Short- and Long-Term Behavior of Depleted Uranium In the Environment of Southern Region of Iraq

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Abstract

The Iraqi environment had been extensively exposed to depleted uranium (DU) contamination in the course of military operation in 1991 and 2003. Burning of tanks and armored vehicles led to the formation of large quantities of fine aerosol containing predominantly poorly soluble uranium oxides. The DU aerosol is deposited on soil surface, transported far from the vicinity of the target, or resuspended in the air by the wind action. Therefore, data from within the country on the behavior of DU in the environment are essential and would offer first-hand basis on many topics of the impact of DU contamination on the environment and health. In the 1st part of the study nearly ten years after military operation, transport of DU from contaminated soil of the southern region of Iraq had been investigated in undisturbed soil columns taken from four locations of battlefield at Basra Governorate. In the 2nd part of the study, uptake of DU was assessed in a field experiment utilizing tomatoes. The plots were mixed with a contaminated soil and the recommended agricultural practices were used during the growing season. At the end of season, samples of the aboveground and belowground were digested and analyzed for their content of DU. Samples of soil, effluent, and plant were analyzed for DU using Solid Scintillation and Liquid Scintillation Counters. Results indicated that no DU had been detected in the eluted samples even after heavy leaching with a variety of displacing solutions. At the same time, no DU had been migrated from soil to above- and below-ground parts of the tomatoe plants. These data suggest that the DU occurred in soil as metal or oxides remains as insoluble forms even after ten years of weathering under desert conditions. It is expected therefore, that the wind action is the main mechanism dominated the transport of DU in the environment of southern region of Iraq.

Keywords: depleted uranium, southern Iraq, desert areas, military munitions, soil columns, leaching, plant uptake.

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1 Introduction

The Iraqi environment had been extensively exposed to depleted uranium (DU) contamination in the course of military operation in 1991. It is expected that additional contamination had occurred in 2003. Details about the quantity, nature of weapons fired and locations of hit targets are found in numerous studies [e.g., 1, 2, 3]. Some sources suggest that nearly 300 tons of DU ammunitions was dropped in the aircraft rounds and tank-fired shells in southern Iraq over an area of about 20,000 km² during the 1991 Gulf War [4]. It is believed that information about 2003 Gulf war have not been fully disclosed.

Depleted uranium is highly toxic and radioactive byproduct of the uranium enrichment process and has a half-life of 4.5 billion years. Burning of tanks and armored vehicles led to the formation of large quantities of fine aerosol containing predominantly poorly soluble uranium oxides. The DU aerosol is deposited on soil surface, transported far from the vicinity of the target, or resuspended in the air by the wind action [5]. When a DU bullet impacts a hard object (armour or rock), it is crushed into fragments, burned (18-70%) and oxidized into dust. Uranium oxides (U_3O_8 , UO_2 and UO_3) are being formed. The latter oxide is the only one soluble in water, forming uranyl (UO_2)2+ ions. The oxide aerosol resulting from impact has 50-96% of respirable-size particles (diameter less than 10 mm) and 17-48% of these particles is soluble in water [6].

Just as natural weathering causes metallic iron to crumble and rust, uranium spontaneously oxidizes when in contact with air and water, either in the atmosphere or within the pores of soils and sediments, in the chemical process known as corrosion [7]. The extent and end-products of this chemical weathering depend on several site specific factors, including the chemical composition and size distribution of the impact particles, precipitation, humidity, as well as soil properties such as pH and moisture content. Soils that are high in organic matter are observed to possess a higher affinity for uranium, thus causing less uranium to be released into the groundwater and plant roots [7].

A soil's affinity for uranium is characterized by its uranium 'sorption' (a term that refers to processes occurring at the solid-solution interface), which ultimately affects the extent of potential plant uptake and groundwater contamination [7]. It is known that the pH of soil affects its uranium sorption: Uranium is more mobile in alkaline soils than in acidic soils due to the formation of stable oxy-anions with oxygen and carbon [8]. Soils that are high in organic matter are observed to possess a higher affinity for uranium, thus causing less uranium to be released into the groundwater and plant roots [7]. Semi-arid soils have less affinity for uranium than moist soils, increasing uranium mobility from the surface to deeper soil.

