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Determination of the Radon Concentration of Gas 222Rn Emitted From Sludge Samples Located in the Radioactive Waste Collection Warehouse in Khader Al-Maa South of Basra City – Iraq

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Abstract

Thirty-one samples of sludge were taken, stored at the natural radioactive waste collection site, NORM, in Khidir Al-Maa, south of Basra city, resulting from the operations of extracting and filtering crude oil in oil fields in Basra Oil Company, to determine gas concentrations Radon ^{222}Rn and other radioisotopes in Sludge models. The rapid electronic technology was adopted through the RAD7 device, the active method, and the gamma analysis technique using HPGe High Purity Germanium Detectors to determine the specific activity of radioactive elements. The measurements for this study showed 98463±7031 Bq.m^{-3} in sample no. S10 a barrel of sludge was brought from the third Degassing Station plant in the northern Rumaila oil field and least concentrated is 8317±594 Bq.m^{-3} in sample no. S14 sludge belonging to the sixth Degassing Station / West Qurna-1, the study also revealed the presence of high concentrations of the specific effectiveness of radioactive isotopes (^{214}Pb, ^{212}Bi, ^{226}Ra) and (^{212}Pb, ^{212}Bi, ^{228}Ac) of the uranium-238 series and Thorium-232 series in a sludge form selected from a barrel returning to the third gas isolation plant - North Rumaila Oilfield and stockpile at the collection site, where the specific activity value of radium was ^{226} Ra 133851 Bq.k^{-1} of the uranium-238 series and for lead ^{212}Pb 56432 Bq.k^{-1} of For uranium-238 series compared to their concentrations, 32 Bq.k^{-1} 45 Bq.k^{-1} in a natural soil model. The effective dose to which on-site workers would be exposed in contact was calculated and found to be 196.92 Svh^{-1}, a dose that exceeded the proposed 50 Sv. h^{-1} dose limits by the US EPA. The probability of developing cancer due to exposure to radon per million people was found and it was found to be a high value compared to the suggested value (170-230) per million people previously by ICRP.

Keywords: Sludge, Rn, Radium, RAD7, HPGe High Purity Germanium Detectors, Effective dose.

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"تحديد تركيز غاز الرادون ^{222}Rn المنبعث من نماذج الحمأ الموجودة في مخزن تجميع النفايات المشعة في خضر الماء جنوب مدينة البصرة–العراق"

الخلاصة:

جمعت في هذه الدراسة (31) عينة من الحمأ (Sludge) المخزون في موقع تجميع النفايات المشعة الطبيعية المنشأ NORM في خضر الماء، جنوب مدينة البصرة- العراق، الناتج عن عمليات استخراج وتصفية النفط الخام في شركة نفط البصرة، لتحديد تراكيز غاز الرادون ^{222}Rn ونظائر مشعة أخرى كالراديوم ^{226}Ra والاكتنيوم ^{228}Ac في نماذج من الحمأ. اعتمدت طريقة الكترونية سريعة للقياس من خلال جهاز متطور الكترونيا يدعى RAD7 وتقنية تحليل طيف كاما باستخدام كاشف الجرمانيوم عالي النقاوة HPGe High Purity Germanium Detectors. بينت نتائج الدراسة هذه إن اكبر تركيز لغاز الرادون في الحمأ (Sludge) هو $98463+7031\pm Bq.m^{-3}$ في عينة رقم (S10) برميل حمأ جلب من محطة عزل غاز الثالثة / حقل الرميلة الشمالي النفطي واقلها تركيزا $8317+594\ Bq.m^{-3}$ في عينة رقم (S14) حمأ عائدة لمحطة عزل الغاز السادسة / حقل غرب القرنة-1، كما بينت الدراسة وجود تراكيز مرتفعة للفعالية النوعية للنظائر المشعة (^{214}Pb, ^{212}Bi, ^{226}Ra) و (^{212}Pb, ^{212}Bi, ^{228}Ac) التابعة لسلسة اليورانيوم -238 و سلسلة الثوريوم -232 في نموذج حمأ منتخب من برميل عائد إلى محطة عزل الغاز الثالثة – حقل الرميلة الشمالي النفطي والمخزون في موقع التجميع،حيث بلغت قيمة الفعالية النوعية للراديوم^{226}Ra $133851\ Bq.k^{-1}$ التابع لسلسة اليورانيوم -238 وللرصاص^{212}Pb $56432\ Bq.k^{-1}$ التابع لسلسة اليورانيوم -238 مقارنة بتراكيزهما، $32Bq.k^{-1}$ $45Bq.k^{-1}$ في نموذج التربة الطبيعي. تم حساب الجرعة المؤثرة التي قد يتعرض لها العاملين في الموقع في حالة التماس مع الحمأ ووجد انها $196.92\mu Svh^{-1}$ وهي جرعة تعدت حدود الجرعة المقترحة $50\mu Sv\ h^{-1}$ المقترحة من قبل EPA في الولايات المتحدة. قدرت احتمالية الإصابة بالسرطان عند التعرض لغاز الرادون لمليون شخص ووجد أنها عالية مقارنتاً بالحدود المقترحة (170-230) لمليون شخص من قبلICRP.

