Iranian Journal of Medical Physics

ijmp.mums.ac.ir

Radiological Hazard Assessment of Radionuclides in Sediment and Water Samples of International Meighan Wetland in Arak, Iran

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ARTICLE INFO

Article type:
Original Article

Article history:
Received: Mar 13, 2019
Accepted: Jun 08, 2019

Keywords:
Cancer
Dosage
Natural Radiation
Sediment
Water

ABSTRACT

Introduction: There are natural and artificial radioactive nuclei in our environment, as well as in the structure of the living organism. Currently, industrial and municipal pollution has also an impact on increasing the level of radiation. The present study investigated the effect of inlet water from Arak Wastewater Treatment Plant on international Meighan Wetland and assessed the radiological indicators of sediments and water samples in this area.

Material and Methods: In this study, the specific activity of radionuclides in water and sediment samples taken from the water entry areas of the international Meighan wetland was determined using a high purity germanium detector (Baltic Scientific Instrument LTD, 005- Latvia). Radiological indices for collected samples were calculated, and the topographical maps of radiation dose distribution were plotted using Surfer software (version 13).

Results: Specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in sediment samples were in the range of 14.44-26.58, 22.78-34.56, 360.84-447.79, and 0.7-13.03Bq/kg, respectively. The average values of the external hazard index for sediment samples were calculated at 0.25.

Conclusion: According to the obtained results, it can be concluded that pollution is more embedded in the Treatment Plant's basin, and a small amount goes to the wetlands. Radioactivity in the research area is normal, and topographic maps show that the distance from the entrance reduces the activity of radium and increases the activity of cesium. Assessment of hazard indicators showed that radiation levels in this area are not dangerous to human health.

►*Please cite this article as:*
Pourimani,R, Fardad R, Khalili H. Radiological Hazard Assessment of Radionuclides in Sediment and Water Samples of International Meighan Wetland in Arak, Iran. Iran J Med Phys 2020; 17: 107-113. 10.22038/ijmp.2019.39081.1512.

Introduction

Ionizing radiation is an integral part of nature. Most of these rays are generated through the decay of natural and artificial radionuclides. Natural radionuclides, including ^{235}U, ^{232}Th, ^{238}U, and their radioactive daughters, as well as other isotopes, such as ^{40}K, are detected in the Earth's crust. [1]. They originate from the beginning of the Earth's formation, and their quantities change on the Earth depending on the type of soil, rock, or geographical area [2].

Average values of uranium content and thorium in the Earth's crust are estimated at 2.7 [3] and 10.5 mg kg^{-1} [4], respectively. Amount of potassium has been measured at 2.8% as the eighth element in terms of abundance in the Earth's crust [5]. Nuclear radiation emitted from these elements is harmful to the tissues of the body and may cause diseases, such as cancer or genetic abnormalities [6].

According to the United Nations Scientific Committee on the Effects of Atomic Radiation reported in 2000 (UNSCEAR), the mean annual absorption of natural ionizing radiation dose is 2.4 mSv, out of which 0.5 mSv refers to the presence of radionuclides in soil, rock, water, and building materials, and 1.2 mSv originates from radon gas breathing. Rest of the absorbed dose stems from cosmic rays and radionuclides in the diet [6].

Therefore, determining the number of radionuclides in soil, rock, and water in each locality is very important for the identification of the exact sources of radioactivity in human ecosystems and ability to estimate the dose absorbed by individuals living in the region. The aim of this study was to determine the specific activities of ^{226}Ra, ^{232}Th,^{40}K, and ^{137}Cs radioisotopes in sediment and water samples at the entrance to international Meighan Wetland from Wastewater Treatment in Arak, as a metropolis, Iran, and assessed radiological impact on this region.

Materials and Methods

Sampling and Sample Preparation

Meighan Wetland with an area of 25,000 ha is located 15 km northeast of Arak and south of

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Davoudabad, Iran, adjacent to the villages of Rahzan, Dehnamak, and Tormad in Markazi province in Iran. The area under water varies between 10,000 and 12,000 hectares depending on annual rainfall.Its average height is 1,670 m above the sea level, and the annual rainfall in this region is 258 mm.

