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Research Article

^{137}Cs Concentration In Soils Collected From Bulgaria-Turkey Border Region

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Abstract

The Chernobyl accident caused artificial radioactive contamination, especially artificial ^{137}Cs radionuclide contamination. Due to having long half-life, ^{137}Cs still continues to remain in soils. In the present study, the surface soil samples were collected from various locations in Bulgaria-Turkey border. The activity concentrations of ^{137}Cs in collected soil samples were determined. In order to investigate the radiological hazard; the dose rates and the annual effective dose rates were estimated. The activity concentrations of artificial ^{137}Cs radionuclide in soils were found to range from 1.71±0.18 Bq kg^{-1} to 6.99±0.66 Bq kg^{-1}. The mean values of absorbed dose rates, outdoor and indoor annual effective dose rates were found to be 0.10, 4.19, and 16.77 µSv y^{-1}, respectively. The dose rates and the annual effective dose rates were below the worldwide limit values reported by UNSCEAR and ICRP. In addition, statistical data were estimated, frequency distribution and Q-Q plot were used to assess ^{137}Cs distribution.

Keywords: ^{137}Cs, Artificial radionuclides, Soil, Activity concentration, HPGe detector.

Bulgaristan-Türkiye Sınır Bölgesinden Toplanan Topraklarda ^{137}Cs Konsantrasyonu

Öz

Chernobyl kazası yapay radyoaktif kirliliğe, özellikle yapay ^{137}Cs radyonüklid kirliliğine neden olmuştur. ^{137}Cs, uzun yarılanma ömrüne sahip olması nedeniyle hala topraklarda bulunmaktadır. Bu çalışmada, yüzey toprak örnekleri Bulgaristan-Türkiye sınırında farklı konumlardan toplanmıştır. Toplanan topraklarda ^{137}Cs aktivite konsantrasyonları hesaplanmıştır. Radyolojik tehlikeyi belirlemek için doz oranları ve yıllık etkin doz oranları tahminleri yapılmıştır. Topraklardaki yapay ^{137}Cs radyonüklid aktivite konsantrasyonları 1.71±0.18 Bq kg^{-1} ile 6.99±0.66 Bq kg^{-1} aralığında bulunmuştur. Soğurulan doz oranları, açık hava ve bina içi yıllık etkin doz oranlarının ortalama değerleri sırasıyla 0.10, 4.19, and 16.77 µSv y^{-1} bulunmuştur. Doz oranları ve yıllık etkin doz oranları, UNSCEAR ve ICRP tarafından raporlanan dünya genelindeki limit değerlerinden daha düşüktür. Ek olarak, istatistiksel veriler hesaplanmış, ^{137}Cs dağılımını belirlemek için frekans dağılımı ve Q-Q çizimi kullanılmıştır.

Anahtar Kelimeler: ^{137}Cs, Yapay radyonüklidler, Toprak, Aktivite konsantrasyonu, HPGe dedektör.

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1. Introduction

Natural and artificial radionuclide contents in soil are the main sources of radiation. Natural radionuclide contents have existed since the beginnig of the universe in the earth's crust. ^{226}Ra, ^{232}Th, ^{40}K and ^{238}U are main radionuclides to exposure natural radiation and can be found in all types of rocks, sand, granite (Mavi and Akkurt, 2010; Aközcan, 2012; Günay, 2018). On the other hand, artificial radionuclides have been spread out to environment by nuclear weapon tests and nuclear accidents (El Samad et al 2013).

The Chernobyl disaster that occured on 26 April 1986 was the most serios nuclear reactor accident. Although, there have been nuclear reactor accidents in the history such as Fukushima Daiichi, Windscale Piles and Three Mile Island accidents, especially the Chernobyl disaster changed the destiny of humanity.

The Chernobyl accident caused artificial radioactive contamination across a large area in the Earth's environment (Obha et al. 2021; Hasegawa et al. 2015). As with many European countries, Bulgaria and Turkey have been affected by the artificial radioactive contamination in relatively high degree (Zhiyanski et al. 2008; Ozyar 2003). Thrace and North-Eastern Blacksea Region are mainly affected areas in Turkey. Radioactive clouds reached to North-Eastern Blacksea Region in 28th April and reached to Thrace region in 1st May (Ozyar 2003).