1. Introduction:

The oil and gas industry is one of the most important and most concentrated industries for natural radioactive materials (NORM), as its workers are exposed to the danger of (NORM) [1] which appear remarkably as radioactive waste accompanying the extraction and refinement of crude oil. These radioactive wastes exist in various forms, such as sludge and scale deposits in pipes and valves, as well as faces accompanying the water associated with crude oil separation production processes, and generally contain quantities of palpable concentrations, especially of radium isotopes Ra, that may cause the environment to be exposed to pollution and then radiation exposure to workers who, they work near these contaminated sites. NORN is present in oil and gas formation basins, like other mineral elements, with varying concentrations. These radioactive materials come out during the search for crude oil from deep underground depths with production fluids. These materials are

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

deposited in oil production lines and points within a single production line, and usually .The highest effectiveness of ^{226}Ra is in the network of oil exit valves from the well and its branches to other oil equipment, and its concentration increases as scaly deposits in oil transportation pipelines, especially at the sites of bends and protrusions, and it is deposited very heavily in the crude oil insulators. The appearance of these substances in squamous sediments and sludge naturally leads to an increase in radioactive levels due to the accumulation of these substances in them [2-5] Oil and the general population come from radon gas of natural origin, which is a colorless, tasteless, and odorless gas that is seven and a half times heavier than air. Obstruction, which makes the level of radiation exposure high for large groups of the public. The inhalation of radon gas or contact with it is a certain risk, because this gas is considered the largest contributor to the exposure of the general population to natural radiation, and this gas may contribute in the range of 50-55% of the total dose that a person may be exposed to from all natural sources of radiation [6-8] Exposure to this gas causes a serious health and environmental problem, and many studies have proven a close relationship between exposure to radon gas emitting alpha particles and lung cancer, that the process of inhalation and the entry of air saturated with radon gas into a person's lungs leads to precipitation to deposition. A large amount of it is on the walls and lining of the respiratory system and thus leads to the absorption of large quantities of doses by the bronchi [9] two measurement methods were used in this study called the first direct method through a fast advanced device called RAD7, and the second using a high purity germanium detector HPGe High Purity Germanium Detectors. The aim of this research is to determine the concentration of radon gas and other radioactive isotopes such as radium ^{226}Ra and ^{228}A, in samples of sludge stored at the radioactive waste collection site in Khader Al-Maa - south of Basra city.

2. Study station

The study station is located in the radioactive waste collection site - Khader al-Maa - Basra city - Iraq, Figure (1) near the Kuwaiti border and 17 km away from the center of Basra city. The site is one of the largest temporary storage site for natural

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

radioactive waste in the city of Basra and Iraq in general. It covers an area of 250 x 120 square meters, containing more than 25,000 barrels of slag contaminated with radioactive materials of natural origin. The capacity of one barrel is 220 liters, as well as hundreds of thousands of tons of solid radioactive waste. The site is surrounded by concrete blocks.

