Original Article

Measuring Radionuclides Concentration in Rice Field Soils using Gamma Spectroscopy in Northern Iran

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Abstract

Background: A few elements of soil are radioactive. Soil can transfer radionuclide into plants feeding human. Sometimes their levels are as high as to be concern of human healthy. Rice has an important share for Iranian foods especially in north of Iran. Therefore we decided to obtain radionuclides concentration emitting $\gamma$ rays in Lahiyan City (Northern Iran) rice fields using $\gamma$ spectroscopy.

Methods: Twenty eight samples from rice field's soils and 12 samples from superficial soils were collected at a square of $10^*10$ m$^2$ to get 2kg weight. To make dry samples were put into oven at 105°C for 24h. Then they were milled and 950 gr of each sample was transferred to Marinelli container with 1000 cc volume, sealed and left for 40 days to get secular equilibrium. After measuring Ph, Electric conductivity and organic carbon, $\gamma$ spectroscopy was done to get sample gamma spectrum at 2000-6000 sec using HpGe detector.

Results: It was found $^{226}$Ra activity in rice fields of 29.273±0.72 Bqkg-1 and city soil of 31.02±1.1 Bqkg-1 and also $^{232}$Th activity of 37.47±1.12 Bqkg-1 for rice fields' soils and 40.47±1.68 Bqkg-1 for city soil were in standard mode.

Conclusion: $^{40}$K activities mean value according to UNSCEAR; 2000 was found a little greater than standard. A little value of $^{137}$Cs was found in Lahiyan rice fields and city soils that could be as a result of Chernobyl accident. In except of $^{137}$Cs, for three other under studied city soil elements, activities were greater than that of rice fields.

Keywords: Radionuclides, Rice fields, Spectroscopy, Iran

Introduction

Soil is composed of organic and inorganic compounds (1). A few elements of those are radioactive (2). These elements are classified into two groups: 1- radionuclides with half life longer than age of earth including $^{40}$K, $^{238}$U, $^{232}$Th, 2- radionuclides with half life shorter than age of earth including $^{137}$Cs. The first group could be found in soil naturally but the second group only could have artificial sources. Van Rooyen et al. (3) has released a table of some natural radionuclides activity in a soil field size of 1km *1 km*1m (Table 1). Of course soil can transfer radionuclide into plants feeding human. Sometimes their levels are as high as to be concern of human healthy.

Some scientists around the world measured soil radioactivity in Mditrean beaches, north of Serbesten, Germany, Japan, India and Italy (4-9). These studies were local and the results could help to make a world map of radionuclide distribution in fields providing food sources.

In Iran Abdi et al. got radionuclide distribution of $^{238}$U, $^{232}$Th, 40K and $^{137}$Cs in Persian Golf for Hormozgan Province (10). In addition, Samavat

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et al. obtained radionuclide concentration of $^{137}$Cs and $^{226}$Ra in food program of Ramsar population (11).

Gamma emitting from soil radionuclide is a part of background irradiation exposing population of 1.5 – 3.5 msv/y and sometimes up to 50 msv/y. However in a few regions in Iran, India and Europe there are reports to get more than 50 msv/y such as 260 msv/y for Ramsar City in Iran (2).

Rice has an important share for Iranian foods especially in north of Iran. Therefore we decided to obtain radionuclides concentration emitting $\gamma$ rays in Lahijan City rice fields using $\gamma$ spectroscopy and compare with UNSCEAR 2000 values in Table 2.

