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Investigation of depleted uranium contamination in south west of Iraq

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Background: The application of DU emanation for the first time contaminated certain areas in the south west region of Iraq after the second Gulf war (1991). These contaminated areas were discovered in 1994. Radioactive contamination was detected using the nuclear enterprise PCM5/1 in soil samples collected from two regions near by grazing lands. This study was done for assessment of DU contaminated soil in the regions under study. ***Materials and Methods:*** Portable detector was used for radiation measurement of the contaminated area. Samples from each region were selected and taken to Baghdad kept in plastic bags for gamma ray spectroscopy measurement. Gamma-ray spectroscopy system consists of high purity germanium (HPGE) detector surrounded by appropriate shield. The measurement of detector efficiency using (GDR) computer programs, supplied by Canberra Company was used to analyze gamma-ray spectrum. The activity of ^{234}Th, ^{235}U, ^{238}Pa and other natural isotopes were measured. ***Results:*** The measurement by gamma -ray spectrometry system showed that six samples were heavily contaminated with DU, because the presence of ^{243}Pa and ^{235}U peak and the percentage ratio between ^{235}U/^{238}U were less than 0.005, when both international mathematical methods, namely IAEA and Kosovo, were used. Because of existence of radiation equilibrium between ^{234}Th and ^{234}Pa, the measurements should be accurate. ***Conclusion:*** The result showed that six of the samples were heavily contaminated with DU and there is a good agreement between the two methods. Because of the accuracy and ease of the Kosovo method, it is recommended for future investigations. **Iran. J. Radiat. Res., 2005; 3 (3): 109-115**

Keywords: *Depleted uranium, soil contamination, south west of Iraq, Kosovo Method.*

INTRODUCTION

^{235}U is used for production in nuclear weapons and electricity. Depleted Uranium (DU) is a low cost and readily available material. There is now more than 0.5 million tone of DU accumulated as nuclear waste. It has high density (19.05 g/cm^3), 2.54 times heavier than iron, with a high penetrating power[1], so it can be used for civilians and military industry. The main civilian application of DU includes counter-weights in aircraft, calorimeter, detectors, flywheels, and sinker bars[2]. In military it is used as penetrating and tanks heavy amour. The highly flammable metal fill with DU small bullet fired by aircraft is shown in figure 1. The weight of a small bullet is approximately 300 g of DU hitting the ground in a straight line; one to three meters a part, depending on the angle of the approach[3].

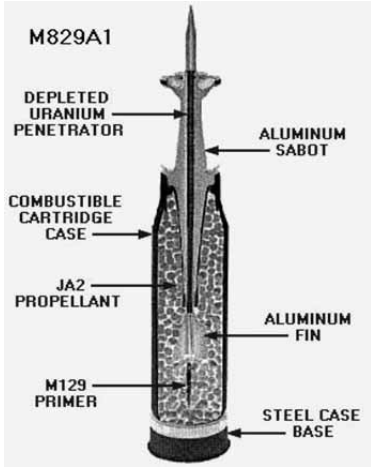

Figure 1. Schematic diagram of a DU round (WHO 2001).

Some of the bullets that miss their targets fall on the ground and could be considered as serious environmental pollutants that may contaminate food, surface water resources and ground water because the corrosion

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penetrations in soil/water may be rapid (0.01-0.05 cm/y). Crancon[4] in his study at French research site for DU penetrations development found that DU bullet reach to depth of 30 cm after 30 years and 10% of its DU contents became soluble at ground water. The highest penetrations caliber weight is 1Kg if fired from a tank.

Al-Kinani *et al.*[5] found seven soil samples contaminated with DU near Basra city, south of Iraq. The ratio of ^{235}U / ^{238}U ranged between 0.00412-0.00351. Al-Dory[6] showed that radiation equilibriums between ^{234}Th and ^{226}Ra in un-contaminated soil exist with a ratio between 0.68-1.16. For contaminated soil samples, the radiation equilibrium between ^{234}T and ^{234m}Pa ranged between 0.928-0.956 indicating that the measurements were accurate. The aim of this work was to investigate DU contamination in the south west of Iraq by adapting the IAEA and Kosovo mathematical methods used for assessment of contaminated soil samples

MATERIALS AND METHODS

Depleted uranium has been detected in two regions located in the south west of Iraq by using PCM5 portable radiation detector device (NE-UK). The detection rate of 2-5000 count/second, make this device suitable for detection of gamma and beta emitting contaminants. It is consisted of scintillation material with ZnS sheet and FAG portable detector for dose measurement, provided by IAEA (with reading range between 0.00001-10 mSv/hour, German made). The samples of soil were collected at hot points and kept inside plastic bags for spectroscopy measurement. Control (background) soil samples were collected from region located far away from the destroyed tanks where the reading of desert by portable detector ranged between 0.07-0.09 μSv/h .

