DISTRIBUTION OF ^{137}CS IN THE SURFACE SOIL FROM SELECTED AREAS IN IRAQ

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ABSTRACT

Environment of Iraq was affected by global and regional fallout from the nuclear weapon tests and nuclear accidents. This paper presents the results of studying the deposition levels and distributions of ^{137}Cs, in soil samples collected from 281 sites throughout the Iraqi territory, between the years 1999 and 2003. The objective of this work is to provide regional radioactivity background levels for cesium in surface soils of Iraq. Activity concentration of ^{137}Cs, in the collected surface soil samples were measured. A gamma spectrometric system based on a pure germanium detector was used. The minimum and maximum activity concentrations of ^{137}Cs in the collected soil samples ranged between below detection limit (BDL, about 0.5 Bq/Kg) to 175 Bq/Kg dry soil, with average of 21.9 Bq/Kg. The maximum concentration of ^{137}Cs (175 Bq/Kg) was found in a soil sample collected from Um Al-Waz area near Akashat, within the western part of Iraq. The average concentration of ^{137}Cs in surface soils of the northern part of Iraq, which was affected by Chernobyl accident that occurred in Ukraine in the year 1986, was about 15 Bq/Kg, while it was about 7 Bq/Kg in the surface soils of the southern part of Iraq. The highest average was in the soils of the western part of Iraq, about 33 Bq/Kg. The concentrations of ^{137}Cs in soil samples, with depth were measured in some selected sites. Migration of ^{137}Cs to depth more than 10 cm was not recorded.

توزيع تراكيز السيزيوم 137 في الترب السطحية من مناطق مختارة في العراق

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المستخلص

تتأثر بيئة العراق بالمتساقطات الإشعاعية الجوية الناتجة عن التفجيرات النووية أو حوادث المفاعلات سواءً كانت الإقليمية منها أو الدولية. جرى في هذا البحث قياس تراكيز السيزيوم 137 في نماذج منتخبة من التربة السطحية في 281 موقع في عموم العراق للفترة من 1999 – 2003 لأجل تتبع أنماط توزيع السيزيوم في التربة السطحية لمناطق واسعة من العراق، ولتكون هذه القياسات قاعدة بيانات مهمة يمكن الاستفادة منها في الدراسات اللاحقة. استخدمت منظومة تحليل أطياف گاما المستندة إلى كاشف جرمانيوم عالي النقاوة في قياس تراكيز الفعالية للسيزيوم 137. بينت الدراسة الحالية إن توزيع السيزيوم 137 في التربة السطحية للعراق غير متجانس، فهو يختلف من منطقة لأخرى ومن مكان لآخر ضمن المنطقة الواحدة. تراوحت تراكيز الفعالية للسيزيوم 137 في نماذج التربة بين أقل من حدود الكشف (بحدود 0.5 بكريل/كغم) إلى 175 بكريل/كغم تربة جافة وبمعدل 21.9 بكريل/كغم. وقيس أعلى تركيز للسيزيوم 137 في نموذج تربة من منطقة أم الوز، قرب عكاشات ضمن المنطقة الغربية من العراق، وكان بحدود 175 بكريل/كغم تربة جافة. بلغ معدل تراكيز السيزيوم 137 في تربة المنطقة الشمالية من العراق والتي تأثرت بحادثة تشرنوبيل في عام 1986 بحدود 15 بكريل/كغم، بينما بلغ معدل وجود السيزيوم 137 في تربة المنطقة الجنوبية من العراق بحدود 7 بكريل/كغم. وبلغ أعلى معدل لتراكيز السيزيوم 137 في نماذج التربة في المنطقة الغربية من العراق وكان بحدود 33 بكريل/كغم. جرى في هذا البحث أيضاً قياس تراكيز السيزيوم في نماذج التربة لأعماق مختلفة في بعض المواقع، ولم يسجل أي تسرب للسيزيوم لأعماق أكثر من 10 سم.