In different seasons, the water level in the swamps changes even by 140 cm. Since 2003, purified water from Arak Wastewater Treatment Plant, with an average of 1,000 l per sec, also pours into wetlands, which had a major impact on the region's ecosystem. Amount of water flowing in from this treatment plant is approximately 31,000,000 m^3 per year [7]. In this study, 10 sediment samples and 10 water samples were collected at the entrance of international Meighan Wetland. The sampling site is depicted in Figure 1, and the geographic coordinates of the sampling points are listed in Table 1.

The sediment samples were collected at a depth of up to 5 cm; at this point, the water sample was obtained from the surface. The water samples were collected in 1.5-liter polyethylene bottles, and their PH reduced to 2 by adding nitric acid to prevent the absorption of radionuclides through the bottle wall [8]. The sediment samples were placed in an oven at 100 °C for 8 h for the complete removal of the moisture and then milled and passed through a 40 mesh screen (0.403 mm^2) to obtain a completely homogeneous powder.

The sediment and water samples were packed in 300 cm^3 polyethylene and 800 ml Marinella containers, respectively. All the samples were sealed and stored in the laboratory for 60 days; this time is necessary for the equilibrium of radium-radon activities in uranium series. Spectrometry of each sample was carried out in duration of 86,400 sec.

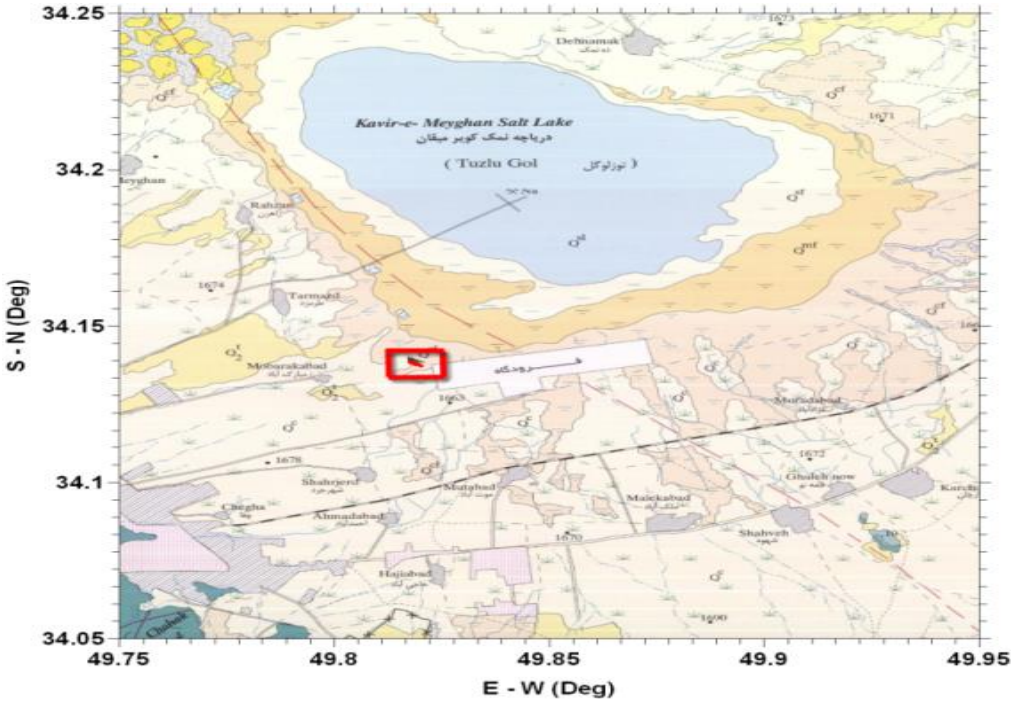

Figure 1. Geographical map of studied region in Meighan Wetland marked by red square

Table 1. Geographical coordinate and code of collected samples of Meighan Wetland

Sediment sample code (S) Water sample code (W)	Longitude (N)	Latitude (E)
S1, W1	34.13855	49.81765
S2, W2	34.13860	49.81769
S3, W3	34.13829	49.81787
S4, W4	34.13819	49.81799
S5, W5	34.13804	49.81844
S6, W6	34.13787	49.81874
S7, W7	34.13771	49.81898
S8, W8	34.13762	49.81936
S9, W9	34.13763	49.81965
S10, W10	34.13729	49.82021

Gamma-ray spectroscopy

Spectra of the samples were recorded using a P-type coaxial high-purity germanium detector model GCD-30195BSI produced by BSI Company (Baltic Scientific Instrument LTD, 005- Latvia) with an energy resolution of 1.95keV for gamma line 1332.52 keV belonging to ^{60}Co. The energy and gamma spectrometry efficiency calibration was measured using the standard sources of a mixture containing radionuclides ^{241}Am, ^{152}Eu, and ^{137}Cs, as well as RGU-1, RGK-1 and RGTh-1 reference materials, with exact known specific activities of natural radionuclides and quality control performed at 0.05 probability.

