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Background: An amount of artificial radionuclide has been released into the environment as fallout, resulting from atmospheric nuclear weapon tests, nuclear accidents such as Chernobyl and together with air currents have polluted the world. Materials and Methods: 37 surface soil samples of Tehran province were collected in the period between June and September 2008, by implementing methods and standard instruments. The concentration of the artificial radionuclides (137Cs) in the soils of Tehran province were determined by gamma spectroscopy (HPGe), and the data were analyzed both quantitatively and qualitatively. The results have been compared with other radioactivity measurements. Results: The concentration of 137Cs found in top soils (0-5 cm), in the depth of (12.5-17.5 cm) and in the depth of (27.5- 32.5 cm), ranged from 0.29-28.82 0.3-19.81 Bq.kg⁻¹, 0.8-7.43 Bq.kg⁻¹, Bq.kg⁻¹, respectively. The total average of 137Cs activity, in the above depths was, 11.30, 4.87 and 1.8 Bq.kg⁻¹, respectively. Conclusion: Considering the obtained results from high average values of 137Cs in Europe and its relation to Chernobyl accident, and the higher average concentration of ¹³⁷Cs in Tehran province. can also be attributed to Chernobyl accident due to the proximity of Iran to the place of the accident's occurrence. Iran. J. Radiat. Res., 2009; 7 (3): 141-149

Keywords: Cs-137, soil sample, gamma spectroscopy, HPGe, Tehran province.

INTRODUCTION

Due to destructive interference of human being in his environment, artificial and natural radionuclides are found in the nature. An amount of artificial radionuclides have been released into the environment as fallout resulting from atmospheric nuclear weapon tests, nuclear accidents such as Chernobyl, and together with air currents have polluted the world ⁽¹⁾.

Among the various radionuclides released to the environment, we have focused on the distribution of ¹³⁷Cs, because of its long half life (about 30.17 year), which will remain in the environment with all its destructive effects ⁽²⁾.

Deposition of radioactive fallout including ¹³⁷Cs at any site is related to factors such as, latitude, precipitation and local topography. ¹³⁷Cs is strongly absorbed and retained by soil particles and it can enter into the diet of human beings, and other leaving creatures. Maintaining ¹³⁷Cs in surface layers of clay soil is considerable ^(3, 4). Therefore, the access to distribution of ¹³⁷Cs in Tehran province soils has been the main objective of this research.

In this study, points of sampling were chosen by VSP (Visual Sample Plan) software, GPS (Global Positioning System) and considering of Tehran province dominant wind current. The chosen points were marked by a digital map and their coordinates were entered in to GPS system. The points on the Tehran province map, as shown in figure 1 were marked and used while collecting soils. Tehran province is approximately located between 35°:43' N latitude and 51°:25' E longitude. Table 1 shows the geographical coordinates of the sampling point and the labels of different samples of different townships.

MATERIALS AND METHODS

Sampling procedure

Thirty-seven soil samples were collected from several regions all over Tehran province. The samples were collected from the depths of (0-5 cm), (12.5- 17.5 cm) and (27.5- 2.5 cm), using Template method ⁽⁵⁾.

