Radiological Risk Assessment of Oil waste in the Oilfields at Zubair district of Basrah

Mohammed Khudheir
Ph.D. Philosophy Soil Science. Geophysical, Ministry of Science and Technology, Iraq
Email: khudheirmohamad@yahoo.com

ABSTRACT The specific activities of natural radionuclides in several samples were collected from the areas that exposed to triple-aggression which is Zubair province at Basrah governorate exactly. Those samples were studied and those concentrations calculated by using Gamma spectrometry system based on HpG detector with efficiency 40% was used for analysis of NORM. Then radiological risk of the natural radioactivity was estimated for the study area that caused from exist the radionuclides in surface soil and water samples. The results have been revealed the contamination that occurring in some sites of Zubair district belonging to the natural radioactive material. Activity concentrations in Bq/kg of $^{238}$U, $^{232}$Th, and $^{40}$K with mean values of 95.8 Bq/kg, 31.6 Bq/kg, and 482.18 Bq/kg respectively. The outcome of soil samples were appeared higher values when compared with international calibrations. Whereas the results of Gamma absorbed dose (nGy/h), the exposure dose rate (µR/h) have mean values non-detected like; 52.5, 12.8 respectively, where under the international average by UNSCEAR 2008. Since the Radium equivalent activity (Ra$_{eq}$) has value 295.8 Bq/kg is higher than the standardizations.

Keywords: Natural Radioactivity; Radiological Risk; Gamma ray Spectrometry; Zubair Province.

1. INTRODUCTION

Naturally occurring radioactive material (NORM) is exhibited through the Ground's crust and can be enhanced by activities related with production of oil and gas. Then can be focused in oil production wastes like sludge, mud, pipe scale, created water [2]. Gamma radiations emitted from naturally happening radionuclides also termed earthly background radiation indicates the primary external source of radiation of the human body. Natural environmental radioactivity and the related external exposure due to gamma irradiation based mostly on the geographical and geological conditions [3, 4].

Natural radioactivity arises mostly from the original radioisotopes like $^{40}$K and the radionuclides from $^{238}$U and $^{232}$Th series and their decay products. NORM is commonly nuclides with long half-life and occurs since the beginning of the Earth formation [2].

NaI (TI) technical were utilized for diverse materials like soil, food, cleaning materials by several researchers [10]. The primary target of this project is to investigate natural radioactivity in soil samples within Zubair district especially the oilfields stations and evaluate the radiological reasons.
and features such as absorbed dose average, Radium equivalent activity, annual effective dose equivalent and hazard for all samples research [7].

[8] Gamma radiation from the waste has a very wide range. $^{226}$Ra which is a major isotope of concern has activity concentration from undetectable to 1000 kBq/kg. The natural radioactivity greater than the background radiation, has been noticed in the oil and gas recovery waste in 1930s [13]. The regulatory agencies started to pay attention to the NORM related with hydrocarbons production at 1980s.

Radioactive decay of $^{238}$U and $^{232}$Th products some series of daughter radioisotopes of diverse elements and of different physical features with respect to their half-lives, kinds and energies of emitted radiation, and modes of decay [1, 2]. Radioactivity materials such as Uranium and Thorium occur in trace concentrations in rock formations. Natural radioactivity can be transported from the reservoir to the surface with the oil and gas products being recovered. The NORM indicating by $^{40}$K, and $^{238}$U, $^{232}$Th, can be found in diverse quantity in the rock formation in the basin depending on the category of rock. Also the NORM may be transfer to the surface through production the oil and gas [4].

These NORMs may be accumulating as scale, brine, and sludge due to the variation of physical and chemical circumstance of oil and water formation on the surface [12]. Most of NORM deposits such as scale and sludge in pipes and equipment or as residues in brine water tanks. The product of oil and gas will increase the accumulation of NORM within the stages of produces which lead to additional radiological risk to the people in the oilfield [11].

There are more industries which produce oil and gas within Zubair district at Basrah to determine this district if have NORM contamination or not, NORM survey, sampling and analysis must to be achieve to know which site have to be contaminated with NORM or industrial radioactive like depleted Uranium.

