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Research article/Araştırma makalesi

Soil radioactive pollution in Falluja-Iraq

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Abstract

Applying the CR-39, Solid State Nuclear Track Detector, the radioactivity of the soil in Fallujah City was measured at the ground depths 0-20, 20-40 and 40-60 cm respectively. The soil samples were collected during summer 2014. At the surface, 0-20 cm depth, the measured radioactivity corresponding to depleted uranium concentration of 1.1-2.5 ppm, showed lower values as compared with the tolerated average value of 2.8 ppm (DU). The detected activity values at the 20-40 cm and 40-60 cm depths were not negligible, (1.0-2.3) ppm and (0.9-2.0) ppm respectively. The results indicate the diffusion of the radioactive material, accepted as depleted Uranium, down the soil layer within the 3 levels. Adding the ppm values at the three depths together yields radioactivity values of (3.1-6.8) ppm, which are greater than the IAE tolerated value of 2.8 ppm. The 'accumulated activity' represents the initial value for the contamination. The formation process dates back to the year 2005, of the 2nd Falluja confrontation. The period required for the diffusion down the soil extends over 9-10 years. The high numeric value of the radioactive contamination 3.0-6.8 ppm DU can permit us to understand the origin of the increase in cancer disease cases, women repetitive abortions, malformations and generic deformations of newly borne babies, following the year 2005, as reported by the health administrations of the city.

Key words: Falluja, Iraq, soil, radioactivity, CR-39

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Falluja-Irak'ta toprak radyoaktif kirliliği

Özet

CR-39, Katı Hal Nükleer Yol Dedektörünü uygulayarak, Felluce Şehri'ndeki toprağın radyoaktivitesi, sırasıyla 0-20, 20-40 ve 40-60 cm'lik toprak derinliklerinde ölçülmüştür. Toprak örnekleri 2014 yılının yaz mevsimi boyunca toplanmıştır. Yüzeyde, 0-20 cm derinliğinde, 1.1-2.5 ppm'lik tükenmiş uranyum konsantrasyonuna karşılık gelen ölçülen radyoaktivite değeri, tolere edilen 2.8 ppm ortalama değerine kıyasla daha düşük değerler göstermiştir. 20-40 cm ve 40-60 cm derinliklerinde tespit edilen aktivite değerleri, sırasıyla, 1.0-2.3 ppm ve 0.9-2.0 ppm olarak göz ardı edilemezdi. Sonuçlar, tükenmiş uranyum olarak kabul edilen radyoaktif malzemenin, 3 kattaki toprak tabakasında aşağı doğru yayıldığını göstermektedir. Üç derinlikte ppm değerlerinin toplanması, 3.1-6.8 ppm'nin radyoaktivite değerlerini verir, bu da 2.8 ppm'lik IAE tolere edilen değerden daha büyüktür. "Birikmiş etkinlik", kirlenmenin başlangıç değerini temsil eder. Formasyon süreci, 2. Felluce çatışmasının 2005 yılına kadar uzanmaktadır. Toprak aşağı difüzyon için gerekli süre 9-10 yıl boyunca uzanır. Radyoaktif kontaminasyon 3.0-6.8 ppm DU'nın yüksek sayısal değeri, 2005 yılından itibaren, şehrin sağlık yönetimi dikkate alındığında, kanser hastalarında görülen artış, kadınların tekrarlayan düşükleri, yeni doğan bebeklerin sakat olması ve genel deformasyonlarını anlamamızı sağlayabilir.

Anahtar kelimeler: Felluce, Irak, toprak, radyoaktivite, CR-39

1. Introduction

The major sources of radioactive material are the products of nuclear fission and naturally occuring radionuclides. These are carried by the particles ranging from less than 1 μ up to a few mm. One of the main routes in their incorporation in to food chain are plants. Plants are the first organisms which get effected by the radioactivite pollution created by

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naturally produced radioisotopes and products of nuclear fission. They form one of the primary inlets in the food chain. However, it is very difficult to put forward the damages in these silent observers, unless the radioactivity levels exceed the natural ones. Radionucleid accumulation is seen to reach a very high level in a short time in lichens which are accepted as the best bioaccumulators. Lately much attention has been paid towards the researches concerning the determination of species which can be used as indicators in radioactive pollution. However, their damaging effects on the plants cannot be detected unless the levels are not above the natural ones. The sensitivity of the plants is related to the size of nucleus and growth form of the plants. Climatic conditions, particularly rain plays a great role in the deposition and removal of the radionuclides from the plants surfaces. (Ozturk et al., 1987, 1994).