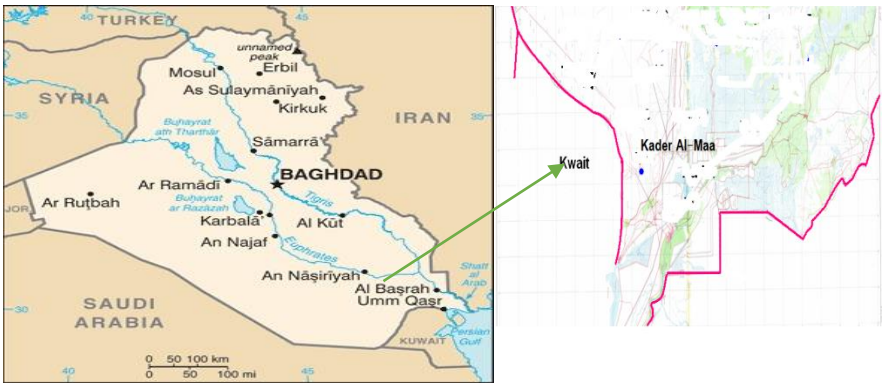

Fig. (1): Map of Basra Governorate showing the study areas.

3. Experimental work and materials used

Sludge samples were taken from the radioactive waste collection and storage site - Khader al-Maa in Basra city - Iraq in April 2021. When collecting these samples, they were subjected to drying at a temperature of 110°C for twenty four hours by using a thermal oven, and sieved with a 2µm sieve to filter them from Impurities and foreign bodies, then ground into a very fine powder using a grinder, and samples were prepared with scientifically approved specifications for the purpose of determining the concentration of radon and other radioactive isotopes Figure (2).

Fig. (2): The Sludge in the radioactive waste collection store - Khaddar alma in the study area.

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

3.1 Advanced radon Technology

RAD7 technology, which is a fast electronic device that is adopted to monitor radon in air, soil and water Figure (3). The work of this technique is based on the RAD7 device, which records the measurement of radon gas concentrations emitted from the samples in this study in real time. Where the device withdraws a quantity of the generated gas Grab from above the sample and sends it to the LOCAS cell to determine the concentrations of radon gas in the ground slag models, after making sure that there is no water vapor associated with the gases, and the process of pumping the device continues for five minutes after which the device starts for five Another minutes. This counting process represents a determination of an elemental concentration of ^{218}Po- (3.05 min), ^{212}Po (164 μs), so the measurement process is after (540-900) seconds, and this device is characterized by its ability to determine the energy of particle alpha electronically, which can distinguish between isotopes of elemental radon Polonium-^{218}Po-Polonium-^{214}Po-between radon^{222}Rn and oxon^{220}Rn, the RAD7 detector must be dried (purging) with new air for 600 seconds by connecting the drying unit in a closed loop with the RAD7 device. Desiccant, when the humidity level appears to us less than 6% in this case we start the test, the pump works for 300 seconds during which the radon is extracted from the sample and it is sent to the measuring room in RAD7 and then the RAD7 device stops for a period of more than 300 seconds in order to reach the state of equilibrium and then The process is repeated for four cycles, at a rate of 300 seconds per cycle, thus the total test duration reaches 1800 seconds, and at the end of each operation, RAD7 provides us with the required information and data.

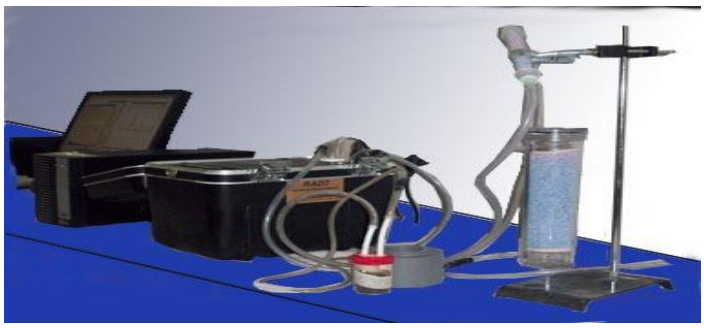

Fig. (3): The shape of the RAD7 device used in the search.