Table 1: Five natural radionuclides activity in a soil field size of 1 km $\times$ 1 km $\times$ 1 m (3)

| Nuclide | Activity (GBq) | Mass (kg) | Typical crustal activity concentration (Bq/kg) |
|---------|---------------|-----------|------------------------------------------|
| $^{238}U$ | 40 GBq | 2800 | 25 |
| $^{232}Th$ | 65 GBq | 15200 | 40 |
| $^{40}K$ | 630 GBq | 2500 | 400 |
| $^{226}Ra$ | 80 GBq | 2.2 g | 48 |
| $^{222}Rn$ | 9.5 GBq | 14 $\mu$g | 10 |

Table 2: Natural radionuclide content in soil (2) concentration in soil (Bq/kg)

| Nuclide | Activity | Mass | Typical crustal activity concentration (Bq/kg) |
|---------|----------|------|------------------------------------------|
| $^{40}K$ | Mean | 140-850 | 35 |
| $^{226}Ra$ | Mean | 17-60 | 30 |
| $^{232}Th$ | Mean | 11-64 | 30 |

Materials and Methods

City location

Lahijan City has been placed in Gilan Province in north of Iran with 584.3 km$^2$ square (Fig. 1). This city is located on latitude of 37° and 11° north. Rice is one of the most important products of the city.

Preparing samples

At first topography map from Lahijan City was prepared. Sampling was being done by a metal ring with 5 cm height and 14 cm diameter. Twenty eight samples from rice field's soil and 12 samples from superficial soil were collected using GPS. Sampling was done according to rice fields scattering so whole city was covered. Each sampling was being made at a square of 10*10 m$^2$ to get 2 kg weight and saved in a plastic pack. Labeling was done for each pack. To make dry samples were put into oven at 105°C for 24h. Then they were milled. In next step 950 gr of each sample was transferred to Marinelli container with 1000 cc volume, sealed and left for 40 days to get secular equilibrium.

Soil Texture

Solid part of soil is composed of organic and inorganic compounds. Relative abundance of sand, silt and clay in soil named as soil texture. These particles are separated and classified by their sizes according to International Community of Soilology (Table 3) (12). Therefore in this study samples were sifted using screens with required sizes. It was to classify particles as
sand, silt and clay with their relative abundances.

Table 3: Soil particles size under International Community of Soilogy rules

| Particle | Size (mm) |
|----------|-----------|
| Large    | 0.02-0.2  |
| Sand     | 0.2-2     |
| Small    | 0.002-0.05|
| Clay     | μ < 2     |

**pH of soil**
To determine pH of soil, suspensions with soil to water ratio of 10 gr soil and 20CC, 50CC and 100CC water were made. These suspensions were shook for 30 minutes and left silent for next 30 minutes to get sink suspended particles. Using pH meter (Genway, relative error; ±0.5) pH of soil was measured.

**Electrical Conductivity (EC) of soil**
Electrical conductivity or inverse of electrical resistance is related to salts or ions concentration directly. Increase of EC means increase of salts. The unit of EC is Simense/m in SI units.
To get EC, 20 gr of soil sample was passed through 2mm screen and poured into Eurlen Mayer 250cc. After adding 100CC water, suspension was being shook for 30 minutes and finally EC was measured by EC meter (Hanna, relative error; ±0.1).

**Organic Carbon**
Organic carbon does not change soil texture but improves physical and chemical properties of soil. To determine percent of organic carbon at first 10 gr of soil sample was crushed by camelian mortar, and then passed through 0.5 mm screen. One gr of screened soil was poured into Eurlen Mayer 500cc while 10CC potassium bichromat, 1 normal was added to it. To make uniform scatter of particles, shaking was done. Then 20CC H2SO4, 90% were poured into container and shaking was repeated for 1 minutes. This mixture was left for 30 minutes, then at first 250cc water and after cooling 10 drops Fero artophenatrolin as determinant was added. Titration was made by fero ammonium sulfate changing color from dark green in the end of titration to red.

**γ Spectroscopy**
γ Spectroscopy was done to get sample gamma spectrum at 2000-6000 sec using HpGe detector (Ortec) with energy resolution of 0.5 kev at 69.5 kev. Detector energy calibration was done by 241Am and 226Ra for energy range of 60-3000 kev.