Radionuclides existing in soil can be dissolved in solution, or ion exchanged in reaction, complexed with soil organics or precipitate as pure or mixed solids. Their immobility in uppermost soil layers represents a problem for environment and human health, due to their easy integration in the food chain (Cazzola et al., 2004; Gavrilescu et al., 2009). Igwe et al. (2005) have proposed a scheme for radionuclides movement in the soil, a major part is released into the environment which either accumulates in the upper layer of soils, inducing a risk for the ecosystems and health (Gavrilescu et al., 2009).

During the last six decades uranium mining and milling has lead to much damage to the environment through abandoned radioactive waste accumulation and its improper disposal, dumping of the wastes after uranium prospections. Large amounts of both high- and low-level uranium-containing wastes is generated from fuel fabrication and reprocessing as well as research and development producing negative effects. These influence the environmental quality mainly surface-ground waters, and soils. Its potential risk in soil contamination is a global problem. These also pollute large land areas endangerring the catchments of drinking water (Gavrilescu et al., 2009). Uranium also generates an important issue against human health (Gongalsky, 2003). Its solubility in soil depends on pH, redox potential, temperature, soil texture, organic and inorganic compounds, moisture and microbial activities (Rivas, 2005). Soluble forms move with soil water, which are absorbed by plants or aquatic organisms or volatilized (Igwe et al., 2005). In view of this, the contamination by uranium has severe negative biological effects on important groups of the soil food web (IAEA, 2005a, b). Its contamination in soil and water has been identified globally at many sites, therefore measures for preventing their assimilation by plants needs to be considered as the first step in the remediation of contaminated sites (Navratil, 2001; Gongalsky, 2003; Cazzola et al., 2004; Gavrilescu et al., 2009). Different *in situ* technologies like alkaline leaching with carbonate and hydrocarbonate ions, or acid leaching were used commercially in a large number of deposits (IAEA 2001, 2002a, b; Normon and Raforth, 1998; Groudev et al., 2007).

The political, economical and environmental reasons have lead to a stopping of all commercial-operations for uranium leaching (IAEA, 2005a, b). Inspite of these preventive and remedial actions many natural ecosystems have sufferred heavily from radioactive element pollution due to uranium recovery, mainly through the seepage of acid drainage waters (Savchenko, 1996; Gupta, 2006; Gavrilescu et al., 2009). Later are still a persistent environmental problem at many abandoned sites, because the soils alongside the water flow path are polluted with radioactive elements, and these have become unsuitable for agricultural use, as such soil remediation is an important task at such sites (AbdEl-Sabour, 2007). The depleted uranium too poses a problem at such places, because this has increased public health concerns due to the chemical toxicity of DU at elevated doses. There is need to develope U removal methods from contaminated sites (Gavrilescu et al., 2009). The remediation of radionuclide-contaminated soils is a matter of high priority. If these soils are left untreated, they will pose a great threat for human health and will be hazardous for our environment. For this purpose methods can be used to extract uranium from its ores to achieve remediation of contaminated soils and water (Merritt, 1971; Roh et al., 2000).

^{238}U, ^{235}U and ^{234}U are the three isotpes of uranium usually occurring in the soil. Their relative abundance is 9.27, 0.720 and 0.0055 percent respectively. Uranium is found in the soil (80-90%) in the +VI oxidation states as uranyl cations (UO_2^{+2}), a predominant species of U in soil under acidic conditions (Ebbs, 1997). Its behaviour in soil is similar to other heavy metals and toxicity mimics that of Pb. Later is toxic to kidneys and some insoluble U-compounds are carcinogenic (Ibrahim et al., 2015). The behaviour of U in soil is complex and the metal speciation (especially pH-dependent) is very difficult to investigate. According to Baes (1982) rare amounts of uranyl cations are present in available forms because of high solid-liquid distribution (Kd).

