concentrations of ^{226}Ra and ^{232}Th compared to this study (13,20,35). ^{40}K activity concentrations of Rize were also higher compared to studies conducted at other parts of Turkey (8, 12, 14-17,19, 21, 22).

Table 1. Radioactivity concentrations of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs in soil samples.

	Number of sampling	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)
Rize Centrum	12	66.50±11.54	48.97±8.29	842.38±45.64	240.32±6.99
Derepaşarı district	12	79.40±12.53	44.49±8.40	876.98±45.70	118.26±5.11
Ardeşen district	12	58.19±11.37	24.29±6.10	654.25±38.54	244.15±6.82
Pazar district	12	65.57±11.11	28.29±5.72	645.94±35.27	374.18±7.78
Fındıklı district	12	50.99±11.44	26.59±6.94	627.95±41.15	322.84±8.65
İkizdere district	10	92.28±9.65	47.25±5.43	871.72±34.11	87.68±4.99
Kalkandere district	12	163.14±21.29	89.57±17.32	708.62±33.82	376.69±18.18
Çayeli district	12	149.64±22.06	68.24±15.54	634.78±36.35	317.58±37.31
Çamlıhemşin district	8	67.86±10.82	24.71±7.39	810.78±28.21	82.49±9.73
Hemşin district	8	101.17±14.46	19.58±4.01	1159.51±61.06	114.78±20.00
Güneysu district	10	85.67±10.34	125.53±22.74	302.40±15.48	481.81±30.07
İyidere district	12	48.54±6.30	65.40±5.20	1123.54±36.57	75.80±6.30
Total district	132	85.75±11.77	51.08±9.42	771.57±37.65	236.38±13.49

Table 2. Radium equivalent, representative level index, external hazard index, absorbed dose rates, annual effective doses and the excess lifetime risks of cancer in soil samples in Rize.

	Ra_{eq} (Bq/kg)	I_{yr}	H_{ex}	D (nGy/h)	AEDE (mSv/y)	Life time total dose(mSv)	ELCR ($\times 10^{-3}$)
Rize Centrum	201.39	1.49	0.54	104.25	0.127	7.30	0.45
Derepaşarı district	210.55	1.56	0.57	104.61	0.128	7.32	0.45
Ardeşen district	143.30	1.07	0.39	76.38	0.093	5.35	0.33
Pazar district	155.76	1.15	0.42	85.73	0.105	6.00	0.37
Fındıklı district	137.37	1.02	0.37	76.06	0.093	5.32	0.33
İkizdere district	226.97	1.67	0.61	110.80	0.135	7.76	0.48
Kalkandere district	345.79	2.46	0.93	170.73	0.209	11.95	0.73
Çayeli district	296.10	2.10	0.80	145.89	0.178	10.21	0.63
Çamlıhemşin district	165.63	1.24	0.45	82.67	0.101	5.79	0.35
Hemşin district	218.45	1.64	0.59	109.46	0.134	7.66	0.47
Güneysu district	288.46	2.03	0.78	147.14	0.180	10.30	0.63
İyidere district	228.57	1.73	0.62	114.61	0.140	8.02	0.49
Total district	218.20	1.60	0.59	110.69	0.136	7.75	0.48
World ⁽⁵⁾	-		<1	60	0.070	4.90	0.29

Table 3. ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs activity concentrations, absorbed dose rates (D), annual effective dose equivalents (AEDE) and excess lifetime risks of cancer (ELCR) in various studies.

Area	²²⁶ Ra (Bq/kg)	²³² Th (Bq/kg)	⁴⁰ K (Bq/kg)	¹³⁷ Cs (Bq/kg)	D (nGy/h)	AEDE (μSv/y)	ELCR (x10 ⁻³)
Rize (Present study)	85.75	51.08	771.57	236.38	110.69	136	0.48
Kırklareli ⁽¹⁴⁾	37	40	667	8	71	87	0.51
Manisa ⁽¹²⁾	-	27	340	-	54	66	-
Istanbul ⁽¹⁶⁾	-	37	342	1.8-81	49	65	-
Marmara ⁽¹⁷⁾	22.5	26.6	443	0.9-154	44.73	54.86	-
Kestanbol ⁽¹⁸⁾	130.93	192	1207	0.37-36	219	269	-
Kastamonu ⁽¹⁹⁾	37.4	27.17	431.43	8.02	52.76	65	-
Çanakkale ⁽²⁰⁾	-	204.69	1171	0-6.57	178-448	-	-
Kazdağı ⁽⁸⁾	21.7	21.1	297.5	0.1-28	44	67	-
Sanliurfa ⁽²²⁾	-	24.95	298.6	9.08	38.24	60.09	-
Firtına ⁽²³⁾	15-116	10-105	105-1235	19-232	77.4	-	-
Eastern Black Sea ⁽²⁴⁾	12-120	40.9	622.8	169.7	77.18	93.30	-
Giresun ⁽²⁵⁾	33	43	733	318	-	92	-
Iran ⁽³⁵⁾	1188	64.92	545.10	10.41	612.37	750	0.30
India ⁽³⁶⁾	57.34	52.83-105.81	95.33-160.30	-	72.35-108.65	72.7-133.2	-
China ⁽³⁷⁾	-	71.5	672	-	124	152	-
India ⁽³⁹⁾	50.58	63.13	268.92	-	66.89	490	-
Nigeria ⁽⁴⁰⁾	-	-	1190	1	52-414	200	-
Worldwide ⁽⁵⁾	35	45	420	-	60	70	0.25

