Assessment of nuclear radiation pollution in uranium mining-impacted soil

Raad Obid Hussein Houmady

Department of Physics, College of science, Kufa University, Najaf, Iraq

E-mail: Raad_Obid@yahoo.co.uk

Abstract

Activities associated with mining of uranium have generated significant quantities of waste materials containing uranium and other toxic metals. A qualitative and quantitative study was performed to assess the situation of nuclear pollution resulting from waste of drilling and exploration left on the surface layer of soil surrounding the abandoned uranium mine hole located in the southern of Najaf province in Iraq state. To measure the specific activity, twenty five surface soil samples were collected, prepared and analyzed by using gamma- ray spectrometer based on high counting efficiency NaI(Tl) scintillation detector. The results showed that the specific activities in Bq/kg are 37.31 to 1112.47 with mean of 268.16, 0.28 to 18.57 with mean of 6.68 and 132.25 to 678.33 with mean of 277.49 for ^{238}U, ^{232}Th and ^{40}K respectively. Based on these values, radium equivalent activity in Bq/kg and absorbed dose rate one meter above the ground surface nGy/h were calculated and found to be vary 52.72 to 1189.84 and from 25.02 to 553.01. The indoor and outdoor annual effective dose rate in mSv/y ranged from 0.12 to 2.71 and from 0.03 to 0.67 respectively. To evaluate the dangerous of the study area, the external (H_{ex}) and internal (H_{in}) hazard indexes are calculated and found to be ranged 0.14 to 3.21 and from 0.24 to 6.22. For the purpose of assessing the seriousness of the study area, results were compared with the world wide average. This comparison indicated that the study area is not safe from the radiological protection point view.

Key words

Natural radioactivity, Gamma-spectroscopy, mine, Najaf/ Iraq.

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تقييم التلوث الاشعاعي النووي في التربة المتأثرة بتعدين اليورانيوم

رعد عبيد حسين حمادي

قسم الفيزياء، كلية العلوم، جامعة الكوفة، النجف، العراق

الخلاصة

تم في هذا البحث قياس النشاط الاشعاعي الطبيعي للنويدات المشعه ^{40}K ^{232}Th and ^{238}U, في الطبقه السطحيه للتربه المحيطه بفوهة منجم اليورانيوم في محافظة النجف والناجمه عن مخلفات الحفر والتنقيب باستخدام مطيافية اشعة كاما ذو كاشف أيوديد الصوديوم ذات بلوره مطعمه بالثاليوم Na(Tl) ابعادها "2 × 2 ". تراوحت قيم النشاط الاشعاعي النوعي ومعدلاته بوحدات Bq/kg من 37.31 الى 1112.47 وبمعدل 268.16 ومن 0.28 الى 18.57 وبمعدل 6.68 ومن 132.25 الى 678.33 وبمعدل 277.49 لكل من ^{40}K و ^{232}Th و ^{238}U على التوالي. كما تبين ان الفعاليه المكافئه للراديوم (Ra_{eq}) بوحدات Bq/kg ومعدل الجرعه الممتصه (AD) بوحدات nGy/h على ارتفاع واحد متر عن سطح الارض تتراوح مابين 52.72 الى 1189.84 وما بين 25.02 الى 553.01 على التوالي. ووجد ان قيم معدل الجرعه السنويه المؤثره الداخليه والخارجيه بوحدات mSv/y تراوحت من 0.12 الى 2.17 ومن 0.03 الى 0.67 على التوالي. ولغرض تقييم الخطوره للمنطقه تم حساب معاملي الخطوره الخارجي والداخلي وكانت ذات مدى من 0.14 الى 3.2 وبمعدل 0.08 ومن 0.24 الى 6.22 وبمعدل 1.53 على التوالي. قورنت النتائج مع المعدل العالمي وبينت المقارنه ان المنطقه المدروسه غير امينه من وجهة نظر الخطوره الاشعاعيه.

