ASSESSMENT OF DEPLETED URANIUM CONTAMINATION IN SELECTIVE IRAQI SOILS

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Abstract

The aim of this research was to measure the radiation exposure rates in three selected locations in southern part of Iraq (two in Nassireya, and one in Amara) resulted from the existence of depleted uranium in soil and metal pieces have been taken from destroyed tank and study mathematically the concentration of Depleted Uranium by its dispersion from soil surface by winds and rains from 2003 to 2007.

The exposure rates were measured using inspector device, while depleted uranium contamination in soil samples and tank's metal pieces were detected with Solid State Nuclear Track Detectors (SSNTDs).

The wind and rain effects were considered in the calculation of dispersion effect on depleted uranium concentration in soil, where the wind effect were calculated with respect to the sites nature and soil conditions, and rain effect with respect to dispersive-convective equation for radionuclide in soil. The results obtained for the exposure rates were high near the penetrated surface, moderate and low in soil and metal pieces. The Depleted Uranium concentration in soil and metal pieces have the highest value in Nassireya .The results from dispersion calculation (wind & rain) showed that the depleted uranium concentration in 2008 will be less than the danger level and in allowable contamination range.

Introduction

When measuring isotopic ratios in environmental samples it is important to realize that uranium may sometimes become depleted (or enriched) in some of its isotopes due to natural processes such as chemical weathering. Depleted Uranium (DU) is a by-product from the process used to enrich natural uranium ore for use as fuel in nuclear reactors and nuclear weapons.

In weapon use ,when penetrator impact on ground surface ,a portion of its DU mass is transformed into aerosols or fine particles and thrown into the surrounding air .These aerosols and fine particles are normally depleted in measurable quantities on the surroundings ground or on other surfaces within about 100m from impact.

After initial deposit, it is possible that fine DU dust particles are resuspended into the atmosphere together with soil-dust by wind or human activities, leading to secondary air contamination. These particles are then deposited once more on the surrounding ground and other surfaces.

If the deposition takes place on surfaces other than soil that are exposed to rain and other metrological phenomena, the surface deposit will be partly washed off. Surface deposits on soil will penetrate into the topsoil layer with time. The dispersion of DU in soil is governed by convective-dispersive equation [1].

Jones in 1991 used track detectors in evaluating uranium concentrations in samples of surface soil in USA [2].

Alhilli, studied in 1998 the effect of using radioactive weapons in soil and air in selective locations in the south of Iraq [3].Mohammed study (1996) consists of measuring the natural radioactivity for surface soil samples for different locations and depths [4]. Tawfiq in (1996) indicated the concentration of α-emitters in soil samples in al-Tiwetha city near of Baghdad, by using track detectors [5], Also Tawfiq in 1996 indicated the uranium concentrations in soil samples in northern Rumela in the south of Iraq, using the same way of detecting [6].

Methods and Materials Instruments Used in Work

The locations were selected and the samples were collected with respect to the environmental basics and United Nation Environment Program (UNEP) recommend-ations. These instruments below were used for collecting samples:

1. Polyethylene bags.
2. Hand trowel.
3. Auger.
4. Portable detector instrument (inspector).

And for self safety, special suit was used from, dress, gloves, glasses, mask and boots.

Soil Sampling

Inspector device was used to locate the contaminated spots, three sites locations were visited, the first location was in Nassireya which were contained destroyed tank, measurements were taken for the penetrators hole in tank and for the whole tank body, and for the soil inside the tank, soil and pieces of metal were collected from there.

The second location in Nssireya near (Ur company), the location was used for collection parts of destroyed tanks and vehicles. Soil and large pieces of metal were taken from the location.

Third location in Amara (kumait), the visited location; contains destroyed tank, the soil sample and pieces of metals were taken. The three locations were far from the public homes in the two cities.all locations shown in Fig.(1,2) and three respectively.

The soil samples were collected with a depth of (0-30 cm) and put in a strong nylon bags (5 Kg), the weight of samples were about (1-4 kg).