In field monitoring of depleted uranium deposits, Crançon et al. [9] indicated that uranium retention in soil is controlled by the <50 micron mixed humic and clayey coatings in the first 40 cm. In the same study even after 100 pore volumes elution, 60% of the total input uranium is retained in the first 2 cm of the column. Retardation factor of uranium on E horizon material ranges from 1300 (column) to 3000 (batch). Other studies have shown that Kd values for uranium can range over several orders-of-magnitude and are highly dependent on important characteristics of the soil such as clay content, texture, pH, and the amount of organic material [10, 11]. Therefore, uranium showed slow migration and soils contaminated with depleted uranium is difficult to remediate, and thus pose a long-term potential risk to the environment and humans. In this respect, Langmuir [12] showed that in the near-surface environment, a soil's chemical and physical

characteristics, including pH, redox potential (Eh), cation exchange capacity, (CEC), pCO2, amount and type of dissolved solutes, organic matter, porosity, and permeability control depleted uranium behavior and distribution. Sorption of uranium onto clays or other mineral surfaces can impede its mobility in sediments. In reducing environments, uranium (IV) is generally insoluble. In oxidizing conditions however, uranium corrodes rapidly, forming U(VI) compounds that for the most part, are fairly soluble and mobile [13]. However, in unsaturated soil U(VI) in the presence of arsenates, vanadates, silicates and phosphates, forms insoluble minerals [12]. In examining environmental monitoring data of ground and surface water, soils, and surface water sediments collected from 1992 to 2004, results show limited transport of depleted uranium within the impact area and no transport at the installation boundary [14].

Uranium behavior in soils is controlled by actions and interactions between physico-chemical and biological processes that also determine its bioavailability [15]. Plants are known to absorb uranium through their roots, and soil sorption affects the rate of uptake in a complex nonlinear manner which cannot be understood without specific site data. On the one hand, a high uranium sorption rate corresponds to more uranium trapped within the upper soil layer, and therefore more uranium accessible to a plant's root structure [7]. However, for highly immobilizing soils, plants absorb uranium more efficiently through their leaf stomata, in which case resuspended surface uranium in the atmosphere could dominate [16].

In greenhouse experiments, six plant species were tested for their ability to accumulate depleted uranium in their aboveground biomass from deployed munitions contaminated soil [17]. They found an increase in leaf and stem DU with the application of citric acid and glyphosate; however, the application of manure generally decreased or had no effect on DU uptake. In a field experiment nearby uranium mining activities, Neves et. al. [18] found that the uptake of uranium by lettuce tissues was positively correlated with soil uranium content, but non-significant differences were obtained from contaminated soils irrigated with different water quality. Also, lettuce bioconcentration is more related to available uranium species in water than to its uranium concentration. In assessment of uptake of depleted uranium derived from weathered munitions in a greenhouse experiment utilizing three common grass species, Meyer et al. [19] observed that concentration ratios (plant tissue DU concentrations divided by soil DU concentrations) decreased with increasing soil DU concentrations, but increased as more moisture was applied. Al-Kharouf et al. [20] in studying soil-plant transfer factors found that the green parts (leaves, stems and roots) of the studied crops tend to accumulate uranium about two orders of magnitude higher than the fruits. Ulery et al., [21] screened plant species for potential phytoextractors (Phytoremediation) that could accumulate uranium from soil in arid and semi-arid regions. They found that none of the 50 species tested would be considered hyperaccumulators. Stojanovic et. al., [22] found a linear dependence between the content of uranium in soil and in corn plants tissue indicates that corn could be used for phytoremediation of uranium-contaminated soils.

Therefore, data from within the country on the behavior of DU in the environment are essential and would offer first-hand basis on many topics of the impact of DU contamination on the environment and health. The objectives of the study were to investigate (i) the transport of DU in soil columns taken from areas of southern Iraq that were contaminated with DU, and (ii) the uptake of DU by tomatoes plants in field experiment conducted at the desert area of southern Iraq.