Many artificial radionuclides have been released to the environment due to the Chernobyl accident. Radiocaesium is the most important one that soils were heavily contaminated by fallout (Rosen et al. 1999). Radiocaesium includes the two long-lived isotopes of ^{134}Cs (half-life is 2.07 years) and ^{137}Cs (half-life is 30.17 years) (Mori et al. 2013). Being a longer-lived isotope, ^{137}Cs will continue to remain in soils and will be detected for many years.

Artificial ^{137}Cs radionuclide bounds and fixes in soil, transfers to plants and the food chain. Exposure to ^{137}Cs can have unhealthy consequences that increases the risk of cancer (Krstic and Nikezic 2006; Paul et al. 2014; Günay and Abamor 2019).

This study focuses on the investigation of contaminated soils due to the Chernobyl accident after 35 years. The soil samples were collected from various locations in Bulgaria-Turkey border to evaluate the activity of artificial ^{137}Cs radionuclide. The surface soil samples counted for ^{137}Cs using High-Purity Germanium detector (HPGe). To understand the health effect of ^{137}Cs contamination due to Chernobyl disaster after 35 years, the dose rates and the annual effective doses were estimated. Statistical analysis of artificial ^{137}Cs activity concentrations were applied to indicate the distribution of radiologic data set.

2. Material and Method

2.1. Sample Preparation

Soil samples were collected from the surface layer (10 cm depth) at 17 different locations in Bulgaria-Turkey border region. The study area is shown in Figure 1. The collected surface soil samples were dried at 105 °C for 24 h in an oven. Dried samples were sieved through a 2 mm mesh to remove impurities. The soil samples were placed in 500 mL polyethylene containers and waited for more than a month in order to reach secular equilibrium before gama spectrometric analysis.

2.2. Sample Measurements

A coaxial high-purity germanium gamma ray detector (Ortec GEM70P4-95) was used to obtain artificial ^{137}Cs activity concentrations. GammaVision-32 software programme was used to analyze activity concentrations. The detector has a 70% relative efficiency in multi-layered coating. The calibrations were carried out using a certified cylindrical shape multinuclide standard source. Source has 1300 mL volume with 1.0 g cm^{-3} density. The activity levels of ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, ^{60}Co were used to perform the energy and efficiency calibrations and energy range was between 80 and 2500 keV.

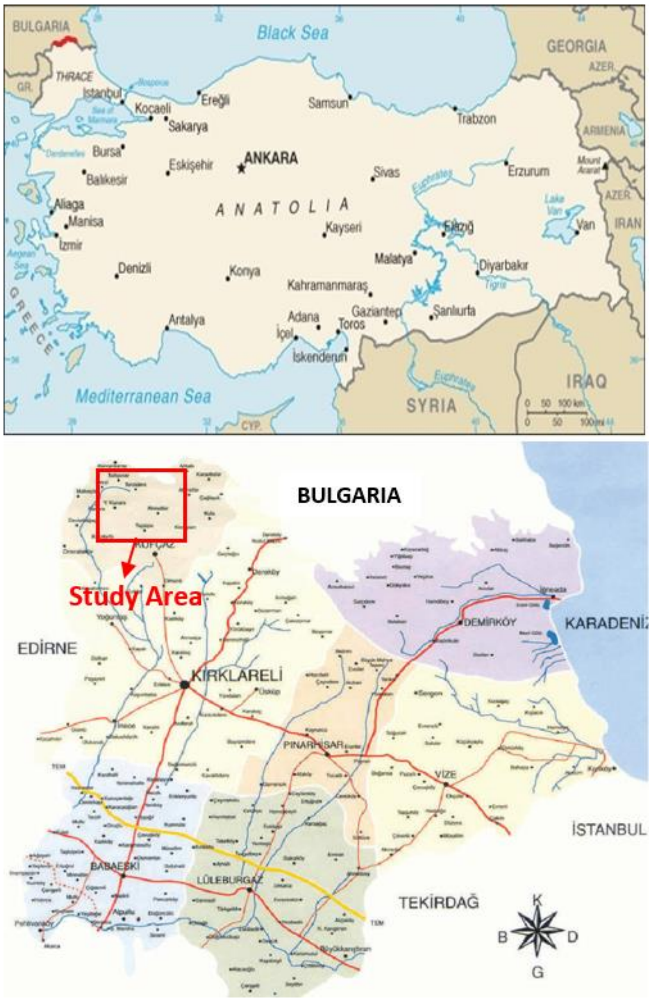

Figure 1. Study area

2.3. Determination of Soil Radioactivity

After equilibrium, each soil samples were counted for 160000 s. The activity of ^{137}Cs was assessed from the gamma-ray determined using 661.66 keV peak. The activity concentrations of ^{137}Cs in the surface soil samples were estimated by;