The detector is placed in the center of a 10 cm thick lead covered with an inner copper layer of 2 mm thickness to reduce the effects of background radiation (i.e., cosmic rays, as well as low energy photons and electrons) [9]. In the present study, background radiation correction was performed with the spectrum recorded for the empty container under the same conditions. Based on the registered spectra, the specific activities of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined in the samples. Absolute efficiency of the detector is calculated using equation 1 [10].

$$\varepsilon\,(\%) = \frac{N_i}{Act \times P_n\,(E_i) \times T} \times 100 \quad (1)$$

Where N_i is a pure counting under the photopeak with the energy E_i, *Act* is the known activity of the radioactive nuclei contained in the standard container in terms of Bq, $P_n\,(E_i)$ is the gamma emission probability with energy E_i per decay in term of percentage, and T is the recording spectrum of the sample in second [10].

Activity measurement

Equation 2 was used in order to calculate the specific activity.

$$Act = \frac{Net\ Area}{\varepsilon \times B.R\ (\%) \times t \times m} \times 100 \quad (2)$$

Where *Act* is the radionuclide specific activity in Bqkg^{-1}, *Net Area* is the subpeak count in full energy corresponding to specific energy E_i, ε is the efficiency of the detector for gamma-ray energy E_i, *B.R* is a branching ratio in percentage, t is the duration of the recorded spectrum in second, and m is the sample mass in kg [11].

Specific activity of ^{226}Ra samples was measured using a ^{214}Pb gamma ray with the energies of 351.93 keV and a gamma ray of ^{214}Bi with the energy of 609.31 keV. To determine the specific activity of ^{232}Th, two ^{228}Ac gamma rays were used, including one with the energy of 911.21 keV and intensity probability of 26.6% and the other with the energy of 968.97 keV and intensity probability of 17.4%. In addition, the specific activity of ^{40}K was determined by the gamma rays of this radionuclide as 1460.70 keV [11].

Radiological hazard parameters

Radium equivalent (Ra_{eq})

Overall, 98.5% of the radiological effects of the uranium series are associated with ^{226}Ra and its progeny radionuclides. Therefore, the radium equivalent activity parameter was introduced by UNSCEAR to determine the total level of environmental radiation and possibility of comparing the radiation level of the samples. Radium equivalent activity is an indicator of the total radioactivity of ^{226}Ra, ^{232}Th and their daughters, and ^{40}K in samples that are expressed in terms of radium equivalent activity and is calculated using Equation 3[12]. The global average value for soil and rock is 131.69 Bqkg^{-1} [12].

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \quad (3)$$

Absorbed gamma dose rate (D)

The absorbed dose rate in the air is calculated based on the gamma rays emitted from the radionuclides of soil, rock, and water at a height of 1 m above the terrestrial level using equation 4 [13]. Global average of absorbed dose rate for terrestrial materials is 55nGyh^{-1} [12].

$$D(nGyh^{-1}) = 0.427A_{Ra} + 0.662A_{Th} + 0.00432A_K \quad (4)$$

Annual effective dose equivalent (indoor and outdoor)

The annual effective dose equivalent absorbed by humans due to external gamma radiation in mSvy^{-1} has been estimated by equation 5, where D is the amount of absorbed dose rate in the air at a height of 1 m from the ground [14].

$$AED_{indoor}\,(mSvy^{-1}) = Dose\ rate(nGyh^{-1}) \times 8760\,(hy^{-1}) \times 0.8 \times 0.7\,(SvGy^{-1}) \times 10^{-6}, \quad (5)$$

where with 8,760 h of exposure in 1 year, 0.8 is a factor of spent-time in the interior, and 0.7 is the coefficient of conversion of absorbed dose rate in the air to the effective dose for humans. External annual effective dose equivalent was calculated using equation 6 [14].