2 Materials and Methods

2.1 Study Site

The investigated area is part of southern desert of Iraq and an extension of Syrian Desert and known as Al-Hajarah in the western part and as Al-Dibdibah in the east. Al-Hajarah has a complex topography of rocky desert, wadis, ridges, and depressions. Al-Dibdibah is a more sandy region with a covering of scrub vegetation. Elevation in the southern desert averages between 100 to 400 meters. Geologically, the desert region consists of undisturbed limestone with some shale of Cretaceous, Eocene, Miocene, and Pliocene origin. This is a part of the Arabian Shield, which forms the western part of the Arabian Peninsula. The oldest (Cretaceous) part of this shield in Iraq is at the southwest corner (Rutba District) surrounded by Eocene limestone and marls. The only extensive zone of sand dunes in Iraq stretching over 270 km lies from south of Najaf in the south to west of Basra in the southeast, forming a belt of dunes 15 to 20 km wide. The climate is semi-arid with average temperatures range from higher than 48 $^{\circ}$ C in July and August to below freezing in January. Most of the rainfall occurs from December through April and averages between 100 and 180 mm annually. The summer months are marked by two kinds of wind phenomena. The southern and southeasterly, a dry, dusty wind with occasional gusts of 80 kilometers per hour (50 mph), occurs from April to early June and again from late September through November. This wind is often accompanied by violent dust storms that may rise to heights of several thousand meters. From mid-June to mid-September the prevailing wind is from the north and northwest and steady.

The desert soils of Iraq are calcareous, usually gypsiferous, often with a cover of recent unsorted material as a result of wind erosion, and contain a low organic content (generally below 0.5%). The soils are often covered with a thin layer of gravel, or with a gypsum or limestone crust. The subsoil starts at a depth of a few centimeters, and has high lime content. Essential physical and chemical characteristics of the soils are given in Table 1 measured using standard procedures [23]. Some locations are cultivated to vegetables (cucumber, tomato, pepper, and many others) using the groundwater as source for irrigation. The groundwater levels nearly 15-20 m below ground surface.

| | Locations | | | | |
|-----------------------|------------|------------|------------|---------------|--|
| Character | (1) Safwan | (2) Jabal | (3) Lihase | (4) Centre of | |
| | | Synam | | Destroyed | |
| | | | | Tanks | |
| Sand (g/kg) | 810 | 812 | 780 | 815 | |
| Silt (g/kg) | 115 | 118 | 132 | 122 | |
| Clay (g/kg) | 75 | 70 | 88 | 63 | |
| Textural Class | Loamy Sand | Loamy Sand | Loamy Sand | Loamy Sand | |
| Salinity (dS/m) | 2.8 | 2.5 | 2.8 | 1.8 | |
| pH | 7.8 | 7.9 | 8.1 | 7.9 | |
| Organic matter (g/kg) | 2.5 | 2.4 | 3.7 | 1.7 | |
| Carbonate (g/kg) | 165 | 156 | 167 | 172 | |

Table 1: Some physical and chemical characteristics of the 0-30 cm soil depth for the four locations Safwan, Jabal Synam, Lihase, and Centre of Destroyed Tanks.

2.2 Transport of DU in Soil

2.1.1 Sampling of undisturbed soil columns

To study vertical migration of DU in the soil, soil columns were taken from four locations at Basra Governorate: Safwan, Jabal Synam, Lihase, and Centre of Destroyed Tanks where these areas were the theatre of battlefield (Figure 1). Sampling of undisturbed soil columns was performed using Perspex tubes 0.75 m long and 0.12 m inside diameter. In the sampling procedure, a sharp edge hollow auger head was screwed at the end of the Perspex tube. The tube was then gently



Figure 1: Map of Iraq, location map for Basra Governorate.

and slowly driven to 0.50 m depth using sledgehammer and plywood. A circular fitted metal mounted on the soil with tri-pod was used to guide the Perspex tube. The column was then dug out of the ground, replaced the hollow auger head by Perspex base of the same tube diameter and closed on both ends with plastic bags. The column was carefully brought to the laboratory and a funnel was fixed at its end. Eight columns were sampled for leaching study in which duplicate soil columns were taken from each location.

