Determination of the activity concentration levels of the artificial radionuclide ^{137}Cs in soil samples collected from Qatar using high-resolution gamma-ray spectrometry

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The goal of this study was to establish the first baseline measurements for radioactivity concentration of the artificial radionuclide ^{137}Cs in soil samples collected from the Qatari peninsula. The work focused on the determination of the activity concentrations levels of man-made radiation in 129 soil samples collected across the landscape of the State of Qatar. All the samples were collected before the most recent accident in Japan, “the 2011 Fukushima Dai-ichi nuclear power plant accident”. The activity concentrations have been measured via high-resolution gamma-ray spectrometry using a hyper-pure germanium detector situated in a low-background environment with a copper inner-plated passive lead shield. A radiological map showing the activity concentrations of ^{137}Cs is presented in this work. The concentration was found to range from 0.21 to 15.41 Bq/kg. The highest activity concentration of ^{137}Cs was observed in sample no. 26 in North of Qatar. The mean value was found to be around 2.15 ± 0.27 Bq/kg. These values lie within the expected range relative to the countries in the region. It is expected that this

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contamination is mainly due to the Chernobyl accident on 26 April 1986, but this conclusion cannot be confirmed because of the lack of data before this accident.

Keywords: Activity concentration; Artificial radionuclide; HPGe detector; Gamma spectrometry; ^{137}Cs .

1. Introduction

The radionuclides released from nuclear power plant accidents can disperse into the atmosphere and travel at a great altitude and eventually fall to the earth. The rain washes down the airborne radionuclides, and they may accumulate in selective areas in the environment according to the soil type. The artificial radionuclide ^{137}Cs is strongly adsorbed by soil particles with a high content of clay, as are the other natural radionuclides.¹ It is also noticed by Q. He and H. Kato²⁻³ that the relaxation mass depth increases as the content of clay in the soil increases,⁴ which is due to their high composition of aluminum silicates (anion) that easily adsorb radionuclide cations on their surface. Hence, since clay particles are characterized as fine sized grains, their surface will have also greater specific areas and accordingly will adsorb more.⁴

Although migration of ^{137}Cs is relatively low in soil,¹ it has a relatively long half-life that makes long-term monitoring of the activity concentration in river water, ground water and vegetation essential, especially in an area known for its elevated levels of Cs.¹ The investigated samples in this study were collected before the 2011 Fukushima Dai-ichi nuclear power plant accident as a part of a comprehensive study aimed at establishing a baseline map for the natural and artificial radionuclides across the landscape of the State of Qatar.

A baseline value for the radioactivity concentration in Qatari soil is primary data to further estimate the natural background radiation dose and annual effective dose equivalent for individuals living in the State of Qatar. Such data can also be used to assess the biological effect of natural radiation and its behaviour in the environment.

Cs has a biological half-life of about 110 days.⁵ Its concentration in biological materials is relative to the K content, and it can be absorbed readily and uniformly in the human body. Uptake in fish depends on the potassium ion content in water.⁵ It is chemically similar to potassium (Alkali metal), and it resembles potassium in the human body. Cs is more soluble in marine water than fresh water.⁶ It distributes in the whole body similarly and does not deposit in a specific organ.⁵

This study will be the reference map to detect any significant future release of the artificially created fission product ^{137}Cs into the environment, since relatively high amounts of radioactive cesium were detected following the most recent NPP accident in Japan using an atmospheric transport model, and measurement data from 43 stations worldwide is estimated to be around 35 PBq.⁷ Other studies have reported ~15 PBq.⁸

2. Experimental Arrangements

2.1. Soil sampling and preparation

A systematic sampling plan, which has been widely accepted and potentially increases the accuracy of soil tests, was followed in this study. It was also preferred for the sample mapping procedures.⁹ However, upon the sampling field visit and following the preliminary analysis of the results of this study, the most suitable approach was found to be a combination of systematic grid sampling and systematic random sampling. This was due to restrictions in collecting the samples from some of the grid points. The regular distance between sampling points was chosen to be 5 km. Thus soil samples were collected from sampling points located at regular intervals of a 5 per 5 km² square grid using the Qatar National Grid (QNG) system. Since the State of Qatar has a total area of 11,437 km², the 129 soil samples that were collected in this study across the country were enough to cover the whole landscape.

