Assessment of Soil Contamination in Area Surrounding Tuwaitha Nuclear Facilities

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Abstract

The wide spread looting of the Tuwaitha Nuclear Facilities as well as damaging of some buildings in 2003, had offered possibilities of contamination of soil environment inside the Site. The objective of the present work was to investigate soil contamination to help in future decontamination programs. A total of 25 soil surface samples (including one reference sample) covered different locations in the Site were collected in March 2011. High purity Ge detector was used for gamma spectrometry of soil samples. Data of total and spectral gamma for U series, Th series, ^{40}K and ^{137}Cs are presented. Slight variations were observed in specific activity of the U series (^{214}Bi or ^{214}Pb and ^{226}Ra) among measured soil samples where the range was 10.3-12.7 for ^{214}Bi as compared with 12.2-33.4 Bq/kg for ^{226}Ra. Values of both ^{214}Bi and ^{226}Ra are in the range of reference sample specific activity indicating that no evidence of contamination had occurred in the investigated area. Results of activity concentrations of thorium series (^{228}Ac or ^{208}Tl, ^{212}Pb, and ^{212}Bi) are in the range of reference sample and close to those values given worldwide for natural uranium in soil. The levels of ^{40}K in soil are within the natural abundance of this isotope in the soil where the range was 207.6-266.1 with 220.3 Bq/kg for the reference sample. On the other hand, ^{137}Cs specific activities showed great variation among measured samples. The minimum value for ^{137}Cs was 0.6 and the maximum 7.6 compared with 0.8 Bq/kg for the control soil sample. The non-uniformity of radioactivity concentration of ^{137}Cs suggest the presence of contamination in some locations although this level is considered as an acceptable level and no hazardous effect will be generated.

Key words: Tuwaitha site, soil contamination, natural isotopes, U series, Th series, K-40, Cs-137.

تقييم تلوث التربة في المساحة المحيطة بالمنشآت النووية في التويثة

المستخلص

إن اعمال النهب الواسع التي حدثت في المنشآت النووية في التويثة فضلاً عن تدمير بعض البنايات في عام 2003 وفر احتمال تلوث التربة المحيطة بالمنشآت النووية. يهدف العمل الحالي الى دراسة تلوث التربة المحيطة بهذه المنشآت للمساعدة في برامج ازالة التلوث في المستقبل. تم جمع 25 عينة تربة تمثل مواقع مختلفة في المنشآت (من ضمنها عينة من تربة المرجع بعيداً عن التويثة) في آذار 2011. استخدم عداد الجيرمانيوم عالي النقاوة لتحليل طيف أشعة كاما في عينات التربة. بينت نتائج النشاط الإشعاعي النوعي لسلسلة اليورانيومU والثوريومTh وكذلك البوتاسيوم ^{40}K والسيزيوم^{137}Cs. لوحظ وجود تغايرات بسيطة في النشاط النوعي لسلسلة اليورانيومU (^{214}Bi أو ^{214}Pb و ^{226}Ra) في عينات التربة المقاسة حيث وصل المدى الى 10.3–12.7 للبيزموث^{214}Bi مقارنة بـ 12.2–33.4 بكريل/كغم للراديوم ^{226}Ra. كانت قيم ^{214}Bi و ^{226}Ra ضمن مدى عينة المرجع والذي يؤشر الى عدم وجود تلوث التربة بهذه النظائر. أشارت نتائج النشاط الإشعاعي لسلسلة الثوريوم Th (^{228}Ac أو ^{208}Tl و ^{212}Pb و ^{221}Bi) أن المدى كان ضمن عينة المرجع وهي قريبة من القيم المذكورة عن اليورانيوم الطبيعي في العالم. كانت مستويات البوتاسيوم ^{40}K ضمن التواجد الطبيعي لهذا النظير في التربة حيث وصل المدى الى 207.6–266.1 مقارنة مع 220.3 بكريل/كغم لعينة المرجع. من جهة أخرى، أظهر النشاط النوعي الإشعاعي للسيزيوم ^{137}Cs تغاير كبير في العينات المقاسة. حيث كانت أقل قيمة 0.6 وأعلى قيمة 7.6 مقارنة بـ 0.8 بكريل/كغم لعينة المرجع. يعود عدم الإنتظام في تركيز الـ ^{137}Cs الى وجود تلوث في بعض المواقع رغم أن هذه المستويات تكون مقبولة ولا تسبب خطورة.

الكلمات الدالة: موقع التويثة، تلوث التربة، النظائر الطبيعية، سلسلة اليورانيوم ، سلسلة الثوريوم، البوتاسيوم–40، السيزيوم–137.

Introduction

The Tuwaitha Nuclear Facilities (TNF), about 20 km south of Baghdad, had been damaged or destroyed in 1981 (Israel attack), 1991 (First Gulf War), and 2003 (US military operation). The TNF had partially enriched uranium, along with quantities of highly radioactive medical and industrial isotopes, sensitive equipment, chemical stores, and radioactive waste materials. The wide spread looting of the Site as well as damaging of some buildings in 2003, had offered possibilities of contamination of soil environment inside the Site.