Fig.(1) :Location (1).

Fig.(2) : Location (2).

Fig.(3) : Location (3).

Lab. Work

The soil samples were dried by exposing to air for four days as recommended [7] with respect to good store conditions. After drying soil samples, the impurities (like small metal pieces and plants) were removed and then crushed and sieved to about 2mm diameter [8,9] before the beginning in the treatment work, the samples were left for 28-30 days to reach the equilibrium state for the radionuclides that exist in soil [7,10].

Experimental Procedure for uranium concentration measurement in soil

The soil samples were prepared as previous suggesting .0.5 g of soil samples were mixed with 0.1 g of methylcellulose powder ($C_6H_{10}O_5$) used as a binding material. The mixture was pressed by using a mechanical compressing device with force equal to (5 tons) into a pellet of 1 cm diameter

and 1.5mm thickness.

The pellets were covered with CR-39 detector and put in a plate of par affin wax at a distance of (5 cm) from neutron source (Am-Be), with flounce of thermal neutron (3.024×10^9 $n.cm^{-2}$) and flux (5×10^3 $n.cm^{-2}.s^{-1}$), to obtain induced fission fragments from the

After the irradiation time (7 days), (CR-39) detectors were etched in (6.25N) NaOH solution at temperature of 60 ^oC for (6 hr), then the induced fission tracks density were recorded using the optical microscope.

The metal's samples were cut in small pieces and irradiated as the above procedure for soil. The density of fission tracks (ρ) in the samples was calculated according to the following relation [11].

Track detectors (ρ)=Average number of total pits(tracks)/Area of field view.

The uranium concentrations in soil samples were measured by comparison between track densities registed on the detectors of the sample pellet and that of the standard geological sample pellets from the relation [12, 13, 14]:

Cx(sample)/ ρx(sample)=Cs(standard)/ ρs(standard).....(1)

Cx=Cs.(ρx/ ρs).

Values of the standard concentration and densities were taken from the best fit of the straight line shown in Fig.(4).

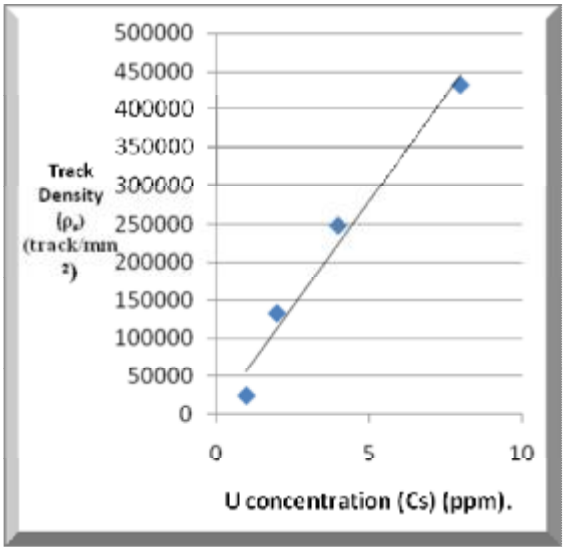

Fig.(4) : Relation between standard track density and standard U concentration.

Where:

Cx, Cs are the uranium concentration in unknown sample and standard respectively in ppm.

ρx, ρs are the track density of unknown sample and standard sample respectively in (track/mm^2).

Results and Discussions

Exposure Rate.

Inspector device was used in sampling procedure, the exposure rates for samples collected are shown in Table (1).

Table (1)
Exposure Rates

Samples	Exposure Rate (mR/hr)
S1	1.2
S2	0.12
S3	0.1
Ss1, Ss2, Ss3, Ss4 and Ss5	0.75
Various locations on tanks body	3.637,0.287 and 0.8

It appears from the results for soil samples, S1 (1.2 mR/hr) have the highest value, because it was collected from inside the tank.

For the metal samples it was very hard to take the metal near the penetrated surface, because of its high contamination, so the metal pieces collected were found burying in soil, inside and outside the tank.