Approximately 1.5–2 kg of soil was collected at ~ 5 to 15 cm depth levels at each sample point. The upper sandy surface was avoided. The correlation between the activity concentration of the radionuclides and sample's depth as well as grain size were investigated in another study by the author.¹⁰

After collection, each sample was transferred to a 2 mm sieve fitted in a collecting pan. The post-sieved samples were then put in labelled polyethylene bags, sealed, packed in a box, and shipped to the UK for analysis. The final sample preparation and all the gamma-ray measurements to determine the sample radioactivity were performed in the low-background radiation detection laboratories of the Centre for Nuclear and Radiation Physics, situated within the Department of Physics at the University of Surrey.

Prior to final measurement in the laboratories in Guildford, all of the samples were placed in a drying oven. The drying temperature was set to 60°C for 24 hours to ensure that any significant moisture was removed from the samples. A 500 µm mesh was then used to sieve the samples that were then weighed and transferred to 500 ml labelled Marinelli beakers.

2.2. Detector calibration

The soil samples in the current study were analyzed using a high-resolution, low-background gamma-ray spectrometry system based on a coaxial hyper-pure germanium detector (HPGe). A crystal of 50 mm diameter was operated under a high voltage bias of +3000 V (DC).

The gamma-ray spectra that were analyzed were created through converting the event energy into a pulse height spectrum. The signal processing was done by connecting the detector to a preamplifier and a standard spectroscopy shaping amplifier (Canberra 2022). The selected shaping time was 4 µs. The applied coarse gain and fine gain were 10 and 0.5, respectively. Finally, the resultant spectral data was analyzed directly using Canberra Genie software.¹¹

The performance of the HPGe detector used in the current work was characterized by the energy calibration and its absolute full-energy peak detection efficiency. The initial energy calibration of the detector was carried out using full-energy peaks from a standard source (^{152}Eu) with an activity of 3.02 kBq. The spectrum was acquired for 1200 s. The range of energies (121.78 keV, 344.27 keV, 778.90 keV, 1112.05 keV and 1407.92 keV) was selected to calibrate the initial energy scale of the program.¹² In order to avoid any drift in the later soil sample measurements, the system was recalibrated on a weekly basis during the sample measurements.

The absolute efficiency calibration of the detector was carried out using full-energy peaks from standard sources of NG3 (a mixed source containing ^{241}Am , ^{57}Co , ^{60}Co , ^{85}Sr , ^{88}Y , ^{109}Cd , ^{137}Cs , ^{139}Ce , and ^{203}Hg). The sources were placed surrounding the germanium detector with the radionuclides dispersed in gel matrices within Marinelli beakers of geometries identical to that of the evaluated samples. The activities of the sources ranged from 6×10^2 to 1.5×10^4 Bq. The activity of ^{137}Cs was 2.68×10^3 Bq for an active volume of 550 ml and was spread homogeneously in a gel matrix of density 1.6 g/cm^3 . This was taken to be representative of the density of the samples measured, and therefore, corrections for gamma-ray self-attenuation within the samples were accounted for in the initial efficiency measurements using this Marinelli-housed source. The efficiency calibration spectrum was acquired for 86,400 s (=1 day). A range of discrete gamma-ray energies were covered using this standard source. A Marinelli beaker with the same geometry and filled with de-ionized water was used on a weekly basis during the measurements period to determine the background spectrum observed by the germanium detector. The counting time of the ambient background spectrum was also 86,400 s and was used to determine the minimum detectable activity (MDA) in the system.

2.3. Samples analysis

The samples were placed directly over the front face of the detector. The acquisition time for each sample was 1 day. The counting geometry of the samples and the standard sources used for efficiency calibration were kept constant. The gamma-ray peak associated with decay from ^{137}Cs at 662 keV was used to determine the activity concentrations for this nucleus. Background contributions were subtracted from the peak areas for the measured samples.