Not much is known about the uptake and translocation of uranium by the plants under diverse soil conditions. Generally U contents of plants growing under U-contaminated environments have been investigated (Whicker and Ibrahim, 1984; Ibrahim and Whicker, 1988). In some cases the uptake of U by field and garden crop having importance to humans and animals has been studied (Sheppard et al., 1985, 1989; Ibrahim et al., 2015). The soil to plant transfer is usually known as transfer factor (TF), which varies with plant species and plant part. The roots generally are reported to contain higher uranium than stems, leaves and shoots (Apps et al., 1988; Ebbs et al., 1998). Leafy vegetables too show higher U-TFs followed by root, fruit and grain crop plants. The TFs are reported to exceed a value of 0.01 rarely, with the exception of some plants growing on very highly contaminated (acidic) U-mining soils. The TFs of different crops also depend on the soil pH and some are very sensitive to pH. The sage brush has shown the highest TF under natural conditions rarely grown at pH below 4 (Ibrahim et al., 2015). The free UO_2^{+2} is most readily taken up and translocated by plants, being present at a pH of 5.5 or less, and acidification of uranium-contaminated sites is necessary for phytoextraction. This species is also responsible for binding soil solids with organic matter; as such a reduction in plant

uptake has been reported. Some soil amendments in addition to acidification may increase the availability of U by the formation of complexes (Sheppard et al., 1984; Ebbs, 1997; Ibrahim et al., 2015).

Fallujah is a city of ca 750,000 inhabitants, located about 70 km northwest of Baghdad, Iraq (Figure 1). It was the scene of heavy military operations in the years 2004 and 2005. In the years following these operations peculiar health abnormalities appeared among its population (Anonymous, 2017). In the following years high number of child blood cancer cases, genetically deformed malbirths, repetitive abortions of pregnant women and multifold increase in adult cancer cases were reported by the local health authorities (Anonymous, 2013). A thorough inspection of the causes of this catastrophic health situation was called for (Anonymous, 2012). For this purpose an intensive study of the soil contamination was needed. The soil analysis aimed primarily at the estimation of radioactive as well as heavy metal contaminations. The subject of this paper deals primarily with the soil radioactive contamination.

Solid state nuclear track detectors (SSNTD) are appropriate and economic device for the quantitative estimation of nuclear radioactivity (Fleischer et al., 1975). Both organic and inorganic materials are used for their production. The organic detectors are mostly composed of polymeric resins (Szydlowski et al., 1999). When exposed to radioactive material, they get chemically changed due to the collision with α or ß particles. The spots of collision appear on the resin sheet and can be counted applying a proper magnifying microscope (Durrani and Bull, 1987). Among the SSNTD, CR-39 seems to be most favored for such studies (UNSCEAR, 1994).

Figure 1. Location of Fallujah City in Iraq

2. Materials and methods

The soil samples were randomly collected from 50 different locations, designated relative to schools and mosques, in the city. From each location 5 different samples were collected from the following depths; 0-20, 20-40, and 40-60 cm. After removal of "garbage" material, the samples were dried under 105°C for 24 hours and then submitted for the radioactivity analysis. CR-39 SSNTD, Landauer, England; optical microscope with 400 x magnifying power, Nicon, Japan and Heraeus furnace with a temperature range of 0-250 °C were used during the evaluation of soil samples. The sensitive balance used was of the type Sartorious-BP 3015. ^{241}Am-Be neutron source of an activity 5.92 x 10^{11} Bq and a neutron flux of $5x10^3$ n $cm^{-2}.s^{-1}$ were used for the irradiation. 1 gm pellet of the sample material was prepared through pressing with a 15 atm. press. It was placed in front of a 1 cm^2 sheet of CR-39, both placed in the radiation container with the neutron source (Figure 2a). The neutron irradiation continued for 7 days, after which the CR-39 was removed and immersed in a (6.25N) NaOH bath at 60°C for 5 hours (Figure 2b). The tracks formed in the Cr-39 sheet were inspected applying the optical microscope. The measured track density (spot/cm^2) was used to evaluate the amount of radioactivity contamination, calculated in ppm of natural uranium according to a correlation curve (Figure 2c).