Measurements for some spots in tanks body were taken, the highest value of the spots were near the penetrated surface (3.637 mR/hr).

All the results exceeded the allowed level of radiation exposure rate (35 mR/y or 0.004 mR/h) [15], this value differs from place to another depending on the geographical nature of regions and soil. The results were exceeded the allowed value for hundred times (180-900) times. This exposed the receptors (who will be

near this soil or tank metal for a long time) to danger on their life.

The very high values have been taken from places near the damaged region in the tank and the relatively low were from places far from the tank.

Dispersion of Radionuclides Pollutants Calculation

Radionuclides pollutants exposed to natural phenomena (winds & rain) which lead to transportation from its original location to other locations and contaminated large areas with time.

It was important to make (reverse analysis) to the uranium concentration data to find the expected original concentration values (concentration at 2003), by using the equation [3]:

$C_o=C_1+C_2\,xn+C_3xn$ .....(2)

where

C_o= Original expected concentration.

C_1= Samples concentration in 2003.

C_2= Concentration dispersed yearly by Winds.

C_3= Concentration dispersed yearly by Rains.

n= no. of years.

The pollutants dispersion was calculated in the three sites, the concentration unit was converted to µg/m^2 by using the specific activity as the equation below:

$Cx1= [C_o/Sp.A]x\ \rho_{soil} x\ D_p\ x\ 10^6$(µg/gm) .....(3)

Where:

Cx1: Radionuclide Concentration (µg/m^2).

C_o: Original Radionuclide Concentration (Bq/kg).

Sp.A: Specific Activity for U = 12354 Bq/gm [3].

ρ_{soil}: 1171.2 Kg/m^3 (calculated in laboratory).

Dp: Soil Depth = 0.3m.

Soil Contamination

Radioactive contamination concentrated in the surrounding of destroyed tank where about 70% of DU converted to aerosols dispersed in this region [16]. This region was about 8 m^2, its width about 5 m.

Dispersion of Pollutants by Winds (Saltation &Creep) and (Suspension).

In this region of Iraq the soil have a high sand percentage. For this reason the movement by (saltation & creep) represent 75% from total movement and for (Suspension) about 25%[17,18].

With loss of (43.2 Kg) for 1m per year, while the annual movement was about 75m [19, 20]. The calculation of dispersion by winds was performed as follows:

The concentrations for the three sites were converted from Bq/kg to µg/m^2 using eq (1), named with C×1 and for the same three samples concentrations in (µg/Kg), named with C×2.

For one year:

$C\times3$ (µg) = 43.2 (Kg/m^2/y) × 8 m^2 × C×2 (µg/Kg)..... (4)

$C \times 4 = 0.25 \times C \times 3$..... (5)

$C \times 44 = C \times 4/8$..... (6)

$C \times 5 = 0.75 \times C \times 3$..... (7)

$C \times 55 = C \times 5/8$..... (8)

Cxx (µg/m^2) = Cx1-Cx44-Cx ..... (9)

Where:

Cx3: Total amount of soil lifted by wind per year (µgm).

C×4: Soil lifted by suspension (µg).

C×44: Soil lifted by suspension (µg/m^2).

C×5: Soil lifted by saltation and creep (µg).

C×55: Soil lifted by saltation and creep, (µg/m^2).

Cxx: Concentration after wind effect.

The concentrations of DU obtained were 15.822, 8.252 and 5.712 ppm in 2003 reaching at 2007 with concentrations of 12.284, 6.406 and 4.434 ppm for site 1, site 2 and site 3 respectively. It was regarded that uranium concentrations on the soil surface decreases gradually from 2003 to 2007 in the three sites, Figs. (5, 6) and (7). As these calculations the expected concentration of DU (in 2008) on the soil surface will be less than the accepted level of 0.3 to 11.7 ppm of depleted uranium [21].