$$AED_{outdoor}\,(mSvy^{-1}) = \text{Dose rate}\,(nGyh^{-1}) \times 8760\,(hy^{-1}) \times 0.2 \times 0.7\,(SvGy^{-1}) \times 10^{-6},$$

where the coefficient of 0.2 is a fraction of spent-time in the external environment. Global average values of the annual effective dose equivalent of the indoor and outdoor environment were 0.41 and 0.06 mSvy^{-1}, respectively, and for safety environment, it should be lower than 1 mSvy^{-1} [12].

Excess lifetime cancer risk

Excess lifetime cancer risk (ELCR) caused by gamma radiation from terrestrial radionuclide is calculated using equation 7 [15].

$$ELCR\ (in\text{-}out) = AEDE\ (in\ and\ out) \times DL \times RF \quad (7)$$

Where AEDE is the total annual effective dose equivalent (indoor and outdoor), DL is the of average life duration (i.e., 70 years), and RF is the risk factor. The International Committee on Radiation Protection determined the coefficient of 0.05Sv^{-1} for possible effects of radiation on individuals [16]. The global average of ELCR is 0.29×10^{-3}, and its maximum value is 10^{-3} [12].

Gamma index

The gamma index is used to estimate the risk of gamma radiation associated with natural radionuclides in the study area. The quantity of these indicators should be less than one for environmental safety. Its value is obtained from equation 8 [12].

$$I_\gamma = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \leq 1 \quad (8)$$

Internal and External Hazard Index

External and internal risk indicators are used to assess the gamma radiation risk caused by natural

radionuclides and inhalation of radon gas, respectively. The quantity of these indicators should be less than one for non-significant hazards and environmental safety. These indicators are calculated using equations 9 and 10 [12].

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 < 1 \quad (9)$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 < 1 \quad (10)$$

Annual Gonadal Dose Equivalent

The UNSCEAR studied some of the body's organs, such as the thyroid glands, lung, bone marrow, bone, testicles, and breast. According to the committee's opinion, the annual equivalent dose of these glands is calculated using equation 11 [12]. Global average value of annual gonadal dose equivalent is estimated at 300 μSvy^{-1} [12].

$$AGED(\mu Svy^{-1}) = 3.09\, A_{Ra} + 4.18 A_{Th} + 0.314 A_K \quad (11)$$

In equations 3, 4, and 8 to 11, A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in $Bqkg^{-1}$, respectively.

Results

Specific activity measurements of natural and artificial radionuclides and radium equivalent in sediment and water samples are shown in tables 2 and 3. Results of calculated radiological parameters for sediment samples are listed in Table 4. The ELCR value was calculated on the basis of the annual effective external dose equivalent, and the results of the calculated dose absorbed rate in the air in comparison with the global average are depicted in figures 2 and 3.

Table 2. Specific activities of radionuclides and radium equivalent in sediment samples

Sample code	Radionuclides specific activities ($Bqkg^{-1}$)				Ra_{eq} ($Bqkg^{-1}$)
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	
S1	22.15±1.01	22.78±1.11	439.47±11.58	0.70±0.16	88.56±2.57
S2	22.32±0.75	22.79±1.03	432.53±11.42	< 1.51	88.21±2.40
S3	26.58±1.27	24.14±1.02	382.81±10.70	< 1.35	90.57±2.61
S4	24.04±1.18	32.00±0.95	411.62±11.15	6.35±0.52	101.48±2.51
S5	24.80±1.28	30.50±0.95	417.32±11.11	8.53±0.54	100.54±2.55
S6	24.87±1.12	33.23±1.45	383.77±10.65	5.36±0.51	101.94±2.86
S7	24.11±1.00	23.80±1.17	394.07±10.76	9.60±0.54	88.48±2.62
S8	14.44±0.60	25.14±0.73	447.79±10.53	8.86±0.37	84.88±2.06
S9	22.77±0.85	34.56±0.95	420.18±11.13	8.06±0.53	104.55±1.92
S10	22.54±0.58	23.65±0.65	360.84±8.70	13.03±0.40	84.14±1.96
Average	22.86±0.96	27.26±1.50.01	409.04±10.78	6.05±1.31	93.34±6.80