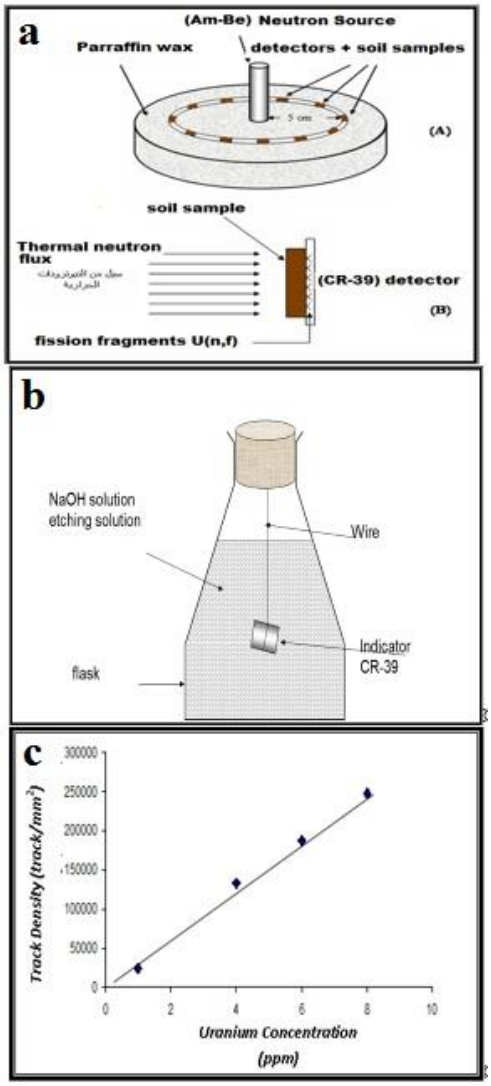

Figure 2. (**a**) the radiation measurement container applied for the estimation of soil radioactivity; (**b**) NaOH bath used for etching the irradiated Cr-39 sheets; (**c**) correlation curve of the measured track density, CR-39, with the Uranium concentration (ppm).[**)] Prof. N. F. Tawfiq, Dept. of Physics, Al-Nahrain University, personal communication

3. Results

The table 1 shows measurement results for most of the chosen sampling locations. The amount of radioactive contamination calculated as ppm values of Uranium, are lower than the internationally permitted values of 2.8 ppm (IAEA, 2005a, b), showing that at the depths of 20 and 40 cm the amount of contamination is not negligible.

Table 1. Measured radioactive contamination, calculated as ppm, Uranium, at some sampling sites in Fallujah

Sample Location	**Depth (cm) - U (ppm)**		
	20 cm	**40 cm**	**60 cm**
Alhhurriya School	1.118	0.952	0.786
Almaamun School	1.433	1.276	1.173
Aljamhurriya School	1.391	1.214	1.147
Alturath Alaraby School	1.251	1.048	0.873
Alkhaleel School	1.250	1.170	0.987

The low U-ppm values at the soil surface, 0-20 cm, suggest that the contamination in the city is not threatening and might be neglected, Figures 3a and 3b.

