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Radiological hazard Assessment Due to Natural Occurring Radioactive Materials (NORM) in Oil and Gas Production Industry –East Baghdad Oil Field

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Abstract

Concentration of NORM then radiological hazard indices were investigated and assessed in different stages of oil and gas production industry that belongs to East Baghdad Oil Field. Sixteen samples of different types of materials were collected representing oil and gas production from first stage to final stage. The samples are prepared then sent to Radiation Protection Center (RPC) / Ministry of Environment for radioactivity analysis using gamma ray spectrometry system based on HPGe. The results show that max value of Ra-226 is 252.4 Bq/kg in sediment sample collected from the final stage of water treatment before it is transferred to disposal well and min value is 1.2 Bq/kg in formation water. The max. activity of Th-232 is 79.4Bq/kg also in sediment sample and the min value 2.9 Bq/Kg in crude oil in storage tank while, the max. value for K-40 is 529.6 Bq/Kg in burning stage (burn pit) and the min value is 2.8Bq/kg in Pre-treatment crude oil. The ambient exposure dose values near the sampling locations were recorded using dosimeter type GMC-300. The results are corresponding with the values of NORMs in the locations. The ambient gamma rates in the study area range between (0.04-0.22) µSv/h corresponding to (57-314 nGray/h). Most locations have values above the average worldwide value of (58 nGray/h). NORMs are caused absorbed dose in max. contribution (79%) to the total ambient gamma dose. Radium caused the max. contribution among the others. All the hazard indices indicate that most of the sampling locations have indices acceptable and with no hazard.

Keywords: NORM, oil and gas, radiological Hazard, Radiation, Gamma spectrometry

تقييم المخاطر الاشعاعية نتيجة للمواد المشعة المتواجدة طبيعيا في مصنع انتاج النفط والغاز – حقل نفط شرق بغداد

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الخلاصة

جرى قياس تراكيز النويدات المشعة المتواجدة طبيعيا ومعاملات المخاطر الاشعاعية لموقع مصنع انتاج النفط والغاز التابع لحقل شرق بغداد. تم اختيار نماذج مختلفة تمثل مراحل انتاج النفط والغاز في موقع المصنع وحصّرت النماذج وقيس النشاط الاشعاعي بمختبرات مركز الوقاية من الاشعاع/وزارة البيئة باستخدام منظومة تحليل اطياف كاما المستندة الى عداد الجرمانيوم عالي النقاوة. بينت النتائج ان اعلى قيم لنشاط الراديوم-226 كانت 252.4 بكريل/كغم في نموذج الرواسب من المرحلة النهائية من عملية معالجة المياه وفصل النفط قبل انتقاله الى ابار التصريف بينما اقل قيمة للراديوم تم قياسها في نموذج المياه المكمنية(1.2

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بكريل/كغم) . بينما تراوحت تراكيز الثوربوم-232 والبوتاسيوم- 40 بين (2.9-79.4) بكريل/كغم و (2.8 -529.6) بكريل /كغم على التوالي. تتراوح جرع التعرض كاما المحيطية في منطقة الدراسة بين(0.04-0.22)مايكروسيفرت/ساعة والتي تعادل (57-314)نانوكراي/ساعة. كانت معظم مواقع النمذجة ذات جرع اشعاعية تقترب أو أعلى من معدل الجرع العالمية(58 نانوكراي/ساعة). كانت مساهمة النويدات المشعة المتواجدة طبيعيا في اعلى قيمها تمثل(79%) من الجرعة المحيطية الكلية وكانت مساهمة الراديوم هي الاكبر من بين النويدات الاخرى. بينت معاملات المخاطر الاشعاعية ان معظم المواقع ذات قيم معاملات مقبولة ولا تشكل مخاطر اشعاعية.

Introdaction

Naturally occurring radioactive materials (NORMs) of uranium series, thorium series and potassium-40 are present everywhere in the earth crust. Concentration of these radionuclides depends on the composition of the soil and rocks. All these nuclides produce radiation doses to all human beings. The NORM occur in oilfields and reservoir rock contains small amounts of natural uranium and thorium and their radioactive daughters [1].