$$A\ (Bq\ kg^{-1}) = C / (\varepsilon \times I\gamma \times m) \quad (1)$$

In equation, A is the specific activity (Bq kg^{-1}), C is the net counting rate (CPS), ε is gamma-ray detection efficiency, Iγ is the emission probability, m is the mass of the soil sample in kg.

2.4. Estimation of Dose Rates

To estimate the absorbed dose rate in outdoor air (nGy h^{-1}) due to artificial ^{137}Cs radionuclide, the following equation was used:

$$D = 0.03xA \quad (2)$$

where, D is the absorbed dose rate (nGy h^{-1}), A is the specific activity and 0.03 (nGy h^{-1}/ Bq kg^{-1}) is the dose conversion factor for ^{137}Cs activity (Rafique, 2014; Kaynar, 2018; UNSCEAR, 2000)

The annual effective dose rate (E) in µSv y^{-1} was estimated by following relation:

$$E=Q \times T \times OF \times D \times 10^{-3} \qquad (3)$$

In the relation, Q is the conversion factor (Q = 0.7 Sv Gy^{-1}) to convert the absorbed dose in outdoor air to the annual effective dose, T is the number of hours in a year (8760 h), OF is the occupancy factor for outdoor air (0.2) and for indoor air (0.8) (Rafique, 2014; Kaynar, 2018; UNSCEAR, 2000).

3. Results and Discussion

The contour map of ^{137}Cs activity concentrations in soil samples collected from Bulgaria-Turkey border is given in Figure 2. The contour map was plotted using Surfer programme. The activity concentrations of ^{137}Cs are given in Bq kg^{-1} d.w. The lowest ^{137}Cs concentration was found at location 41° 56′99″ N, 27° 8′79″ E and the highest one was found at 41° 50′57″ N, 27° 10′77″ E.

The activity concentrations of ^{137}Cs in the soil samples collected from Bulgaria-Turkey border are summarized in Table 1. In Figure 3, ^{137}Cs activity concentrations of in soil samples are compared with each other. As given in Table 1 and Figure 3, the highest activity concentration value of fallout ^{137}Cs was observed as 6.99±0.66 Bq kg^{-1} d.w for BR4 sample, whereas the lowest activity concentration value of fallout ^{137}Cs was observed as 1.71±0.18 Bq kg^{-1} d.w for BR10 sample.

High ^{137}Cs activity concentration results in surface soil samples can be explained by collected areas are rich in organic matter (Karadeniz and Yaprak 2007).