Next to air and water, soil is generally considered as the main environmental component. Handling

of potential pollution problems in soil must be based on the prediction of the functioning of soil. Soil may act as a sink for deposited fallout radionuclides [1] and/or contaminants reached from many possible sources. Also, soil that is heavily contaminated with pollutants will be a source for contamination [2]. Therefore, the dual functions of soil in this process, acting as sink and being a source of pollutants are connected. Consequently, predictions on the retaining factors and decontamination of radionuclides are possible with detailed investigation of the complicated system.

Soil is a valuable environmental monitoring medium because it can accumulate contaminants from both current air emissions and resuspended materials. Hence, soil sampling and analysis evaluates long-term contamination trends and monitors environmental radionuclide inventories [3]. Radioactivity observed in soil could be originated from fall-out radionuclides (^{137}Cs and ^{90}Sr), natural isotopes (Uranium series and ^{40}K), fission products (^{137}Cs, ^{60}Co, and others), or contaminants reached soil due to accidents or mishandling of radioactive materials [1].

Cesium-137 was introduced into the environment mainly through atmospheric nuclear tests, in the 1950s and 1960s and the Chernobyl accident in 1986 [4] where significant amounts of fallout ^{137}Cs were deposited worldwide on the landscape. Cesium-137 is a gamma and beta emitter with a relatively long half-life of 30.2 years. Therefore, ^{137}Cs is expected to persist for some time. However, it is relatively immobile in the environment and is expected to sorb strongly to soils and sediment, especially those with high clay or organic content [5]. In this respect, Fahadet. al. [2] found most of ^{137}Cs in the liquid waste applied to soil columns taken from Tuwaitha Site and leached for 180 days with water, remained in the upper 10 cm of soil. Also, 96% of ^{137}Cs applied to soil columns in the form of CsCl and in concentration of 18.5 MBq/column remained in the upper 1.0 cm even leached with 240 cm of water for 115 days [6]. Results of these experiments suggest that the Tuwaitha soil is highly reactive to ^{137}Cs and other radionuclides and when they present, they will be strongly adsorbed by soil particles in essentially nonexchangeable or irreversible forms.

Natural uranium (U) occurs in soils in typical concentrations of a few parts per million. Uranium-238 is the most abundant isotope in natural uranium (fraction by weight in natural uranium is 99.28%) and decays into other radioactive elements [7]. In surface soils, its concentration ranges from 0.1 to 20 mg/kg with a world average value of 2.8 mg/kg [8]. Natural uranium

consists of a mixture of three radioactive isotopes which are identified by the mass numbers ^{238}U (99.2836% by mass), ^{235}U (0.711%) and ^{234}U (0.0054%).These radionuclides have very long half-lives: 4.5 x 10^9, 7 x 10^8 and 2.5 x 10^5 years, respectively [9]. Uaranium-238 decays into series of isotopes, ^{234}Th, ^{234}Pa, ^{234}U, etc. down the decay chain [10].

Potassium is an important constituent of fertile soil and is an essential nutrient for plant growth; it is widely distributed in nature and is present in all plant and animal tissues [11]. Potassium-40 is a naturally occurring radioactive isotope of potassium which has a very long half-life of 1.248 ×10^9 years [12]. It decays to ^{40}Ca by emitting a beta particle with no attendant gamma radiation (with branching time of 89%) and to the gas ^{40}Ar by electron capture with emission of an energetic gamma ray (with branching time of 11%).

This paper presents results of soil contaminations with radionuclides in area surrounding Site of the Tuwaitha Nuclear Facilities. The information presented here will aid in understanding the real situation of contamination of the area and in future decontamination programs.

Materials and Methods

Study Site:

The area under investigation is located 20 km southeast Baghdad, Iraq in the Tigris valley with coordinates of 33° 12" latitude and 44° 30" longitude and elevation of 39.3 m. Average annual precipitation in the area is about 150 mm, and the prevailing wind direction is from the northeast.

The soils of the area are recent alluvium without any horizons differentiation. These are entisols with an ochricepipedon as result of plowing. Organic matter content in the upper soil surface did not exceed 1.5% and lime content was relatively high and may be observed as a fine fraction in non-active form. The salinity status depends upon many factors including environmental conditions, soil texture, landscape features, and groundwater table [13].

Water table depth is in the range 130-400 cm and is not a fixed level; it rather increases in summer to the greatest depth from the soil surface, while it decreases in winter [13]. The fluctuation in water table levels may be due to i) absence of efficient drainage system in the area and ii) effect of water levels in the Tigris and Diyala rivers on the groundwater level.

Sampling and characterization of soil:

Sampling of soil was carried out on an area of nearly 2 hectare surrounding nuclear facilities and buildings in March 2011. Surface soil samples (0-25 cm depth) were taken by auger and transferred to laboratory. A total number of 24 samples were collected from the area with one sample taken far from the investigated area (Zaafraniya, 1.0 km south of the Tuwaitha Site) to represent a reference sample (control). Location map of the samples is given in Figure 1. List of samples locations is given in Table 1.