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

3.2 Gamma spectrum analysis technique using HPGe high-purity germanium detector.

The technique of gamma spectrum analysis is adopted by using the high-purity germanium detector CANBERRA connected to a multi-channel spectral analyzer and an IBM calculator for the purpose of recording gamma-ray spectra and results, and connected to a liquid nitrogen flask for the purpose of cooling to a low temperature, which is a low temperature that is adopted to maintain the crystallization of the detector during work. This technique is an important and widely used technique in the detection of isotopes that emit gamma rays. Standard sources, type ^{60}CO, possessing two gamma lines (1332, 170) Kev and cesium ^{137}CS at power 662 Kev were used for the purpose of obtaining the amount of energy per channel and in order to calculate the real area under the peak, 1kg of sludge from each model was placed in the (Marineli Beaker), which is a plastic cup designed in a manner that makes the used model close to the effective detector area in order to give a high counting efficiency of gamma rays emitted from the radioactive nuclei in the sample The natural background radiation is subtracted from the area of all the samples under study to obtain the net area under the peak.

4. Results and Discussions

The outputs of the results were included in Tables (1) and (2) where Table (1) shows radon concentrations in samples of sludge at the radioactive waste collection site - Khader Al-Maa, south of Basra city, where the table shows the minimum concentration of radon 8317 + 594 Bq.m^{-3} in the sample of sludge transferred from the sixth degassing station, West Qurna, and that the maximum value of radon concentration was 98463 + 7031 ± Bq.m^{-3} in the sludge sample belonging to the waste of the third degassing plant from the northern Rumaila oil field in the assembly store, which is sample No. S10 , Figure (4,shows the Relationship note between the concentration of radon gas emitted from sludge samples in study area at the NORM radioactive waste collection site in the city of Basra - southern Iraq, measured by RAD7 technology, as for the concentrations of other radioactive isotopes such as radium-226 in slag samples in The location of the study area Table

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

(2). We note that there is a very clear increase, as we have very high concentrations of radium-226, reaching 133,851Bq.K^{-1}, which is one of the radionuclides in the decay series of uranium-238 due to its environmental behavior and its long half-life. He is 1620 years old and his health risk lies in his deposition In bones [10] they were higher than the suggested global average of 32Bq.kg^{-1}, and also recorded high specific efficacy values for ^{212}Pb above the suggested limits of 45Bq.kg^{-1} by [11] UNSCEAR, 1993 It is likely that the reason for this increase in gas concentrations Radon ^{222}Rn and radioisotopes (^{214}Pb, ^{214}Bi, ^{226}Ra) belonging to the uranium-238 series and radioisotopes (^{212}Pb, ^{212}Bi, ^{228}Ac) belonging to the thorium - 232 in sludge samples selected from the radioactive waste collection site - Khidr Alma, we also showed an increase in the concentration of other radionuclides compared to their concentration in sludge with the results of previous international studies, Table No. 3 [12].

U-238 and Th-232 in the sludge models selected from the radioactive waste collection site - green water in the study area, since the sludge present at the site resulted from production and maintenance operations accompanying the work in the crude oil production and separation plants, Basra Oil Company - southern Iraq, and that this is a natural product because oil It is brought from the depths of the earth, and contains quantities of natural radioactive materials NORM, and the reasons for the emergence of varying concentration from one model to another we suggest that to the nature of the geological formation of the fields from which the models were brought. The effective dose was calculated upon contact with the slag, considering that 1Bq.m^{-3} is equivalent to 0.025mSv y^{-1} according to [13-14] EPA, CEC, meaning that 1Bq/m3=2.854x10-3µSv h^{-1}, and from that we conclude that the greatest concentration of radon is 98463+7031±Bq. m^{-3} in the sludge samples under study is equivalent to an effective dose of 196.92 µSvh^{-1} on contact, which is a dose higher than the permissible dose level of 50 µSv h^{-1} [15] recommended by the EPA in the United States Figure (6) The relationship between gas concentration Radon and effective dose of sludge samples at the collection site.

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

The probability of developing cancer resulting from exposure to radon gas was estimated per million individuals by multiplying the annual effective dose by mS.y (18×10-6). The probability of infection recorded in this research, when adopting the maximum dose of radon concentration 196.92μSvh^{-1} results in a high value for each million people, which is considered a very dangerous value based on the proposed limits, which are (170-230) per million people [16-18].