**Results**
Table 4 and 5 reveal activities of 226Ra, 232Th, 40K and 137Cs in rice field's soil samples and Lahijan city soil samples.
In rice fields soil samples, mean activity of 226Ra, 232Th, 40K and 137Cs were obtained 29.73(24.15-37.71), 37.47(29.25-51), 555.43(424.7-651.5) and 10.47(2.76-18.83) Bqkg⁻¹ consequently. For Lahijan City soil samples those values were gotten as 31.02(26.1-39.38), 40.47(32.17-50.24), 599.11(432.98-695.65), 7.46(2.55-13.11) Bqkg⁻¹ consequently. However there is no world reported value for 137Cs since this element because of its short half life relevant to earth age is only as an artificial type. Therefore it should be zero.
### Table 4: Radiometric results for $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in rice fields soil samples

| No. | Ra-226 (Bq/kg) | Th-232 (Bq/kg) | K-40 (Bq/kg) | Cs-137 (Bq/kg) |
|-----|----------------|----------------|--------------|----------------|
| 01  | 32.32±1.46     | 36.37±1.90     | 581.52±8.73  | 9.05±0.35      |
| 02  | 30.86±1.25     | 36.42±1.89     | 462.49±6.43  | 16.23±0.53     |
| 03  | 28.12±1.35     | 32.90±1.43     | 516.66±7.04  | 3.41±0.18      |
| 04  | 25.31±0.82     | 34.20±1.10     | 565.91±5.68  | 8.36±0.22      |
| 05  | 28.80±1.10     | 35.52±1.30     | 599.03±6.52  | 8.67±0.27      |
| 06  | 27.07±1.68     | 29.25±2.04     | 424.7±7.66   | 14.68±0.59     |
| 07  | 24.30±0.92     | 30.33±1.30     | 425.68±5.01  | 14.35±0.35     |
| 08  | 31.40±0.97     | 41.76±1.34     | 446.26±4.77  | 18.83±0.43     |
| 09  | 36.37±1.26     | 47.81±1.58     | 562.27±6.23  | 15.05±0.36     |
| 10  | 30.21±0.89     | 35.73±1.10     | 510.29±5.15  | 18.12±0.41     |
| 11  | 34.74±1.18     | 47.55±1.61     | 620.84±6.84  | 10.78±0.33     |
| 12  | 37.71±1.58     | 51.00±2.38     | 534.94±7.99  | 8.38±0.39      |
| 13  | 29.43±1.28     | 34.68±1.61     | 565.72±7.65  | 7.54±0.30      |
| 14  | 37.71±1.58     | 51.00±2.38     | 534.94±8.00  | 8.38±0.39      |
| 15  | 28.03±1.07     | 35.41±1.55     | 651.5±7.22   | 7.8±0.26       |
| 16  | 27.54±0.86     | 36.08±1.01     | 609.51±5.99  | 7.36±0.21      |
| 17  | 31.05±1.41     | 36.46±1.97     | 625.23±8.83  | 2.76±0.26      |
| 18  | 27.43±1.36     | 32.61±1.99     | 592.12±9.28  | 9.82±0.40      |
| 19  | 30.38±1.30     | 37.61±1.67     | 595.65±7.88  | 10.96±0.38     |
| 20  | 24.77±1.27     | 35.19±1.56     | 575.86±7.32  | 10.19±0.34     |
| 21  | 27.23±0.88     | 34.61±1.08     | 592.28±5.91  | 11.74±0.30     |
| 22  | 27.96±1.43     | 34.73±2.10     | 628.5±7.76   | 8.35±0.38      |
| 23  | 29.40±1.39     | 34.56±1.82     | 594.79±7.95  | 12.23±0.42     |
| 24  | 24.15±1.59     | 32.66±2.06     | 559.14±9.07  | 11.92±0.49     |
| 25  | 36.09±1.47     | 46.84±2.01     | 574.3±7.61   | 8.95±0.33      |
| 26  | 28.73±1.73     | 38.61±2.33     | 444.65±7.82  | 15.61±0.61     |
| 27  | 27.97±1.55     | 36.53±2.12     | 574.15±8.58  | 6.78±0.36      |
| 28  | 27.44±1.82     | 32.75±1.86     | 583.11±10.0  | 8.63±0.37      |