Oil and gas industry, and oilfield are examples of NORM sources in Iraq. Most of these sites and industries have been not radiologically assessed. They may cause high radiation exposure to workers, in addition to their environmental impact during the production process of petroleum in East Baghdad Oil Field. These radionuclides might be transferred by different means to the surrounding environment.

Radioactive decay of ²³⁸U and ²³²Th produces several series of daughter radioisotopes of different elements and of different physical characteristics with respect to their half-lives, modes of decay, and types and energies of emitted radiation [2]. Various radioactive wastes are produced in the oil and gas industry. Sludge, drilling mud, and pipe scales are example of materials that can contain elevated levels, if NORM and the radioactive materials move from site to site as equipment and materials that may be reused. A common understanding of the radiation hazards and protection principles within the petroleum industry would lead to efficient and increasingly safer operation.

The concentration of NORM then radiological hazard indices would be investigated and assessed in different stages of oil and gas production industry belong to East Baghdad Oil Field. **Study Area Description**:

The study area covers about 25 km² represents farmlands surrounding the oil and gas production industry (O&GPI) which belong to East Baghdad Oil Field (EBOF) under the responsibility of Midland Oil Company. It is located northern Baghdad city (Figure-1).

The super-giant East Baghdad Oilfield is, situated in Baghdad and Salah al-Din governorates, discovered in 1976. The field has been delineated and developed with 80 wells. Production started in 1980 over a small section of the field using pilot plant facilities. The field includes oil and gas production industry which consists of two lines of production; one for oil and the other for gas. Figure-1 shows stages of oil and gas production in this industry.

Recently, although there are 97 drilled wells but the industry produces about 10000 oil Barrels/day from 40 produced wells.

The field has a faulted, NW-SE trending structure, with oil production to-date coming mainly from the late Cretaceous Tanuma and fractured Khasib carbonates and from the early Cretaceous Zubair sandstone Formation.

Field Work and Methodology

Samples Collection

The samples include different types of materials which were selected and collected from inside the O&GPI. Sixteen samples of different types of materials were collected represent oil and gas production from first stage to final stage as shown Figure-2 and in Table-1. Each sample was put in nylon bag of 3 kg capacity and label with code No., type of sample, location and date of sampling. These samples include sludge (SL), scale (SC), sediment (SE), and crude oil (O) (two samples were taken from pre-treatment oil and two samples were taken from oil after treatment (storage tank)), one sample of water was taken from formation water (W) and one sample of soil (S). The samples are then sent to laboratories for preparation then sent to Radiation Protection Center (RPC) / Ministry of Environment for radioactivity analysis.



Figure 1- Oil and Gas Production Industry within East Baghdad Oilfield, showing sampling locations in sketch map of stages of oil and gas production in EBOF.

Code No	Material	Location of Sample			
SL1	Sludge	First Stage (1 st separator)			
SL2	Sludge	Second Stage (2 nd separator)			
SL3	Sludge	Third Stage (3 rd separator)			
SL4	Sludge	Gas Stage (dehydrator)			
SL5	Sludge	Wet oil stage (dehydrator)			
SC6	Scale (outside the adapter)	Desalter & dehydrator & stabilizer			
SE7	Sediment result from Skamming Oil	Final Stage treatment water (waste water)			
SC8	Scale (inside the pipe)	pre-injection water			
SC9	Scale (burn pit)	Burning stage			
O10	Crude oil	Storage tank			
011	Crude oil	Storage tank			
012	Crude oil	Well Oil (pre-treatment oil)			
013	Crude oil	Well Oil (pre-treatment oil)			
W14	Formation Water	Oil Well			
S15	Soil	Near gas dehydrator			
S16	Soil	Near separators			

Table 1- Samples code No., type and location of sample taken from different stage of oil & gas production in EBOF.

Measurements

Geiger Muller Counter type GMC-3000 is used for measuring radiation exposure dose rate in microsevert per hour (μ Sv/h). It measures ambient doses between 0.01-1000 μ SV/h.