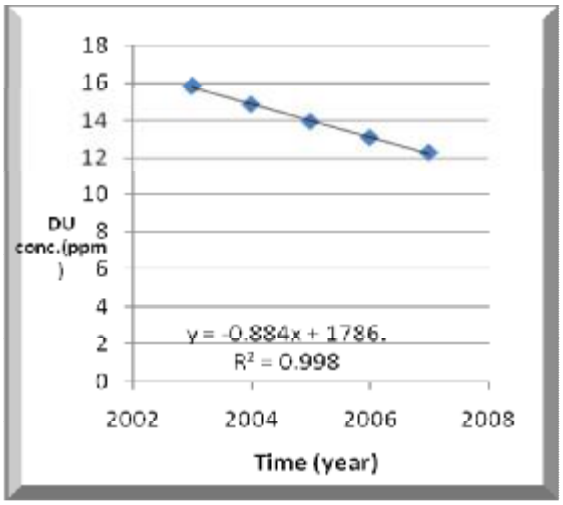

Fig. (5) : Relation between DU concentration & time (wind effect , site 1).

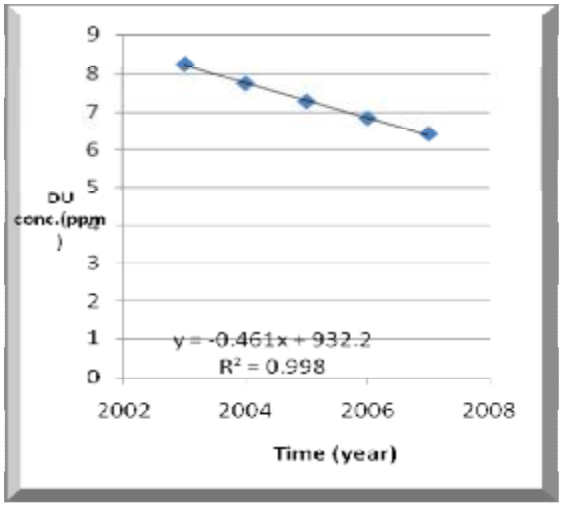

Fig.(6) : Relation between DU concentration & time (wind effect , site 2).

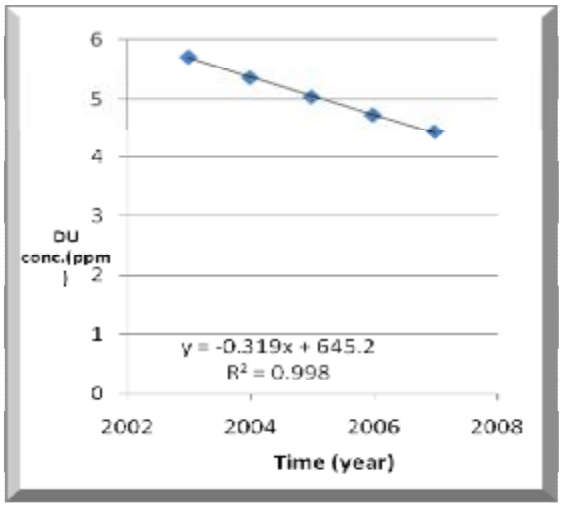

Fig. (7): Relation between DU concentration & time (wind effect, site 3).

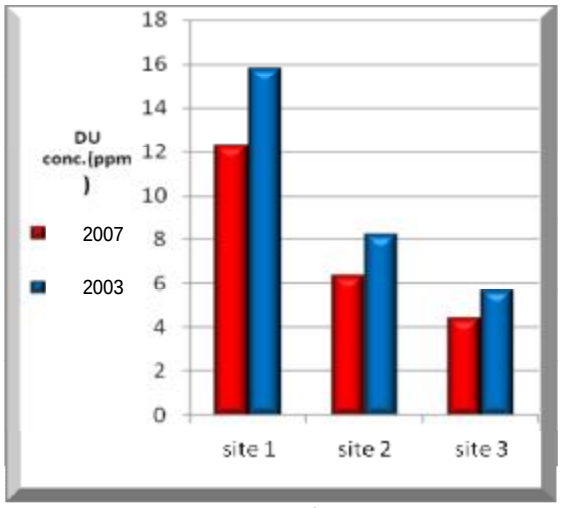

Fig.(8) : The removal of Depleted Uranium (wind effect) in the three sites.