Some physical and chemical characteristics of the investigated soils (Location 10 and 15) are given in Table 2. For characterization of soils, standard procedures of soil analysis have been used [14].

Measuring of radioactivity (gamma-ray spectrometry):

Soil samples were air dried, ground by rubber hammer, and screened through a 2.00 mm sieve. Exactly 1000 g were taken from each bulk soil sample and placed in plastic sheet for gamma spectrometry.

For gamma spectrometry analysis of soil samples, high purity germanium detector, Canberra was used for this purpose. The relative efficiency of the system was 30% with energy resolution of 2 keV for 1.33 MeV gamma line of ^{60}Co. The system was calibrated using reference source ^{152}Eu Marnelii beaker geometry for the reference and samples. The counting time was 60 min for each soil sample. Analysis of gamma spectrometry was accomplished by the Faculty of Radiation Protection Center, Ministry of Environment.

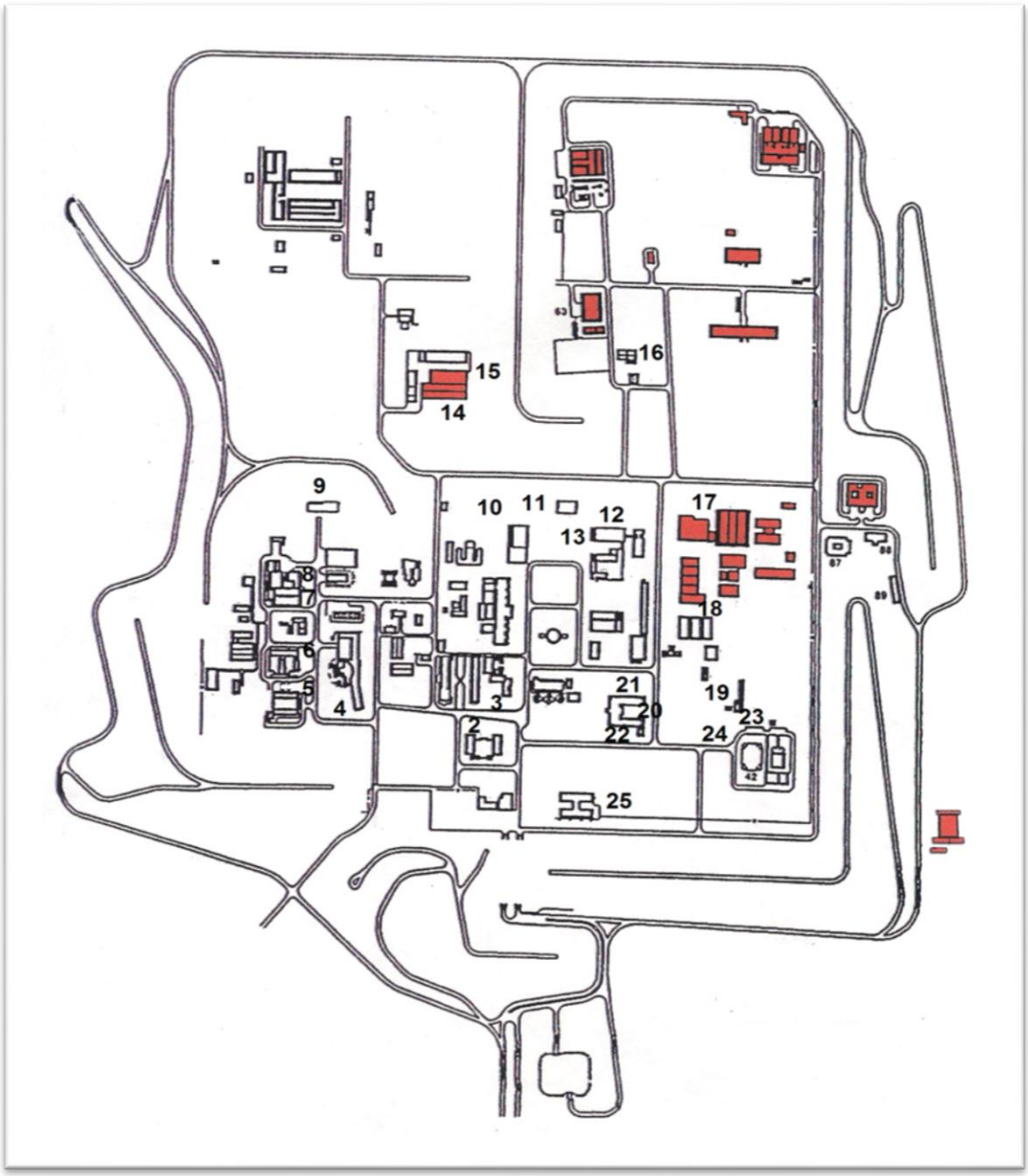

Fig. 1. Lay out of the Tuwaitha Site where the numbers indicate the location of soil sampling.

Table 1. Number of soil sample and the corresponding building or activity at the Tuwaitha Site.