Gamma ray spectrometry system based on HPGe was used to analyze NORM in the samples. Concentration in Bq/kg of Radium-226, Th-232, and K-40 were measured in all samples. The specific activities were averaged from gamma-ray photo peaks at several energies. The gamma-ray lines at 180.5keV from ²²⁶Ra and/or lines at 295.2 keV and 351.9 keV from ²¹⁴Pb and at 609.3 keV and 1764.5 keV from ²¹⁴Bi were used to determine the specific activity of ²²⁶Ra. The gamma-ray lines of 338.4 keV, and 911.2 keV from ²²⁸Ac, the 727.3 keV from ²¹²Bi and 583.2 keV and 2614.5 KeV from ²⁰⁸Tl were used to determine the specific activity of ²³²Th. The specific activity of ⁴⁰K was measured directly by its own gamma-ray line at 1460.8 keV. The energy calibration and efficiency were calibrated by using a standard source of a multi energy made by the American Canberra Company. The Marinilli geometrical shape was used to measure the activity of the samples. The measurements were achieved in laboratories of Radiation Protection Center/ Ministry of Health and Environment/Iraq.

Preparing Samples

Surface soil, sludge and scale and sediment samples should be dried for a sufficient period of time at a fixed temperature to acquire a constant dry weight using electric oven with temperature (105 °C). Large gravels and sand are removed using sieve of (18mm). Samples are crushed to fine powder.

All samples (surface soil, sediment, sludge, scale, crude oil and formation water) are then thoroughly homogenized. (0.5-1) kg of each sample is taken and storage for 25 days in completely sealed Marinilli beaker to achieve equilibrium between radon and its daughters [3].

Estimation of Hazard Indices

Many hazard indices are used to indicate the level of radiation and its radiological impact according to level values recommended by IAEA, ICRP and UNSCEAR and other agencies. The following hazard indices were used in current study

Gamma Absorbed Dose Rate

The outdoor air-absorbed dose rates due to terrestrial gamma rays at 1m above the ground level can be calculated from ²²⁶Ra, ^{232Th}, and ⁴⁰K concentration values in soil assuming that the other radionuclides, such as ¹³⁷CS, ⁹⁰Sr.and the ²³⁵U decay series can be ignored as their contributions are expected to be negligible to the total dose from environmental background [4].

The gamma dose rate (D) in the outdoor air at 1m above the ground level can be calculated by the equation [4]

$\sqrt{nC_{11}/h} = 0.462C_{11} + 0.621C_{11} + 0.041C_{11}$	(1)	۱.
$V((V/V)) = V_4 O_2 V_{P_0} + V_0 O_2 (V_{T_0} + V_0 U_4) V_4$)
$(10 j/1)^{-0.1020 Ka} + 0.0210 III + 0.011 CK$	(+)	/

Where D is the dose rate in (nanogray per hour) and C_{Ra} , C_{Th} , C_K are the radioactivity in (Bq/kg) of ²²⁶Ra, ²³²Th, and ⁴⁰K respectively.

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent received outdoor by a member is calculated from the absorbed dose rate by applying dose conversion factor of 0.7 Sv/Gy and the occupancy factor for outdoor. In current study, maximum of 8 hours/day for 6 days per a week (2503h per year) are assumed as working time for workers inside the oil and gas production plant. Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 VGy^{-1} , which is used to convert the absorbed rate to human effective dose equivalent [5]. AEDE is determined using the following equation [5]

AEDE (outdoor) (nSv/v) = Absorbed dose (nGv/h)×0.7Sv/Gv×2503h/year(2) **Radium Equivalent Activity (Ra eq)**

Radium equivalent activity (Ra eq) is a common index used to compare the specific activities of materials containing 226 Ra, 232 Th, and 40 K by a single quantity which takes into account the radiation hazards associated with them [6]. The following formula is used to acquire Ra eq: [4, 5]

 $Ra \ eq = C \ Ra + 1.43C \ Th + 0.077C \ K.$ (3) Where: CRa, CTh, CK are the radioactivity concentration in Bq/kg of²²⁶Ra, ²³²Th, and ⁴⁰K respectively. The material whose Ra eq concentration exceeds 370Bq/kg should be avoided due to radiation hazards [7].

External Hazard Index (Hex)

Many radionuclides occur naturally in terrestrial soil and rock and upon decay; these radionuclides produce an external radiation field to which all human beings are exposed. In term of dose, the principal primordial radionuclides are ²³²Th, ^{238U} and⁴⁰K.