### Table 5: Radiometric results for $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in Lahijan city soil superficial samples

| No. | Ra-226 (Bq/kg) | Th-232 (Bq/kg) | K-40 (Bq/kg) | Cs-137 (Bq/kg) |
|-----|----------------|----------------|--------------|----------------|
| A   | 30.06±0.89     | 40.28±1.28     | 558.56±5.58  | 6.60±0.19      |
| B   | 26.10±1.20     | 32.17±1.88     | 432.98±6.11  | 11.91±0.43     |
| C   | 34.57±1.51     | 46.60±2.04     | 504.68±6.69  | 5.85±0.30      |
| D   | 31.58±1.51     | 46.91±1.97     | 679.67±9.12  | 2.63±0.24      |
| E   | 32.84±1.65     | 50.24±2.22     | 684.15±8.91  | 12.73±0.47     |
| F   | 27.10±1.60     | 33.95±2.60     | 579.85±10.33 | 7.01±0.46      |
| G   | 27.87±1.14     | 35.48±1.83     | 598.25±8.03  | 11.26±0.39     |
| H   | 29.19±1.25     | 39.48±2.10     | 621.25±8.48  | 13.11±0.42     |
| I   | 30.96±1.87     | 35.09±2.31     | 592.80±9.53  | 2.55±0.12      |
| J   | 28.04±1.21     | 37.94±1.68     | 623.26±7.83  | 10.98±0.36     |
| K   | 27.88±1.04     | 33.70±1.18     | 637.71±6.37  | 5.52±0.20      |
| L   | 29.93±1.54     | 38.49±2.14     | 534.13±8.55  | 8.87±0.43      |
| M   | 38.73±1.81     | 48.61±2.43     | 644.65±7.91  | 2.61±0.53      |
| N   | 39.38±1.13     | 47.61±1.45     | 695.65±7.76  | 2.82±0.30      |
Discussion

This study measured natural radioactivities of three γ emitter element, $^{226}$Ra, $^{232}$Th, $^{40}$K and also artificial $^{137}$Cs in Lahijan City rice field's soil using gamma spectroscopy with HpGe detector. For comparison city natural soil was studied too. All of those values were compared to standard values published by UNSCEAR 2000. According to Tables 2–4, $^{226}$Ra activity was found in rice fields $[29.73(24.15-37.71) \ \text{Bqkg}^{-1}]$ and city soil $[31.02(26.1-39.38) \ \text{Bqkg}^{-1}]$ were less than standard values. For $^{232}$Th, the resultant values of $37.47 \ (29.25-51) \ \text{Bqkg}^{-1}$ for rice fields' soils and $40.47 \ (32.17-50.24) \ \text{Bqkg}^{-1}$ for city soils were in standard mode. However 6 samples for each rice fields and city soil were greater than standard mean values but less than higher limits of that. In the case of $^{40}$K activities mean value according to UNSCEAR; 1988 is almost in the range of standard. However UNSCEAR; 2000 finds it a little greater than standard. Since $^{137}$Cs should have found zero, the values more than that could be as a result of Chernobyl accident. World mean values for natural radionuclides of $^{226}$Ra, $^{232}$Th, $^{40}$K are 35, 30, 400 Bqkg$^{-1}$ consequently (UNSCEAR, 1993).

In except of $^{137}$Cs, for three other under studied city soil elements, activities were greater than that of rice fields. It could be related to irrigation method. Manjilban in altitudes of Roudbar City irrigates Lahijan rice fields. These latitudes could be a source of $^{137}$Cs. The lower activities of three other natural elements in comparison to city soil could get back to plough and to irrigate rice fields. These operations could reduce natural radioactivity.