2. MATERIALS AND METHODS

Sample Collection and Preparations

Soil samples were taken a rounded the sites of high radiation from Zubair district of Basrah governorate, depending on radioactivity survey by using mobile devices, Fig. (1). Soil Samples were dried in electrical oven at $100^\circ$C and then crushed with electric mill to be homogenised with more powdered, and then allowed to pass through a soft mesh [1], [12]. For gamma measurements the soil samples were weighted, packed in Marnelle beakers that is of the sensor geometry, and sealed to prevent radon gas leakage, and then put it in ray spectra of Gamma device for period $3600$sec to define increasing in radio actives of every sample based on dry weight with units (Becquerel/kg), as in Fig. (1).

![Figure (1) Photo for Germanium system mobile (field) and constant (laboratory) using Nitrogen to measure radioisotopes](image-url)
3. **Experimental Setup**

Radioactivity measurements were conducted by using gamma radiation spectroscopy system based on High purity Germanium detector the resolution of it 1.8 kev and efficiency 40% for $^{60}$Co at 1333.7 kev. [12, 13] Analyses of result do by gamma vision program supplied by (Alpha Spectra, Inc. 12112/3), connected with a multi-channel analyser (MCA) (ORTEC – Digi Base) with range of 4096 channel associated with ADC (Analog to Digital Convertor) unit by interface and desperation energy (FWHM) in the peak 1.33 kev for $^{60}$Co is 7% mm. the gamma rays levels were measured by integral counting [16]. Spectroscopic measurements and are analysed by a computer program titled (MAESTRO-32) software into the PC in the laboratory as it is connected to parts of system analysis. Air samples were also collected to measures the concentration of alpha, beta emitters and radon gas by using CCAM aerosol monitor [17].

The field work includes collecting samples from surface soil, air, and water, were collected from the different contaminated sites within the fields of oil and gas production. Soil samples were dried for a sufficient period of time at a fixed temperature (105°C) to acquire a constant dry weight using electrical oven [14]. Plants and large stones sand were cleaned if any while soil and water sample used as it is. All samples are then thoroughly homogenized, then (0.5 – 1) kg of each soil sample, (0.5-1 L of water) was taken and stored for 25 days in completely sealed Marnelle beaker to achieve equilibrium between radon and its daughters [4].

[12] Gamma spectrometry system based on HpG with efficiency 40% was used for analysis of NORM in the samples. Concentration in Bq/kg $^{226}$Ra, $^{232}$Th, and $^{40}$K were measured in all samples. The specific activities were averaged of gamma radiation plots at several energies. The gamma-ray lines of 338.4 kev, and 911.2 kev from $^{228}$Ac, the 727.3 kev from $^{212}$Bi, and 583.2 kev and 2614.5 kev from $^{208}$TI were used to determine the specific activity of $^{232}$Th. The gamma-ray peaks at 186.0 of $^{226}$Ra and/ or at 295.2 kev and 351.9 kev from $^{214}$Pb and at 609.3 kev from $^{208}$TI were used to determine the specific activity of $^{226}$Ra. The specific activity of $^{40}$K was measured directly by its own gamma-ray line at 1460.8 kev. The energy calibration and efficiency was calibrated using a standard source of a multi energy made by the American Canberra Company [15]. The Marnelle geometrical form was utilized to estimate the radioactivity of samples and in the source of standardization, as in Table (1).

| No. | Isotopes | Energy (Kev) |
|-----|----------|--------------|
| 1   | Ra-226   | 185.0        |
| 2   | Pb-212   | 238.2        |
| 3   | Pb-214   | 351.9        |
| 4   | Bi-214   | 609.3        |
| 5   | Cs-137   | 661.6        |
| 6   | Ac-228   | 911.2        |
| 7   | K-40     | 1460.8       |