2.1.2 Calibration of scintillation counters for λ and β radiation:

Uranium and DU can be detected by measuring the different types of radiation (i.e., alpha, beta and/or gamma radiation) emitted [24]. For measuring DU, Solid scintillation Counter (SSC) (Compu-Gamma, well type with NaI detector and multichannel analyzer, LKB-1218, Sweden) was fully computerized and calibrated to receive low energy λ radiation from contaminated soil with DU. Soil sample of 2 g was placed in vial and measured for its activity using the following equation:

$$\mathbf{A} = \left(\frac{\mathbf{c}}{\mathbf{Eff}}\right) \cdot \frac{1000}{2}$$

(1)

Where,

A = activity in Bq/kg. c = counts/sec. Eff = efficiency of the solid scintillation counter (90%).

A calibration curve of the SSC for measuring radioactivity of soil samples contaminated with DU is given in *Figure 2*. Channels 5-110 receive low energy λ from 238U and 235U of DU and the latter is imbalance due to isolation of Radium and its products.

Liquid scintillation counter (LSC) with 256 channels analyzer (Fully computerized LKB 1287, Sweden) was calibrated to receive β radiation from 238U, 234Th, 234U, 230Th, 226Ra, and 210Ld corresponding to solution contaminated with DU. The solution (5 mL) was normally mixed with proper cocktail of PPO, POPOP, and Dioxane (15 mL) and measured for its activity. *Figure 3* illustrates the channel numbers (110-208) for measuring the peak of radioactivity of effluent samples contaminated with DU.



Figure 2: Calibration of Solid Scintillation Counter to read energy peak from DU at channels 5-110.



Figure 3: Calibration of Liquid Scintillation Counter to read energy peak from DU within Channels 110-208.

2.1.3 Leaching of DU

A contaminated soil with DU that had been weathering in the soil for approximately 10 years was collected from the surface of location 4 (Centre of Destroyed Tanks). The contaminated soil with average activity of 40556 Bq/kg was used for leaching experiments. In the 1st. part of the study, 170 g of the contaminated soil were mixed with the upper undisturbed soil columns. The soil columns (a total of eight) were then leached with distilled water (150 cm for each) to simulate the

rainfall and then replaced with groundwater (200 cm for each) brought from the same well used for irrigation. During leaching, effluents samples of all columns were collected every 10 days during the observation period of nearly 3 months and their radioactivity was measured using LSC.

In the 2nd. part of the leaching experiment, 25 g of the contaminated soil were placed in glass column connected from its bottom with sintered glass and leached sequentially with 250 mL from each of EDTA (Ethylene diaamine tetra acetic acid), DTPA (Diamine triamine penta acetic acid), and CaCl₂ (1.0 M). Each solution was applied in 10 portions (25 mL for each portion). Similar to the 1st. part, effluent samples were collected during leaching and measured for their radioactivity by LSC.

2.2 Uptake of DU by Tomatoes

2.2.1 Plantation of tomatoes

A field experiment on plant uptake of DU was carried out during the season 2001-2002 after nearly 10 years of environmental contamination with DU. The study area was at Agricultural Experiment Station, Ministry of Agriculture in Burgisiya, close to Safwan, location 1 of the study area. Characteristics of the soil used for growing tomatoes are very close to those given in Table 1. A plot of 4 x 0.75 m2 was selected for growing tomatoes, the major vegetable crops at the area. The plot was divided into 4 sections and received 4

treatments. The treatments included the application of 300 g of a contaminated soil with DU collected from Location 4 (Centre of Destroyed Tanks). The total radioiactivity of the contaminated soil was 40556 Bq/kg and applied as follow:

C: Treatment 1 (control, no application of contaminated soil).

MS: Treatment 2 (mix the contaminated soil with the upper 10 cm of soil surface).

BS: Treatment 3 (broadcast the contaminated soil on the soil surface).

HS: Treatment 4 (adds the contaminated soil on the soil surface and make harrowing)

Selection of the different ways in application the contaminated soil to the subplots was in fact followed the tradition practices by farmers in cultivation of tomatoes and others vegetable crops. It is expected that the DU may reach the top soil of the cultivated land through two main mechanisms: direct deposition of DU aerosol and transport of DU mixed with soil particles by wind action. Tomatoes seeds were grown in nursing place on 20-10-2001 and transfer to the subplots following the common practice by farmers. All agricultural practices including irrigation and fertilization were made according to recommendations.