Introduction

Uranium mines tailings are one of the important sources of natural radionuclides from uranium decay chain in the environment [1]. Human has always been exposed to ionizing radiation because of the multiplicity of its sources as found in the soil that we walk up on, the air we breathe, the water we drink, the food we eat and the building that we occupy [2]. So, radiation is all around us, it naturally exists in our environment and it has been since the birth of our planet. Exposure to ionizing radiation is generally considered undesirable at all level because even a small amount of a radiation substance may produce a damaging biological effects and that ingested and inhaled radiation can be a serious health risk [3]. It may lead to somatic infirmities, like cancer and genetic defects such as mutation and chromosome aberrations [4]. The radiological impact is due to the gamma-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters. The ^{238}U and ^{232}Th series and ^{40}K isotope, have very long half-life up to 10^{10} years, include gamma energies greater than 1.022 MeV which is the threshold of pair production (electron-positron) [5]. Thus, these radionuclides have a non- negligible radioactivity and require particular attention [6]. In 1988 they were classified as a known pulmonary carcinogenic human by the International Agency for Research on Cancer (IARC) [7]. Also, from the view point of biological effect of radiation protecting, United Nations Scientific Committee on the Effects of Atomic Radiation established that the world mean dose from natural radiation sources of normal area is estimated to be 2.4mSv/y while for all man-made is about 0.8mSv/y [3]. Therefore, 75% of the radiation dose received by humanity is come from natural radiation sources [8]. Activities associated with mining or processing of uranium for nuclear fuels and weapons have generated significant quantities of waste materials containing ^{238}U, ^{232}Th, ^{40}K and other toxic metals [9, 10]. These radionuclides and other metals can find their way into the soils around the mine and water resources in the vicinity of the facilities involved in mining, milling, ore separation, and purification. Leaching of metals, non-metals, and radionuclides from surface impoundments of tailings generated during uranium mining and milling often leads to groundwater contamination [11]. Soil is to being the final repository of nuclear waste, especially that surrounding and near the mines [12]. The mine tailings were predominantly sandy material, and strong winds were conducive for their transport from actual mining and milling areas to adjacent lands [13]. Some of these metals have been reported to produce severe diseases like cancer, kidney failure, liver damage and neurological and genetic malfunctions [14-17]. For these considerations, monitoring of ground and surface water surrounding uranium mines has great significance from a human health perspective. So, uranium mines are one of the important sources of natural radionuclides from uranium decay chain in the environment [18].

In Iraq, the uranium mine was constructed in 1990. It situated at latitude 31° 52′ 254" N and longitude 22° 26′ 221" E in the southern of Najaf city. It located near Al-Manathera villages and in agricultural area raised concerns with the potential enhancement of radioactivity in agricultural products and with the radiation exposure of populations through food chain transfer of uranium series radionuclides.

The reasons that led to propose the subject of this study are the using of simple and primitive methods in exploration, excavation, drilling and mining in addition

to proliferation of remnants of drilling and mining in the soil surrounding the mine hole without taking into account the requirements of radiological prevention, control and occupational safety which lead to increasing the exposure of workers, farmers, plants and living close to the mine, to radiation. The more dangerous is the proliferation of waste in the soil surface surrounding the mine hole, making it affected by the erosion conditions such as winds, rainfalls and flooding which are the major factors effects the natural environmental radioactivity and the associated external exposure due to gamma radiation. In addition to nomination of radionuclides in the inner layers of the soil it is causing a mixing with groundwater which is one of the important sources to irrigate crops. Soil is to being the final repository of nuclear waste, especially that surrounding and near the mines [1]. Uranium, thorium and potassium contamination in the soils and ground water is a global problem a cause to their dangerous damage [13]. As well as the mine became abandoned, non-controlled after its closure in 1998 by Union Nations. Therefore, the assessment of gamma radiation dose from uranium-mines activities is of particular importance.

So, the aim of this work, which is the first of its kind in the study region, is to assess the quality and quantity of ^{238}U, ^{232}Th and ^{40}K radionuclides resulting from the remnants of drilling and exploration of uranium mining in order to assessment of radiological impact as well as to draw the radiological baseline information for soil surrounded the mine mouth in Najaf Province.

Methodology

1. Sampling and background

The area surround the abandoned uranium mine, was identified on the form of a square of side length 200 meter centered shaft. It was divided as square grid with dimension of 50×50 meter as a distance between each two points. The sampling locations were systematically selected and mapped in Fig.1.

