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Assessment of Radioactivity Levels of Wastewater Treatment Plants Sludges of Western Antalya

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***Abstract*:** *To determine whether the use of wastewater treatment sludge in agricultural areas poses any radiological risk or not, both natural ^{226}Ra, ^{232}Th, ^{40}K and artificial ^{137}Cs radionuclide activity concentration levels of the sludges, collected from wastewater treatment plants located in Beldibi, Çamyuva, Göynük, Hurma, Kemer and Tekirova districts of Antalya were assessed. Measurement results of the wastewater sludge samples indicate the existence of natural (^{226}Ra, ^{232}Th, ^{40}K) and artificial radionuclides (^{137}Cs) from the Chernobyl accident and other nuclear activities. The calculated mean ^{226}Ra (4.2 Bq kg^{-1}), ^{232}Th (6.5 Bq kg^{-1}) and ^{40}K (115.0 Bq kg^{-1}) and ^{137}Cs (0.9 Bq kg^{-1}) activity concentrations of the sludges were observed to be below the world average. Findings were found to be consistent with the published results in the literature for Türkiye and other countries. The absorbed gamma dose rate (D: 6.2 - 16.2 $nGyh^{-1}$), radium equivalent activity (Ra_{eq}: 12.7 - 34.5 Bq kg^{-1}), annual equivalent dose (AED: 7.7 – 19.9 µSv h^{-1}), internal ($H_{in}<1.0$) and external ($H_{ex}<1.0$) hazard indices were calculated by using sample activity concentrations. The radiological risk indices (D, Ra_{eq} and AED) are in the permissible limits published by IAEA. As a result, it was observed that the use of wastewater treatment sludge in agricultural areas would not create any risk in radiological terms.*

Keywords: *Radioactivity levels, treatment sludge, WWTP*

1. Introduction

Radioactive pollution in water that has emerged in recent years is also a serious concern for human health. Recently, radioactive contamination and related health effects have been reported in many parts of the world. Leakage of naturally occurring radioactive materials (NORM) from soil sediments into the aquifer causes groundwater pollution [1]. In anthropogenic sources of radioactive pollution, nuclear weapons research, nuclear disasters, nuclear power plants and the discharge of radioactive waste are major sources of contamination of radioactivity, while the application of radioisotopes in industries and scientific laboratories are minor sources. Mining activities of radioactive elements such as uranium and thorium also pollute surface and groundwater. Radioactive contamination is more common in groundwater than in surface waters due to greater exposure to the radioactive elements found in rocks. A number of common radionuclides ^{3}H, ^{14}C, ^{40}K, ^{210}Pb, ^{210}Po, ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{232}Th, and $^{234,235,238}U$ are found in surface and ground waters. Uranium, thorium and actinium are the three NORM series that pollute water resources. ^{40}K and ^{7}Be are the most common radioactive elements in sludge from sewage treatment plants.

Urban wastewater sludge is an end product of urban wastewater treatment and contains many pollutants left over from wastewater treatment. Sewage sludge is a concentrated solids suspension, which consists mostly of organic solids loaded with mineral salts and whose density can vary in slurry or dry form depending on the treatment technique. Today, the agricultural use of sewage sludge is accepted as an economical alternative disposal method compared to other disposal methods. The common finding of the studies carried out to date is that sludge has an economic value in plant cultivation, but pollutants that can mix into the sludge and significantly limit their use.

In recent years, the use of sewage sludge in agriculture has been made safer with legal regulations regarding the use of sewage sludge in agricultural lands. However, studies on radioactive contamination of sewage sludge are very limited. In this study, sludge from treatment plants in western Antalya region will be evaluated in terms of radioactivity pollution.

2. Materials and Methods

Sludge samples were collected from wastewater treatment plants (WWTP) located around Beldibi, Çamyuva, Göynük, Hurma, Kemer and Tekirova districts of Antalya province of Türkiye on a monthly basis for one year (Figure 1). The sludges were numbered and labeled after they were transferred to the sample preparation laboratory of Akdeniz University, Faculty of Science, Department of Physics. Foreign substances and impurities in each sludge sample were removed. Before the measurements, all sludge samples were stored (air-dried) 4–7 d until they reached a constant weight in a ventilated room. All samples were homogenised with the grinding machine and then sieved through a 2-mm mesh in the sample preparation laboratory. The sieved samples were then filled into hermetically sealed (6cm x 5cm) 150 cc polyethylene cylindrical containers, labelled,weighed and stored for 4 weeks in order to reach secular equilibrium between ^{226}Ra and ^{222}Rn prior to counting. Approximately 5 g of sludge from each sample were put in 6-cm diameter cylindrical containers and dried at 80^{0}C for 14 h to determine the moisture rate of the samples.