Sample Number	Location in the Tuwaitha Site
1	Reference sample (Control) taken from Zaafaraniya, 1.0 km south of Baghdad
2	Restaurant
3	Radiochemistry Labs
4	Tamuz-2, zero power reactor (destroyed)
5	Laboratory Work Building
6	Hot Laboratory (LAMA)
7	RWTS, North
8	RWTS, Northwest
9	Waste Storage for Tamuz-1 Reactor
10	Tamuz-1 (IRT-5000), West
11	Tamuz-1 (IRT-5000), Northwest
12	Isotope Production, West
13	Isotope Production, South
14	Chemical Waste Treatment, East
15	Chemical Waste Treatment, North
16	Sewage Station
17	Health Physics
18	Green Houses
19	Water Treatment Station
20	Agriculture and Biology, North
21	Agriculture and Biology, West
22	Agriculture and Biology, East
23	Main Library, West
24	Main Library, Southwest
25	Administration Building, North

Table 2. Some physical and chemical characteristics of the soils.

Characteristics	Sample Location 1 (Reference)	Sample Location 10	Sample Location 15
Clay (g kg^{-1})	312	293	315
Silt (g kg^{-1})	490	303	490
Sand (g kg^{-1})	198	304	195
Soil Textural Class	Silty Clay Loam	Clay Loam	Silty Clay Loam
Electrical conductivity (dS/m)$^{+}$	3.1	2.5	2.6
pH^{+}	8.0	8.1	8.1
Lime (g kg^{-1})	255	386	246
Organic Matter (g kg^{-1})	12.6	11.4	14.0
Cation Exchange Capacity (cmol kg^{-1})	26.2	27.6	25.9
Soluble Cations (mg kg^{-1})			
Ca^{++}	138.1	81.2	122.4
Mg^{++}	50.5	36.9	48.6
Na^{+}	56.8	12.8	45.5
K^{+}	27	12.5	28

+ Measurements were made on saturation extract.

Results and Discussion

Uranium and thorium series in soil samples:

Data on specific activity of uranium and thorium series measured by high purity Ge detector are presented in Table 3. Total gamma and spectral gamma provide a direct measurement of uranium progeny isotopes in the soil [10]. Uranium undergoes radioactive decay to lead via a series of radioactive progeny or daughter radionuclides. Some nuclides emit gamma radiation as the nucleons and electrons reconfigure to a more stable form during or shortly after an alpha or beta decay [10]. Only data of 10 samples (selected randomly) are given in Table 3 including the reference sample (sample taken from area far from the investigated one). It is evident that slight variations were observed in specific activity of the U series (^{214}Bi or ^{214}Pb and ^{226}Ra). For these 10 locations, the minimum specific activity for ^{214}Bi was 10.7 Bq/kg (nearly 0.86 mg/kg in unit of mass concentration) (Sample 21) and

a maximum of 12.7 Bq/kg (nearly 1.02 mg/kg) (Sample 12); the reference (or the control) sample was 12.0 Bq/kg (nearly 0.97 mg/kg). The ^{226}Ra showed higher values than ^{214}Bi in those 10 samples. Specific activities of ^{214}Bi and ^{226}Ra in all samples (25 samples) are best evaluated by the statistical parameters given in Table 4. Less variation was obtained for ^{214}Bi than for ^{226}Ra where the range (minimum and maximum) was 10.3, 12.7 for ^{214}Bi as compared with 12.2, 33.4 Bq/kg for ^{226}Ra. Values of both ^{214}Bi and ^{226}Ra are in the range of reference sample specific activity indicating that no evidence of contamination had occurred in the investigated area. The current values of both isotopes are close to those values given worldwide for natural uranium in soil. United Nation SCEAR [15) reported values in the range from 0.3 to 11.7 mg/kg of uranium in soil worldwide. Bleise et al. [16] showed that values for concentration of uranium in soil were from 0.7 to 11 and to 15 mg/kg in farmland soil due to use of phosphate fertilizers. Also, Gilday and Edick [17] reported values of 0.9-2.3 for ^{226}Ra (U chain) and 1.1-2.7 pCi/g for ^{228}Ra (Th chain) in soil of New York State, USA.

Results of activity concentrations of thorium series (^{228}Ac or ^{208}Tl, ^{212}Pb, and ^{212}Bi) for only 10 samples (locations) are given in Table 3. Statistical parameters (mean, min, max, and standard deviation) of these isotopes for all samples are given in Table 4. Apparently, variations among samples for the same isotope were minimal. At the same time, values are close to the control sample indicating that the radioactivity of these isotopes was in the range of background readings. For instance, the minimum and maximum specific activity of ^{228}Ac was 9.0 and 14.6 as compared to 12.8 Bq/kg for the control sample. Similarly, the ^{212}Pb showed values of 7.8 and 15.7 for minimum and maximum with 9.1 Bq/kg for the control sample. The magnitude of the specific activity of U and Th series (^{226}Ra, ^{214}Pb, ^{228}Ac, ^{212}Pb, and ^{212} Bi) for the 25 soil samples is given in Figure 2. In general, ^{226}Ra showed the highest magnitudes among the measured 25 soil samples followed by ^{228}Ac, ^{212}Pb, and ^{212}Pb with the lowest magnitude was for ^{212} Bi.