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INTRODUCTION

Cesium (Cs) is the second heaviest element of the Group I alkali metals and displays a high degree of chemical similarity with the other alkali metals and with K, in particular (Bowen, 1979). There are at least 21 isotopes of Cs, mostly in the range of atomic masses 123 – 144. The only natural isotope of Cs is the stable isotope ^{133}Cs.

^{137}Cs is the important Cs isotope, it is a fission product, produced from weapons testing at a rate of 6.3 PBq per megaton (0.17 MCi per megaton), also produced from nuclear reactor accidents (Eisenbud and Gesell, 1997). ^{137}Cs is one of the most important isotopes in the environmental studies, that is formed in relatively large amounts in atomic weapons tests and nuclear accidents. Its physical half-life is 30.2 years, so it remains in the upper 5 cm of the surface soil in the environment for a long time (Lewyckyj *et al*., 1996; Eisenbud and Gesell, 1997 and Zhiyanski *et al*., 2005).

After the accident at the Chernobyl nuclear power plant in 1986, soil activities of Cs radioisotopes at the most heavily contaminated site in the UK were 0.12 Bq of ^{134}Cs cm^{-3} and 0.26 Bq of ^{137}Cs cm^{-3}. Soil activities of ^{137}Cs in excess of 30 Bq cm^{-3} (6.2×10^{-7} µg of ^{137}Cs g^{-1} of soil) persist over large areas in Russia and Ukraine (reviewed in Philip and Martin, 2000) and concentrations of $^{134/137}$Cs, at least of this order have also arisen from the testing and production of nuclear weapons. For example, in the Marshall Islands and the Mayak nuclear complex (Graham and Simon, 1996 and Aarkrog *et al*., 1997).

It has been well established that cesium is so tightly bound by the clay minerals of the soil, but root uptake is slight, and foliar absorption is, therefore, the main portal of entry of ^{137}Cs to the food chains during periods of active fallout, so the external exposure doses from cesium are higher than those are caused by ingestion of cesium. Because of its solubility and close physico-chemical similarity to potassium, cesium can be considered as one of the most hazardous radionuclides in the environment and one of the dangerous products of nuclear fission. It is a source of gamma radiation and also is a carcinogen (UNSCEAR, 1988; IAEA, 1996; UNSCEAR, 2000). Graham and Simon (1996) concluded that 80% of the ^{137}Cs from Chernobyl is located within the upper 15 cm of soils and its migration is very slow and is strongly dependent on soil type (Barisic *et al*., 1999). Owing to long half lives of ^{137}Cs and the abundant deposited fallout, especially in the northern hemisphere, ^{137}Cs can be detectable in environmental samples for many years (UNSCEAR, 1988 and UNSCEAR, 1993). The uptake of cesium from soil has been shown to be inversely proportional to the potassium content of soils in which there is a potassium deficiency (Anderson and Roed, 1994; Eisenbud and Gesell, 1997 and Agudo, 1998).

Cesium was used by many researchers in different geological, hydrological and ecological studies (Ritchie *et al*., 1974; Alberts, *et al*., 1989; Fawaris and Johanson, 1994; Antonopoulos-Domis *et al*., 1995 and Albrecht *et al*., 1998)

This paper presents the results of studying the ^{137}Cs deposition levels and distributions in soil samples collected from 281 sites throughout Iraq between the years 1999 and 2003. The objective of this work is to provide a data base of regional radioactivity background levels for cesium in surface soils of Iraq.

MATERIALS AND METHOD

Soil samples (each one about 4 Kg) from an undisturbed area of (1 × 1) m and at a depth of (0 – 5) cm were collected from 281 sites in Iraq (Fig.1), during the period from 1999 to march, 2003. Samples were dried at room temperature, placed in an oven at 105° C for 24 hour, and gravels and pieces of grass were removed by hand. Samples were crushed and sieved through a 1 mm sieve and homogenized.