Migration of Pollutants in Soil Profile.

The concentration of migrated uranium was calculated by using eq.(11),assuming a single radionuclide of uranium [3]

$$C_{i,j+1} = \left[\left(\frac{-V.\Delta t}{\Delta X.Rf}\right) - \left(\frac{\lambda.\Delta t}{Rf}\right) + 1\right] C_{i,j} - \left(\frac{V.\Delta t}{\Delta X.Rf}\right) C_{i-1,j} \quad \text{..... (11)}$$

where

ΔX= space segment.

Δt= time interval.

$Rf = (\theta + \rho K)$

i = Represent the location.

j – Represent the time.

where the soil profile represented by a column of (ΔX = 0.3 m), (Δt = 1 year), (ρ_{soil}=1171.2 Kg/m^3), (θ =0.2 ml/ml) and (k=1230 ml/g) [3]. The concentrations were found to be 15.822 ppm, 8.2ppm and 5.712 ppm in 2003. In 2007 the concentrations were 15.489 ppm, 8.078 ppm and 5.591 ppm for site 1, site 2 and site 3 respectively.

The results showed that the concentrations were also decreased from 2003 to 2007 for the three sites, Figs. (9, 10) and (11).

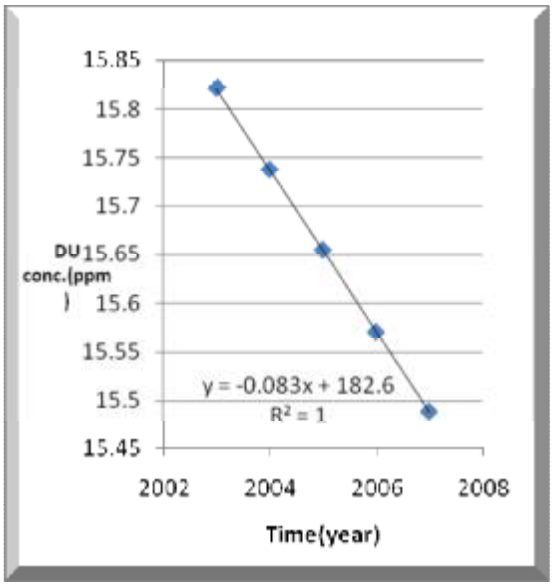

Fig.(9) : Relation between DU concentration and time (rain effect, site 1).

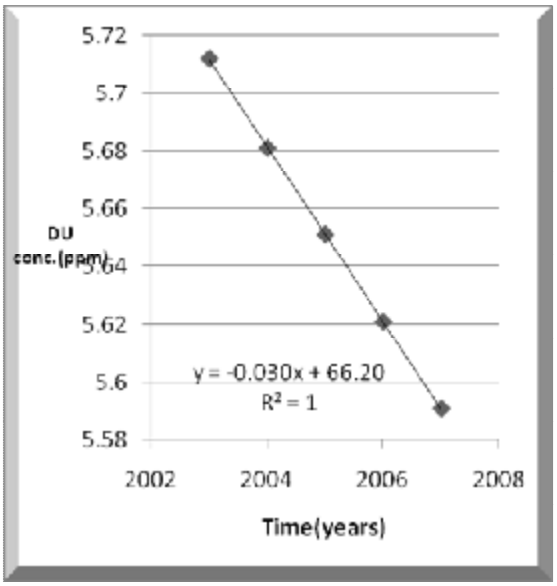

Fig.(11) : Relation between DU concentration and time (rain effect, site 3).