The external hazard index (Hex) is defined as [7]:

respectively. The value of this index must be less than unity for the radiation hazard to be negligible (Sam and Abbas, 2010). Hex equal to unity corresponds to the upper limit of Raeq 370 Bq/kg [4].

Internal Hazard Index (H_{in})

In addition to the external hazard, Radon (²²²Rn), a radioactive noble gas with a half-life of about 3.8 days and daughter product of ²²⁶Ra, accounts for half of the radiation dose to the general population [8] and is currently considered as a major source of lung cancer [9]. The internal exposure to Radon and its daughter products are quantified by the internal hazard index (H_{in}) which has been calculated by the following relationship [4]:

 $H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \le 1.$ (5) Where: A_{Ra} , A_{Th} and A_K are the mean activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in (Bq. kg⁻¹). H_{in} should be less than unity for a radiological safety [10].

Results and Discussion

Radioactivity of NORMs inside Oil and Gas Production Industry in (EBOF)

The activities of NORMs in all samples inside the O&GPI have been presented in Table-2. The table shows that the max value of Ra-226 is 252.4 Bq/kg in sediment sample (SE7) collected from the final stage of water treatment (Figure-2) before it transfer to disposal well and min value is 1.2 Bq/kg in formation water (W14). The max activity of Th-232 is 79.4 Bq/Kg in sediment sample (SE7) and the min value 2.9 Bq/kg in Pre-treatment crude oil (O12) while, the max value for K-40 is 529.6 Bq/kg in burning stage (burn pit) and the min value is 2.8 Bq/kg in crude oil (O11) in storage tank.

During the extraction of crude oil and gas, NORM is coming with produced water and accumulates in sludge, scale and sediment. It is logical results that radium has max level in sediment samples collected from the final stage of water treatment because most of radium is dissolved in formation water and transferred with the associated oily water then it is deposited as salts with other elements such as barium and are being enhanced upon unique conditions (due to change in temperature, pressure, acidity etc.) [11, 12] especially with increasing salinity [13].

Scale and sludge have significant level of radium. Usually scale has more radium concentration than sludge. For comparison, the worldwide average concentration of Ra-226, Th-232 and K-40 soil are 32 Bq/kg, and 45 Bq/kg and 412 Bq/kg, respectively [14]. It seems that all the maximum values of the NORMs (Ra-226, Th-232 and K-40) are above the average worldwide. The average value of Ra-226 in sludge and scale are (16.8 ± 13.1) , (94.9 ± 46.8) Bq/kg while oily sediment sample contains and 252.4 Bq/kg of radium. So, the average value of Ra-226 in sediment and scale are above the average worldwide while average radium content in sludge is below the average value worldwide. Accordingly sludge in current location is not a source of contamination for soil in sampling sites. In the other hand, sample SL2 have radium contents above the average worldwide in soil. While the average value of Th-232 concentrations in sludge, scale are (11.2 ± 3.5) , (19.8 ± 9.9) and 79.4 Bq/kg in sediment sample. Only that in sediment sample seems to be above the average worldwide. K-40 average concentrations are (145.8 ± 86.6) , (60.4 ± 31.5) and 120.0 Bq/kg in sludge, scale and sediments. Other samples have NORM concentration below the average value in worldwide. In compression the average in current study is below values in study achieved by Al-Talib [15] in oilfield in northern region of Iraq and below the average of Ra-226 in sludge in Oman (547B/kg).

The activity of Ra-226, Th-232 and K-40 were plotted as colored classed position map overlaying the base map (sketch) of the O&GPI using software program Surfer-11 as shown in Figures-2, 3 and 4. More than 25% of overall sites have radium-226 concentrations above average the worldwide in soil. So it should to take in consideration to monitor these sites. According to Figure-4, sites that have thorium concentrations above the average worldwide in soil represent 12.5% of the overall sampling sites. These sites are burning pit and water treatment & well deposal sites. While Figure-5 shows that only two sites have K-40 concentrations above the average value of the worldwide in soil. From the overall sites the sites: separators, dehydrators, water treatment and burning pit are sites of relatively high contents of NORMs.