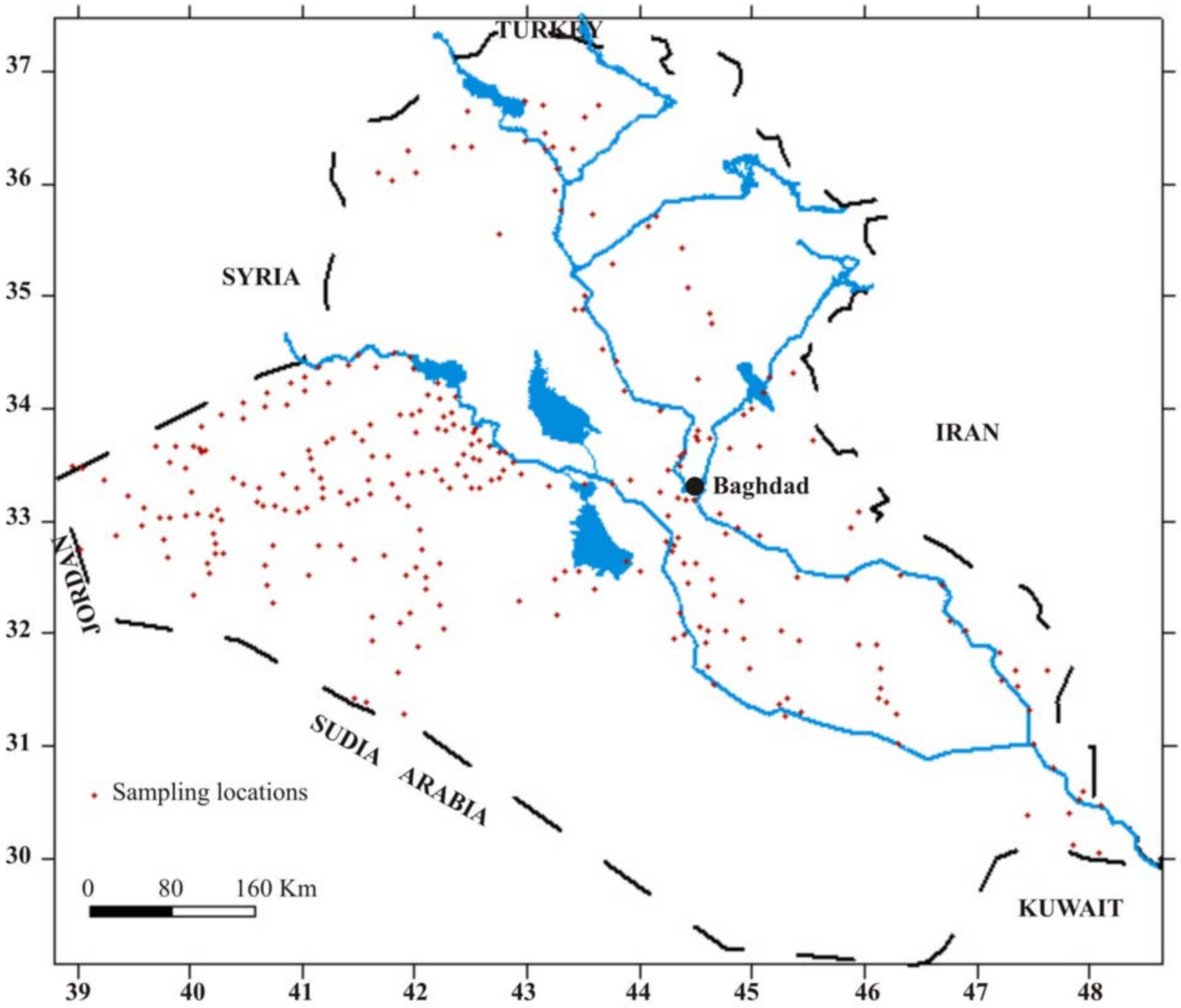

Fig.1: Locations of the collected soil samples of the current study

The radioactivity of ^{137}Cs in one kilogram of soil sample was measured, for up to 10800 sec, with gamma spectrometric system based on highly pure germanium detector with efficiency of 40%. The detector was shielded by a 10 cm thick lead with a fixed bottom and a moving cover. One-liter Marinelli beaker geometry was used for the measurements. A Europium-152 (Eu-152) standard source was used for energy and efficiency calibration. The computer code GDR-4 was used. Global positioning system (GPS) from e-Trex, German company was used to determine the geographic coordinates of each sampling site.