The two regions were Al-Nukhaib, in Karbala province, at south west of Iraq near Saudi border, located in the longitude between 42°-44° and latitude between 32°-34°, and Al-Salman, in Al-Muthanna government, which is located in the longitude between 44°-46° and latitude between 30°-32°. Figure 2 shows the location of sites under investigation on the Iraq map. Many of soil samples were collected from these regions. Five samples from Al-Nukhaib and five samples from Al-Salman region were chosen to do this study. Table 1 shows the location,

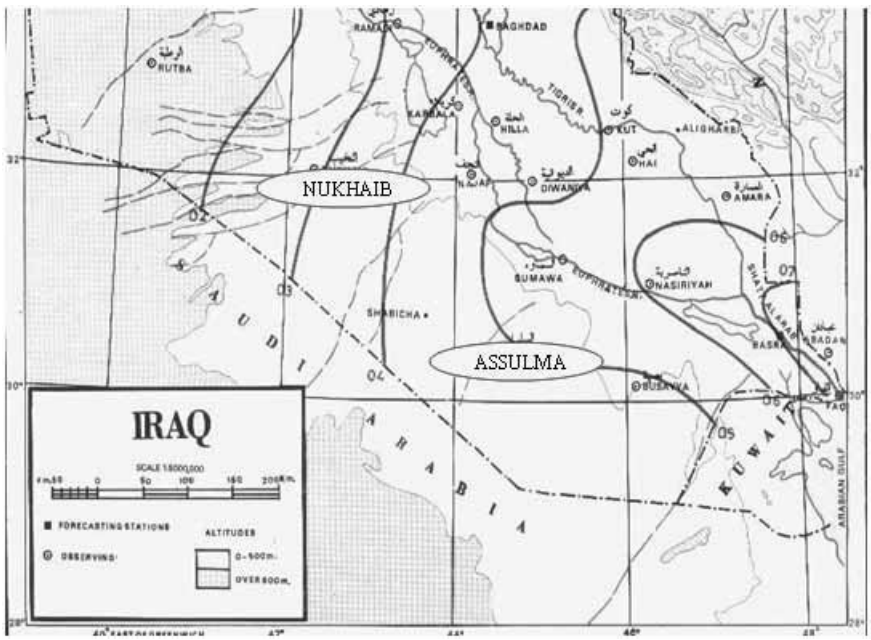

Figure 2. Iraq map (IMO, 2001), the regions under investigation are shown.

Table 1. The location, type and code of collected samples.

Samples Number	The Locations	Samples Type	Samples Code	Notes
1	Al-Nukhaib	Soil	SA_1.SPM	Contaminated
2	Al-Nukhaib	Soil	SA_2.SPM	Not - Contaminated
3	Al-Nukhaib	Soil	SA_3.SPM	Not - Contaminated
4	Al-Nukhaib	Soil	SA_4.SPM	Contaminated
5	Al-Nukhaib	Soil (Background)	SA_5.SPM	Not - Contaminated
6	Al-Salman	Soil	SA_6.SPM	Contaminated
7	Al-Salman	Fired Soil	SA_7.SPM	Contaminated
8	Al-Salman	Small Particles of DU	SA_8.SPM	Contaminated
9	Al-Salman	Soil (Background)	SA_9.SPM	Not - Contaminated
10	Al-Salman	Bullet	SA_{10}.SPM	Contaminated

type, and the code of samples.

After preparation of soil samples, they were stored in Marinelli Beaker for one month to reach secular radioactive equilibrium. Each sample was put on the crystal detector for a period of 36000 seconds. The high purity germanium (HPGe) detector was surrounded by a lead shield of 12 cm in thickness to protect the detector from background radiation, and lined inside with cadmium (0.8 mm) and copper (~0.4 mm) sheets to minimize the X-rays emitted due to interaction of gamma radiation with Pb. ^{152}Eu source was used for energy and efficiency calibration because of its suitable half life and the wide range of gamma ray energies produced during it's decay process (121.8 keV - 1409.1 keV). The efficiency of the detector was 40% with a resolution of 2.2 keV for ^{60}Co at 1.332 MeV energy.

The radionuclide were detected by using PCA, GDR computers programs commonly used for identification of radionuclide by gamma ray spectroscopy (Canberra company). The net area under the curve for each decay isotopes of U-235, U-238, and K-40 were calculated.