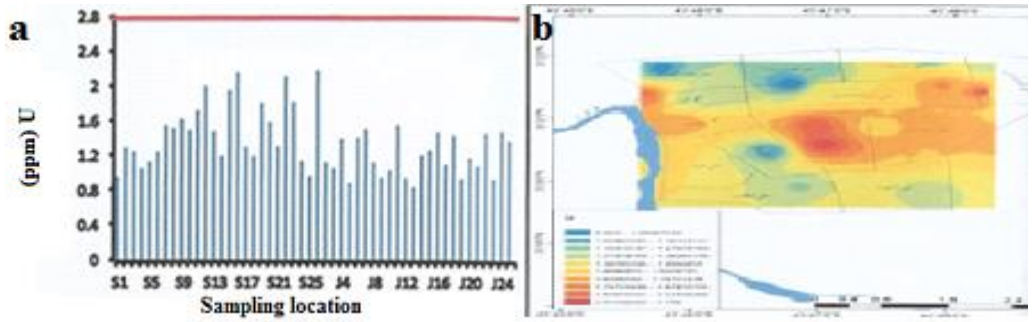

Figure 3. **(a)** Plots of the measured U concentration at 50 different soil sampling locations taken at the depth 0-20 cm; (**b**) Area **c**ontours of the measured U concentration at 50 different soil sampling locations taken at the depth 0-20 cm.

However, the situation is different on considering the sums of all three concentrations at the three depths (Figure 4). The calculated cumulative concentration values are all higher than the average tolerated (2.8 ppm) value of U (Table 2). Based on the cumulative concentration values we can conclude that the initial U concentrations, compelled in the years 2004-2005, were several folds higher than 2.8 ppm. Figure 5 describes the concentration change relative to the soil depths. The solid lines represent the measured concentration values, the dotted lines represent the calculated concentration values applying the gradient method. By this calculation we defined the concentration gradient as the difference quotient of the concentration (ppm) over the change in depth (cm).

C_i = the concentration at the depth d_i (1)
C_{i+1} = the concentration at the depth d_{i+1} (2)
$\Delta^c = C_i - C_{i+1}$ (3)
$\Delta^d = d_i - d_{i+1}$ (4)
$\blacktriangledown_{i-i+1} = \Delta^c / \Delta^d$ is the gradient of concentration change.
$C_{i+1} = C_i (1 - \blacktriangledown_{i-i+1})$ (5)

Table 2. Cumulative soil radioactive contamination values (ppm, Uranium) at some sampling sites in Fallujah

	Depth (cm) - U concen. (ppm)			
Sample Location	**20 cm**	**40 cm**	**60 cm**	**Sum**
Aljumhurriya school	1.391	1.214	1.147	**3.752**
Alturath alaraby	1.251	1.048	0.873	**2.299**
Alkhaleel School	1.250	1.170	0.987	**2.420**
Alzaytoona School	1.472	1.322	0.952	**2.794**
Alfalluja School	1.713	1.563	1.378	**4.654**
Alsuudad School	1.712	1.548	1.293	**4.553**

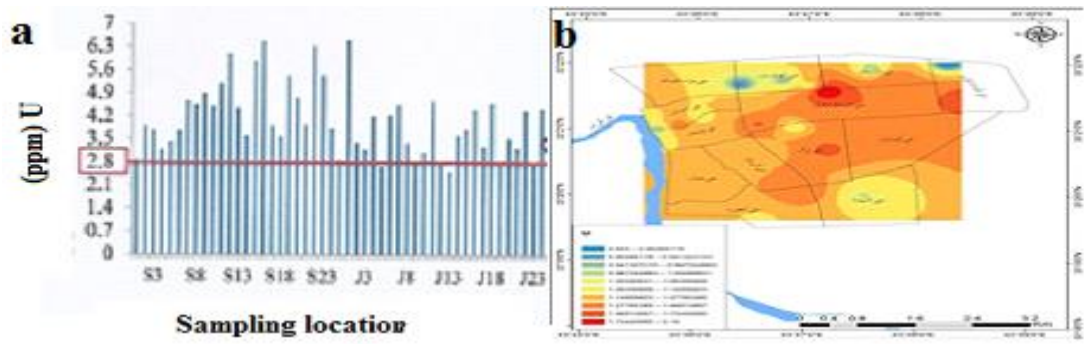

Figure 4. **(a)** Plots of the calculated cumulated U concentrations at 50 different soil sampling locations; (**b**) Area **c**ontours of the cumulative U concentration at 50 different soil sampling locations