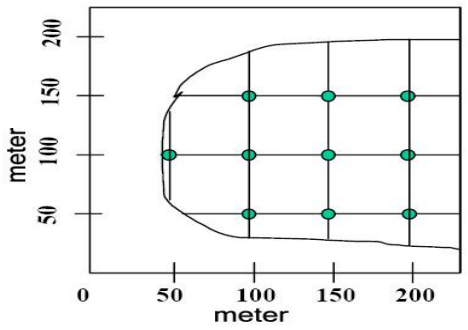


Fig.1: Distribution of samples.

A total of twenty five surface soil samples were collected from twenty five vertexes by digging a hole at a depth of 10 cm before the ground surface at each position using hand auger. The samples were transported to the laboratory in sterile polypropylene sacs and crushed, oven dried at temperature of 100 C° for 24 h and meshed perfectly to pass 0.2 mm mesh. One kilogram from each sample was homogenized in sterile pestle and motor inside a laminar flow hood and then pooled to create a single well mixed composite sample .Each sample was packet and sealed in a marinelli beaker of one liter volume. This marinelli beaker was used as sampling and measuring container. Before use, the containers were washed with hydrochloric acid and rinsed with distilled water. The measurements were performed six weeks later to ensure secular equilibrium [18,19,20]. In order to reduce the background effect, the detector was housed in a lead castle shielded of two layers starting from inside with copper (2 mm thick) followed by lead 10 cm thick from outside. The cosmic rays, photons and electrons are reduced to a very low level by the lead layer. This interaction will produced

X-ray with low energy which can be suppressed by the copper layer [21]. The X-rays can be also come from radioactive impurities like antimony in the lead. In order to minimize the effect of the scattered radiation from the shield the detector was located in the center of chamber. The specific activities were performed by gamma rays spectrometry employing a NaI(Tl) (BICRON supplied by U.S.A) of 2"×2" crystal dimension and coupled with 4096 channels analyzer of CASSY type. For energy calibration we requiring a spectrum from radioactive standard sources of known energies like ^{137}Cs, Eγ = 662 keV and ^{60}Co , $E_{\gamma 1}$ = 1173 keV and $E_{\gamma 2}$ = 1332 keV. The resolution of the system was of 6.73 for the 662 keV of ^{137}Cs photopeak. The counting efficiency of the system was measured by using standard sources (^{22}Na, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba and ^{137}Cs). The efficiency was 3.5% for 1332 keV of ^{60}Co. The activity of ^{238}U was estimated at 1765 keV gamma transition energy of ^{214}Bi (17% possibility). Also the activity of ^{232}Th was measured at 2614 keV gamma transition energy of ^{208}Tl (100% possibility) whereas ^{40}K activity was determined using the 1460 keV gamma ray line (10.7% possibility). In order to subtract the background from each measurement, an empty marinelli beaker (with the same geometry) was measured. The accumulation time for each sample was 20000 s just to obtain gamma spectrum with appropriate to the detector statistics for the same geometrical conditions.

Theoretical Part

1. Natural activity concentration measurement

The specific activity of radionuclides were calculated using the following equation [22,23,24,25,26].

$$Specific\ \ Activity\ (Bq/kg) = \frac{CPS \times 100 \times 100}{Eff \times B.I. \times m} \pm \frac{SD_{CPS} \times 100 \times 100}{Eff \times B.I. \times m}$$

where, CPS = Net count rate per second, B.I. = Branching Intensity, Eff = Efficiency of the detector, m = Sample mass in kg and SD_{CPS} = Standard Deviation of net count rate.

2. Radium equivalent activity (Ra_{eq})

Radiation quality indexes incorporate a series of judgments and concentration data into a single parameter that is indicative of the pollution level. The widely used radiation index, to assess the radiological hazard of soil, is called the radium equivalent activity (Ra_{eq}). It is a weight sum of activities of the three natural radionuclides ^{238}U, ^{232}Th and ^{40}K based on the estimation that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma ray dose rate. The radium equivalent activity (Ra_{eq}) was calculated by using the following relation [3, 6, 24, 27].

$$Ra_{eq}(Bq/kg) = A_U + 1.43A_{Th} + 0.077A_K$$

where A_U, A_{Th} and A_K are the specific activities of Uranium, Thorium and potassium respectively. The worldwide average of Ra_{eq} is equivalent to the annual dose equivalent 1.5 mSv/y which is assumed to be the maximum permissible dose to human from their exposure to natural radiation from soil in one year.