2.4. Measurements of activity concentration

The activity concentration of the ^{137}Cs radionuclide found in the soil samples was determined using Eq. (1) and expressed in Bq/kg:

$$Ac = C_{net} / \gamma \times \varepsilon(E_\gamma) \times m, \quad (1)$$

where C_{net} are the net peak counts, γ is the absolute gamma decay intensity for the specific energy photopeak, $\varepsilon(E_\gamma)$ is the absolute photopeak efficiency of the germanium detector at this energy, and m is the mass of the sample in kg.

3. Results and Discussion

One hundred and twenty nine soil samples were analyzed in order to estimate the levels of the activity concentrations of ^{137}Cs . The background spectrum was subtracted from all the spectra of the analyzed samples in the current work.

^{137}Cs exists in soil naturally only in trace amounts following the spontaneous fission of ^{238}U . Any measured values above the MDA in the current work would therefore arise from the artificially produced fission of ^{238}U . Nuclear accidents, bomb tests and nuclear weapons are the origins of ^{137}Cs contamination. Hence since the investigated samples in the current study were collected before the 2011 Fukushima Dai-ichi nuclear power plant accident, it is expected that this contamination is mainly due to the Chernobyl accident of 26 April 1986. The reported released amounts of ^{137}Cs during this accident were about 9 PBq.¹³ Since it has a half-life of about 30 years, it can be traced for several years. Table 1 shows all the results of A_c in Bq/kg resulting from this study. Around 30 samples show no indication of ^{137}Cs contamination below the MDA. The measured activity concentrations of ^{137}Cs across Qatar ranged from 0.21 to 15.41 Bq/kg. The mean value was found to be 2.15 ± 0.27 Bq/kg. The highest activity concentration of ^{137}Cs was observed in sample no. 26 in the north of Qatar. The sample collected from the same sampling point (sample 26) for a depth of 0–5 cm and investigated in the earlier study (Ref. 10) was reported to have a lower level of activity concentration among the other five sample points investigated in the same study; it was about $11.57 \pm 0.54_{\text{Stat.}}$ and $0.84_{\text{Syst.}}$.¹⁰ Unlike the other samples that have shown expected levels inversely proportional with depth, they were mostly in correlation and decreasing with depth and consistent with scientific expectations. This can be attributed to many key factors such as soil composition in this area, more clay content in sub-surface soil, and effects of metrological factors and rain wash off in the Northern areas of Qatar.

Figure 1 shows the histogram of the activity concentration of the artificial radionuclide ^{137}Cs in the Qatari soil. Figure 2 shows the distribution of the activity concentration of ^{137}Cs for the total number of investigated samples on the map of Qatar. Taking into account the ^{137}Cs decay half-life of 30.2 y and the sampling depth in the current work ($5 \approx 15$ cm), which is a deeper layer and dilutes the ^{137}Cs concentrations,¹⁴ the resultant values are lower than the observed values for those reported in other countries, such as the reported values in Jordan in 2002, which ranged from 7.75 to 576 Bq/kg for a sampling depth of 0–2 cm and a mean value of 88 ± 31 Bq/kg.¹⁵ They are however comparable to those determined in Algeria in 1996 which reported a mean value of 4.2 Bq/kg.¹⁶ Only in the areas where low levels of ^{137}Cs were found, an agreement was observed between our results in the current work and another study conducted in 2012 on natural radioactivity levels in Qatari soil after our study,¹⁷ but it is not clear whether their investigated samples were collected before or after the 2011 Fukushima Dai-ichi nuclear power plant accident.

Table 1. Activity concentrations of ¹³⁷Cs in Bq/kg resultant from the 129 soil samples investigated in this study.