2.2.2 Sampling of soil and plant parts for measuring activity:

Sampling of soil from subplots was made after application the contaminated soil and before planting tomatoes. Two samples for the depths 0-10 cm and 10-20 cm were taken from each subplot. The soil was dried, ground, and sieved through 1.00 mm sieve and 2 g was taken for measuring activity by SSC.

At harvest on 20-3-2012, sampling was made for plant roots, vegetative parts (leaves and stems), and fruits. The plant samples were oven dried at 600 C for 24 h and sieved through 1.00 mm sieve. Two g of plant samples were taken and measured for γ radiation by SSC. Also, for plant digestion, 10 mL from acids mixtures (HNO3:HCIO4:H2SO4 in the ratio 10:4:1) were added to 0.25 g taken from each plant tissue (Page et. al., 1982). The mixture was then dried on sand bath and mixed with 100 mL distilled water. For measuring radioactivity, 2 mL were taken from the solution and mixed with 10 mL of Lomagel cocktail and measured for β by LSC.

3 Results and Discussion

3.1 Leaching of DU in Soil

In general, the same behavior of DU was observed for the four Locations. Therefore, only data of Location 1, Safwan will be presented. Effluents samples collected during leaching of soil columns and measured by Liquid Scintillation Counter (LSC) were in the range of background count rates (27-30 counts/min) (Fig. 4). It is evident that no movement of DU had observed in soil columns although they were heavily leached with 150 cm of distilled water (simulate the rainfall) and 200 cm of saline water (taken from wells at the investigated area). This result agrees with numerous studies indicated the slow movement of natural U and DU particles or fragments in soil [24, 25]. The depleted uranium compounds that are produced after impact of ammunitions with tanks are settled on the ground locked up in soil compounds and subjected to weathering process. The extent and end-products of this chemical weathering depend on several site specific factors, including the chemical composition and size distribution of the impact particles,

precipitation, humidity, as well as soil properties such as pH and moisture content [26]. It seems that the soils under investigation with the chemical and physical characteristic given in Table 1 are highly reactive to DU. The existing of appreciable amounts of clay, silt, carbonate, organic matter as well as oxides of Mn and Fe in the soils would precipitate DU and stop or slow migration of DU [12]. Furthermore, maximum sorption of uranyl ions on natural materials (e.g., organic matter; iron, manganese and titanium oxyhydroxides, zeolites, and clays) occurs at a pH of 5.0-8.5 [12]. An important point in considering uranium migration in soils is that when UO22+ is reduced to U4+ by humus, peat, or other organic matter or anaerobic conditions, it is essentially immobilized. It should also be noted that phosphates and sulfides usually precipitate uranium and hence stop migration [24].

It is well documented that facilitated transport is usually attributed to the contaminant being bound to particles such as colloids, or having enhanced solubility due to the presence of complexants, ligands, and/or chelators [24]. However, data presented in Figure 5 showed different behavior on the role of the chelating agents EDTA and DTPA in displacement of DU. No DU was detected in the effluents samples even the contaminated soil was leached sequentially with 750 mL of the two chelators and CaCl2 solutions. On the contrary of this result, Fahad [27, 28] observed an increase in Cs137 and Zn65 mobility in soils leached with EDTA and DTPA. These soils have similar physical and chemical characteristics to the investigated soils including pH, type of clay minerals, organic matter, and the content of carbonate and oxides of Fe and Mn. Also, Wallace et al., [29] and Nishita et al., [30] observed an increase in mobility of uranium series found in solid and liquid waste with the application of ammonium acetate and calcium chloride solutions.



Figure 4: Count rate of effluent samples collected from soil columns leached with Distilled water and saline water from well.



Figure 5: Count rate of effluent samples collected from contaminated soil with DU leached sequentially with EDTA, DTPA, and CaCl₂.