3. The gamma absorbed does rate in air (AD)

Gamma dose rate in air one meter above the ground is used to describe the terrestrial radiation in (nGy/h) or (pGy/h) unit. It is calculated on guideline provided by UNSCEAR (2000).

$$AD(nG/y) = 0.462A_U + 0.621A_{Th} + 0.0417A_K$$

where 0.462, 0.621 and 0.0417 are the conversion factors for ^{238}U, ^{232}Th and ^{40}K respectively.

4. Annual effective doses

The annual effective dose rates were estimated using a conversion coefficient from absorbed dose in air to effective dose (0.7 Sv.Gy per year). Based on outdoor occupancy of 80% and 20% for indoor it was determined as following [3,28,29,30].

In door (nSv) = absorbed dose nGy/h×8760h×0.8×0.7SvGy/y
Outdoor (nSv) = absorbed dose nGy/h×8760h×0.2×0.7SvGy/y

5. Hazard index

To reflect the external exposure, a widely used hazard index, called the external hazard index (H_{ex}), which is defined as following [27]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}$$

The internal hazard index (H_{in}) is given by equation below[27].

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}$$

The values of the index must be less than the unity in order to keep the radiation hazard to be insignificant.

Results and Discussion

The spectra of twenty five surface soil samples surrounded the abandoned uranium mine hole have been analyzed. The specific activity (Bq/kg) of ^{238}U, ^{232}Th, ^{40}K and Radium equivalent activity (Ra_{eq}) (Bq/kg) are given in Table 1. The specific activity varied from 37.31 to 1112.47 (Bq/kg) (mean = 268.16) 0.28 to 18.57 (mean = 6.68) and 132.25 to 678.33 (mean = 277.49) for ^{238}U, ^{232}Th and ^{40}K respectively.

The obtained results are compared to the worldwide average recommended by UNSCEAR, 2000 which are 30, 35 and 400 Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively [6]. It was found that all values of ^{238}U specific activities are higher than the worldwide average whereas those of ^{232}Th are less than it. For ^{40}K, it is clear that the specific activities, with the exception of five samples, are found to be less than worldwide average.

Obviously, the results demonstrate that the maximum specific activity value of ^{238}U is higher than the corresponding value of worldwide average by factor of 37. The large variation between the specific activities obtained for ^{238}U and other two radionuclides can be easily ascribed to the high content of uranium in the neglected waste of drilling and exploration operations on the surface soil surrounding the mine. The contour maps (radiological maps) of the activity distribution of ^{238} U, ^{232}Th and ^{40}K in the study area are shown in Fig.1: (a), (b) and (c). From Fig.1: (a), we can observe three regions with a highest specific activity values of ^{238}U situated at northeast, east and south-west portions of the hole mine. In contrast, Fig.1: (c) indicates that high concentrations of ^{40}K occupies the same positions of ^{238}U while for ^{232}Th there are no placements have activities require attention as shown in Fig.1: (b).

Table 1: Specific activity, radium equivalent activity and absorbed dose.

Sample code	specific activity (Bq/kg)			Ra_{eq} (Bq/kg)	AD nGy/h
	^{238}U	^{232}Th	^{40}K		
S11	72.17±4.43	3.38±0.69	253.19±8.79	96.49	46.00
S12	59.37±4.02	0.28±0.20	184.47±7.51	73.97	35.29
S13	213.24±7.62	5.63±0.89	238.84±8.54	239.68	111.97
S14	39.76±3.29	0.28±0.20	174.70±7.30	53.61	25.82
S15	480.12±11.43	8.02±1.06	415.06±11.26	523.54	244.10
S21	138.07±6.13	2.81±0.63	176.23±7.34	155.65	72.88
S22	645.42±13.26	10.13±1.19	521.04±12.61	700.02	326.20
S23	312.64±9.23	6.47±0.95	409.87±11.19	353.45	165.54
S24	249.18±8.24	4.08±0.76	283.43±9.30	276.83	129.47
S25	37.31±3.19	3.66±0.72	132.25±6.36	52.72	25.02
S31	122.82±5.78	2.39±0.58	153.63±6.85	138.06	64.63
S32	285.95±8.82	12.94±1.35	219.60±8.19	321.36	149.30
S33	122.82±5.78	5.35±0.87	153.52±6.85	142.29	66.46
S34	273.97±8.64	6.47±0.95	211.96±8.05	299.54	139.43
S35	78.43±4.62	4.08±0.76	232.12±8.42	102.13	48.44
S41	142.43±6.23	4.92±0.83	205.85±7.93	165.31	77.44
S42	324.07±9.39	7.03±0.99	264.49±8.99	354.48	165.11
S43	860.29±15.31	13.65±1.39	678.33±14.39	932.04	434.21
S44	175.38±6.91	3.80±0.73	229.67±8.38	198.49	92.96
S45	1112.47±17.41	18.57±1.62	459.96±11.85	1189.84	553.01
S51	167.48±6.75	7.32±1.01	302.06±9.60	201.20	94.51
S52	276.69±8.68	10.41±1.21	156.98±6.92	303.66	140.84
S53	139.16±6.16	7.32±1.01	158.21±6.95	161.81	75.43
S54	85.78±4.83	7.88±1.05	201.27±7.84	112.54	52.91
S55	288.94±8.87	10.27±1.20	320.38±9.89	328.29	153.22
minimum	37.31	0.28	132.25	52.72	25.02
maximum	1112.47	18.57	678.33	1189.84	553.01
mean	268.16	6.68	277.49	299.09	139.61