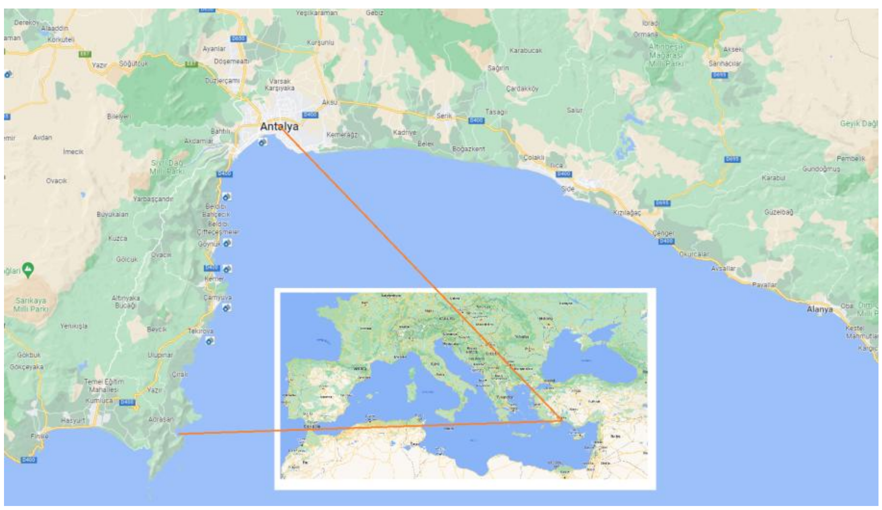

Fig. 1: Study area

Radioactivity measurement was conducted by using a p-type, coaxial, electrically cooled, high-purity germanium gamma-ray detector AMATEK-ORTEC with Full Width Half Maximum (FWHM) at 122 keV for ^{57}Co and 1.85-keV FWHM at 1332 keV for ^{60}Co. It is connected to an NIM consisting of ORTEC bias supply, spectroscopy amplifier, analogue-to-digital converter and a computer. The detector was placed into a 10-cm thick lead shield with an inner surface covered by a 2-mm thick copper foil to shield from the x-rays originating in lead. Data acquisition and analysis were carried out with MAESTRO32 software.

All samples were placed to the front face of the detector and counted for 50 000 s. Background intensities were obtained with an empty beaker for 50 000 s under the same conditions before and after measurement of the samples. Then, the average of the background counts was subtracted from the sample spectrums. ^{238}U and ^{232}Th activity concentrations were determined from their daughter products indirectly, while ^{137}Cs and ^{40}K were determined directly by their gamma-ray peaks. To determine the activity concentration of the ^{238}U nuclide, daughter nuclides ^{214}Pb and ^{214}Bi were used, while ^{228}Ac concentration was chosen for the parent ^{232}Th. The gamma transitions of 351.9 keV ^{214}Pb and 609.3 keV ^{214}Bi were used to determine the concentrations of ^{238}U. The gamma transition of 911.2 keV ^{228}Ac was used to determine the concentration of ^{232}Th. 661.6 keV and 1461.0 keV gamma transitions were used to determine the concentration of ^{137}Cs and ^{40}K, respectively.

Details of the activity and dose calculations were presented by [2].

3. Results and Discussion

Measurement results of the wastewater sludge samples indicate the existence of natural (^{226}Ra, ^{228}Ac, ^{40}K) and artificial radionuclides (^{137}Cs) from the Chernobyl accident and other nuclear activities. Activity concentration levels of the sludges were presented in Figure 2.