#	Sample no.	Grids (X,Y)	Density (g/cm ³)	¹³⁷ Cs	Stat	Syst	#	Sample no.	Grids (X,Y)	Density (g/cm ³)	¹³⁷ Cs	Stat	Syst
1	10	195000 485000	1.38 ± 0.05	*			33	162	195000 435000	1.50 ± 0.05	12.27 ± 0.56	0.89	
2	12	205000 485000	1.32 ± 0.04	6.10 ± 0.48	0.44		34	164	205000 435000	1.57 ± 0.05	1.11 ± 0.17	0.08	
3	X14	215000 485000	1.78 ± 0.11	7.84 ± 0.90	1.4		35	166	215000 435000	1.59 ± 0.05	15.03 ± 0.66	1.1	
4	26	185000 475000	1.52 ± 0.05	15.41 ± 0.67	1.1		36	168	225000 435000	1.65 ± 0.05	1.54 ± 0.21	0.11	
5	28	195000 475000	1.25 ± 0.04	14.58 ± 0.62	0.80		37	169	230000 435000	1.45 ± 0.05	1.32 ± 0.19	0.10	
6	30	205000 475000	1.38 ± 0.05	8.88 ± 0.49	0.65		38	X194	165000 425000	1.44 ± 0.05	*		
7	32	215000 475000	1.28 ± 0.04	4.21 ± 0.30	0.31		39	X196	175000 425000	1.36 ± 0.05	*		
8	X50	175000 465000	1.72 ± 0.08	3.69 ± 0.22	0.25		40	198	185000 425000	1.44 ± 0.05	0.61 ± 0.23	0.03	
9	52	185000 465000	1.60 ± 0.05	3.71 ± 0.29	0.27		41	200	195000 425000	1.56 ± 0.05	*		
10	54	195000 465000	1.44 ± 0.05	3.10 ± 0.36	0.23		42	202	205000 425000	1.61 ± 0.05	0.71 ± 0.17	0.05	
11	56	205000 465000	1.43 ± 0.05	0.32 ± 0.15	0.02		43	204	215000 425000	1.66 ± 0.05	0.88 ± 0.16	0.06	
12	58	215000 465000	1.38 ± 0.05	8.83 ± 0.52	0.64		44	206	225000 425000	1.49 ± 0.05	1.65 ± 0.23	0.12	
13	60	225000 465000	1.68 ± 0.06	2.44 ± 0.16	0.18		45	X208	235000 425000	1.35 ± 0.05	2.74 ± 0.25	0.12	
14	X62	235000 465000	1.38 ± 0.05	0.86 ± 0.18	0.06		46	X228	155000 415000	1.31 ± 0.05	1.12 ± 0.16	0.07	
15	84	175000 455000	1.44 ± 0.05	0.43 ± 0.19	0.03		47	230	165000 415000	1.51 ± 0.05	0.39 ± 0.15	0.03	
16	86	185000 455000	1.64 ± 0.05	0.51 ± 0.16	0.04		48	232	175000 415000	1.45 ± 0.05	1.02 ± 0.08	0.07	
17	88	195000 455000	1.31 ± 0.05	4.76 ± 0.37	0.30		49	234	185000 415000	1.59 ± 0.05	1.53 ± 0.12	0.11	
18	90	205000 455000	1.57 ± 0.05	1.37 ± 0.21	0.10		50	236	195000 415000	1.44 ± 0.05	*		
19	92	215000 455000	1.44 ± 0.05	5.44 ± 0.32	0.40		51	238	205000 415000	1.47 ± 0.05	0.61 ± 0.19	0.04	
20	94	225000 455000	1.52 ± 0.05	0.66 ± 0.16	0.05		52	240	215000 415000	1.34 ± 0.05	1.41 ± 0.21	0.09	
21	96	235000 455000	1.26 ± 0.04	6.25 ± 0.37	0.45		53	242	225000 415000	1.43 ± 0.05	0.86 ± 0.24	0.03	
22	X100	170000 445000	1.51 ± 0.05	1.00 ± 0.14	0.07		54	266	155000 405000	1.69 ± 0.06	1.28 ± 0.22	0.09	
23	X120	175000 445000	1.33 ± 0.05	0.84 ± 0.17	0.06		55	268	165000 405000	1.20 ± 0.04	1.14 ± 0.16	0.06	
24	122	185000 445000	1.52 ± 0.05	0.29 ± 0.17	0.02		56	270	175000 405000	1.34 ± 0.05	*		
25	124	195000 445000	1.45 ± 0.05	*			57	272	185000 405000	1.43 ± 0.05	*		
26	126	205000 445000	1.56 ± 0.05	1.60 ± 0.20	0.12		58	274	195000 405000	1.66 ± 0.06	*		
27	128	215000 445000	1.26 ± 0.04	0.58 ± 0.23	0.04		59	276	205000 405000	1.58 ± 0.05	0.34 ± 0.04	0.02	
28	130	225000 445000	1.47 ± 0.05	0.87 ± 0.17	0.06		60	278	215000 405000	1.60 ± 0.05	*		
29	132	235000 445000	1.29 ± 0.04	1.06 ± 0.20	0.07		61	280	225000 405000	1.55 ± 0.05	0.49 ± 0.06	0.03	
30	X156	165000 435000	1.34 ± 0.05	0.48 ± 0.20	3.86		62	X282	235000 405000	1.59 ± 0.05	2.10 ± 0.23	0.15	
31	158	175000 435000	1.27 ± 0.04	1.06 ± 0.21	0.07		63	304	155000 395000	1.60 ± 0.06	1.83 ± 0.22	0.07	
32	160	185000 435000	1.47 ± 0.05	*			64	306	165000 395000	1.43 ± 0.05	0.31 ± 0.18	0.02	