RESULTS AND DISCUSSION

The activity concentrations of ^{137}Cs in the collected soil samples ranged between below detection limit (BDL, about 0.5 Bq/Kg) to 175 Bq/Kg dry soil, with average of 21.9 Bq/Kg. There are three levels of ^{137}Cs concentrations in the soils of Iraq. The average concentration of ^{137}Cs in surface soil of northern part of Iraq was about 15 Bq/Kg, while it was about 7 Bq/Kg in the surface soils of the southern part of Iraq. The highest average was in the soils of the western part of Iraq, about 33 Bq/Kg. Contour map was compiled from the ^{137}Cs activity concentrations with the Kriging grid method using a computer program surfer 7.0 (Fig.2).

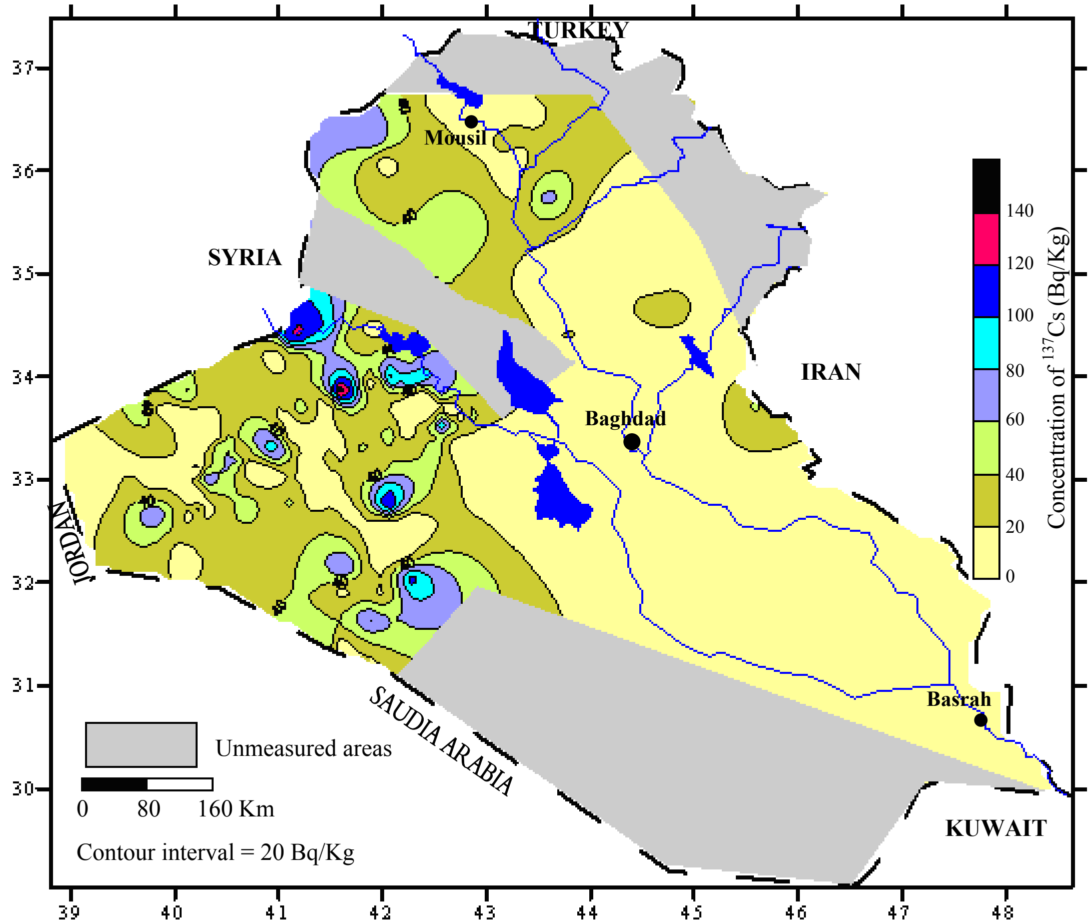

Fig.2: Distribution of ^{137}Cs in surface soils of the studied areas

Generally, the distribution of the pollutants; such as cesium in the surface soil depends on many factors: Weather conditions (rain rate, speed and directions of wind, time and position of the contaminated cloud), geological conditions (type of soil, texture, weathering and erosion, transport and dilution of pollutants with rain), and distance to the pollution source (Adriano *et al*., 1981; Eisenbud and Gesell, 1997; Agudo, 1998; Mahmoud *et al*., 2003 and Zhiyanski *et al*., 2008).