The absence of DU in the effluents samples of the leaching experiments with water and chelating solutions (Figs. 4 and 5) may be explained on the basis of DU forms that exist in the investigated areas of southern Iraq. It is believed that most of DU in these soils is present as oxides or remains as metal particles which are basically insoluble. Moreover, the physical and chemical weathering remain very slow since moist conditions are hardly found with seasonal rainfall is below 100 mm and potential evaporation is very high reaching nearly 2000 mm annually. Upon weathering, non-oxidized small particles may be adsorbed to clay minerals and humus. The surfaces of remaining DU fragments in soil exposed to the atmosphere will slowly oxidize to uranium oxides. The chemical form of DU impact aerosols has been measured to be predominantly UO2 and U3O8 and these oxides are only sparingly soluble, but will gradually form hydrated uranium oxides in moist conditions [31]. Observations within tanks revealed a particle size distribution with two modes - one within the range of 0.1 and 2 or 3 μ m (respirable range) and another between 3 and 30 µm [32]. Immediately after impact, thermal plumes from the burning wreckage and surface winds control the dispersion of this cloud of tiny particles, resulting in eventual environmental contamination via surface deposition in the vicinity of the impact zone [31]. The current data suggest that most of DU found in the areas of battlefields is in fine particles of metal or oxides or fragments deposited on soil surface immediately after burning of the tanks hit by DU rounds or as a result of deposition of resuspension of the contaminated soil transported by wind actions. However, the deposition flux was depended on climatic factors at time of explosion particularly rainfall, wind speed, and wind direction. Precise measurements of long range surface deposition under controlled atmospheric conditions have either not been carried out or made public [31]. Numerical simulations under idealized atmospheric conditions estimate more than an order of magnitude difference in deposition flux at a couple of kilometers from the release point, compared to that a few meters near the release zone [33].

3.2 Plant uptake of DU

Data presented in Table 2 and Figure 6 clearly indicate that the method of application the contaminated soil (170 g of contaminated soil contained weathered munitions) to the subplots before planting tomatoes caused varied distribution of DU in the 0-10 cm and 10-20 cm soil layers. The broadcasting of the contaminated soil with DU (BS) showed the highest radioactivity at the 0-10 cm soil depth for both before (Table 2) and after (Fig. 6) planting of tomatoes. At the same time, the control treatment (C) showed values very close to the background count rates. The differences in the magnitude of DU distributed between layers may be attributed to the degree of incorporated the contaminated soil with the field soil. It is expected that most of the contaminated soil remained at the soil surface for the broadcasting practice.

The uptake of uranium by tomatoes above- and below-ground expressed as counts rate is given in *Table 3* (counts rate by SSC) and *Table 4* (counts rate by LSC). Surprisingly, only the roots showed counts rates above the background indicating the presence of DU in the samples of roots. The mean of total counts rate was 20803 for the roots as compared with 18080 for the vegetative part and 17760 for the fruits (*Table 3*). The latter counts rates for the vegetative parts and fruits were in the range of background reading (17765). Similarly, the radioactivity of roots, vegetative parts, and fruits measured by LSC (*Table 4*) showed the same trend. The mean of total counts rates were 15472 for the roots decreased to the background levels of 13424 for the vegetative parts and 13632 for the fruits.



Figure 6: Counts rates (c/min) of soil samples taken from subplots received contaminated soil with DU before planting tomatoes and measured by Solid Sc. C. Ciunter

Therefore, the presence of DU in the roots samples with no DU was detected in the aboveground tissues indicated that no uptake or translocation of uranium ions had occurred under the current experiment. It is believed that the activity detected associated with the roots was a very fine particle of DU (metal or oxides) incorporated with soil particles that remained adhered to the root system measured for total radioactivity. In fact,

the method used for separation roots from soil was the washing with water and this method seemed inefficient in removing all contaminants including soil or DU fine particles. In this respect, several studies have indicated that uranium is transported poorly from soils to plants [34, 35]. Also, it has been postulated that uranium uptake by plants may be limited to the outer membrane of the root system and may not occur on the interior of the root at all [36, 37]. In several experiments conducted to screen for potential phytoextractors that could accumulate uranium from soil in arid and semi-arid regions, Ulery et al. [21] found none of fifty species tested in a variety of experiments (in the lab, greenhouse, and field) would be considered hyperaccumulators according to accepted definitions. In experiment on the uptake of munitions-derived DU by three grass species, Meyer et al. [19] found that both aboveground and belowground uptake was dependent on the soil DU concentration, and on the moisture regime utilized during the experimental duration. Uptake was enhanced under higher moisture regimes where concentration ratios decreased with increasing soil DU concentrations, but increased as more moisture was applied.