Table 3. Specific activities of U series and Th series for 10 soil samples including the reference sample.

No.	Location No.	Specific Activity (Bq/kg)				
		U series		Th series		
		Bi-214 or Pb-214	Ra-226	Ac-228 or Tl-208	Pb-212	Bi-212
1	Reference (Control)	**12**	**21.8**	**12.8**	**9.1**	**8.6**
2	2	11.5	20.7	11.2	8.2	5.8
3	11	12.4	31.5	11.7	9.1	8.1
4	12	12.7	19.5	9.0	7.8	5.3
5	14	11.4	25.8	10.7	9.2	7.3
6	15	11.7	23.9	14.6	9.7	7.3
7	18	11.3	22.8	11.2	9.9	8.6
8	19	11.8	20.5	11.2	9.2	8.8
9	21	10.7	18.2	11.9	8.7	7.04
10	22	11.5	22.4	13.1	9.9	5.1

Table 4. Statistical parameters of the specific activity of uranium and thorium series for the 25 soil samples.

Statistical Parameter	Specific Activity (Bq/kg)				
	U series		Th series		
	Bi-214 or Pb-214	Ra-226	Ac-228 or Tl-208	Pb-212	Bi-212
mean	11.6	23.0	11.7	9.6	7.1
max	12.7	33.4	14.6	15.7	10.2
min	10.3	12.2	9.0	7.8	4.7
Stand. dev.	0.75	4.82	1.30	1.60	1.55
(Control)	**12**	**21.8**	**12.8**	**9.1**	**8.6**