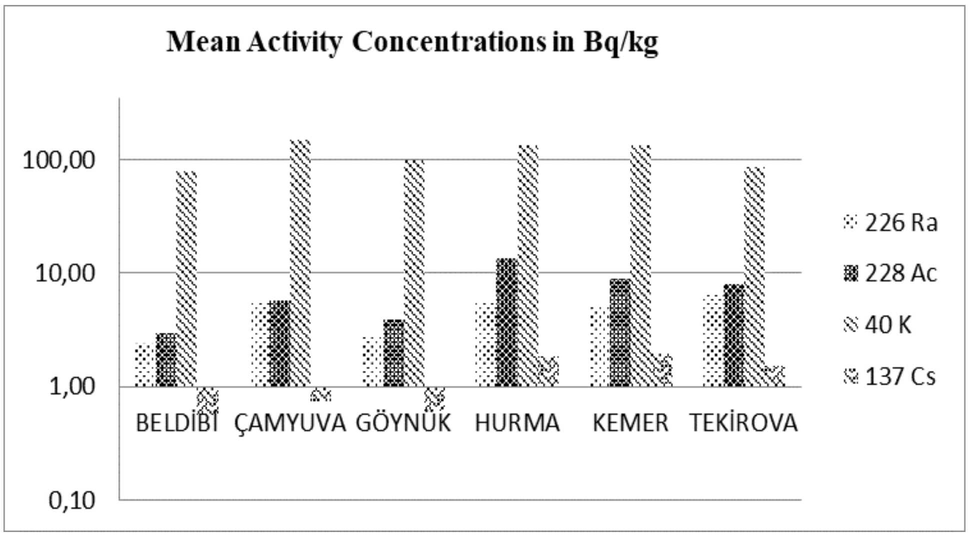

Fig. 2 – ^{226}Ra, ^{228}Ac, ^{40}K and ^{137}Cs activity concentrations of Sludge in Bq/kg

The calculated ^{226}Ra activity concentrations were ranged from BDL to 4.0 with a mean (2.4 Bq kg^{-1}) for Beldibi district, from 1.5 to 14.4 with a mean (5.7 Bq kg^{-1}) for Çamyuva district, from 0.5 to 5.1 with a mean (2.9 Bq kg^{-1}) for Göynük district, from BDL to 17.8 with a mean (5.2 Bq kg^{-1}) for Hurma district, from BDL to 6.0 with a mean (0.9 Bq kg^{-1}) for Kemer district and from 2.2 to 13.8 with a mean (6.5 Bq kg^{-1}) for Tekirova district.

The calculated ^{228}Ac activity concentrations were ranged from 2.2 to 4.0 with a mean (3.0 Bq kg^{-1}) for Beldibi district, from 2.1 to 12.0 with a mean (5.8 Bq kg^{-1}) for Çamyuva district, from 3.1 to 4.6 with a mean (3.9 Bq kg^{-1}) for Göynük district, from 9.6 to 17.4 with a mean (13.4 Bq kg^{-1}) for Hurma district, from 2.9 to 16.0 with a mean (9.0 Bq kg^{-1}) for Kemer district and from 2.6 to 11.4 with a mean (8.1 Bq kg^{-1}) for Tekirova district.

The calculated ^{40}K activity concentrations were ranged from 62.4 to 105.6 with a mean (78.7 Bq kg^{-1}) for Beldibi district, from 117.7 to 175.9 with a mean (150.2 Bq kg^{-1}) for Çamyuva district, from 74.3 to 123.0 with a mean (97.8 Bq kg^{-1}) for Göynük district, from 95.6 to 211.1 with a mean (133.2 Bq kg^{-1}) for Hurma district, from 50.1 to 322.0 with a mean (135.7 Bq kg^{-1}) for Kemer district and from 48.1 to 134.3 with a mean (86.6 Bq kg^{-1}) for Tekirova district.

The calculated ^{137}Cs activity concentrations were ranged from BDL to 1.0 with a mean (0.6 Bq kg^{-1}) for Beldibi district, from 0.5 to 1.3 with a mean (0.7 Bq kg^{-1}) for Çamyuva district, from BDL to 1.0 with a mean (0.8 Bq kg^{-1}) for Göynük district, from BDL to 3.6 with a mean (3.5 Bq kg^{-1}) for Hurma district, from BDL to 4.2 with a mean (1.9 Bq kg^{-1}) for Kemer district and from BDL to 3.2 with a mean (1.7 Bq kg^{-1}) for Tekirova district.

The calculated ^{226}Ra, ^{232}Th (^{228}Ac), ^{40}K and ^{137}Cs activity concentrations of the sludges were observed to be below the world average [3]. Findings were found to be consistent with the published results in the literature for Türkiye and other countries (Table 1).

The calculated ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs activity concentrations of the sludges were observed to be below the world average [3]. Findings were found to be consistent with the published results in the literature for Türkiye and other countries (Table 1).

TABLE I: Radionuclide activity concentrations of soil samples from Literature (in Bq/kg).