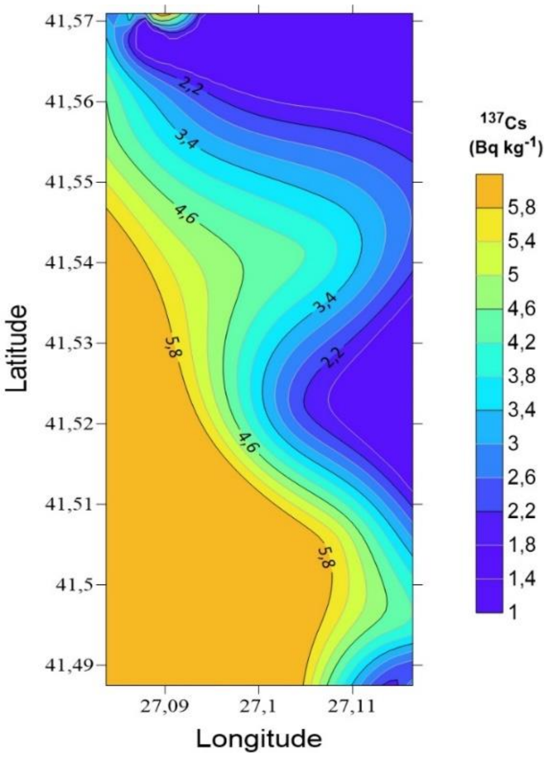

Figure 2. The distribution map of ^{137}Cs activity concentrations

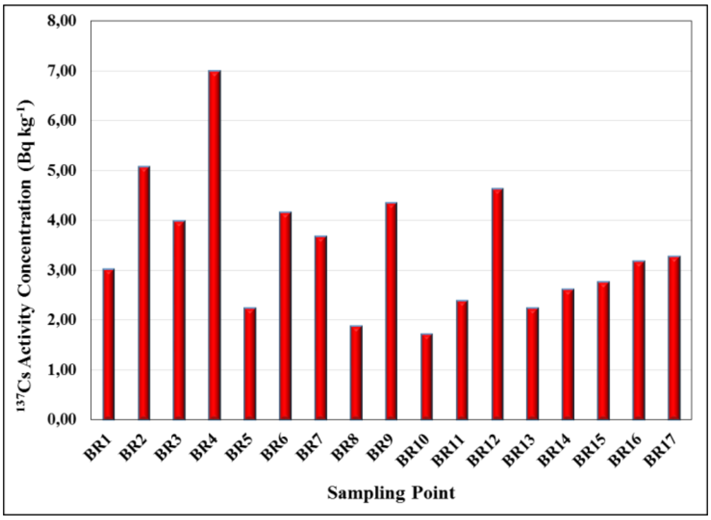

Figure 3. The activity concentrations of ^{137}Cs

The absorbed dose rates (D) in outdoor air (nGy h^{-1}) and the annual effective dose rates (E) in µSv y^{-1} were calculated using activity concentrations of artificial ^{137}Cs radionuclide in surface soil samples and estimated values were given in Figure 4 and 5. The absorbed dose rates ranged 0.05 nGy h^{-1} to 0.21 nGy h^{-1}. The mean value of absorbed dose rates was found as 0.10 nGy h^{-1}. The determined values showed that the annual effective dose rates for outdoor air varied from 2.10 µSv y^{-1} to 8.57 µSv y^{-1} with a mean value of 4.19 µSv y^{-1}. In addition, the annual effective dose rates for indoor air varied from 8.39 to 34.29 µSv y^{-1}, with a mean value of 16.77 µSv y^{-1}. The estimated absorbed dose rates in outdoor air, annual effective dose rates in outdoor and indoor air are given in detail for each sampling point in Table 1. The determined annual effective dose rates are lower than the worldwide standard value (70 µSv y^{-1}) reported by UNSCEAR.

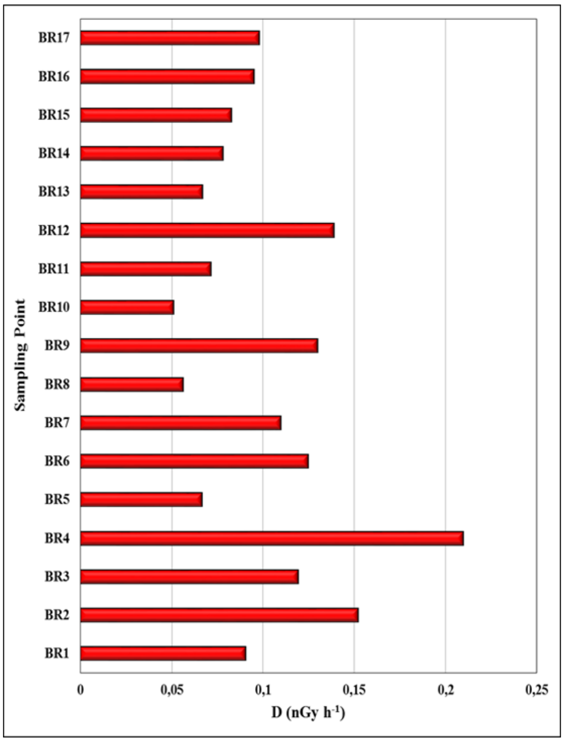

Figure 4. Estimated mean the absorbed dose rates due to ^{137}Cs in Bulgaria-Turkey border

Table 1. The activity concentrations of ^{137}Cs, dose rates and annual effective doses due to ^{137}Cs (Bq kg^{-1})