S: Sediment sample code Ra_{eq}: Radium equivalent

Table 3. Specific activities of radionuclides in water samples

Sample code	Radionuclides specific activities (Bql^{-1})				Ra_{eq} (Bql^{-1})
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	
W1	2.66±1.16	< 0.91	21.30±2.80	< 0.20	4. 30±1.08
W2	< 1.90	< 3.83	< 1.92	< 0.53	< 8.30
W3	< 1.21	< 2.10	30.47±2.90	0.16±0.10	2.34±0.13
W4	< 2.10	< 4.35	50.98±6.70	0.31±0.25	3.93±0.20
W5	< 1.97	< 4.11	12.57±3.57	< 0.81	0.96±0.15
W6	< 1.17	< 2.13	35.25±2.78	< 0.30	2.71±0.13
W7	< 1.28	< 1.47	< 3.17	< 0.26	< 3.62
W8	< 1.75	< 3.64	19.73±3.37	< 0.40	1.52±0.14
W9	< 2.04	< 4.16	45.20±6.34	< 0.34	3.48±0.19
W10	< 1.64	< 3.70	< 6.45	< 0.41	< 7.43
Average	---	----	21.55±11.54	≤0.32±0.27	1.92±1.10

W: Water sample code Ra_{eq}: Radium equivalent

Table 4. Radiological parameters of sediment samples

Sample code	D ($nGyh^{-1}$)	$AGED$ (μSvy^{-1})	I_γ	H_{in}	H_{ex}	$AEDE_{out}$ ($mSvy^{-1}$)	$ELCR_{out}$ ($\times 10^{-3}$)
S1	43.52±1.16	301.64±7.71	0.67	0.30	0.24	0.05	0.19
S2	43.30±1.08	300.04±6.72	0.67	0.30	0.24	0.05	0.19
S3	43.87±1.17	303.21±7.41	0.67	0.32	0.24	0.05	0.19
S4	49.23±1.12	337.25±7.54	0.75	0.34	0.27	0.06	0.21
S5	48.80±1.14	335.13±7.66	0.75	0.34	0.27	0.06	0.21
S6	49.20±1.29	336.25±8.54	0.75	0.34	0.28	0.06	0.21
S7	43.07±1.20	297.71±7.82	0.66	0.30	0.24	0.05	0.18
S8	45.16±0.93	290.33±6.17	0.65	0.27	0.23	0.06	0.19
S9	50.76±1.07	346.77±7.12	0.78	0.34	0.28	0.06	0.22
S10	40.87±0.88	281.80±5.88	0.63	0.30	0.23	0.05	0.18
Average	45.78±2.82	313.01±23.07	0.70	0.32	0.25	0.06	0.20

S: Sediment sample code AGED: Annual gonadal dose equivalent ELCR: Excess lifetime cancer risk

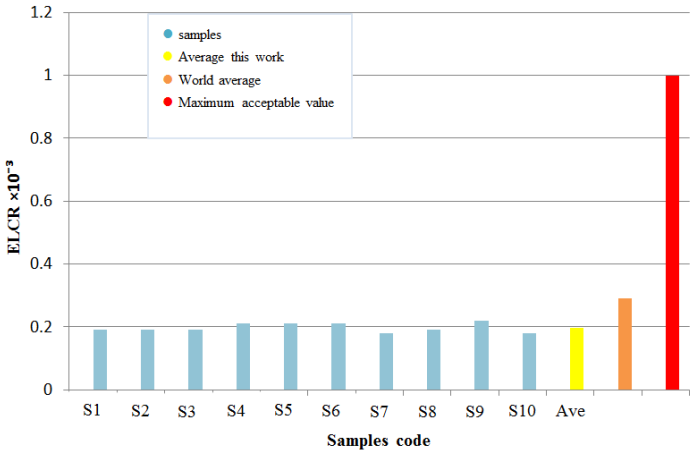

Figure 2. Excess lifetime cancer risk (ELCR) of sediment samples in comparison with maximum acceptable value