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| Sample code | Site | GPS coordinates | Alt(m) |
|-------------|---------------------|-------------------|--------|
| S001 | Lashgarak (I) | 51.70150-35.86832 | 2800 |
| S002 | Lashgarak (II) | 51.53354-35.96699 | 2800 |
| S003 | Rineh | 52.07669-35.85727 | 3000 |
| S004 | Roodehen | 51.75907-35.73762 | 1866 |
| S005 | Sarbandan | 52.32778-35.64694 | 2665 |
| S006 | Arjmand (I) | 52.62531-35.74773 | 2110 |
| S007 | Arjmand (II) | 52.55092-35.75814 | 2100 |
| S008 | Pirdeh | 52.81778-35.68754 | 2530 |
| S009 | Shangoldeh | 52.42197-35.78739 | 2400 |
| S010 | Sefidargoleh | 53.09712-35.79046 | 2945 |
| S011 | Yam | 51.78658-35.36978 | 1000 |
| S012 | Javad Abad | 51.69475-35.19797 | 836 |
| S013 | Varamin | 51.58461-35.45075 | 977 |
| S014 | Arad | 51.76667-35.31525 | 857 |
| S015 | Asara | 51.11824-36.09546 | 3500 |
| S016 | Gajereh | 51.31842-36.05314 | 2765 |
| S017 | Mahdasht (I) | 50.90134-35.52739 | 1228 |
| S018 | Mahdasht (II) | 50.96441-35.73711 | 1228 |
| S019 | Sharghe Tehran | 51.67856-35.70783 | 1600 |
| S020 | Lavasan Bozorg | 51.90361-35.77233 | 2400 |
| S021 | Shahryar | 51.16330-35.64164 | 1100 |
| S022 | Robat karim | 51.09011-35.46894 | 1100 |
| S023 | Tehran | 51.42628-35.62172 | 1200 |
| S024 | Eshtehard | 50.45989-35.73117 | 1630 |
| S025 | Gomrokan | 50.69038-35.61284 | 1227 |
| S026 | Karaj (Part 2) (I) | 50.77378-35.80309 | 1257 |
| S027 | Karaj (Part 2) (II) | 50.86556-35.99667 | 1675 |
| S028 | Hashtgerd | 50.74352-35.97528 | 2500 |
| S029 | Abyek | 50.64517-36.11414 | 2560 |
| S030 | Fashand | 50.94339-36.21442 | 2500 |
| S031 | Karaj (Part 1) (I) | 51.07237-35.93467 | 2100 |
| S032 | Ghale boland | 51.81147-35.14908 | 797 |
| S033 | Istgah Kavir | 52.15878-35.17394 | 1000 |
| S034 | Tajrish | 51.28223-35.78699 | 1700 |
| S035 | Karaj (Part 2) (II) | 51.24222-35.86058 | 2632 |
| S036 | Gharpooz Abad | 50.29572-35.76375 | 1260 |
| S037 | Gazorkhan | 50.50469-36.41678 | 1645 |

Table 1. Sample identification.

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Figure 1. The map of a part of the Tehran province and the location of the sampling sites.

Sample preparation

The samples were dried in an oven at the temperature of 200°C for about 24h. Then the dried soils were grounded by jaw and planet mill, and passed through a 50 mesh sieve (with 0.2 mm aperture diameter). Then, 950 gr of the homogenized samples were placed in the Marinelli Beaker for spectroscopy and analysis $^{(6)}$.

Spectroscopy and analysis

The samples were measured by using two p-type coaxial High Purity Germanium (HPGe) (EG&ORTEC) gamma spectrometers with relative efficiency (38.5%) and (55%). The resolution of these detectors at 1.33 MeV of 60Co were 1.98 keV and 1.8 keV, respectively. Both detectors were connected to standard electronics and the spectra were accumulated in 4K MCA (multi channel analyzer) Made by IAP (Institute of Applied Physics)). The detectors were surrounded by a 10-cm thick lead shield to reduce the background from the surrounding material and cosmic rays. The detectors were calibrated by using certified gamma-ray standard sources. The counting time was prolonged as necessary to ensure at least 95% of statistical significance. The background spectra was measured frequently under the same

conditions of sample measurements and used to correct the calculated sample activities. The characteristic gamma peak for the determination of ¹³⁷Cs was 661.66 keV.

After electronic instruments adjustment, the energy calibration in the region of 60–3000 KeV was done. The background rate for the mentioned detectors was 2.5 and 2.9 count/second, respectively. Analysis and collecting data for both detectors were fulfilled with OMINGAM Ortec software.

Determining the efficiency of HPGe detectors is the main factor for using of these detectors to specify quantity values of active samples. The use of convenient standard samples is a reliable method to determine the efficiency of the specified regions of energy. Science determining the efficiency of HPGe detectors is confined to the production of standard ¹³⁷Cs samples, to ensure the validity of determining efficiency, the standard samples of ¹⁵²Eu, ²⁴¹Am, ¹³⁷Cs and ¹³³Ba, in the same configuration of the analyzed samples were prepared ⁽⁷⁾, and after making sure of the accuracy of the production of the standard samples, the efficiency of 661.66 keV peak of ¹³⁷Cs was used to determine quantity efficiency. The preparation of these standard samples was accomplished by

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spiking 50 gr of $Al_2O_3^{(3)}$, by a suitable volume of standard solutions and adding this dried spiked Al_2O_3 to the 900 gr of sieved soil (50-mesh), as a complete homogenized compound. After collecting the spectrums of the standard samples, they were processed by the mentioned software. In calculating the efficiency of photo peaks, equation (1) was applied.