#	Sample no.	Grids (X,Y)	Density (g/cm ³)	¹³⁷ Cs	Stat	Syst	#	Sample no.	Grids (X,Y)	Density (g/cm ³)	¹³⁷ Cs	Stat	Syst
65	308	175000 395000	1.51 ± 0.05	2.58 ± 0.24	0.14		98	446	235000 365000	1.50 ± 0.05	1.12 ± 0.28	0.08	
67	312	195000 395000	1.52 ± 0.05	0.72 ± 0.19	0.05		99	X472	155000 355000	1.68 ± 0.06	0.23 ± 0.12	0.02	
68	314	205000 395000	1.50 ± 0.05	0.90 ± 0.20	0.07		100	474	165000 355000	1.77 ± 0.06	2.63 ± 0.22	0.19	
69	316	215000 395000	1.60 ± 0.05	0.93 ± 0.28	0.07		101	476	175000 355000	1.46 ± 0.05	*		
70	318	225000 395000	1.37 ± 0.05	*			102	478	185000 355000	1.53 ± 0.05	0.51 ± 0.19	0.04	
71	320	235000 395000	1.64 ± 0.06	*			103	480	195000 355000	1.38 ± 0.05	0.40 ± 0.18	0.01	
72	346	155000 385000	1.74 ± 0.06	0.58 ± 0.14	0.04		104	484	215000 355000	1.71 ± 0.06	3.34 ± 0.23	0.24	
73	348	165000 385000	1.64 ± 0.05	0.70 ± 0.16	0.05		105	486	225000 355000	1.67 ± 0.06	3.25 ± 0.25	0.24	
74	350	175000 385000	1.61 ± 0.05	*			106	488	235000 355000	1.76 ± 0.06	0.95 ± 0.17	0.07	
75	352	185000 385000	1.62 ± 0.05	0.82 ± 0.17	0.06		107	508	165000 345000	1.77 ± 0.06	*		
76	354	195000 385000	1.58 ± 0.05	*			108	510	175000 345000	1.69 ± 0.06	0.58 ± 0.27	0.04	
77	356	205000 385000	1.63 ± 0.05	1.41 ± 0.18	0.10		109	512	185000 345000	1.60 ± 0.05	1.33 ± 0.20	0.10	
78	358	215000 385000	1.67 ± 0.06	*			110	514	195000 345000	1.59 ± 0.05	0.87 ± 0.18	0.06	
79	360	225000 385000	1.49 ± 0.05	0.27 ± 0.18	0.02		111	516	205000 345000	1.46 ± 0.05	0.63 ± 0.19	0.05	
80	362	235000 385000	1.51 ± 0.05	0.71 ± 0.18	0.05		112	518	215000 345000	1.58 ± 0.05	1.27 ± 0.20	0.09	
81	388	155000 375000	1.36 ± 0.05	0.58 ± 0.29	0.04		113	520	225000 345000	1.71 ± 0.06	0.50 ± 0.16	0.04	
82	390	165000 375000	1.48 ± 0.05	0.89 ± 0.29	0.06		114	542	165000 335000	1.68 ± 0.06	0.23 ± 0.12	0.02	
83	392	175000 375000	1.58 ± 0.05	0.36 ± 0.14	0.03		115	544	175000 335000	1.71 ± 0.06	2.29 ± 0.20	0.17	
84	394	185000 375000	1.53 ± 0.05	*			116	546	185000 335000	1.75 ± 0.06	*		
85	396	195000 375000	1.63 ± 0.05	0.88 ± 0.16	0.06		117	548	195000 335000	1.58 ± 0.05	1.60 ± 0.23	0.12	
86	398	205000 375000	1.41 ± 0.05	0.42 ± 0.22	0.03		118	550	205000 335000	1.77 ± 0.08	*		
87	400	215000 375000	1.63 ± 0.05	*			119	552	215000 335000	1.39 ± 0.05	*		
88	402	225000 375000	1.54 ± 0.05	1.08 ± 0.19	0.08		120	554	225000 335000	1.69 ± 0.06	0.21 ± 0.13	0.02	
89	404	235000 375000	1.61 ± 0.05	1.26 ± 0.19	0.09		121	X574	165000 325000	1.33 ± 0.05	*		
90	X430	155000 365000	1.01 ± 0.04	1.60 ± 0.22	0.10		122	576	175000 325000	1.80 ± 0.06	*		
91	432	165000 365000	1.58 ± 0.06	0.32 ± 0.22	0.01		123	578	185000 325000	1.46 ± 0.05	0.60 ± 0.18	0.04	
92	434	175000 365000	1.65 ± 0.05	0.25 ± 0.19	0.02		124	580	195000 325000	1.71 ± 0.06	*		
93	436	185000 365000	1.31 ± 0.04	1.00 ± 0.27	0.07		125	582	205000 325000	1.64 ± 0.06	4.87 ± 2	0.31	
94	438	195000 365000	1.50 ± 0.05	0.67 ± 0.16	0.04		126	584	215000 325000	1.67 ± 0.06	*		
95	440	205000 365000	1.71 ± 0.06	0.34 ± 0.15	0.02		127	608	185000 315000	1.80 ± 0.06	*		
96	442	215000 365000	1.68 ± 0.06	*			128	610	195000 315000	1.40 ± 0.05	2.02 ± 0.30	0.15	
97	444	225000 365000	1.51 ± 0.05	0.90 ± 0.23	0.07		129	612	205000 315000	1.73 ± 0.06	*