The ambient exposure dose values near the sampling locations were recorded using dosimeter type GMC-300. The results, shown in Table-2, are corresponding with the values of NORMs in the locations. The doses range between (0.04-0.22) μ Sv/h (57-314 nGray/h) with average (0.094±0.06) μ Sv/h (133.9±78.7)nGy/h. The maximum value is recorded near the 4th stage of oil production process (Desalter & Dehydrator process) (sample SC6) where scale has been deposited. It is logical a result due to this site has high concentration of radium-226. Although, the maximum concentration of Ra-226 was measured in sediment sample selected from final stage of water treatment, but it gives the second highest level of ambient exposure dose due to that the water in the container works as a shield and attenuates the gamma radiation which cause to reduce the exposure rates. Figure-5 shows the distribution of the measured exposure doses overlaying the base map of O&GPI.

ID	Material	Ra-226	Th-232	K-40	Exposure dose
SL1	Sludge	1.8	6.80	12.1	0.07
SL2	Sludge	38.1	14.01	242.1	0.07
SL3	Sludge	24.1	14.40	180.4	0.09
SL4	Sludge	6.1	8.10	118.1	0.09
SL5	Sludge	14.1	12.80	176.1	0.1
SC6	Scale	61.8	12.80	82.6	0.22
SE7	Sediment	252.4	79.40	120.0	0.15
SC8	Scale	128.4	26.80	38.1	0.21
S9	Soil	46.8	43.60	529.6	0.1
O10	Crude oil	2.5	5.50	36.0	0.05
011	Crude oil	5.6	3.30	2.8	0.04
012	Crude oil	2.3	2.90	5.0	0.05
013	Crude oil	5.8	3.90	5.6	0.05
W14	Formation Water	1.2	8.80	52.8	0.04
S15	Soil	13.2	35.20	432.2	0.09
S16	Soil	15.9	25.00	385.5	0.08

Table 2- Activities in Bq/kg of Ra-226, Th-232, K-40 and measured exposure dose in μ Sv/h near the sampling point inside O&GPI



Figure 2- Sketch base map with colored classed post map overlay indicating the concentration of radium-226 in processing stages of O&GPI.



Figure 3- Sketch base map with colored classed post map overlay indicating the concentration of thorium-232 in processing stages of O&GPI.



Figure 4- Sketch base map with colored classed post map overlay indicating the concentration of postassium-40 in processing stages of O&GPI.



Figure 5- Sketch base map with colored classed post map overlay indicating the concentration of postassium-40 in processing stages of O&GPI.

Almost all values are above the worldwide average value of 58 nGy/h [14]. On the other hand, the total exposure rates are corresponding to about (0.1-0.6 mSv/y), so the max value which recorded near Dehydrator desaltors below the max value of (20 mSv/y) recommended by UNSCEAR for workers in radiation field and 1 mSv/y for the population [16].

Hazard indices

The overall results of measuring hazard indices for NORM in all samples inside the O&GPI are shown in Table -3. Herein after is the description of each index separately.

Gamma Absorbed Dose Rate

The results of calculating gamma dose rates (nGy/h) by using equation (1) show that the min. value inside the O&GPI is 3.0 (nGy/h) near sample O12 location (Post treatment oil) while max. value 169.6 (nGy/h) near sample SE7 location (Final stage treatment water) with average of $(35.7\pm42.6 \text{ nGy/h})$. It is a logical result as SE7 contains the highest concentration of radium and thorium among other materials investigated in current study. Although the average value is below the world average value of 59 nGy/h [14], some locations (SE7, SC8 and S9) (Figure.-6) have values above the world average. The main contributor in gamma absorption dose inside the O&GPI is Ra-226 as appears in Figure-6b. **Annual Effective Dose Equivalent (AEDE)**

Annual Effective Dose Equivalent (AEDE) values, which calculated by using equation (2), show that the area inside O&GPI have max value of 0.3 mSv/y in sample SE7 (final stage treatment water) and min value of 0.01 mSv/y Bq/kg in sample O12 (Post treatment oil) with average of 0.06. These values appear acceptable values because it is lower than the world average value (0.45mSv/y)[16]. It is within the range of AEDE in the western part of Iraq (0.03-1.84) mSv/y [17]. So the study area is considered within areas with normal values of AEDE. On the other hand monitoring and action should be taken in the some sites inside O&GPI to reduce this dose as low as possible according to ALARA principle recommended by UNSCEAR, IAEA and ICRP.