This study showed the equivalent activities were to be $126.08 \pm 2.53 \ \text{Bqkg}^{-1}$ for rice fields’ soil samples and $135.02 \pm 4.36 \ \text{Bqkg}^{-1}$ for superficial city soil samples holding under UNSCEAR authorized limits.

Relation among amounts of organic carbon, slit, clay, Ph an EC in rice fields soil and superficial soil samples in Lahijan City and activities of $^{226}$Ra, $^{232}$Th, $^{40}$K, $^{137}$Cs was evaluated by Pearson correlation coefficient.

In rice field soil samples, for $^{226}$Ra, there was direct significant relation with soil organic carbon, humid and electric conductance as 0.852, 0.752 and 0.618 consequently. For $^{232}$Th it was found correlation coefficient with soil organic carbon to be 0.912. However it was -0.461 between $^{40}$K and clay. As a matter of fact in this case increasing clay means less $^{40}$K activity.

In Lahijan superficial soil samples it was significant relation among organic carbon and activities of Ra, Th and K with Pearson correlation coefficient of 0.88, 0.857 and 0.637 consequently. Also it was revealed Pearson correlation coefficient of 0.629 between soil humid and $^{226}$Ra activity.

Ethical considerations

Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors.

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References

1. Merril E, Thomas G (1997). Environmental Radioactivity: from natural, industrial and military sources. Fourth edition. San Diego : Academic Press, p.59.
2. UNSCEAR 2000. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effect of Atomic Radiation, United Nation, New York.
3. Van Rooyen TJ (2002). Lecture notes. The science of radiation protection. Themba LABS, Unpublished.

4. Brai M, Basile S, Bellia S, Hauser S, Puccio P, Rizzo S, Bartolotta A, Licciardello A (2002). Environmental radioactivity at Strombol (Aeolian Islands). Applied Radiation and Isotopes, 57(1): 99-107.

5. Brai M, Bellia S, Hauser S, Puccio P, Rizzo S, Basile S, Marrole M (2006). Correlation of radioactivity measurements, air kerma rate and geological features of Sicily. Radiation Measurements, 41(4):461-470.

6. Bikil I, Slivka, Onki LJ, Krmar M, Veskovi M, Todorovi N, Varga E, Mrdja D (2005). Radioactivity of the soil in Vojvodina (Northern province of Serbia and Montenegro). J Environ Radioactivity, 78(1):11-19.

7. Winkelmann I, Strobl C, Thomas M (2004). Aerial measurements of artificial radionuclides in Germany in case of a nuclear accident. J Environ Radioactivity, 72 (1-2):225-231.

8. Sakaue H, Maruta F, Fujimaki H, Tonouchi T (2007). Distribution of $^{137}$Cs around Kashiwazaki-Kariwa area. Journal J Radioanayl Nucl Chem, 273(1):123-127.

9. Sengupta D, Ghosh A, Mamtani MA (2005). Radioactivity studies along fracture zones in areas around Galudih, East Singhbhum, Jharkhand, India. Applied Radiation and Isotopes, 63(3):409-414.

10. Abdi MR, Faghihia H, Mostajabodavati M, Hasanzadeh A, Kamali M (2006). Distribution of natural radionuclides and hot points in coasts of Hormozgan, Persian Gulf, Iran. J Radioanal Nucl Chem, 270 (2): 319-324.

11. Samavat H, Seaward RD, Aghamiri M, Reza-Nejad SMR (2006). Radionuclide concentrations in the diet of residents in a high level natural radiation area in Iran. Radiat Environ Biophys, 45(4):301-306.

12. Bohn R, McNeal B, Connor GO (1979). Soil Chemistry. First edition. John Wiley and Sons. New York, NY. p.329.