| Treatment | Symbol | Counting Time | Total Count | | | |
|--------------------------|--------|---------------|-------------|--|--|--|
| (DU application) | | (min) | | | | |
| Roots | | | | | | |
| Control | С | 480 | 18720 | | | |
| Mixing with Soil Surface | MS | 480 | 20640 | | | |
| Broadcasting | BS | 480 | 21120 | | | |
| Harrowing | HS | 480 | 20640 | | | |
| Vegetative Parts | | | | | | |
| Control | С | 480 | 17280 | | | |
| Mixing with Soil Surface | MS | 480 | 18240 | | | |
| Broadcasting | BS | 480 | 17760 | | | |
| Harrowing | HS | 480 | 18240 | | | |
| Fruits | | | | | | |
| Control | С | 480 | 17760 | | | |
| Mixing with Soil Surface | MS | 480 | 17280 | | | |
| Broadcasting | BS | 480 | 17760 | | | |
| Harrowing | HS | 480 | 18240 | | | |
| Background | | 480 | 17760 | | | |

Table 3: Radioactivity (count rate) of 2 g plant samples at the end of tomatoes season measured by Solid Scintillation Counter.

| Treatment | Symbol | Counting Time | Total Count | | | |
|--------------------------|--------|---------------|-------------|--|--|--|
| (DU application) | | (min) | | | | |
| Roots | | | | | | |
| Control | С | 480 | 16080 | | | |
| Mixing with Soil Surface | MS | 480 | 16320 | | | |
| Broadcasting | BS | 480 | 14640 | | | |
| Harrowing | HS | 480 | 15456 | | | |
| Background | | 480 | (13344) | | | |
| Vegetative Parts | | | | | | |
| Control | С | 480 | 13152 | | | |
| Mixing with Soil Surface | MS | 480 | 13200 | | | |
| Broadcasting | BS | 480 | 13680 | | | |
| Harrowing | HS | 480 | 13392 | | | |
| Background | | 480 | (13584) | | | |
| Fruits | | | | | | |
| Control | С | 480 | 13584 | | | |
| Mixing with Soil Surface | MS | 480 | 13632 | | | |
| Broadcasting | BS | 480 | 13680 | | | |
| Harrowing | HS | 480 | 13584 | | | |
| Background | | 480 | (13584) | | | |

Table 4: Radio activity (count rate) of plant samples at the end of tomatoes season extracted by acids mixtures and measured by Liquid Scintillation Counter.

Figures 7 and 8 illustrate the radioactivity (c/min) of DU for roots, vegetative parts, and fruits at the end of tomatoes season. It is quite clear that only the roots samples taken from subplots received the DU treatments (MS, BS, and HS) showed the presence of DU. Again, the radioactivity associated with roots is believed to be as a result of physical adsorption or adhering of fine DU particles on surfaces of roots and no uptake mechanism had been taken place. Therefore, it is evident that the DU in soil of the contaminated areas of southern Iraq remained in forms either insoluble or poorly soluble in water. The prevailing dry conditions dominated most of the seasons largely limited the chemical, physical, and biological weathering of DU metals or oxides. These results are in consistent with estimates of mobility which indicated that DU munitions and armour degrade and chemically weather over a period of 100 to 1000-years [24].



Figure 7: Counts rates (c/min) of plant parts taken from subplots received contaminated soil with DU after harvest tomatoes and measured by Solid Sc. Ct.



Figure 8: Counts rates (c/min) of plant parts taken from subplots received contaminated soil with DU after harvest tomatoes and measured by Liquid Sc. C.