Table (1) Rn -222 concentration (Bq\ m^3) in Sludge sample barrels of radioactive waste, radioactive waste collection site - khadir Alma in the study area.

Sample. No	Name of on-site stock Sludge barrel model	Radon concentration (^{222}Rn) in Bq/ m^3	Annual effective dose μSv/ h	Lung Cancer ×10^{-6}
S1	Central Degassing Station South	21314+152	42.62	774.21
S2	Sothern Degassing Station	23760+1697	47.52	863.22
S3	Qurenit -1Degassing Station	30412+2172	60.82	1104.82
S4	Shamei-1 Degassing Station	29543+2110	59.08	1073.21
S5	Ratka Degassing Station	31327+2237	62.65	1133.89
S6	Qurenit-2 Corporation station	33713+2408	67.42	1224.71
S7	Shimea-2 Degassing station	36821±2630	73.64	1337.70
S8	Central Degassing Station North	28765+2054	57.53	1045.06
S9	Second Degassing Station	29433±2102	58.86	1069.22
S10	Thread Degassing Station of North Rumaila	98463+7031	196.92	3544.63
S11	Fourth Degassing Station of North Rumaila	31162+2225	61.75	1121.72
S12	Fiveth Degassing Station of North Rumaila	44572+3183	88.33	1604.56
S13	Central Pump Station	38120+2722	75.54	1372.22
S14	Sexes Degassing station of East Qurna Quran	8317+594	16.63	299.36
S15	Seven Degassing station of East Qurna	9123+651	18.07	328.225
S16	Eight Degassing station of East Qurna	9578±684	18.98	344.78
S17	Zubair Degassing Station	32480±2320	64.36	1169.13
S18	Zubair musharaf Degassing Station	33752±2410	66.88	1214.90
S19	hamar musharaf Degassing station	40134±2866	79.53	1444.70
S20	alrafidia Degassing station	63918±4565	126.67	2301.02

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

S21	qubat safwan Degassing station	15952±1139	31.61	574.21
S22	Zubair Warehouse-1	18245± 1303	36.15	656.68
S23	Zubair Warehouse-2	20763± 1483	41.14	747.29
S24	altuwba Warehouse	19548± 1396	38.73	703.55
S25	Warehouse- ps1	21365± 1526	43.90	797.46
S26	Warehouse in North Rumaila	19688±1406	39.01	708.63
S27	nahr bin Omar Degassing Station	18216±1301	36.10	655.77
S28	majnun Degassing St-1ation	91325±6523	180.98	3287.59
S 29	Majnun-2 Degassing Station	18245± 1303	36.15	656.68
S30	altuwba Degassing Station	30763± 2197	60.96	1107.36
S31	nahr bin Omar Degassing Station	18640± 1331	36.94	662.85

Table (2) Effective concentration of specific activities in a sludge model selected from the waste collection site - khadir Alma - belonging to the Thread Degassing Station of North Rumaila in the study area measured by the gamma spectrum analysis technique HPGe.

NO	Radionuclide's	Radiation chains	Specific activities in Bq/kg
1	Pb-212	Th-232	56432
2	Bi-212	Th-232	33720
3	Ac-228	Th-232	52519
4	Pb-214	U-238	109479
5	Bi-214	U-238	117135
6	Ra-226	U-238	133851

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

Table (3) The effective concentrations of radium ^{226}Ra and radium ^{228}Ra emitted from Sludge and scale sediment models for different international studies [12]

Country/ material	^{226}Ra kBq·kg^{-1}	^{228}Ra kBq·kg^{-1}	^{232}Th kBq·kg^{-1}
Brazil -scale	19.1 to 323.0	4.21 to 235.0	-
Brazil -sludge	0.36 to 367.0	0.25 to 343.0	-
Algeria-Hard scale	1.0 to 950.0	-	-
Algeria-Soft scale	1.0 to 300	-	-
Tunisia- scale	4.3 to 658	-	-
Algeria-sludge	0.069 to 0.393	-	-
Norway- scale	0.3 to 32.3	0.3 to 33.5	-
Norway-sludge	0.1 to 4.7	0.1 to 4.6	-
Brazil-scale	< 839	< 377	-
Brazil-sludge	3,060	2,570	-
Various locations	< 1,000	< 360	< 360