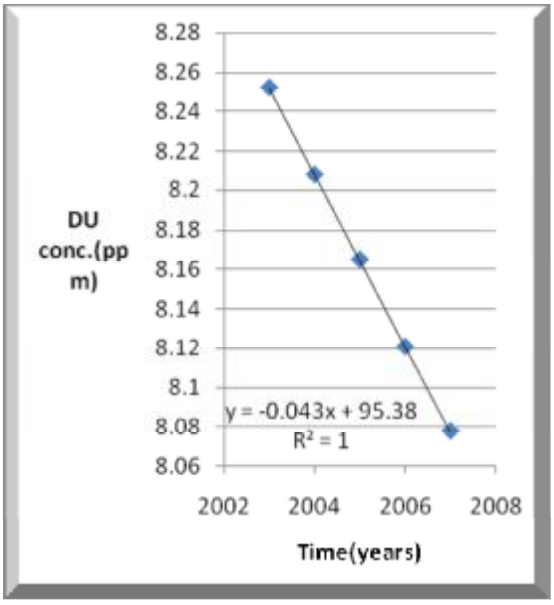

Fig.(10) : Relation between DU concentration and time (rain effect, site 2).

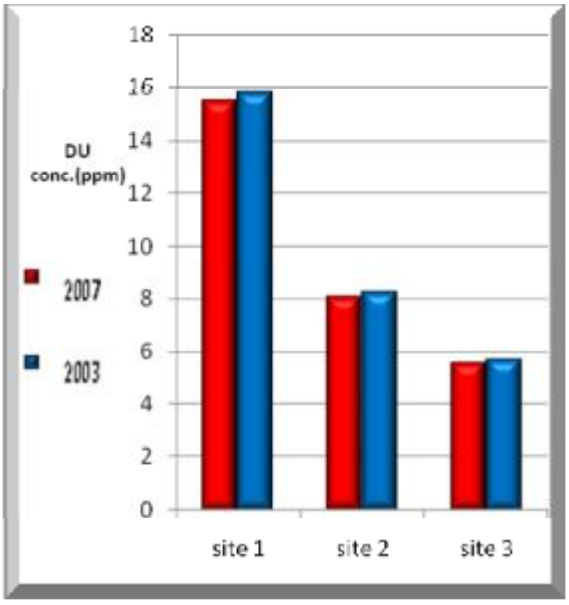

Fig.(12) : Overall removal of Depleted Uranium for three sites (rain effect).

The very slight decrease in DU concentration from the soil surface was observed because of the soil nature (i.e., high adsorption for the nuclide and alkaline condition (pH ≥7,[3])), where the uranium came in contact with organic matter and minerals(less than 100μm) the uranium will initially be sorbed to these minerals and organic matter particularly in PH range (6-8) [1]. After years the contamination might reach

the ground water. The overall removal of uranium by (wind and rain) are shown in Fig.(13).

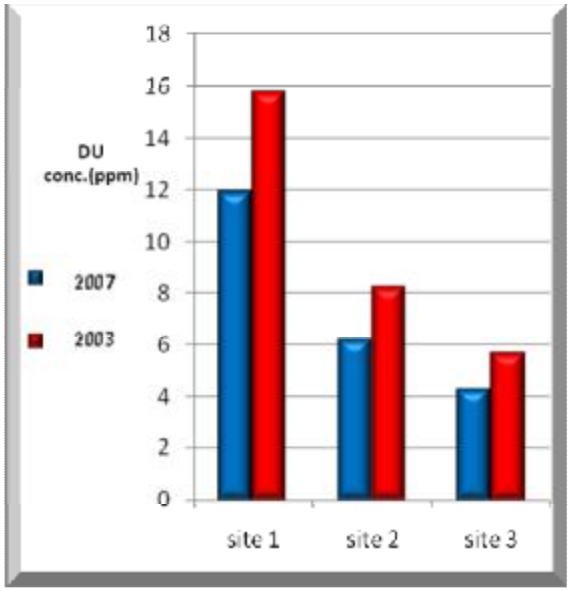

Fig.(13) : Overall removal of uranium by (wind & rain).