Country	**^{226}Ra**	**^{232}Th**	**^{40}K**	**Reference**
Türkiye	29	22	464	[4]
Greece	16	55	305	[5]
Hong Kong	59	95	530	[6]
India	57	87	143	[7]
Pakistan	51	59	665	[8]
Serbia	60	49	379	[9]
Yugoslavia	39	53	554	[10]
Bosnia Herzigova	32	32	331	[11]
Italy	79	48	640	[12]
World Mean	35	30	400	[3]
Present study	8.5	12.1	96.9	

In order to determine the health effects, radiologic risk parameters (D, Raeq,AED, Hex and Hin) of the sludges are calculated and results were presented in Figure 3.

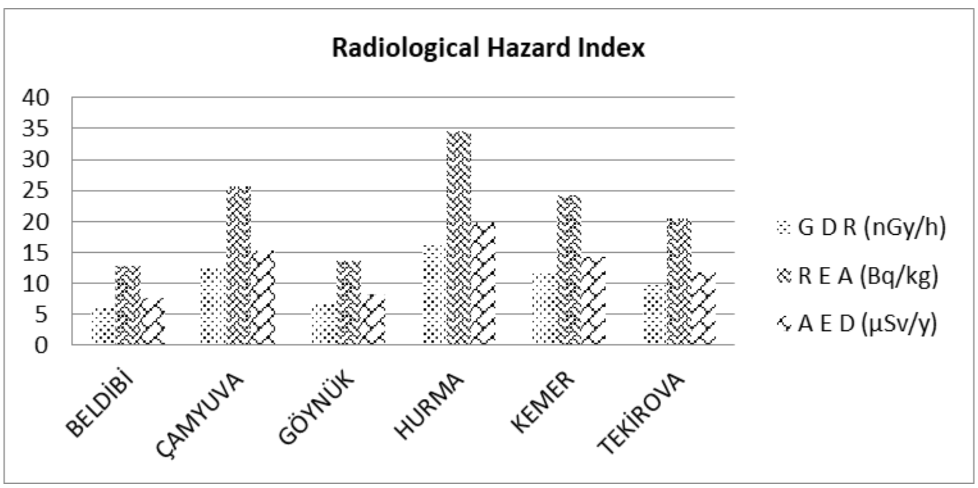

Fig. 3 – Mean D, Ra_{eq} and AED values of Sludges

The calculated mean absorbed gamma dose rate (D), were ranged from 6.2 $nGyh^{-1}$ (Beldibi) to 16.2 $nGyh^{-1}$ (Hurma) with a mean (10.5 $nGyh^{-1}$), radium equivalent activity (Ra_{eq}) were ranged from 12.7 Bq kg^{-1} (Beldibi) to 34.5 Bq kg^{-1} (Hurma) with a mean 21.9 Bq kg^{-1}, annual equivalent dose (AED) levels were ranged from 7.7 µSv h^{-1} (Beldibi) to 19.9 µSv h^{-1} (Hurma) with a mean 12.9 µSv h^{-1}, internal (H_{in}<1.0) and external (H_{ex}<1.0) hazard indices were less then unity. The radiological risk indices (D, Raeq and AED) are all in the permissible limits reported by IAEA.

The absorbed gamma dose rate (D: 6.2 - 16.2 $nGyh^{-1}$), radium equivalent activity (Ra_{eq}: 12.7 - 34.5 Bq kg^{-1}), annual equivalent dose (AED: 7.7 – 19.9 µSv h^{-1}), internal (H_{in}<1.0) and external (H_{ex}<1.0) hazard indices were calculated by using sample activity concentrations. The calculated average values for D, Ra_{eq} and AED are in the permissible limits published by IAEA.

4. Conclusion

Radionuclide concentrations of waste water sludge samples around Antalya were determined by the present study. Findings were lower or comparable to the literature levels for soil samples around the world. Moreover, annual effective dose exposure due to the radioactivity content of sludges was not very high to pose a serious health risk.

We can conclude that the use of wastewater treatment sludge in agricultural areas would not create any risk in radiological terms. In terms of chemical properties, the Cs element is remarkably similar to the element K, which is a plant nutrient and is consumed significantly by plants. Since sewage sludge contains ^{137}Cs even in low concentrations, it is obvious that care should be taken both in its use in agricultural areas and in discharges to the open sea and similar situations.

5. Acknowledgements

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