$$\varepsilon(\%) = \frac{NetArea}{(A ct_{(Bq)})(B R_{(\%)})(t_{(Sec.)})} \times 100$$
(1)

In this equation, e stands for the efficiency of detector, Act is the activity of the standard isotope at Bq.kg⁻¹, B.R is the branching ratio related to concerned energy at percent, t is the time of collecting spectrum of samples at second, Net Area is the net area under the concerned peaks calculated by software. After fitting convenient functions on efficiency data through software, equations (2) and (3) are for efficiency functions of detectors (38.5%) and (55%), respectively.

Efficiency value at energy (661.66 keV) of 137 Cs, in HPGe (38.5%) and (55%) has been 1.41 and 2.03, respectively. The spectrums were collected between 30000-60000s. The peak (661.66 keV), indicated the presence of 137Cs in samples were the analysis of this isotope could be done.

RESULTS

Determination of radionuclide concentration

Equation (4) was applied for calculating the activities of ¹³⁷Cs in soil samples of Tehran province.

.Where, w is the weight of samples.

Table 2, shows the concentration of ¹³⁷Cs in soil samples of Tehran province. The first column is the sample code and the second column is the depth of collecting samples (0-5), (12.5-17.5) and (32.8-37.5) cm, and the last column is the ¹³⁷Cs concentration in the mentioned depths. For all 37 samples, in the 3 depths, the information were given and for almost all samples the depth profile distribution of ¹³⁷Cs showed a tendency to decrease toward deeper layers. The variations of ¹³⁷Cs concentration in the depths of (0-5), (12.5-17.5) and (32.8-37.5) cm is shown in figure 2, and the shape of frequency distribution of ¹³⁷Cs activity concentration are plotted in figure 3, X and Y axises are ¹³⁷Cs concentration (Bq.kg⁻¹) and Frequency, respectively.



Figure 2. The variations of 137 Cs concentration in the depths of (0-5), (12.5-17.5) and (32.8-37.5) cm.

$$\varepsilon(\%) = a + b \times Ln(E) + \frac{C}{Ln(E)} + d \times (Ln(E))^2 + \frac{e}{(Ln(E))^2} + f \times (Ln(E))^3$$
(2)

$$\varepsilon(\%) = \frac{a + c \times (E)^{0.5} + e \times E + g \times (E)^{1.5} + i \times (E)^2}{1 + b \times (E)^{0.5} + d \times E + f \times (E)^{1.5} + h \times (E)^2}$$
(3)

$$Act_{(Bq/kg)} = \frac{NetArea}{\varepsilon (\%)(-B.R_{(\%)})(t_{(sec)})(W)} \times 100$$
(4)