The values of Ra_{eq} varied from 137.37 to 345.79 Bq/kg and average value of Ra_{eq} was found to be 218.20 Bq/kg. The estimated average values of Ra_{eq} in the present work are lower than the recommended maximum value of 370 Bq/kg⁽³¹⁾. On comparing the measured mean values from some of the other studies, it is observed that value of this work is higher than the measured values of 166.3 Bq/kg in Firtına Valley⁽²³⁾. The values of representative level index, I_{γr} varied from 1.02 to 2.46 and average value of I_{γr} was found to be 1.60.

The values of external hazard index, H_{ex} range from 0.37 to 0.93 and average value was found to be 0.59 for the soil samples. The maximum value of H_{ex} must be less than unity. All values estimated of H_{ex} in the present work are lower than unity. The average values of H_{ex} were found to be 0.50 for India, 0.45 for Firtına Valley of Turkey and 0.84 for Xiazhuang Granite Area (China)^(36,23,37).

The absorbed dose rates in air for soil samples the average dose rate was 110.69 nGy/h in Rize. The average D value for soil was

calculated as 71, 77.4, 77.18, 44, 178-448 nGy/h in Kırklareli, in Adana, in Çanakkale, in Firtına Valley, in Eastern Black Sea, respectively^(14,20,21,23,24). The population weighted values give an average absorbed dose rate in air outdoors from terrestrial gamma radiation of 60 nGy/h⁽⁵⁾. This reveals that the mean absorbed dose rate in air outdoors from Rize areas is almost two times higher than that of the worldwide average value.

As shown in table 2 and 3, the calculated values of annual effective dose for the all soil samples ranged from 93 to 209 μSv/y, with a mean of 136 μSv/y, which is higher than the world average value of 70 μSv/y⁽⁵⁾. So, the obtained values are higher than the world average value. The average AEDE value was calculated to be 87 μSv/y in Kırklareli, 66 μSv/y in Manisa, 65 μSv/y in the Istanbul (Turkey), 60.09 μSv/y in Sanliurfa^(14-16,22). These average values are generally lower than our result.

As shown in table 2, when life expectancy was taken as 70 y, the average life time total gamma radiation was calculated as 7.75 mSv, which yielded a lifetime cancer risk of 0.48×10⁻³. The

world's mean value of life time total gamma dose and the excess lifetime cancer risk are 4.90 mSv and 0.29×10^{-3} , respectively (5). The mean of life time total gamma dose and the excess lifetime cancer risk observed in this study are higher than the world's mean values. The average ELCR value was calculated to be 0.5×10^{-3} in Kırklareli, 0.26×10^{-3} in western Mazandaran (Iran) (14,35). Yet, due to the unavailability of related mortality and morbidity statistics, the health hazards of the assessed values on the population were not calculated. Therefore, this study was limited to background radiation levels.

CONCLUSION

The obtained data cover a wide area in Rize. The mean concentrations of the radionuclides ^{226}Ra , ^{232}Th , ^{137}Cs , and ^{40}K in soil samples determined in this study compare suitably with literature values. But the ^{137}Cs activity concentrations in some places are higher than the other results. This can be attributed to the Chernobyl nuclear power plant accident and the atmospheric nuclear weapon tests conducted by several countries. From the measured values, the average values of radium equivalent activity (R_{eq}), representative level index (I_{yr}), external hazard index (H_{ex}), absorbed dose rate in air (D), annual effective dose equivalent (AEDE) and the excess life time cancer risk (ELCR) were calculated. The outdoor air absorbed dose rates (D) due to terrestrial gamma rays for soil have been calculated because of agricultural area and living in the surrounding. It is important to determine background radiation level in order to evaluate the health hazards. Annual effective gamma doses and the lifetime risks of cancer were higher than the world's average. Moreover compared to the World's average, the lifetime risk of cancer doubled for most of the localities.

Conflict of interest: Declared None

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