4. **RESULTS AND DISCUSSION**

After the radioactive survey procedures were performed for all the sites that located nearby the hot points that believed had a large contamination depended on prior studies. The results of measurements for 22 soil samples were collected at varied polluted sites from Zubair district within Basrah governorate, Iraq [1]. Table (2) showed specific activities of the radionuclides were dependant in Bq/kg ranged between 8.6 – 315.4 for $^{238}$U; 14.5 – 44.7 for $^{232}$Th; 34.3 – 1411.8 for
Table (2) Specific activities of radionuclides in soil samples at different locations in the municipal area of the Zubair district

| No | Location          | Specific Activities of the Isotopes in Bq/kg |
|----|-------------------|---------------------------------------------|
|    |                   | U-238 | Pb-212 | Pb-214 | Th-232 | Bi-234 | Ac-228 | K-40  |
| 1  | Um Qaser          | 8.6   | 47.9   | 97.1   | 14.6   | 24.5   | 11.7   | 785.4 |
| 2  | Safwan            | 15.3  | 68.1   | 91.5   | 17.3   | 31.7   | 17.5   | 651.9 |
| 3  | Burjia            | 10.5  | 34.3   | 67.6   | 18.7   | 27.5   | 13.1   | 394.5 |
| 4  | Khrang station    | 189.1 | 764.1  | 978.2  | 22.3   | 42.7   | 1176.3 | 241.3 |
| 5  | Zubair city       | 12.8  | 74.6   | 117.4  | 37.1   | 25.3   | 19.5   | 893.4 |
| 6  | Ahmadi            | 161.3 | 511.7  | 775.5  | 34.9   | 89.2   | 1203.1 | 321.5 |
| 7  | Raffahia          | 17.6  | 106.8  | 306.3  | 25.7   | 15.1   | 15.6   | 595.7 |
| 8  | Sanam mountain    | 76.4  | 233.6  | 411.5  | 29.8   | 38.4   | 18.7   | 368.3 |
| 9  | Al Luhaise        | 15.9  | 126.4  | 286.9  | 27.2   | 14.2   | 9.5    | 581.6 |
| 10 | Al Shamia         | 11.8  | 123.5  | 183.4  | 21.9   | 19.4   | 8.8    | 617.3 |
| 11 | Al Meshraj        | 18.1  | 117.4  | 203.4  | 38.5   | 27.4   | 27.6   | 493.5 |
| 12 | Jwebida           | 48.4  | 414.2  | 178.6  | 32.5   | 165.8  | 132.5  | 352.1 |
| 13 | NR-1 Oilfields    | 261.6 | 843.2  | 1457.8 | 38.7   | 134.8  | 1087.3 | 196.8 |
| 14 | SR-1 Oilfields    | 295.3 | 1266.8 | 1789.2 | 41.3   | 137.5  | 1371.5 | 324.7 |
| 15 | NR-2 Oilfields    | 281.6 | 718.6  | 1248.7 | 34.8   | 196.2  | 1202.4 | 201.2 |
| 16 | SR-2 Oilfields    | 315.4 | 1411.8 | 1978.4 | 44.7   | 281.4  | 1476.3 | 236.5 |
| 17 | Greshan           | 26.9  | 202.5  | 645.3  | 36.4   | 45.2   | 893.5  | 675.2 |
| 18 | Shuiaba           | 46.3  | 167.2  | 117.4  | 19.8   | 34.8   | 245.8  | 752.5 |
| 19 | Collect area      | 174.8 | 391.2  | 817.6  | 40.7   | 79.8   | 768.3  | 241.2 |
| 20 | Hartha            | 68.4  | 106.5  | 622.5  | 43.2   | 35.1   | 321.4  | 491.7 |
| 21 | Al Najmi          | 19.2  | 69.7   | 166.3  | 35.7   | 11.5   | 143.2  | 580.1 |
| 22 | Tanuma            | 31.82 | 149.3  | 195.3  | 39.8   | 15.6   | 737.2  | 611.6 |
|    | Average           | 95.78 | 361.4  | 578.91 | 31.61  | 67.87  | 495.5  | 482.18 |
|    | Max. V.           | 315.4 | 1411.8 | 1978.4 | 44.7   | 281.4  | 1476.3 | 893.4 |
|    | Min. V.           | 8.6   | 34.3   | 67.6   | 14.5   | 11.5   | 8.8    | 196.8 |
|    | Int. Standards    | 32    | 45     |        |        |        |        | 412.3 |