There are two international methods used to calculate the percentage ratio for ^{235}U / ^{238}U:

1. The method that used by International Committee to detect the DU in Kosovo (3) as follow:

A. Conversion of the radioactivity of ^{238}U to mass percentage by dividing to 12.35 Bq/kg.

B. From the mass percentage the percentage of ^{238}U in background sample was subtracted to find the quantity of DU in soil samples.

C. The percentage of depleted uranium (X%) was found by dividing the quantity of DU by the total concentration of ^{238}U.

D. According to the following equation Ru was found:

$$Ru = (0.72 - 0.52\ X) / (99.2745 + 0.5255\ X) \quad (1)$$

Where; X = the quantity of DU in soil samples, and Ru = the percentage between ^{235}U and ^{238}U.

2. The method used by IAEA to detect DU[7], is as follows:

A. Calculate net counts (N_1, N_2, and N_3) in the peaks at the energies of E_1, E_2, and E_3; correct for contribution of background radiation.

B. Calculate (q and F) values.

$$q = c\,\frac{P_2}{P_{1U}}\left(\frac{E_2}{E_1}\right)^a \frac{N_1}{N_2} \quad (2)$$

$$F = 1 - f\,\frac{P_{1Ra}}{P_3}\left(\frac{E_1}{E_3}\right)^a \frac{N_3}{N_1} \quad (3)$$

C. Then applying the following equation:

$r_m = qF$ (4)

Where:

c = Conversion Factor (0.1555).

f = Correction Factor (1.04).

a = Slope of Efficiency Curve.

P_{1U}, P_{1Ra}, P_2, and P_3 = Emission probability of ^{235}U, ^{226}Ra, ^{234m}Pa, and ^{214}Bi respectively.

E_1, E_2, and E_3 = Gamma ray energy of ^{235}U, ^{226}Ra, ^{234m}Pa, and ^{214}Bi respectively.

RESULTS

Contamination with DU was considered if the ratio between $^{235}U/^{238}U$ in samples was less than 0.005. From gamma ray spectrums, the peak of ^{234m}Pa and ^{235}U radionuclide are not seen in non-contaminated samples (figure 3), but in contaminated samples these peaks were easily detected by the gamma spectrometry system. The specific radioactivity of ^{226}Ra is calculated by subtracting the specific radioactivity of ^{235}U which is calculated from the peak energy of 205.3 keV from the total specific radioactivity calculated for ^{226}Ra using the peak energy of 186.21 keV. The result shown in table 2 is the specific radioactivity for ^{226}Ra and ^{234m}Pa in the contaminated samples, and for ^{226}Ra only in the non-contaminated samples[8].

From the specific radioactivity of ^{234}Th and ^{234m}Pa the percentage between $^{234}Th/^{234m}Pa$ are calculated for the contaminated samples (table 3). Existence of equilibrium between radionuclide means that the measurements were accurate. The percentage between $^{235}U/^{238}U$ (Ru) which has been calculated according to Kosovo method is shown in table 4. Also the percentage between $^{235}U/^{238}U$ (r_m) has been calculated using the IAEA method.

DISCUSSION

The ratio between $^{234}Th/^{234}Pa$ was in the range between 0.84-0.99 indicating that these isotopes were at radiation equilibrium for the first daughter nuclide of ^{238}U series. Other radionuclide such as ^{214}Bi, and ^{214}Pb did not reach to equilibrium state with other isotopes, in spite of one month storage of the samples in Marinelli beaker. This might be due to generation of radon gas (^{222}Rn) which can leak from the pores of the plastic container. Therefore, the concentration of ^{214}Bi, and ^{214}Pb found to be less than that of other isotopes. For reaching to radiation equilibrium state of these isotopes, samples must be stored in Marinelli beaker made of quartz to avoid leaking of radon gas[9, 10].

Table 4 shows that the percentage ratio between ^{235}U and ^{238}U range between 0.020-