| Code sample | Depth(cm) | ¹³⁷ Cs(Bq.kg ⁻¹) |
|----------------|-----------|--|
| | 0-5 | 25.75±0.68 |
| S001 | 12.5-17.5 | 2.48±0.19 |
| | 27.5-32.5 | 0.36±0.16 |
| | 0-5 | 11.76±0.91 |
| S002 | 12.5-17.5 | 5.48±0.4 |
| 3002 | 27.5-32.5 | 4.13±0.36 |
| | 42.5-47.5 | <mda*<sup>i</mda*<sup> |
| | 0-5 | 20.47±1.11 |
| S003 | 12.5-17.5 | 14.36±0.39 |
| | 27.5-32.5 | 0.63±0.18 |
| | 0-5 | 22.35±1.23 |
| S004 | 12.5-17.5 | 4.01±0.27 |
| | 27.5-32.5 | 0.19±0.09 |
| | 0-5 | 26.42±0.75 |
| S005 | 12.5-17.5 | 8.12±0.27 |
| | 27.5-32.5 | 0.19±0.84 |
| 7 00 f | 0-5 | 24.79±0.55 |
| S006 | 12.5-17.5 | 3.35±0.38 |
| | 27.5-32.5 | 0.21±0.08 |
| 2007 | 0-5 | 24.08±0.8 |
| S007 | 12.5-17.5 | 5.72±0.36 |
| | 27.5-32.5 | 0.36±0.12 |
| 2000 | 0-5 | 20.03±0.50 |
| S008 | 12.5-17.5 | 3.27±0.3 |
| | 27.5-32.5 | 0.71±0.09 |
| 5000 | 0-5 | 28.82±0.79 |
| S009 | 12.5-17.5 | 6.48±0.34 |
| | 27.5-32.5 | 1.73±0.06 |
| | 0-5 | 14.01±1.96 |
| S010 | 12.5-17.5 | 3.2±0.2 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 1.80±0.24 |
| S011 | 12.5-17.5 | 4.82±0.17 |
| | 27.5-32.5 | 2.98±0.14 |
| | 0-5 | 1.77±0.03 |
| S012 | 12.5-17.5 | 1.16±0.03 |
| | 27.5-32.5 | 1.90±0.11 |
| | 0-5 | 0.76 ± 0.01 |
| S013 | 12.5-17.5 | <mda< td=""></mda<> |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 3.04±0.22 |
| S014 | 12.5-17.5 | 3.78±0.12 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| a c · - | 0-5 | 19.49±0.57 |
| S015 | 12.5-17.5 | 10.75±0.61 |
| | 27.5-32.5 | 12.82±0.62 |
| | 0-5 | 27.47±0.74 |
| S016 | 12.5-17.5 | 12.95±0.53 |
| | 27.5-32.5 | 1.01±0.15 |
| 0017 | 0-5 | 15.27±0.42 |
| S017 | 12.5-17.5 | <mda< td=""></mda<> |
| | 27.5-32.5 | <mda< td=""></mda<> |
| 0010 | 0-5 | 0.64±0.14 |
| S018 | 12.5-17.5 | 3.17 ± 0.21 |
| | 27.5-32.5 | 2.92±0.17 |

Table 2. ¹³⁷Cs concentration in Tehran province soil samples by HPGe (38.5%) and (55%).

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| | 0-5 | 1.62±0.35 |
|------|-----------|---------------------|
| S019 | 12.5-17.5 | <mda< td=""></mda<> |
| | 27.5-32.5 | 0.08±0.18 |
| | 0-5 | 7.55±0.37 |
| S020 | 12.5-17.5 | 6.38±0.21 |
| | 27.5-32.5 | 4.93±0.18 |
| | 0-5 | 1.03±0.13 |
| S021 | 12.5-17.5 | 1.32 ± 0.12 |
| | 27.5-32.5 | 1.4±0.15 |
| | 0-5 | 4.51±0.41 |
| S022 | 12.5-17.5 | 3.47±0.29 |
| | 27.5-32.5 | 2.37±0.22 |
| | 0-5 | <mda< td=""></mda<> |
| S023 | 12.5-17.5 | <mda< td=""></mda<> |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 25.36±0.47 |
| S024 | 12.5-17.5 | 10.65 ± 0.54 |
| | 27.5-32.5 | 1.77±0.3 |
| | 0-5 | 6.92±0.29 |
| S025 | 12.5-17.5 | 1.7±0.13 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 18.55 ± 0.48 |
| S026 | 12.5-17.5 | 13.41±0.49 |
| | 27.5-32.5 | 0.73±0.06 |
| | 0-5 | 11.64±0.58 |
| S027 | 12.5-17.5 | 19.81±0.61 |
| | 27.5-32.5 | 1.41 ± 0.18 |
| | 0-5 | 6.79±0.23 |
| S028 | 12.5-17.5 | 0.44±0.11 |
| | 27.5-32.5 | 0.32±0.13 |
| | 0-5 | 1.21±0.28 |
| S029 | 12.5-17.5 | <mda< td=""></mda<> |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 1.27±0.17 |
| S030 | 12.5-17.5 | 0.95±0.13 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 8.03±0.41 |
| S031 | 12.5-17.5 | 1.36±0.29 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 1.91±0.45 |
| S032 | 12.5-17.5 | 0.44 ± 0.08 |
| | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 11.86±0.23 |
| S033 | 12.5-17.5 | 6.78±0.15 |
| | 27.5-32.5 | 7.43±0.28 |
| | 0-5 | 1.77±0.23 |
| S034 | 12.5-17.5 | 0.82 ± 0.14 |
| | 27.5-32.5 | 0.8±0.13 |
| | 0-5 | 5.41±0.27 |
| S035 | 12.5-17.5 | 1.48±0.24 |
| | 27.5-32.5 | 1.97±0.1 |
| | 0-5 | 5.96±0.28 |
| S036 | 12.5-17.5 | 0.3±0.09 |
| ~~~~ | 27.5-32.5 | <mda< td=""></mda<> |
| | 0-5 | 0.29±0.07 |
| S037 | 12.5-17.5 | 6.92±0.07 |
| 5051 | 27.5-32.5 | < <u>MDA</u> |

Table 2 (Continue). ¹³⁷Cs concentration in Tehran province soil samples by HPGe (38.5%) and (55%).