The results can be compared with the world average activity concentrations that are 412, 32, and 45 Bq/kg for \(^{40}\text{K}, \(^{238}\text{U}, \text{and} \(^{232}\text{Th}\) respectively, as reported by [20, 22]. Most of outcomes in specific activity of \(^{238}\text{U} \text{and} \(^{40}\text{K}\) are higher than worldwide average, since that those areas are attributed to oilfield lands.
Table (3) Activity of Equivalent Radium, Gamma Absorbed Dose in samples selected from study area and total Exposure Dose Rates

| No. | Location          | Ra- Equivalent Bq/kg | Gamma Abs. Dose (nGy/h) | Exposure Dose (µR/h) |
|-----|-------------------|----------------------|-------------------------|----------------------|
| 1   | Um Qasr           | 12.7                 | 8.37                    | 6.13                 |
| 2   | Safwan            | 31.4                 | 13.5                    | 9.15                 |
| 3   | Burjsia           | 25.8                 | 11.6                    | 7.27                 |
| 4   | Khrang station    | 495.4                | 96.8                    | 16.26                |
| 5   | Zubair city       | 38.1                 | 11.8                    | 9.65                 |
| 6   | Ahmadi            | 294.3                | 89.3                    | 16.12                |
| 7   | Rafdhiah          | 42.7                 | 21.7                    | 8.86                 |
| 8   | Snam mountain     | 75.9                 | 38.5                    | 15.03                |
| 9   | Al Iuhaise        | 28.3                 | 12.9                    | 9.05                 |
| 10  | Al Shamia         | 52.9                 | 14.7                    | 8.94                 |
| 11  | Al Meshraj        | 30.1                 | 12.1                    | 8.65                 |
| 12  | Jwebida           | 78.7                 | 26.2                    | 12.06                |
| 13  | NR-1 Oilfields    | 834                  | 85.9                    | 18.45                |
| 14  | SR-1 Oilfields    | 1148                 | 178.1                   | 22.13                |
| 15  | NR-2 Oilfields    | 967                  | 98.6                    | 19.66                |
| 16  | SR-2 Oilfields    | 1185                 | 192.3                   | 23.93                |
| 17  | Greshan           | 65.6                 | 19.8                    | 8.09                 |
| 18  | Shuaiba           | 46.4                 | 13.2                    | 9.17                 |
| 19  | Collect area      | 533.7                | 107.6                   | 17.86                |
| 20  | Lowdan            | 76.6                 | 10.8                    | 9.17                 |
| 21  | Al Najmi          | 32.09                | 13.4                    | 8.07                 |
| 22  | Tanuma            | 413.6                | 78.6                    | 17.23                |
|     | Average           | 295.83               | 52.54                   | 12.77                |
|     | Max. Value        | 1185                 | 192.3                   | 23.93                |
|     | Min. Value        | 12.7                 | 8.37                    | 6.13                 |
|     | Int. Standard     | 50                   | 55                      | 15                   |

The activity concentration of Radium equivalent and gamma absorbed dose and total exposure dose rate in all samples of Zubair district were displayed in Table (3). It's found that value of gamma
absorbed dose varied from (8.4 – 192.3) where the average (52.5) and the total exposure dose rate varied from 6.1 – 23.9 has an average (12.77) [9]. Since that the values correlated with those variables under the levels of international calibrations. But the value of Radium equivalent varied from 12.7 – 1185 Bq/kg has an average (295.8) are higher than of worldwide average in most of the locations, which proves on existing high quantities of the NORM, as in Fig. (2). Whereas several sites has high values of Gamma absorbed and exposure air doses rate such as Khrang station, Ahmadi, southern and northern Rumaila oilfields, collect area, and Tanuma, with the values varied from (8.4 – 192.3) nGry/h with average 52.5 for absorbed dose, while the values varied from (6.3 – 23.9) µR/h has the mean 12.8 as in Fig. (3) Agreed with [15]. It's clear to prior results where lead to expose the people and environment to the huge threats of contamination, which effects in continuous increase of the ecological hazards were resulted from oil and gas production, as agreed with [19, 21].