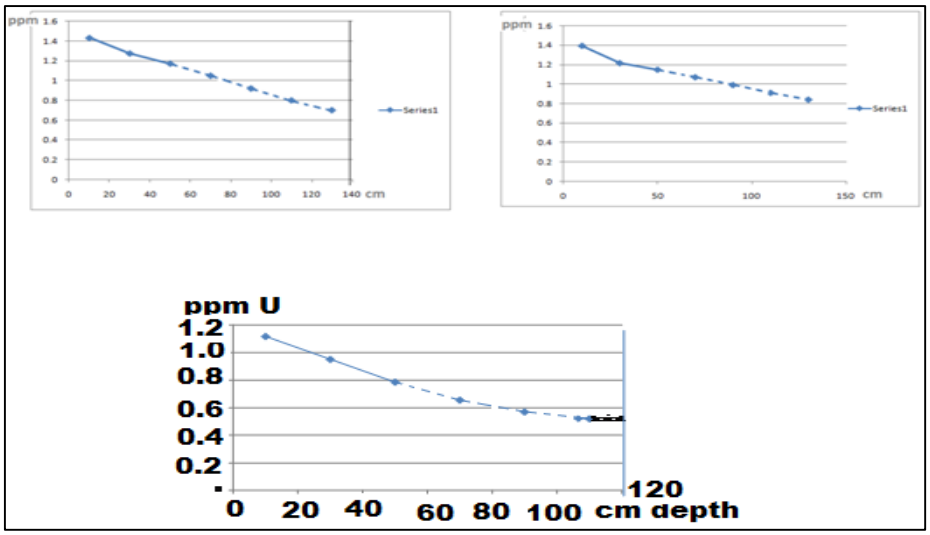

Figure 5. Measured and calculated U concentration as a function of soil depth (cm) from 3 different sample collection locations in Fallujah city

Two interesting results appear in these figures;

a- the diffused concentration values are not negligible even at the depths of 100 cm and

b- the concentration change due to the diffusion might extend to depths of several meters.

A thorough calculation of the total cumulative concentrations at different sampling locations should yield ppm values that are in multiple folds of the tolerated value (2.8 ppm). The cumulative concentration is by definition equivalent to the initial radioactive concentration (U-ppm) at the time of its initiation, i.e. in the years 2004-2005. Such radioactive contamination levels should be considered on discussing the peculiar health abnormalities that were reported by the Fallujah health authorities in the years following 2005.

The nuclear industry, which involved the mining, milling, and fabrication of various U products has been responsible for uranium contamination of surface soils. Its contamination now poses significant health risks for living beings and limits the future use of many sites formerly used for U production and processing. There are large areas of U-contaminated soils in the world, engineering-based remediation methods such as excavation require millions of tons of soils to be disposed of as low-level radioactive waste. This process is expensive, fills up scarce landfill space, and requires additional site restoration. Remediation of U-contaminated soils represents a significant expense to many industries and governmental agencies. According to Huang et al. (1998) development of a cost-effective method to remove U from contaminated soils could accelerate the cleanup process and reduce remediation costs.

4. Conclusions and discussion

Uranium sorption strongly dependends on the pH of the solution because of the changes in solution speciation, and surface species and surface charge as a function of pH (Ibrahim et al., 2015). Presence of uranium in soil generates an important issue against public perception on the risk which contamination poses to the environmental and human health. In spite of some preventive and remedial actions during the uranium recovery, many natural ecosystems have got heavily polluted with radioactive elements (Gavrilescu et al., 2009).

Our results clearly show that significant radioactive contamination has occurred in the city of Fallujah during the years 2004-2005. Diffusion of the radio-active material has taken place into the depths of its soil in the following years. The diffusion is expected to proceed to several meters in depth and will reach the ground water level. Monitoring of the radioactivity of the ground water of the city and its surroundings is important and strongly called for. It is expected that similar diffusion behavior of radioactive contamination in other areas of Iraq must have taken place, therefore continuous monitoring of the activity is needed in Iraq and its neighbouring countries.

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