Soil and Metal Samples.

Three soil samples and five metal samples were detected by nuclear track detectors method, the results are shown in Table (2).

Table (2)
DU concentrations in different samples.

Sample	Description	Conc.(ppm)
S1	Soil Sample(Nassireya),Site1	12.52
S2	Soil Sample(Nassireya),Site2	6.53
S3	Soil Sample(Amara).	4.51
Ss1	Metallic surface	4.78
Ss2	Metallic surface	6.19
Ss3	Metallic surface	6.5
Ss4	Metallic surface	6.16
Ss5	Metallic surface	6.75

S1 have the highest concentration (12.52 ppm) because it was collected from inside the destroyed tank, most of metal samples have approximately the same value of DU concentration (about 6.5 ppm).

Conclusions

The study of DU exposure rates and mathematically dispersion in soil caused by wind and rain for three Iraqi sites, showed that the exposure rate were hundred times higher than the allowed limit and that will lead with the time for any receptor (who exposed continuously to this soil) to health damages, exposure rates were from (0.75 to 3.6 mR/hr).

The DU concentration in soil had the highest value in Nassireya (12.52 ppm), and approximately equal value (about 6.5 ppm) in metal pieces.

While for the dispersion of DU in soil the results were indicated the decrease in DU concentration on soil surface gradually from 2003 to 2007 by the wind effect. The DU concentration were decreased from 15.82 to 12.28ppm, from 8.25 to 6.4ppm and from 5.712 to 4,43ppm at site1, site2 and site3 respectively.

The rain effect was not significant because of high adsorption of this kind of soil in south region of Iraq in addition to its alkaline nature (pH $\geq$7), the DU concentrations were decreased from 15.822 to 15.48 ppm, 8.25to 8.07 ppm and 5.71 to 5.59 ppm at site1, site2 and site3 respectively. The DU concentration will be less than the allowed level in 2008.

References

[1] United Nation Environmental Program, UNEP "*Depleted Uranium in Bosnia and Herzegovina*" Post Conflict Environmental Assessment, 2003.

[2] Jones R.L., "*Uranium in Illinois surface soils*". Soil Science Society of America Journal, Vol.55, No.2, March-April, 1991. pp.549-550.

[3] وليد مجيد رشيد الحلي,"تاثير استخدام الأسلحة الإشعاعية على التربة والهواء في مناطق منتخبة من جنوب العراق", اطروحة ماجستير,1998 .

[4] محمد ، شذى عبد الحسين ، "إيجاد تراكيز المواد المشعة في التربة باستخدام تقنية التحليل الطيفي لأشعة كاما" ، أطروحة ماجستير – كلية التربية / ابن الهيثم – جامعة بغداد ، 1996 .

أسلحة اليورانيوم المنضب على الإنسان والبيئة في العراق, 2002 و ص139- 145.

[16] Depleted Uranium Education Project; "*Metal of Dishonor*"; International Action Center; New York, 1997.

[17] The U.S. Department of Agriculture; "*Soil, the 1957 yearbook of agriculture* ";U.S. Government printing office; 1957.

[18] Chepil, W.S.; Woodruff, N.P.;" *The Physics of Wind Erosion and its Control*"; Advances in Agronomy; Vol.15; 1963, PP 211-302,.

[19] Bagnold, R.A.; "*The Physics of Blown Sand and Desert Dunes*"; Mathues andCo. Ltd.;UK; 1960.

[20] سحر نافع شاكر,"جيومورفولوجية الكثبان الرملية للمنطقة المحصورة بين الكوت - الديوانية – الناصرية", رسالة ماجستير,كلية العلوم,جامعة بغداد, 1985.

[21] De Jesus, A.S.M.;" *The Behavior of Radium in Waterways and Aquifers*" IAEA; Vienna, 1984. PP 87-115.