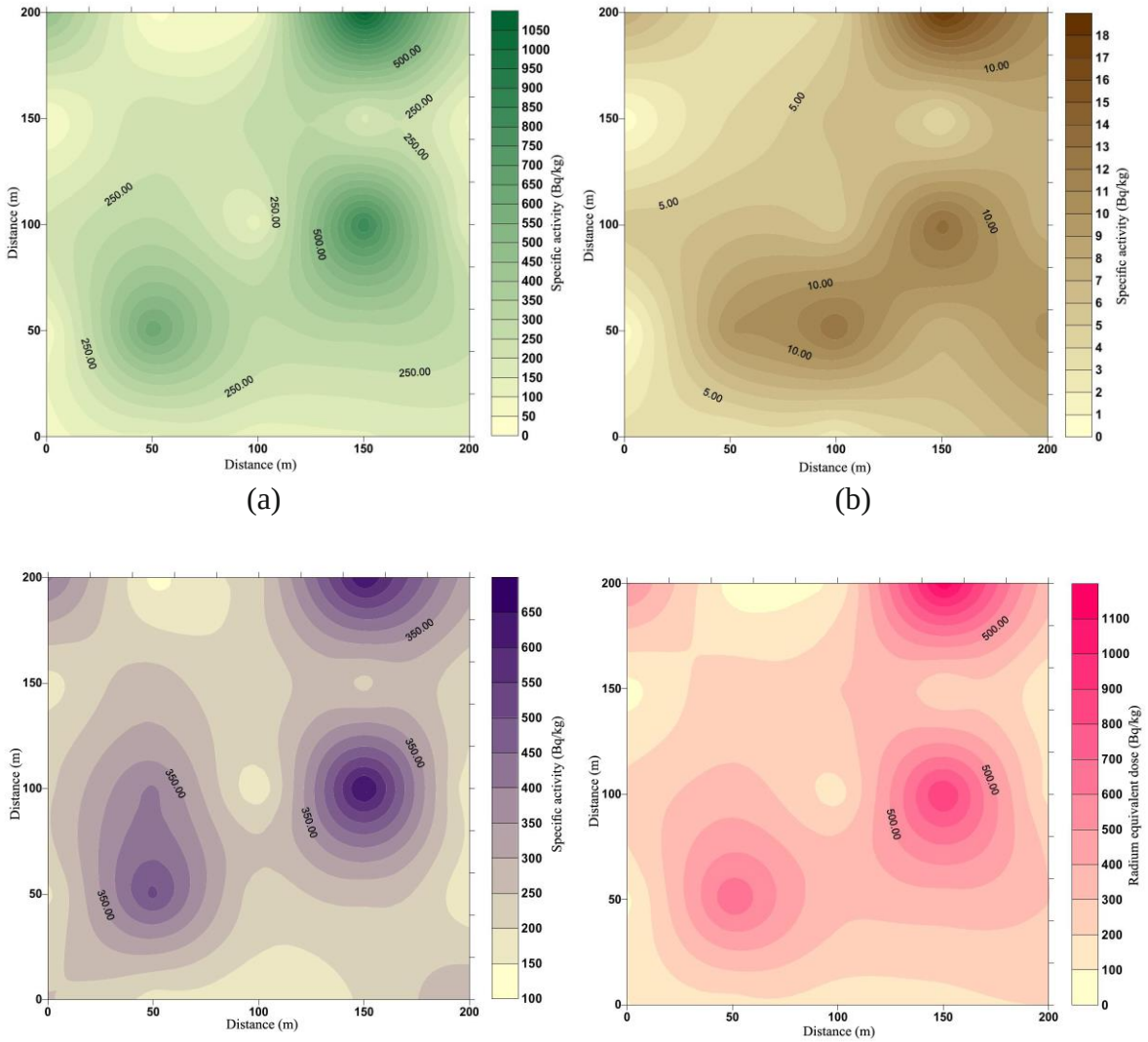
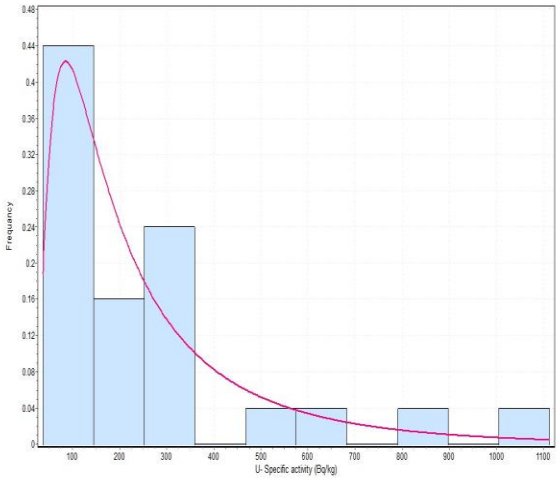