All aforementioned factors may have affected the distribution of the cesium in soils in Iraq. A definite distribution pattern for the higher concentrations in the western part of Iraq trends W – NW, which corresponds with the predominant wind direction during 1990 – 2003, in the area; according to the meteorological reports in Iraq (IMGO, 2002). After 1986, the main source of fallout of ^{137}Cs, especially in the northern hemisphere is Chernobyl nuclear plant accident. Radioactive pollutants were found in soils of the northern part of Iraq in a prior study achieved by Marouf during 1987 (Marouf, 1992).

Marouf *et al*. (1996) noted that the pattern distribution of ^{137}Cs in the soil samples collected from the Western Desert of Iraq was different from that in the soil samples collected from the northern part of Iraq, which is influenced by the Chernobyl nuclear plant accident (Marouf, 1992). ^{137}Cs was associated with ^{134}Cs in the northern part, while ^{134}Cs was BDL in the soil samples taken from the Western Desert of Iraq, in that measuring time. They concluded that the source of the ^{137}Cs in the Western Desert may be not due to Chernobyl accident, but from nuclear explosion test, based on that ^{134}Cs nuclide is not present in bomb fallout, but is formed in reactors by neutron capture in ^{133}Cs. This characteristic has been used to distinguish between ^{137}Cs produced in the reactors and that produced in nuclear weapon tests (Eisenbud and Giessal, 1997).

In the current study, the maximum concentration of ^{137}Cs was found in Um Al-Waz area (N 41.611º and E 33.855º) within Al-Anbar Governorate, western part of Iraq and this value is more than those found in the northern part of Iraq, which is geographically nearer to the areas contaminated with the fallouts caused by Chernobyl accident. So the authors think that the sources of ^{137}Cs in the western part of Iraq were the global fallout from the atmosphere as a result of atmospheric nuclear weapon testing, ground surface and underground nuclear explosions, and accidental release from nuclear facilities; Chernobyl nuclear plant accident is one of them.

In the eastern part of Iraq, the authors also found relatively high concentration of ^{137}Cs in Mandili (about 40 Bq/Kg) near the border with Iran, while it was found to be less than 8 Bq/Kg in the soil sample selected from the same area in prior study (Marouf *et al*., 2000). This means, the atmosphere is still feeding the surface soil with additional cesium, which may come from recent sources (Fukuyama and Fujiwara, 2008).

Practically, the authors noticed, in the field, that the topography is playing a significant factor that influenced the distribution of Cs in the soil. Moreover, we almost found that ^{137}Cs concentrates are higher in depression deposits; as compared with soil samples taken from the elevated surrounding areas. This is attributed to the removal of ^{137}Cs from soil by rain water and its accumulation in the lower sites.

In General, the activity concentrations of ^{137}Cs in the samples of most surface soils in Iraq were normal levels; excluding few sites that have relatively high concentration in comparison with concentrations of cesium in soils in the other neighbor countries. Table (1) shows that these values are corresponded with the activity concentrations in Syria (about 200 Bq/m^2) (Othman, 1990), on the other hand, are less than the activity of ^{137}Cs in the coastal Syrian mountain, in western Syria, which range between (500 – 8000) Bq/Kg (Al-Rayyes and Mamish, 1999). The activity values of ^{137}Cs in western and northern parts of Iraq, in the current studied areas are mostly similar to those of the western, central and southern parts of Turkey (10 – 150 Bq/Kg), on the other hand these values are less than those in the eastern Black Sea (Gokmen *et al*., 1996 and Mahmoud *et al*., 2003). We can also note that the activity values of ^{137}Cs in southern part of Iraq are slightly higher than those in Saudi Arabia (Al-Kahtani *et al*., 2001).