الخلاصة

الهدف من البحث هو قياس التعرض الأشعاعي في ثلاث مناطق منتخبة من جنوب العراق (موقعين في الناصرية وموقع في العمارة) الناجمة عن وجود اليورانيوم المنضب في التربة وقطع معدنية مأخوذة من دبابة مدمرة, وحساب تراكيز اليورانيوم المنضب وأنتشاره من سطح التربة بتأثير الرياح وألأمطار للفترة من(2003-2007). الجرع الأشعاعية تم حسابها بأستخدام جهاز ال(Inspector) .

وتم الكشف عن اليورانيوم المنضب في نماذج التـرب والقطع المعدنية بأستخدام كواشف ألأثـر النـووي فـي الحالـة الصلبة. الرياح وألأمطار أخذت بنظر ألأعتبار فى حساب تـأثير ألأنتشار على تركيز اليورانيوم المنضب في التربة, حيـث تـم أحتساب تأثير الرياح نسبة الى طبيعة المنطقة وظـروف التربـة وأحتسب تأثير الأمطار نسبة الى معادلة ألأنتشار في التربة.

كانت نتائج الجرع ألأشعاعية عالية قرب منطقـة أختـراق أطلاقة اليورانيوم, وكانت معتدلة وقليلة نسبيا في نماذج التـرب والقطع المعدنية.

أعلى تركيز لليورانيوم المنضب في نماذج التربة كانت فـي محافظة الناصرية. وأن هذه التراكير ستكون أقل مـن مسـتوى الخطر بحلول عام 2008.

[5] Tawfiq N.F., Al-Ani N.H.K., Al-Jafari R.A.H. and Hani H., "*Determination of a-particle emitters concentrations in the soil samples around Tuwaitha site using plastic track detector CR-39*". Journal of college of education–University of Al-Mustansiryah, No.1, 1999, 261-264.

[6] ندى فاضل توفيق ، "دراسة مقارنة لبعض كواشف الأثر النووية العضوية واللاعضوية وتطبيقاتها في البيئة". أطروحة دكتوراه – كلية التربية – الجامعة المستنصرية 1996 .

[7] Leung, K.C.; Lau, S.Y.; Poon, C.B.; "*Gamma Radiation Dose From Radionuclides in Hong Kong Soil*"; J.Environ Radioactivity, Vol.11, 1990, PP 279-290.

[8] Lin, Y.; Lin, P.; Chen, C.; Haung, C.;"*Measurement of Terrestrial & Radiation in Taiwan, Republic of China*" Health Physics, Vol.52, No.6, Health Physics Society; June,1987,PP 805-811.

[9] Mollah, A.S.; Rahman, M.M.; Husain,S.R.; "*Distribution of γ-emitting Radionuclides in Soils at the Atomic Energy Research Establishment, Saver, Bangladesh*" Health Physics; Vol.50, No.6, June,1986, PP 835-838.

[10] Kataoka, T.; Ikebe, Y.; Minato, S.; Ishida, K.;"*Detailed Evaluation of Natural Gamma-Radiation Field due to Uranium(U-238) Series*"; Journal of Nuclear Science and Technology; Vol.19; No.6,June,1982, PP482-490.

[11] Amald, O., Custball, N.H and Nielsen, G.A. "*Cs-137in Montarq Soils*", Health Physics, 57 No.6 ,1989 ,PP 955-958.

[12] Fleischer, R.L. , Price, P.B. and Walker, R.M." *Nuclear Tracks in Solids: Principles and Applications*", Pergam-mon press, Berkeley, 1975.

[13] Durrani, S.A. and Bull, R.k."*Solid State Nuclear Tracks Detection: Principles, Methods and Applications*", Pergammon press, UK, 1987.

[14] Bansal, V.,Azam, A. and Prasad, R."*Health Physics*", Vol. 27, (1989), PP 985.

[15] د. بهاء الدين حسين معروف " النشاط الإشعاعي الطبيعي قي العراق " المؤتمر العلمي عن آثار استعمال