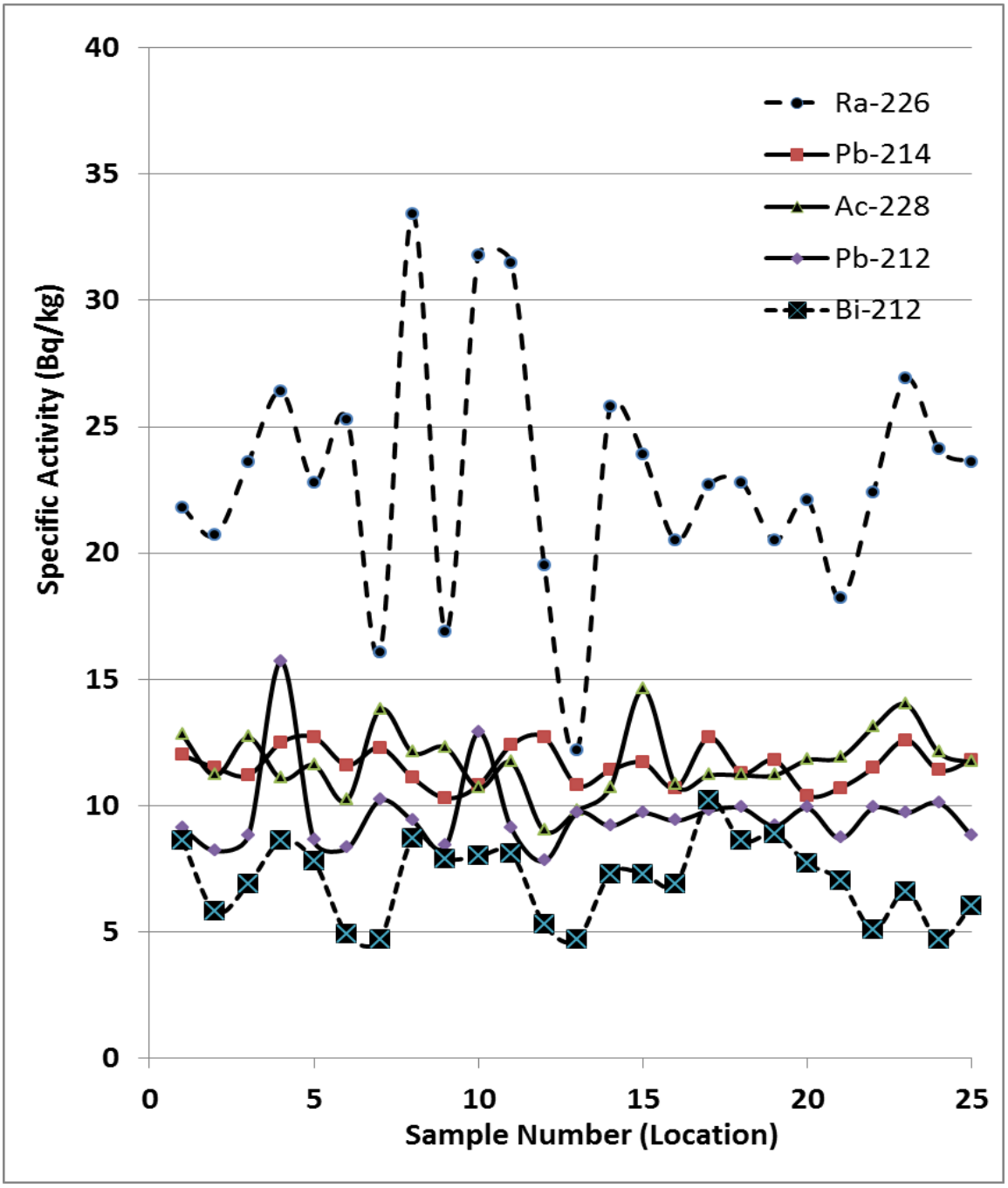

Fig. 2. Magnitudes of the specific activity of U and Th series (Ra-226, Pb-214, Ac-228, Pb-212, and Bi-212) for the 25 soil samples.

Potasium-40 and Cesium-137 in soil samples:

Specific activity of ^{40}K and ^{137}Cs for 10 soil samples selected randomly along with the statistical parameters for the 25 soil samples are given in Table 5. Data on ^{40}K indicate the presence of slight variations in specific activity among measured samples. Most of samples specific radioactivities were very close to the reference sample. The minimum specific activity was 207.6 and the maximum 266.1 with 220.3 Bq/kg for the reference sample. Therefore, it is expected that these levels of ^{40}K in soil are within the natural abundance of this isotope in the soil. Potassium is an important constituent of fertile soil and is an essential nutrient for plant growth and in the human diet. ^{40}K is an important radionuclide in terms of the dose associated with naturally occurring radionuclides and comprises a very small fraction (about 0.012%) of naturally occurring potassium [18].

Zhu et al. [19] showed that ^{40}K is a natural isotope present in soil and an essential plant's nutrient. Under field conditions, plants can suffer from potassium starvation (or potassium deficiency) periodically or constantly throughout the growing season, due to spatial and temporal variations in the potassium status of agricultural soil. Holmgen et al. [20] reported value of 51,800 mCi/km^2 (1,916 kBq/m^2 or nearly 5474 Bq/kg) for ^{40}K in soil of the USA and this value is not high enough to be hazardous.

Unlike ^{40}K, ^{137}Cs specific activities showed great variation among measured samples (Table 5). The minimum value for ^{137}Cs was 0.6 (Sample 7) and the maximum 7.6 (Sample 21) compared with 0.8 Bq/kg for the control soil sample. It is expected that ^{137}Cs was deposited as fallout primarily during the late 1950s and the 1960s and after Chernobyl accident in 1986 and in most environments was rapidly and strongly absorbed by soil particles at the ground surfaces [21]. Figure 3 presents histograms of the magnitudes of both ^{40}K and ^{137}Cs among the 25 soil samples. The greater deviation in ^{137}Cs values among the investigated soil samples is evident. On the other hand, very slight deviation was observed for ^{40}K.

Since the distribution of fallout ^{137}Cs is assumed to be uniform on soil surface, the variability of radioactivity concentration observed in soil samples reflected the presence of an input other than the fallout deposition. Even though the maximum specific activity observed was 7.6 Bq/kg (Location 21), this

level is considered an acceptable level and no hazardous effect will be generated. The locations which have shown the highest radioactivity of ^{137}Cs (much greater than the reference sample) are in ascending order: 21 (Agriculture and Biology Building), 18 (Green houses), 12 (Isotope production), and 4 (Tamuz-2 destroyed reactor). It is believed that the source of ^{137}Cs in Locations 18 and 21 was the looting which occurred in 1991 and 2003. This looting resulted in spreading contaminated soils and/or containers of liquid ^{137}Cs. The high level of ^{137}Cs in Locations 4 and 12 is hard to be explained. In this respect, Holmgen et al. [20] reported an average value of 620 mCi/km^2 (22,940 Bq/m^2 or nearly 65.5 Bq/kg) for ^{137}Cs in the USA. In the arable part of the catchment area of central Europe, Van der Perk et al. [21] estimation was 8756 Bq/m^2 [25 Bq/kg) for ^{137}Cs.

Table 5. Specific activity of ^{40}K and ^{137}Cs for 10 soil samples and the statistical parameters for the 25 soil samples.

No.	Location No.	Specific Activity (Bq/kg)	
		K-40	Cs-137
Data for 10 Samples			
1	**Reference (Control)**	**220.3**	**0.8**
2	2	228.0	2.08
3	11	230.9	0.8
4	12	207.6	3.4
5	14	218.7	0.72
6	15	258.9	0.74
7	18	232.5	4.05
8	19	253.3	2.2
9	21	230.1	7.6
10	22	238.3	1.27
Statistical Parameters for 25 Samples			
mean		236.2	2.1
max		266.1	7.6 (Sample 21)
min		207.6	0.6 (Sample 7)
Stand. dev.		15.91	1.52

Gamma spectral analysis of soil samples:

Spectral analysis of gamma radiation measured by HPGe detector for only four selected soil samples are given in Fig. 4 (Sample 1, Reference sample and Sample 5, Laboratory Work Building) and Fig. 5 (Sample 15, Chemical Waste Treatment-North and Sample 21, Agriculture and Biology-West). In general, nearly the same spectrum was observed for the investigated 25 soil samples. The difference was in magnitude of energy peaks for ^{212}Pb, ^{137}Cs, and ^{40}K. The spectrum given is a natural-gamma energy spectrum, which is caused by the decay of uranium, thorium, and potassium-40, along with anthropogenic radioactive isotopes ^{137}Cs. Natural gamma rays are emitted by isotopes that are the natural products (daughter products) of the uranium decay series, the thorium decay series, and potassium-40 [22]. Uranium and thorium each decay into a series of unstable (radioactive) daughter products. The uranium decay series consists of several unstable elements in nature; this series of unstable isotopes finally decays to a stable (not radioactive) lead isotope. The decay of thorium forms a similar series of unstable elements. Potassium-40 decays into two stable isotopes, argon and calcium [22].

In Figures 4 and 5, the spectrum of gamma energy was plotted versus the count rates (counts the number of gamma emissions associated with each energy level), where the energy ranges from nearly 10 to 1,765 keV (for ^{214}Bi). The isotopes and the corresponding energy peak are presented in Table 6 in ascending order of their decay energy. Post-processing was used to determine the concentration of these isotopes in the investigated soils presented in Tables 2-5.

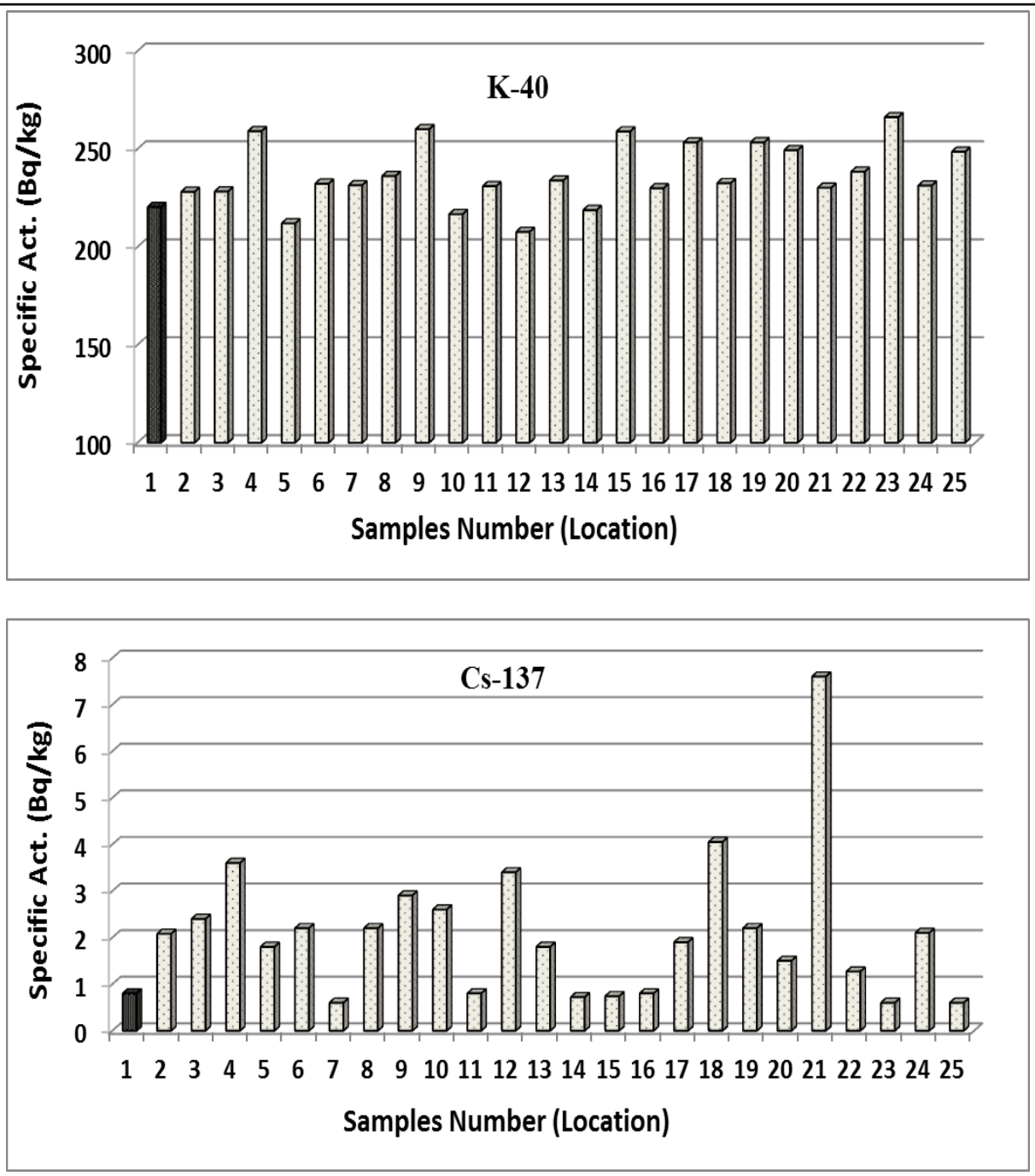

Fig. 3. Magnitude of specific activity of K-40 and Cs-137 among soil samples where sample number 1 refers to the reference (control)