Sample ID	Activity Concentrations (Bq kg^{-1})	Dose Rates (nGy h^{-1})	Annual Effective Dose Rates (µSv y^{-1})	
			Outdoor	**Indoor**
BR1	3.02±0.43	0.09	3.70	14.81
BR2	5.07±0.48	0.15	6.22	24.87
BR3	3.98±0.29	0.12	4.88	19.52
BR4	6.99±0.66	0.21	8.57	34.29
BR5	2.23±0.37	0.07	2.73	10.94
BR6	4.16±0.37	0.12	5.10	20.41
BR7	3.67±0.45	0.11	4.50	18.00
BR8	1.88±0.86	0.06	2.31	9.22
BR9	4.34±0.18	0.13	5.32	21.29
BR10	1.71±0.18	0.05	2.10	8.39
BR11	2.39±0.42	0.07	2.93	11.72
BR12	4.63±0.49	0.14	5.68	22.71
BR13	2.24±0.71	0.07	2.75	10.99
BR14	2.61±0.38	0.08	3.20	12.80
BR15	2.76±0.41	0.08	3.38	13.54
BR16	3.18±0.23	0.10	3.90	15.60
BR17	3.27±0.54	0.10	4.01	16.04
Range	**1.71±0.18-6.99±0.66**	**0.05-0.21**	**2.10-8.57**	**8.39-34.29**
Mean	**3.42±0.44**	**0.10**	**4.19**	**16.77**

Table 2. Comparison of activity concentrations and annual effective dose rates with other studies

Country	^{137}Cs (Bq kg^{-1})	E (µSv y^{-1})	Reference
Saudi Arabia	0.80–3.10	0.00005-0.0008	(Alaamer, 2012)
Lebanon	23.90-119.20	19.3-91.6	(El Samad et al., 2007)
Pakistan	0.076–2.94	0.0123–0.54	(Rafique, 2014)
Hungary	2.30-13.60	-	(Tserendorj et al., 2021)
Ordu (Turkey)	67.40–275.30	2.50–10.10	(Celik et al., 2010)
Black Sea (Turkey)	26.80–774.70	0.98–28.11	(Cevik et al., 2009)
Manisa (Turkey)	3.61–124.14	3.66–13.81	(Kaynar, 2018)
Marmara Region (Turkey)	0.92-153.72	-	(Kılıç et al., 2008)
Ayvacık (Turkey)	4.00-134	-	(Top et al., 2021)
Adana (Turkey)	0.10 to 28.00	-	(Degerlier et al., 2008)
Mersin (Turkey)	1.80-86.20	-	(Karataşlı et al., 2016)
Küçük Menderes Basin (Turkey)	2.31–7.75	-	(Aközcan, 2014)
Büyük Menderes Basin (Turkey)	2.81-20.75	-	(Aslani et al., 2003)
Thrace Region (Turkey)	3.05-46.78	-	(Aközcan et al., 2014)
Anatolian Side of Istanbul (Turkey)	0.74-6.21	-	(Günay and Aközcan 2018)
Istanbul-Sarıyer (Turkey)	1.70-16.43	-	(Günay and Canel 2019)
Bulgaria-Turkey Border	**1.71-6.99**	**10.49-42.86**	**Present Work**