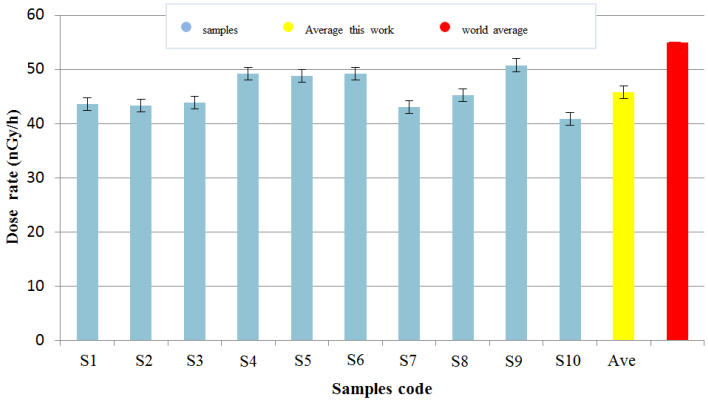

Figure 3. Absorbed dose rate in air in comparison with world average

Discussion

Specific activity of ^{226}Ra in sediment samples ranged from 14.44 to 26.58 Bqkg^{-1}, and it was lower than the minimum detectable activity in all water samples with the exception of W1. Radium content in the inlet water (W1) was measured at 2.66 Bqkg^{-1}. Average specific activity of ^{226}Ra in sediment samples obtained as 22.86 Bqkg^{-1}, which is lower than the global average [12].

The US Environmental Protection Agency has established Maximum Contamination Level for radium in drinking water as 0.185 Bql^{-1}; therefore, in this context, the inlet water exceeds the critical value [17].

Specific activities of ^{232}Th and ^{40}K for sediment samples ranged from 22.78 to 34.56 and from 360.74 to 447.79, with average values of 27.26 and 409.04 in Bqkg^{-1}, respectively, which in the case of ^{232}Th is lower and for ^{40}K with the same as the global average.

For water samples, the specific activity of ^{232}Th was below the detectable limit, and for ^{40}K it varied from <3.17 to 50.98 in Bqkg^{-1}. Global average values of ^{226}Ra, ^{232}Th, and ^{40}K in soil were reported by UNSCAER as 35, 45, and 400 in Bqkg^{-1}, respectively [12]. Radium equivalent for sediment samples ranged from 84.14 to 104.55 with an average of 93.34 in Bqkg^{-1}, and for water samples, it ranged from 0.96 to 3.93 with an average of 1.92 in Bqkg^{-1}. Average specific activity for ^{137}Cs in sediment samples was measured at 6.05 Bqkg^{-1}.

The results of this study indicated that the sediments in this region were contaminated with cesium radionuclides, which is similar to other parts of Markazi province, Iran, and Karbala, Iraq [18-20]. The H_{in} and H_{ex} in sediment samples ranged from 0.27 to 0.34 and from 0.23 to 0.28, respectively, which for all the samples was less than one. In addition, the world average values for H_{in} and H_{ex} were reported as 0.43 and 0.32, respectively [12].

Topographic map of the distribution of radium and cesium radionuclides in sediment samples are shown in figures 4 and 5. This map shows that the radium precipitates at distances close to the inlet of water (S1) and cesium at greater distances. In addition, the topographic map of the cesium distribution in the water sample is shown in Figure 6, which indicates an increase in its pollution in the medium distance. Figure 7 depicts a gamma distribution map of the absorbed dose in the air in this area, which shows an almost increasing amount near the water inlet.

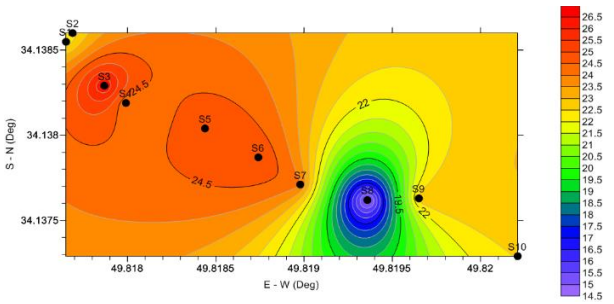

Figure 4. ^{226}Ra distribution map in sediment at the inlet from Wastewater Treatment Plant to Meighan Wetland