Fig. 1. A histogram of activity concentration of the artificial radionuclide ^{137}Cs in Qatari soil.

Fig. 2. A radiological map of Qatar shows the distribution of activity concentration of ^{137}Cs in the current work.

4. Conclusions

The activity concentration of ^{137}Cs measured in Qatar in the current work ranged from 0.21 to 15.41 Bq/kg. Most of the relatively higher results were collected from the North of Qatar, which can be related to the clay content in the soil composition of those areas. The mean value was found to be 2.15 ± 0.27 Bq/kg. The highest activity concentration of ^{137}Cs was observed in sample no. 26 in North West Qatar. It is expected that this contamination is mainly due to the Chernobyl accident on 26 April 1986. This conclusion cannot be confirmed because of the lack of data before this accident.

The resultant values in this study are not directly comparable with the observed values for those reported in other regional countries such as the reported values in Jordan in 2002¹⁵ for a sampling depth of 0–2 cm, but they were in good agreement with those determined in Algeria in 1996 which reported a mean value of 4.2 Bq/kg.¹⁶ It is suggested that the values reported in the current work are lower than those reported in other countries because of the variation in the characteristics of the soil as well as the metrological factors at the time of deposition.¹⁴

The radiological map of the activity concentration of the radionuclide ^{137}Cs has been determined using soil at depth ranges of 5–15 cm. Since ^{137}Cs is likely to be lower at these depths than the activity concentration in topsoil samples, it is suggested that a further study be done on the radioactivity concentrations of ^{95}Zr , ^{95}Nb , ^{134}Cs , ^{137}Cs , ^{90}Sr , and ^{241}Am with a correlation to soil composition and fractions, particularly following the recent (March 2011) Fukushima accident, in order to address this issue in a more statistically significant picture, especially in the sampling points that have reported higher levels of ^{137}Cs than the mean values.

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