Table 1: Comparison of ^{137}Cs activity in the soils in the current study with ^{137}Cs activity in the soils in other countries

Region	Type of soil	Activity of ^{137}Cs (Bq/Kg)	Reference
Iraq	Surface soil	0.5 – 175	Current study
Syria	Surface soil	200 Bq/m^2	Othman (1990)
Syria (Coastal mountains)	Mountains' soil	500 – 8000	Al-Rayyes and Mamish (1999)
Turkey (Eastern Black Sea)	Surface soil	160 – 9260	Gokmen *et al*. (1996)
Turkey (Northwestern part)	Cultivated soil	0.92 – 40	Gokmen *et al*. (1996) and Mahmoud *et al*. (2003)
Turkey (Central part)	Surface soil	20 – 150	Gokmen *et al*. (1996)
Turkey (Southern part)	Surface soil	10 – 110	Gokmen *et al*. (1996)
Saudi Arabia	Surface soil	0.06 – 2.03	Al-Kahtani *et al*. (2001)

Table (2) shows the concentrations of ^{137}Cs in soil samples with depth, which were collected from some selected sites, in Iraq. Transfer of ^{137}Cs to a depth more than 10 cm did not occur. Some researches concluded that transfer factor of Cs in soil increases with increase of the organic contents in the soil (Bergeijk *et al*., 1992 and Zhiyanski *et al*., 2005) and decrease with increasing clay content in soil (Kühn *et al*., 1984) and with increasing of potassium and pH of the soil (Kühn *et al*., 1984 and Wang *et al*., 1997). On the other hand, other researchers noted that transfer factor of Cs in soil is not affected by clay content (Smolders *et al*., 1997), nor pH of soil (Bergeijk *et al*., 1992; Mahmoud *et al*., 2003). Current study, however did not verify these conclusions.

Table 2: Concentration of ^{137}Cs in the soil samples with sampling depth in selected areas (in Iraq)

Coordinates		**^{137}Cs concentration in Bq/Kg**			
N	**E**	**0 – 5 cm**	**5 – 10 cm**	**10 – 15 cm**	**15 – 20 cm**
42.5796	33.5498	108	9	*	*
44.537	32.063	18	*	*	*
42.0694	33.4385	60	10	*	*
46.279	31.275	7	4	*	*
44.307	31.945	9	2	*	*
43.588	35.738	72	5	*	*
43.789	34.424	22	*	*	*
42.0444	33.9833	112	9	*	*
41.755	33.581	24	5	*	*
44.647	34.754	39	6	*	*
42.255	32.0494	114	10	*	*
40.465	33.333	75	8	*	*
45.875	32.934	4	*	*	*
45.951	33.088	10	1	*	*
44.365	32.191	11	1	*	*

* BDL: below detection limit

CONCLUSIONS AND RECOMMENDATIONS

- The current study is useful database for further studies on ^{137}Cs in surface soils of Iraq and the factors controlling its distribution. Further studies are needed to study the concentrations and transfer of ^{137}Cs to plants and surface and ground water. Additional research is required on the calculation and estimation of the external and internal dose corresponding with the concentrations of ^{137}Cs in the soils, as well as to study the influence of soil composition, clay minerals and organic matter on ^{137}Cs concentration.
- It is difficult to use the data in the current study to get decisive conclusion about the sources of cesium in different areas in Iraq, or to give the exact percentage of ^{137}Cs that is derived from the earlier nuclear weapon tests or that from Chernobyl accident. However, generally the cesium content in the soils of Iraq is attributed to the global and regional fallout from the nuclear weapon tests and nuclear accidents.
- The activity concentrations of ^{137}Cs in the samples of most surface soils in Iraq ranged between below detection limit (0.5 Bq/Kg) to 175 Bq/Kg. These values are mostly similar or less than the activity concentrations in some neighbor countries, such as Syria and in Turkey but slightly more than those in Saudi Arabia. It was also found that there is no transfer of ^{137}Cs to depth more than 10 cm in the surface soil.

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