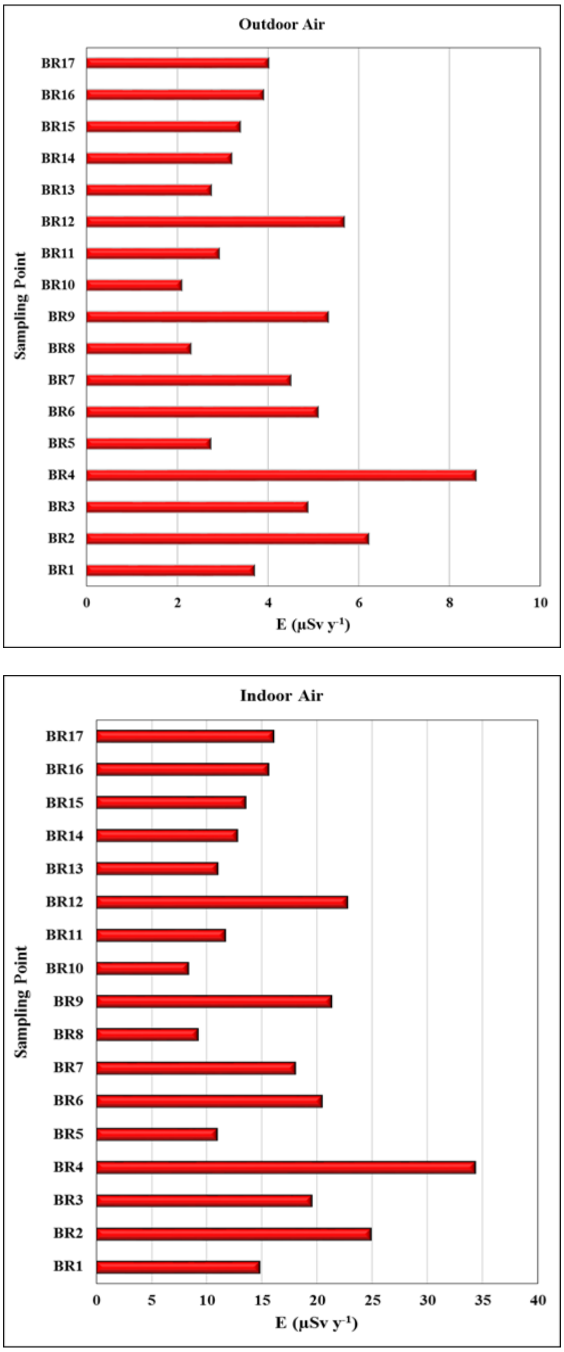

Figure 5. Annual effective doses of ^{137}Cs in Bulgaria-Turkey border

Comparison of the activity concentrations of artifical ^{137}Cs radionuclide in soil samples and estimated indoor annual effective dose plus outdoor annual effective dose rate in this study with other studies was given in Table 2. The measured ^{137}Cs activity concentration was found lower than Black Sea (27–775 Bq kg^{-1}), Manisa (3.61–124.14 Bq kg^{-1}), Ordu (67.40–275.30 Bq kg^{-1}) in Turkey and Lebanon (23.90-119.20 Bq kg^{-1}). Some of concentration levels in this study were found higher than Saudi Arabia, Pakistan, Hungary, Marmara Region (Turkey), Ayvacık (Turkey), Mersin (Turkey), Küçük Menderes Basin (Turkey), Büyük Menderes Basin (Turkey), Thrace Region (Turkey), Anatolian Side of Istanbul (Turkey) and Istanbul-Sarıyer (Turkey). Activity concentration values obtained in the present study was almost in the same range with the results obtained in Anatolian Side of Istanbul (Turkey). The maximum value of annual effective dose (42.86 µSv y^{-1}) in this study was found higher than the values obtained for Saudi Arabia, Pakistan, Ordu (Turkey), Eastern Black Sea (Turkey) and Manisa (Turkey) but lower than Lebanon.

In addition, statistical analysis were performed to understand the behavior of the data of artificial ^{137}Cs activity concentration. Statistical software (SPSS 25.0) was used to obtain information about the data. Mean, median, variance, skewness and kurtosis were calculated and the results are given in Table 3.

Table 3. Statistical data for assessed parameters in soil samples

Variables	Results
Mean	3.42
Median	3.18
Std. Deviation	1.35
Skewness	1.13
Kurtosis	1.62
Minimum	1.71
Maximum	6.99
Range	5.28

As seen in Table 3, mean and median of the data was found as 3.42 and 3.18, respectively. Skewness of the data gives information about the distribution. Skewness of ^{137}Cs was found as a positive value (1.13). Positive value of skewness shows that the peak of the distribution is right of mean value and the data are highly skewed (>1). Kurtosis of the data provides information about the probability distribution rate. Kurtosis was found as a positive value (1.62). Positive value of kurtosis indicates relatively peaked distribution.