Fig. 1: Map distribution of, (a) ^{238}U specific activity, (b) ^{232}Th specific activity, (c) ^{40}K specific activity, (d) Radium equivalent activity Ra_{eq} .

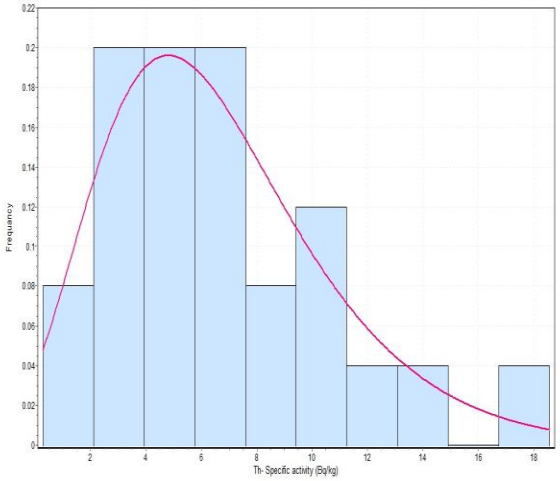
The values of calculated Ra_{eq} for all samples were also presented in Table 1. It may be seen that Ra_{eq} oscillates between 52.72 and 1189.84 with an average of 299.09 Bq/kg. It is observed that the values of Ra_{eq} in twenty one samples were less than the acceptable safe limit of 370 Bq/kg [31,32]. As shown in Table 1, there are four values greater than worldwide average. As a rule, the matter whose Ra_{eq} exceeds 370 Bq/kg is discouraged [27].

Fig.1: (d) demonstrates the distribution of Ra_{eq} and it appears three positions have highest values.

The frequency distribution of the study was represented in Figs. 2(a), 2(b) and 2(c) for ^{238}U, and ^{40}K respectively. It is clear that the statistical distribution is governed by Gaussian distribution. As shown in Fig.2(a), the specific activity value belong to the interval from 3o to 100 Bq/kg have high frequency whereas those for ^{232}Th was from 2 to 8 Bq/kg and for ^{40}K from 130 to 240 Bq/kg have the high frequencies respectively.