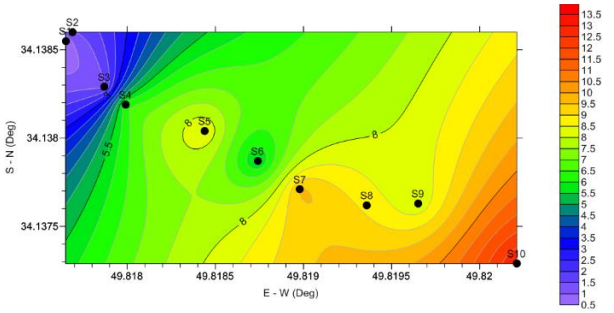

Figure 5. ^{137}Cs distribution map in sediment at the inlet of Wastewater Treatment Plant to Meighan Wetland

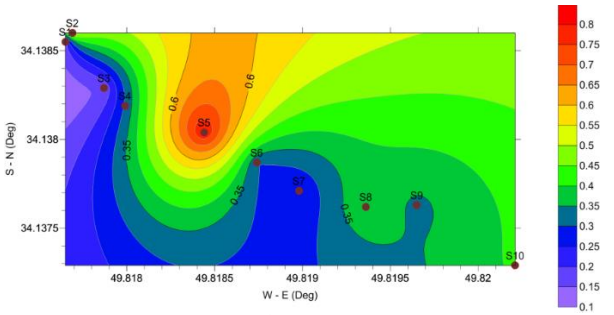

Figure 6. Distribution map of ^{137}Cs in water in the study area

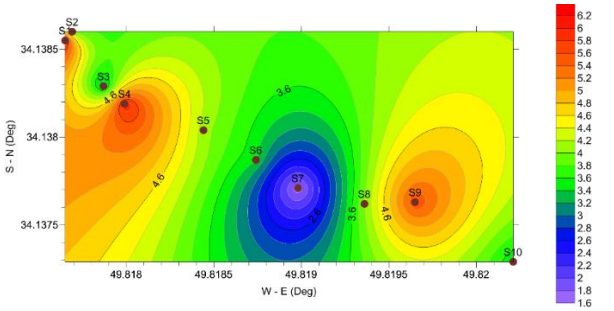

Figure 7. Gamma dose distribution map absorbed by the air in the study area

To compare the obtained results in this study, Table 5 tabulates the reported measurements for the specific activities of radionuclides in some countries. The results of this study are in good consistency with the other mentioned findings and the global average. The results of this study also revealed that the amount of radionuclides in the soil of Gorgan, Iran, was higher than other reported measurements. The exception is iron ore for ^{226}Ra radionuclide.

Table 5. Comparing the results of specific activities of natural radionuclides in this study with soil of some Countries

Country	^{226}Ra	^{232}Th	^{40}K	Reference
China (around coal power plant)	130-241	215-321	811-1571	[21]
Egypt	4-48	8-50	16-187	[22]
France	9-62	16-55	120-1026	[23]
Serbia	26-48	30-58	410-680	[24]
Spain	9-14	11-16	220-460	[25]
Algeria	-	6-32	56-607	[26]
Italy	-	31-37	410-475	[27]
Iraq	29-38	23-46	185-550	[20]
Iran (Gorgan)	30-63	54-84	826-1035	[28]
Iran (Iron ore)	9-271	5-61	25-800	[29]
Iran (around oil power plant)	19-43	25-54	230-700	[19]
Iran (around Arak)	37.27	43.18	604.05	[30]
Iran	14-26	23-34	360-447	This study

Conclusion

This study examined the radioactivity of sediment and water samples at the inlet of purified water from Arak Wastewater Treatment Plant to international Meighan Wetland. The average values of specific activities of radium and thorium in sediment samples were lower than the global average and average value in the soil of the region [30]. However, the specific activity of ^{40}K was equal to the global average. Sediments similar to the soil surrounding the areas were contaminated with artificial radionuclide ^{137}Cs. Radiological maps show that ^{226}Ra precipitated more frequently near the inlet (S1), and ^{137}Cs were deposited at distant points. It seems that ^{226}Ra and ^{232}Th have precipitated in the relaxation basins of the wastewater treatment plant, and fewer of them enter the wetland. Radiological parameters are within the acceptable range and do not pose a threat to the local population.

Acknowledgment

Hereby, the authors would like to extend their gratitude to the Department of Research Council of Arak University of Iran for their financial support for this study.

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