3.3 Critical Pathways of DU in Desert Areas of Southern Iraq

The data presented in Tables 2-4 and Figures 6-8 on mobility of DU in soil and transfer to plant could be used to generate the pathways of DU in the desert environment of southern Iraq and the expected risk to human (Figure 9). As is evident from these data the mobility of DU by leaching in soil or uptake by plant was not observed under the current investigation indicating the importance

of other pathways in transport of the deposited DU on soil surface. Depleted Uranium that deposited will usually reside on land and incorporate into the soil or adhere to plant surfaces, be resuspended in the atmosphere as a result of wind action [37]. Under desert conditions of southern Iraq with the lack of rain, wind is a major transporting factor of the contaminants (Pathway 1). Therefore, dust inhalation (Pathway 2) remains the potential risk to human for the short- and long-term and the critical pathway for transporting of contaminated soil particles with DU to human. Keller et al. [6] have pointed out that upon impact on armour and while penetrating it, a part of the DU-projectile is transformed into an extremely fine powder called aerosol, which predominantly burns into poorly soluble uranium oxides. Nearly 90% are generated in a first phase while the projectile hits the target and in a second phase by subsequent explosions and when the target is set on fire. Dust can partly be raised and carried along by the wind over greater distances.

Other mechanism that had led to intake of DU by population was the edible fungus "Truffles" (Pathways 3 and 4). It can be grown on soil surface of desert areas during fall and winter seasons particularly at rainy ones. The imperfect washing of tissues before cooking may lead to the contamination of food with DU and subsequent ingestion by population. These pathways are considered important during few seasons following the military operation.

Contamination of native plants (Pathway 6) by direct deposition of DU aerosol or the deposition of resuspended contaminated soil particles represents an important environmental compartment that lead to transport DU to animals (mainly camels and sheep) (Pathway 7). On the other hand,

Transport of DU present as metal, oxides, or penetrators from the upper soil layers to groundwater (Pathways 8 and 9) remain a long term process for many reasons. These include the dry conditions prevailing the contaminated areas, the deep groundwater level (nearly 15 m below soil surface), and the presence of compacted layers at nearly 10 m below soil surface. It is expected therefore, that the transformation of insoluble or poorly soluble DU to soluble forms would be limited. At the same time it is difficult to predict how long it would take for this to occur. Further, the lack of transporting agent (rain fall) under desert conditions would limit the transport of DU to deep layers below soil surface. Numerous investigators [9, 39, 40] pointed out that the mobility of uranium in soil and its vertical transport (leaching) to groundwater depend on properties of the soil such as pH, oxidation-reduction potential, concentration of complexing anions, porosity of the soil, soil particle size, and sorption properties, as well as the amount of water available.

Farmers may use the groundwater as source for irrigation in the contaminated areas of southern Iraq to grow vegetables mainly tomatoes (Pathway 10). The moist conditions associated with continuous irrigation may enhance weathering of DU and the solubility of some forms of DU leading to transfer of uranyl ions to plant tissues (Pathway 11). Also, this condition would increase the downward movement of soluble forms of DU. In this respect; Meyer et al. [19] observed that aboveground and belowground uptake by three grass species was dependent on the soil DU concentration, and on the experimental moisture regime utilized during the experimental duration. Uptake was enhanced under higher moisture regimes, suggesting a greater degree of DU solubility and concomitant plant availability.



Figure 9: Expected pathways of DU in southern region of Iraq for the short- and long-term. (Numbers indicate the important of pathways on the short-term)

4 Conclusions

- 1. No Depleted uranium mobility was observed in desert soil of southern Iraq even the contaminated soil subjected to heavy leaching by water or chelating solutions.
- 2. No uptake of DU by tomatoes aboveground had occurred in the field trial. However, the detection of DU observed in roots samples was attributed to the imperfect washing of samples during the process of analysis.
- 3. Results of the current study suggest that DU present in these soils is in metallic or oxides forms which are basically insoluble and no dissolution of DU had occurred even 10 years had passed on weathering of DU forms.
- 4. With limited rainfall, the wind action would play a major role in transporting DU particles in the environment and inhalation of DU dust or contaminated soil resuspended in the atmosphere remain the critical pathway and potential risk to human for the short- and long-term.
- 5. Downward movement of DU to groundwater and the transfer to plant represent important pathways for the long term.

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