Figure (2) Radium Equivalent (Bq/kg) within different sites at Zubair district

Figure (3) Gamma Absorbed and Exposure Doses rates within different sites at Zubair district

The histogram of $^{238}$U concentrations in different sites within Zubair district shows that these are independent on the previous wars that occurred among (1991 – 2003) [23]. But on reverse the sites
that containing on the oilfields were produced the natural occurred radioactive material (NORM). That means in some sites there are high amount of natural Uranium has not been produced from the missiles or ammunitions such as in the Khrang station, Ahmadi, southern and northern Rumaila oilfields due to those fields produced oil and gas continuously, as in Fig. (4).

![Figure (4) Uranium-238 Isotope in Bq/kg with different sites at Zubair district](image)

Figure (4) shows that more than 70% of the sampling locations have Radium contents higher than the worldwide average (50 Bq/kg) of $^{226}$Ra in Iraqi soil that may indicate to the diffusion of radium resulting from oil and gas production operations over most area of Zubair district [17]. The highest concentrations of $^{226}$Ra-eq were noticed in southern Rumaila oilfields with an average of 1185 Bq/kg while the lowest value is detected in Um Qasr of 12.7 Bq/kg. 232-Thorium concentrations are mostly with estimated values in all samples as appear in Fig. (6). It's concentrations in most of samples are below the worldwide average [20]. Concentrations of $^{40}$K are found to be higher than others in Zubair city while lower the values are found in Northern Rumaila oilfields as shown in Fig (7). The differences in the distribution of radium, thorium, and potassium in all samples
attribute to the differences in their geochemistry and mobility under different conditions. The variations in the concentration values of different isotopes in different sites are because of lithological differences and metamorphic rocks resulting for these lithounits have higher average of Uranium contents [19].

![Figure (6) Specific activity of 232-Thorium and 238-Uranium in Bq/kg in various areas of Zubair district](image)

![Figure (7) Specific activity of 40-Potassium in Bq/kg within various areas of Zubair district](image)

The exposure dose rates were measured in those sites to evaluate the safety of population and environment due to 60% of these locations have Ra-eq exceeds the worldwide average, where most of the exposure rates are caused by radium content mong other NORMs, as agreed with [15, 19]. No one of these locations has values of Th-232 and K-40 higher than the international standards in the soil. Thus, most sites displays exposure dose rates exceeds the international calibrations (0.0406 μSv/h) matching to 58 (nGy/h) [15, 19]. Means that, radiation risks that resulting from the NORM are significant for the population living in the oilfields area such as in khrang station, Ahmadi and any site that contains on the oilfields for oil and gas production, as in Fig (8).
5. CONCLUSIONS

Soil sampled had been picked-up, and then analysed to evaluate the source of radioactivity waste if its natural (NORM) or industrial as residuals from the former wars like depleted uranium or other industry isotopes. The specific activity of $^{238}$U, $^{214}$Pb, $^{214}$Bi, and $^{40}$K isotopes were measured by using gamma ray spectrometry in soil samples collected at diverse sites within Zubair district in Basrah governorate are higher than the recommended value. The concentrations of radioisotopes are independent of depth as exhibited from the results. It depends on the lithological dissimilarities of the oilfields.

The average of Gamma dose rate obtained in this study (52.5 nGy/h) is less than worldwide average (55 nGy/h) when compared with those values. Exposure dose rate is lower than background value; exception the sites have oilfield stations, which effects on the population in living within studied area due to the radiation hazard is actual significant. Polluted sites should be monitored further frequently to that no more radiation doses are present due to expose to NORM. Radium equivalent of the samples were extended from $(12.7 - 1185)$ Bq/kg for Um-Qasr and Southern Rumaila Oilfields respectively. Most of the specific activities of radium content are higher than recommended value (50 Bq/kg).

Radioactivity measurements and evaluate of NORM in oilfield is one of most significant facts due to producing oil and Gas are one of the main sources of contamination. The studied sites in Zubair district are moderately not extremely contaminated with NORM, if they compared with other oilfield in worldwide. Although, some sites within studied area; like Ahmadi, Khrang station, Collect area, Northern and Southern Rumaila oilfields have more contaminated with NORM. For radioactivity protection these locations should be checked continuously and the basic control procedures should be taken in consideration by individuals when dealing with these materials. Radioactivity assessment in Rumaila oilfields and other sites at Zubair districts is of highly recommended to ensure no more radiation risk is present in those locations.

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