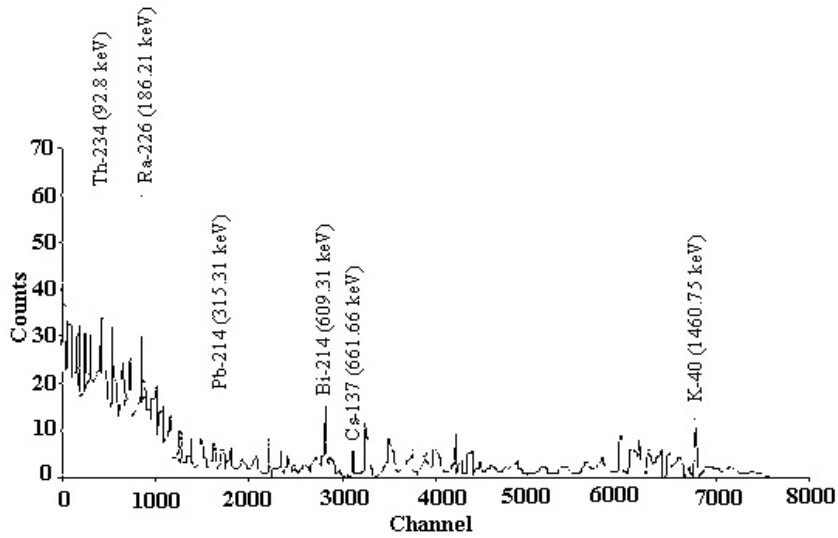

Figure 3. The spectrum of SA5 for un-contaminated soil samples with DU.

Table 2. The specific radioactivity (Bq/kg) for the specified radionuclide's for each sample.

Radio-nuclides	SA_1. SPM	SA_2. SPM	SA_3. SPM	SA_4. SPM	SA_5. SPM	SA_6. SPM	SA_7. SPM	SA_8. SPM	SA_9. SPM	SA_{10}. SPM
Th-234 92.80	13521.6 ± 42.6	74.8 ± 10.6	65.9 ± 11.7	31711.1 ± 148.2	48.5 ± 3.8	1809.3 ± 13.4	430949.3 ± 450.8	895377.6 ± 1216.7	60.9 ± 4.4	1110495.7 ± 5962.1
U+Ra+Pa 186.21	13933.0 ± 38.2	99.7 ± 10.5	88.4 ± 9.8	35550.0 ± 121.3	62.0 ± 3.4	1901.6 ± 17.1	450344.7 ± 205.5	925051.3 ± 1220.4	81.3 ± 3.4	1217001.4 ± 2987.8
U-235 205.31	186.6 ± 11.7	-----	-----	397.0 ± 15.3	-----	20.1 ± 2.7	4053.1 ± 45.0	8617.5 ± 113.6	-----	10665.5 ± 821.5
Pb-214 315.31	17.3 ± 1.4	13.8 ± 0.8	14.5 ± 0.8	25.3 ± 2.4	15.2 ± 0.3	21.2 ± 0.5	40.2 ± 5.8	-----	23.1 ± 0.4	-----
Bi-214 609.31	15.4 ± 1.2	11.3 ± 0.8	12.9 ± 0.8	17.8 ± 2.0	15.3 ± 0.4	19.4 ± 0.6	33.7 ± 5.3	-----	21.3 ± 0.4	-----
Cs-137 661.66	15.0 ± 0.8	5.4 ± 0.4	27.5 ± 0.7	17.0 ± 1.2	1.0 ± 0.1	20.7 ± 0.3	-----	-----	1.4 ± 0.2	-----
Pa-234m 1001.03	13630.1 ± 40.3	-----	-----	37807.6 ± 267.8	-----	2007.0 ± 15.6	445852.8 ± 490.5	939170.7 ± 1221.2	-----	1207872.4 ± 4552.7
K-40 1460.75	193.3 ± 8.8	158.3 ± 7.8	176.4 ± 7.7	253.1 ± 12.5	124.8 ± 2.9	446.9 ± 5.6	273.8 ± 15.4	-----	229.5 ± 5.2	-----

Table 3. The percentage ratio calculated for $^{234}Th/\ ^{234m}Pa$ in samples.

Radio-nuclies		Th-234	U+Ra+Pa	U-235	Pb-214	Bi-214	Cs-137	Pa-234m	K-40
Energy (keV)		92.80	186.21	205.31	351.92	609.31	661.66	1001.03	1460.75
SA_1.SPM	Activity (Bq/kg)	13521.6	13933.0	186.6	17.3	15.4	15.0	13630.1	193.3
	Th^{234}/Pa^{234m}	0.99							
SA_2.SPM	Activity (Bq/kg)	74.8	99.7	-----	13.8	11.3	5.4	-----	158.3
	Th^{234}/Pa^{234m}	-----							
SA_3.SPM	Activity (Bq/kg)	65.9	88.4	-----	14.5	12.9	27.5	-----	176.4
	Th^{234}/Pa^{234m}	-----							
SA_4.SPM	Activity (Bq/kg)	31711.1	35550.0	397.0	25.3	17.8	17.0	37807.6	253.1
	Th^{234}/Pa^{234m}	0.84							
SA_5.SPM	Activity (Bq/kg)	31711.1	62.0	-----	15.2	15.3	1.0	-----	124.8
	Th^{234}/Pa^{234m}	-----							
SA_6.SPM	Activity (Bq/kg)	1806.3	1901.6	20.1	21.2	19.4	20.7	2007.0	446.9
	Th^{234}/Pa^{234m}	0.90							
SA_7.SPM	Activity (Bq/kg)	430949.3	450344.7	4053.1	40.2	33.7	-----	445852.8	273.8
	Th^{234}/Pa^{234m}	0.97							
SA_8.SPM	Activity (Bq/kg)	895377.6	925051.3	8617.5	-----	-----	-----	939170.7	-----
	Th^{234}/Pa^{234m}	0.95							
SA_9.SPM	Activity (Bq/kg)	60.95	81.3	-----	23.1	21.3	1.4	-----	229.5
	Th^{234}/Pa^{234m}	-----							
SA_{10}.SPM	Activity (Bq/kg)	1110495.7	1217001.4	10665.5	-----	-----	-----	1207872.4	-----
	Th^{234}/Pa^{234m}	0.92							