(a)



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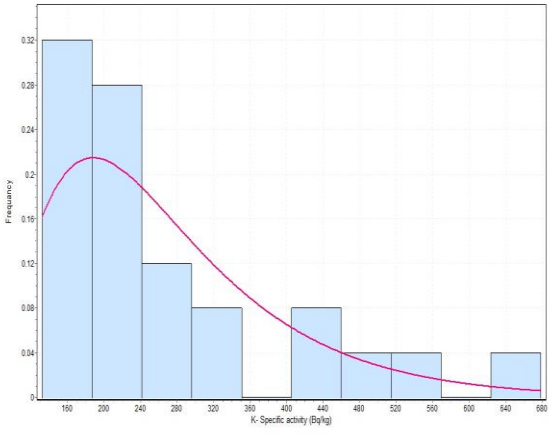

(c)

Fig.2: Frequency distribution of, (a) ^{232}U-specific activity, (b) ^{232}Th- specific activity, (c) ^{40}K- specific activity.

The calculated absorbed dose rate of samples was listed in Table 1. The values ranged from 25.02 to 553.01 with an average value 139.61 nG/h which is nine fold higher than the world average of 15 nG/h recommended by UNSCEAR, 2000. It can be seen that all values were much higher than the world average. Table 2 contains the calculated values of annual effective dose. They were found to be in the range 0.12 to 2.71 mSv/y with an average value 0.68 mSv/y and from 0.03 to 0.67 with an average value of 0.17 mSv/y for indoor and outdoor annual effective dose respectively. In general and as shown in Table 2, for indoor annual effective dose, It is important here to notice that there are fourteen sample have values higher than the word average whereas, the values of the rest samples are close or slightly above of the world average value of soil. In other words, all values of outdoor annual effective dose were below the worldwide average.

The international commission on Radiological Protection (ICRP) has recommended that the annual effective dose equivalent limit of 1 mSv/y for the individual members of the public and 20 mSv/y for the radiation workers [34]. The worldwide average of annual effective dose is approximately 0.5 mSv and the results for individual countries being generally within the 0.3 to 0.6 mSv range [6].

In addition, the calculated values of hazard index for the soil samples were ranged from 0.14 to 3.21 with an average value of 0.80 and from 0.24 to 6.22 with an average value of 1.53 for external (H_{ex}) and internal (H_{in}) respectively as mentioned in Table 2.

Out of 25 positions, 4 for H_{ex} and 13 for H_{in}, have values very higher than unity. Since these values are dispersed randomly within a limited area around the min hole, therefore, according to the report of European Commission in Radiation Protection, the area study is not safe and

posing significant radiological threat to the population [34, 35].

Table 2: Annual effective dose rate and hazard index.

Sample code	Annual dose (mSv)		Hazard index	
	indoor	outdoor	H_{ex}	H_{in}
S11	0.22	0.05	0.26	0.45
S12	0.17	0.04	0.20	0.36
S13	0.54	0.13	0.64	1.22
S14	0.12	0.03	0.14	0.25
S15	1.19	0.29	1.41	2.71
S21	0.35	0.08	0.42	0.79
S22	1.60	0.40	1.89	3.63
S23	0.81	0.20	0.95	1.80
S24	0.63	0.15	0.74	1.42
S25	0.12	0.03	0.14	0.24
S31	0.31	0.07	0.37	0.70
S32	0.73	0.18	0.86	1.64
S33	0.32	0.08	0.38	0.71
S34	0.68	0.17	0.81	1.55
S35	0.23	0.05	0.27	0.48
S41	0.38	0.09	0.44	0.83
S42	0.81	0.20	0.95	1.83
S43	2.13	0.53	2.51	4.84
S44	0.45	0.11	0.53	1.01
S45	2.71	0.67	3.21	6.22
S51	0.46	0.11	0.54	0.99
S52	0.69	0.17	0.82	1.56
S53	0.37	0.09	0.43	0.81
S54	0.26	0.06	0.30	0.53
S55	0.75	0.18	0.88	1.66
min.	0.12	0.03	0.14	0.24
max.	2.71	0.67	3.21	6.22
mean	0.68	0.17	0.80	1.53

Conclusions

The surface soil layer around the uranium mine hole has uranium activities greater than worldwide average; this can mainly due to the leaved waste of drilling and exploration on the surface layer of soil surrounding the mine.

The thorium activities were within normal level in the studied area. Generally, potassium radionuclide in soil samples was in the range of worldwide average.

The absorbed dose rates of studied area are higher than the criterion limit of gamma radiation dose rate with an average of nine times.

Finally, from the radiation protection point of view the studied area is considered to be not safe inhabitants because the values of both internal and external hazard indexes associated with the samples are higher than unity. Thus, the human inside the area are supposed to acquire radiological complication.

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