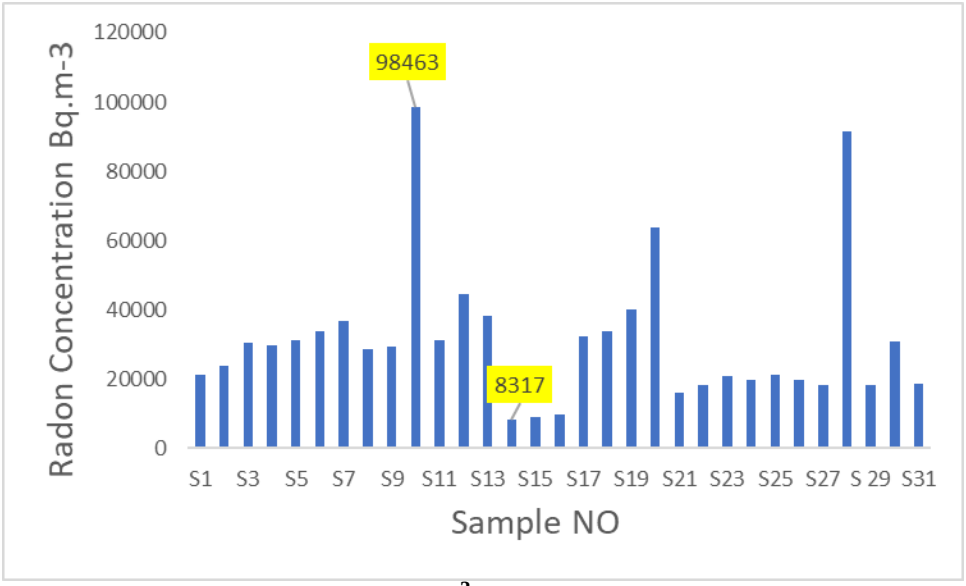

Fig. (4): R-222 concentration ((Bq.m^{-3}) measured in Sludge samples by RAD7 in study area.

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
E- ISSN: 2710-1096

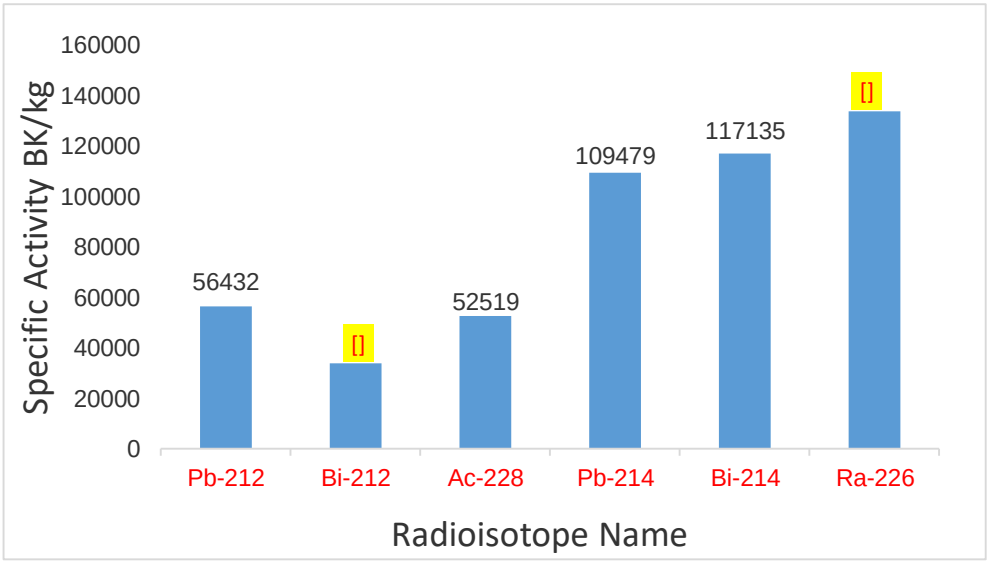

Fig. (5): The values of the specific activity of radioactive isotopes (B.kg^{-1}) in a Sludge sample and solution returning to thread Degassing Station of North Rumaila and stored at the radioactive waste collection site in Khader Al-Maa - the study area.