Table 4. The percentage ratio of $^{235}U/^{238}U$ in samples.

Samples	U-238 Concen-tration mg/kg	[DU] Concen-tration mg/kg	X %	Quantity of Natural Uranium (1-X)	Ru	1/Ru
SA_1.SPM	1094.866	1090.979	99.645	0.00355	0.00200	500.000
SA_2.SPM	6.057	2.170	35.826	0.64174	0.00533	187.617
SA_3.SPM	5.336	1.449	27.155	0.72845	0.00583	171.527
SA_4SPM	2567.700	2563.813	99.849	0.00151	0.00200	500.000
SA_5.SPM	3.927	0.040	1.019	0.98981	0.00720	138.889
The average of background radiation = 48 Bq/kg = 3.887 mg/kg						
Samples	U-238 Concen-tration mg/kg	[DU] Concen-tration mg/kg	X %	Quantity of Natural Uranium (1-X)	Ru	1/Ru
SA_6.SPM	146.259	141.401	96.678	0.03322	0.00220	454.545
SA_7.SPM	34894.680	34889.822	99.986	0.00014	0.00200	500.000
SA_8.SPM	72500.211	72495.353	99.993	0.00007	0.00200	500.000
SA_9.SPM	4.935	0.077	1.560	0.98440	0.00717	139.470
SA_{10}.SPM	89918.680	89913.822	99.995	0.00005	0.00200	500.000
The average of background radiation = 60 Bq/kg = 4.858 mg/kg						

0.0022 for contaminated samples and 0.00533-00720 uncontaminated samples with DU which is in agreement well with other findings[2, 5, 7]. Table 5 shows the ratio $^{235}U/^{238}U$ calculated using both Kosovo and IAEA methods. As seen in figure 4 the calculated ratios were nearly similar according to the IAEA and Kosovo methods. Based on the obtained results, the Kosovo method was found more useful and accurate method for DU contamination measurement.

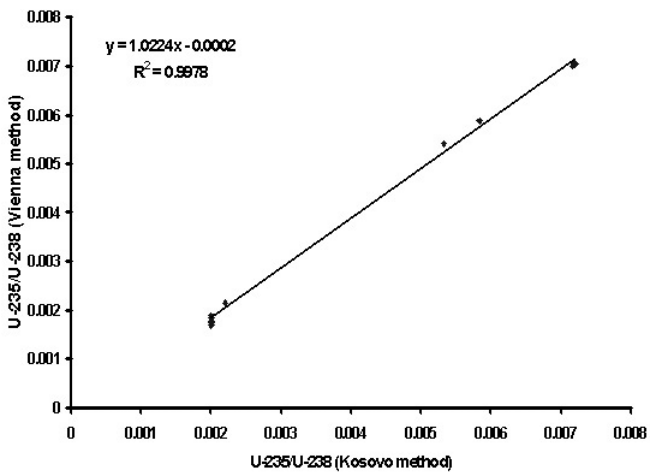

Figure 4. $^{235}U/^{238}U$ according to IAEA and Kosovo method.

Table 5. The ratio of $^{235}U/^{238}U$ calculated according to the IAEA and Kosovo methods.

Samples	235U/238U (KOSOVO)	235U/238U (VIENNA)
SA_1.SPM	0.00200	0.00169
SA_2.SPM	0.00533	0.00540
SA_3.SPM	0.00583	0.00588
SA_4.SPM	0.00200	0.00183
SA_5.SPM	0.00720	0.00704
SA_6.SPM	0.00220	0.00215
SA_7.SPM	0.00200	0.00189
SA_8.SPM	0.00200	0.00176
SA_9.SPM	0.00717	0.00701
SA_{10}.SPM	0.00200	0.00177

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