* MDA:0.09±0.02

Minimum Detectable Activity (MDA)



Figure 3. The shape of frequency distribution of 137 C activity concentration in the depths: a. (0-5), b. (12.5-17.5) and c. (32.8-37.5) cm.

DISCUSSION

High-resolution gamma-ray spectrometry was used to determine the concentration of 137 Cs in soil samples collected in the period between June to September 2008 in Tehran province.

The relatively high contamination of ¹³⁷Cs in Tehran, in comparison with other countries, has been of significant importance to the present study.

The main reason for the accumulation of ¹³⁷Cs in some regions was due to atmospheric fallouts. It seemed that air currents, in their course of movements, accumulate heavy particles, such as ¹³⁷Cs covered with dust in higher altitudes (height of Alborz Mountains). The heights of North East of Tehran were strongly contaminated with ¹³⁷Cs which was at the end of the route of the dominant air currents during the year (with the direction of west to east).

Considering the obtained results from high average values of 137 Cs in Europe and its relation to Chernobyl accident, and the higher average concentration of 137 Cs in Tehran province, can also be attributed to Chernobyl accident due to the proximity of Iran to the place of the accident's occurrence, as shown in figure 4 and 5 ⁽¹⁵⁾. Neighboring countries which were in the same heights and geography with Iran could have the same average concentration of ¹³⁷Cs as Iran presumably, and it was obvious by moving farther from the place of occurrence (Chernobyl accident) to lower height, where the accumulation of ¹³⁷Cs decreased.

The depth profile distribution of ¹³⁷Cs in most of the Tehran province soil samples showed a tendency of decrease toward deeper layers (as figure 4 shows). To confirm the assumption, it was necessary to make a survey of ¹³⁷Cs concentration in different depths (with regard to annual accumulation of soils). To do so the time of the beginning of increasing ¹³⁷Cs concentration was estimated and proved to be related to Chernobyl accident. In addition, it seemed necessary to make a more accurate survey of the course of air currents in the regions behind the Alborz Mountains (Northern provinces).

The range of ¹³⁷Cs concentration in surface soils, was observed to vary in the range 0.29-77.35 (Bq.kg⁻¹), and the average ¹³⁷Cs activity was about 15.09 (Bq.kg⁻¹), which was more than the reported values in countries such as Egypt ⁽³⁾, China ⁽¹⁾, Nigeria ⁽⁸⁾, Alkharje city ⁽⁹⁾, Kuwait ⁽⁶⁾, western areas of Pakistan ⁽¹⁰⁾, Faisalabad in Pakistan ⁽¹¹⁾, Dhaka city ⁽⁴⁾; and also provinces in Iran, such as Booshehr,



Figure 4. 137Cs activity versus of depths.

Khoosestan and Hormozgan ⁽¹²⁾, and it was less than the reported values in countries such as, Coniferous forest ⁽²⁾, Jordan ⁽¹³⁾, Firtina Valley (Rize, Turkey) ⁽¹⁴⁾. The limit of detection was mainly governed by the counting statistics. ¹³⁷Cs in some samples was found to be lower than the minimum detectable activity of the gamma counting systems (<MDA).

CONCLUSION

The results of the measurements are presented in table 2. The range of ¹³⁷Cs concentration in surface soils, was observed to vary in the range 0.29-28.82 (Bq.kg⁻¹), and the average ¹³⁷Cs activity was about 11.3 (Bq.kg⁻¹). The highest concentrations 28.82 (Bq.kg⁻¹) was observed in Shangoldeh and the lower concentrations 0.29 (Bq.kg⁻¹) in Gazorkhan. The depth profile distribution of ¹³⁷Cs in most of the Tehran province soil samples showed a tendency of decrease toward deeper layers.

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