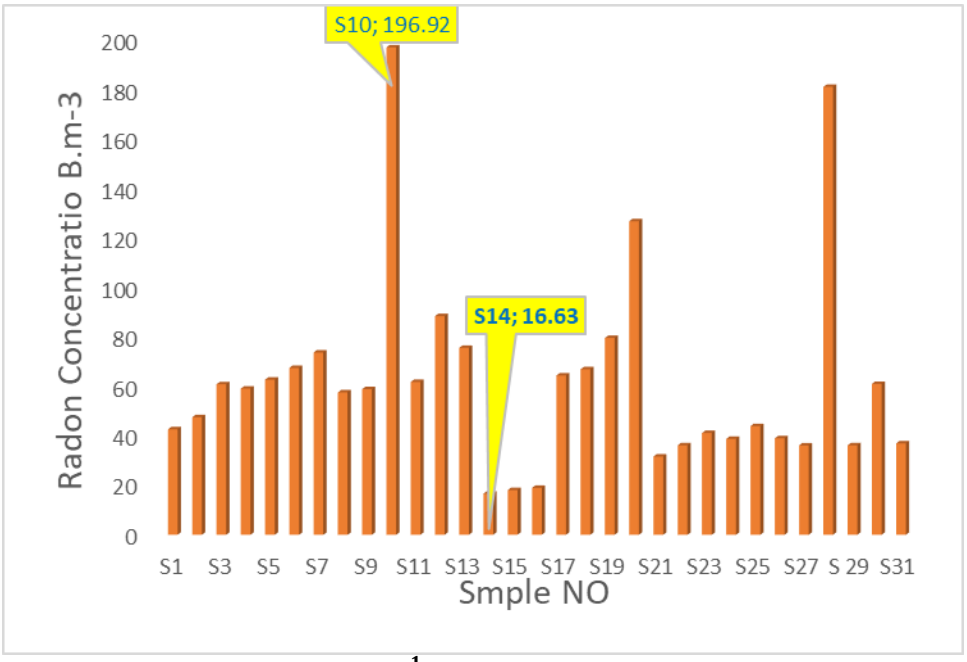

Fig. (6) Effective dose (Svh^{-1}) in Sludge samples in the study area.

Open Access
No. 34 part 2, April 2022, pp. 256-269
P- ISSN: 2220-5381
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5. Conclusions:

This study, for measuring the concentrations of radon gas emitted from Sludge and other radioactive isotopes from the NORM, radioactive waste collection site in the khadir alma - south of Basra city, showed Using advanced RAD electronic technology and High Purity Germanium Detectors (HPGe).

1. The largest concentration of Sludge is 98463 ± 7031Bq.m^{-3}, which appeared in sample (S10) in a ±model from a sludge barrel at the study site transferred from the Thread Degassing Station of North Rumaila field, the concentration of radon gas in the sludge of the Sixth Gas Isolation Station / West Qurna field is the least concentrated of the species 8317 + 5949 Bq.m^{-3}
2. The emergence of an increase in the concentrations of radioactive isotopes (^{214}Pb, ^{214}Bi, ^{226}Ra) (^{212}Pb, ^{212}Bi, ^{228}Ac), which belong to the uranium-238 and thorium-232 series, respectively, ibn high rates compared to their permissible concentration according to UNSCEAR.
3. It was found that the annual equivalent dose in slag samples (Sludge) in the production of slag drums studied is higher than the permissible dose level of 50 µSv h^{-1} proposed by the EPA in the United States.
4. This concentration radon gas and the specific effectiveness of radioisotopes and doses have a negative impact on the health of the workers at the site In case of non-compliance with radiation safety procedures in the workplace.
5. The rsearch added important information and a roadmap about radon concentrations in oil sludge at Khidir alma site for collecting and storing radioactive assemblies in Basra Governorate - southern Iraq.

Open Access
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