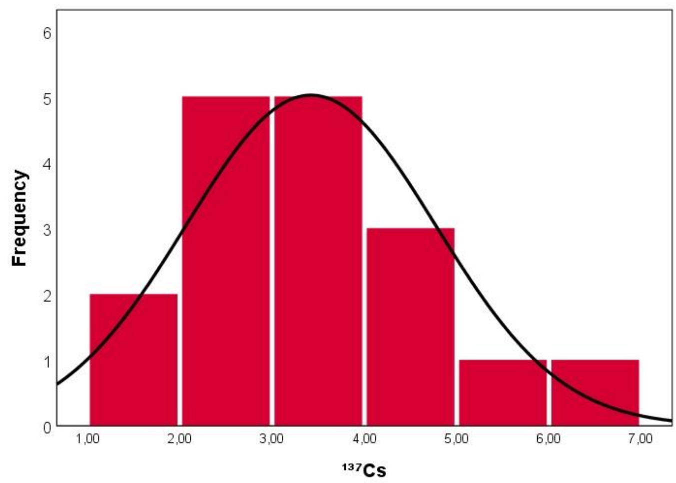

Figure 6. Frequency distribution of ^{137}Cs in soil samples

The frequency distribution of ^{137}Cs in soil samples is given in Figure 6. The frequency distribution shows that radionuclides demonstrate multimodality characteristic with log-normal distribution. The multimodality characteristic of distribution shows the complexity of radionuclide in soil samples. In addition, to identify type of distribution, quantile-quantile plot (Q-Q plot) of ^{137}Cs was examined and given in Figure 7. As seen in plot, the points line approximately along reference line. It can be supposed that the obtained data for ^{137}Cs activity concentration show normal distribution.

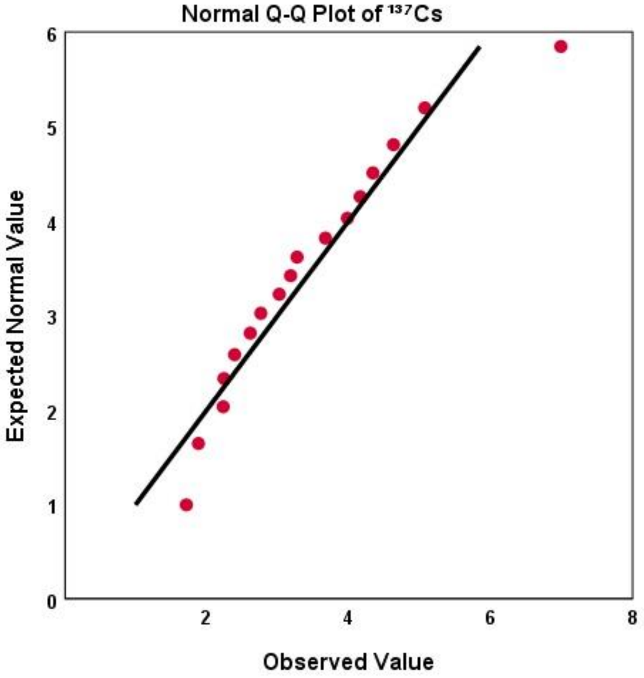

Figure 7. Q-Q plot of ^{137}Cs in soil samples

4. Conclusions and Recommendations

In the present study, the activity concentrations of ^{137}Cs in soil samples collected from Bulgaria-Turkey border were measured. The activity concentration levels of ^{137}Cs in suface soil samples were found to range from 1.71±0.18 Bq kg^{-1} to 6.99±0.66 Bq kg^{-1}. The mean activity concentration value was calculated as 3.42±0.44 Bq kg^{-1}. The absorbed dose rates, the outdoor and indoor annual effective dose rates due to ^{137}Cs for soil samples were estimated. The mean values of absorbed dose rates, outdoor and indoor annual effective dose rates of the soil samples were found to be 0.10, 4.19, and 16.77 µSv y^{-1}, respectively. Estimated data were found lower than the worldwide standard value of annual dose (70 µSv y^{-1}) and recommended annual dose rate limit (1 mSv y^{-1}) (UNSCEAR 2000, Smith, 1991).

In addition, statistical analyis were performed to understand the distribution of ^{137}Cs activity concentration. Skewness of the data showed that the peak of the distribution is right of the mean value and kurtosis of the data indicated relatively peaked distribution. The frequency distribution and also Q-Q plot showed the multimodality characteristic of distribution and normal normal distribution, respectively.

The measurements and analysis showed that the studied regions are contaminated with ^{137}Cs. Results in this study will be indicated in important data for the contaminated soils due to the Chernobyl accident after 35 years.

5. Acknowledge

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