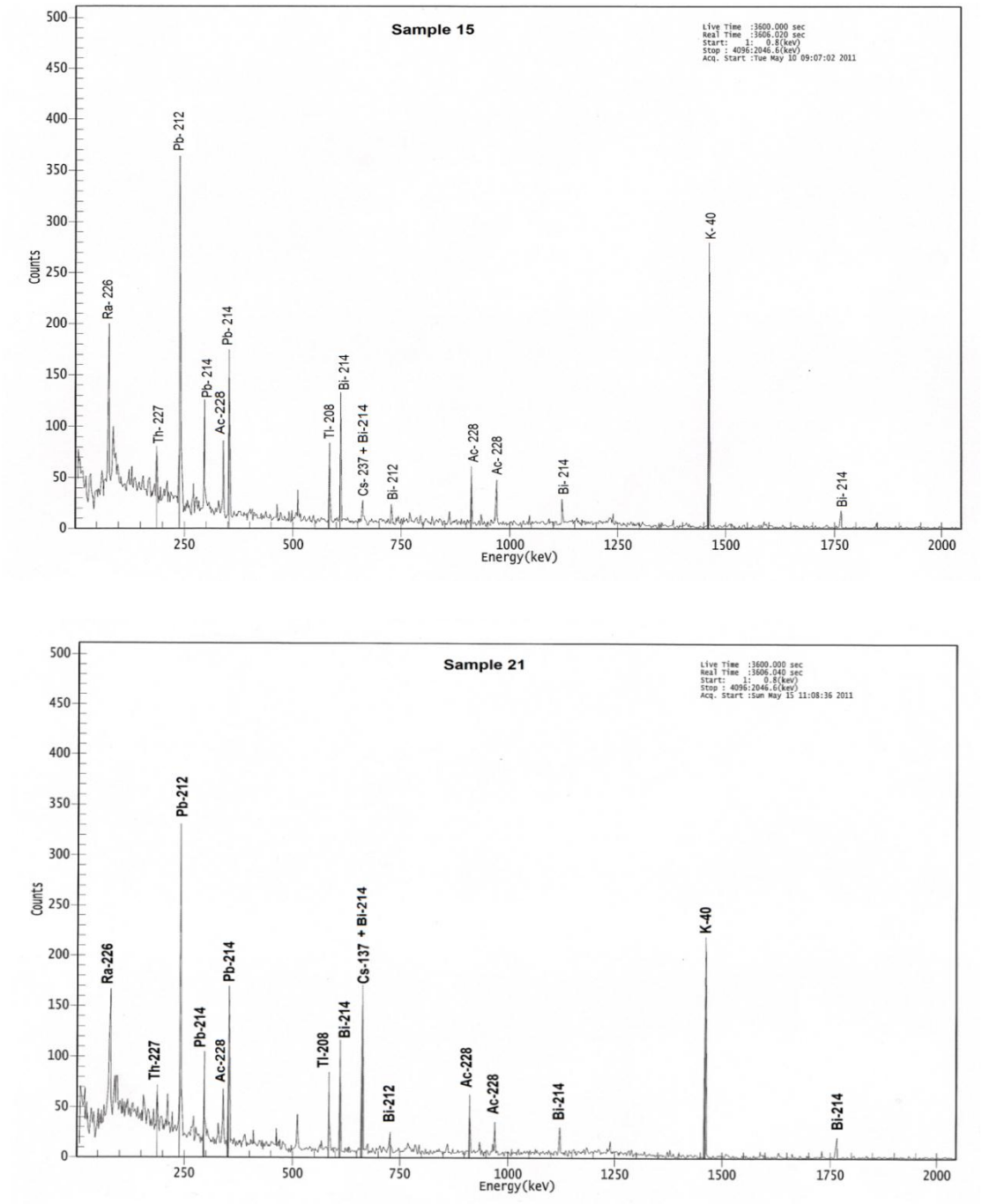

Fig. 5. Gamma spectral analysis of soil sample 15 (Chemical Waste Treatment, North) and sample 21 (Agriculture and Biology, West).

Table 6.Isotopes and the decay energy of gamma radiation of the investigated soil samples and their corresponding energy in ascending order.

No.	Isotopes	Energy (keV)
1	Ra-226	210.1
2	Th-227	210.1
3	Pb-212	239.1
4	Pb-214	296.1
5	Ac-208	339.0
6	Pb-214	352.0
7	Tl-208	583.3
8	Bi-214	609.3
9	Cs-137	661.8
10	Bi-212	727.7
11	Ac-228	911.0
12	Ac-228	969.0
13	Bi-214	1120.0
14	K-40	1460.0
15	Bi-214	1765.0

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