 Table 3- The values of hazard indices result from existing of NORM in samples selected from study area

ID	dose µSv/h (measured)	dose mSv/y (measured)	Gamma dose (nGray/h)	AEDE (mSv/y)	Ra-eq (Bq/Kg)	Hex	Hin
SL1	0.07	0.61	5.4	0.01	12.5	0.03	0.04
SL2	0.07	0.61	36.2	0.06	76.8	0.21	0.31
SL3	0.09	0.79	27.4	0.05	58.6	0.16	0.22
SL4	0.09	0.79	12.6	0.02	26.8	0.07	0.09
SL5	0.1	0.88	21.6	0.04	46.0	0.12	0.16
SC6	0.22	1.93	39.7	0.07	86.5	0.23	0.40
SE7	0.15	1.31	169.6	0.30	375.2	1.01	1.70
SC8	0.21	1.84	77.1	0.14	169.7	0.46	0.81
S9	0.1	0.88	70.0	0.12	149.9	0.40	0.53
O10	0.05	0.44	6.0	0.01	13.1	0.04	0.04
011	0.04	0.35	4.7	0.01	10.5	0.03	0.04
012	0.05	0.44	3.0	0.01	6.8	0.02	0.02
013	0.05	0.44	5.3	0.01	11.8	0.03	0.05
W14	0.04	0.35	8.1	0.01	17.8	0.05	0.05
S15	0.09	0.79	45.4	0.08	96.8	0.26	0.30
S16	0.08	0.70	38.5	0.07	81.3	0.22	0.26



Figure 6- Absorbed doses rates (a) due to exposure to gamma ray emitted from NORM materials in sampling locations. (b) Contribution of Ra-226, Th-232 and K-40 in total absorption dose rates due to exposure to NORM in sampling locations.



Figure 7- AEDE for all samples inside study area.

Radium equivalent (Req)

Radium equivalent in all samples were calculated by using equation (3). The results of the samples selected from inside O&GPI show that the max value is 375 Bq/kg represents sample SE7 (Final stage waste water) While the min value is 6.8 Bq/kg represents sample O12 (Post treatment oil) with average value of 75.5 ± 94 Bq/kg. All values of Ra_{eq}, except that of sample SE7 are acceptable and no hazard because these values are lower than the standard guideline (370 Bq/kg) recommended by UNSCEAR [16], (Figure- 8).

External hazard indices

External hazard index values in all samples which calculated by using equation (4) show that the max value 1.0 Bq/kg in SE7 (final stage treatment water) and min value is BDL. These values appear acceptable values excepting H_{in} value of sample (SE7) because it is less than the unity [4-16]. (Figure-9)

Internal Hazard Index (H_{in})

Internal hazard index values in all samples which calculated by using equation (5) show the max value 1.7Bq/kg in SE7 (final stage treatment water) so it is not acceptable for radiological safety because it is higher than unity the standard guideline (unity) [10, 16]. While the other values in the rest locations are acceptable and with no hazard because it is less than unity (Figure-9).



Figure 8- Ra-eq for all samples selected from the study area



Figure 9- Hex and Hin caused by NORM for samples in study area.

Conclusion

According to the activity of NORM in soil sample in current study, the ambient soil in the studied location consider within soil that has normal background radiation and no anomalies existing.

In scale samples the activities of NORMs are higher than those in sludge and consider within low values of the ranges in worldwide. Although, the NORM activities in sludge are below those activities in other countries but it is necessary to be monitored periodically to ensure that there is no radiological threat to workers and environments.

The highest activity was found in oily-sediment collected from containers for the last stage of oilwater separation. It is higher than the average worldwide so it could be a source of internal and external radiation contamination in the study area.

Other samples including formation water, crude oil or produced oil, all have low concentrations of NORMs and no hazard.

Most locations in the study area have values of ambient gamma rates above the average worldwide value of (58 nGy/h).NORMs are caused absorbed dose in max contribution (79%) of that the total ambient gamma dose. Radium caused the max contribution among the others.

The hazard indices all indicate that most of the sampling locations have indices acceptable and no hazard exist. On the other hand the locations SC6 and SE7 have indices close or above acceptable values so it must to take in consideration in action of radiation protection.

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