Natural Radioactivity in Soil and Bitumen in Al-Marj Spring and Abu-Jir Village, Anbar, Western Iraq

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Abstract

The aim of the present research is to illustrate γ-ray levels emitting from soil and bitumen producing from 238U, 232Th, and 40K along Abu-Jir Fault Zone. in the area extended from Al-Marj valley to Abu-Jir village using scintillometer device. Such study is important in environmental assessment to buildup data base about radioactivity. The concentration of natural radionuclides in the study area was determined to be occurring mostly in the clay minerals and organic matter. A high purity germanium spectrometer was used to detect the activity of these elements which ranged between 100.6±18.1-1526±102, 0-8.4±1.4, and 70.1±10.9-328.2±73 in soil, and 28.2±5.6-94±22.1, 0-2.2±0.5, and 38.4±7.9-70.1±10.9 in bitumen for 226Ra, 232Th, and 40K respectively. The anomaly of the Naturally occurring radioactive material was associated with hydrocarbon accumulation and springs. The source of 232Th and 40K came from the same source rock (illite), while 226Ra was associated with ascending fluids. The obtained results showed that the activity of 226Ra in soil has value above the world averages and other countries.

Keywords: Natural radionuclides; Bitumen; Al-Marj spring; Abu-Jir; Gamma ray

1. Introduction

The study area is characterized by the presence of many hydrocarbon seepages distributed along Abu-Jir Fault Zone (AJFZ), particularly, in the area extending between Hit and Abu-Jir village (Awadh et al., 2013). The outcrops of the interested area are described by rocks of Miocene age represented by carbonate facies of Euphrates Formation (Early Miocene) and evaporate facies of the Fatha Formation (Middle Miocene). Bitumen seepages are interfered and intruded with gypsum of the Fatha Formation in the area extended along Hit-Ramadi Road. At Abu-Jir village, huge deposits of bitumen ascending upward and covering the most area which may contain heavy metals and radionuclides that may be transferred to plants, thereafter may accumulate in plants and animals (Al-Rawi et al., 2014). A lot of heavy metals were found in both water and sediments of the Euphrates River in the study area (Awadh and Ahmed, 2013). Naturally occurring radioactive material (NORM) occurs everywhere and is present in the earth’s crust, buildings, food, drink, and human body itself. The origin of NORM arises mainly from cosmic radiation and terrestrial radioactive material. The major source of natural radioactivity is terrestrial radiation which majority of it belong to radionuclides in 238U series, 232Th series, and 40K, as a result of radioactive decay these elements in rocks and soil, where they can migrate in environment through soil, rocks, air, and water. These elements are present in soil in trace amounts and yields gamma

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radiation in the environment resulting in changes in the background radiation level (Amanjeet et al., 2017). Also, they emit alpha or beta particles which may be taken into body via inhalation or ingestion. There is a direct relationship between terrestrial gamma radiation and the concentration of radionuclides in soil (Ademola et al., 2014). These radioactive elements are favorably bonded to clay minerals and organic matter (OM) by adsorption, but in varies ability ranging from strong bonded in montmorillonite and illite to weak in kaolinite and chlorite, and less bonded to OM (Guo et al., 2021).

The problem of this study is the knowledge of the concentration of radioactive elements in the soil and bitumen and what their relationship with mineralogical content. Very limited previous studies conducted on radioactivity near the study area but limited in formation or with different medium. Some researchers studied only uranium concentration in the Euphrates Formation such Al-Bassam et al. (2006) and Al-Obaidy (2015), which attributed the enrichment of U in the Euphrates Formation to ascending groundwater as a result of tectonic movement. Ali (2015) measured the activity of radioactive elements (226Ra, 232Th, 40K, and 137Cs) in sediments of the Euphrates River and springs and conclude they had high concentration in spring sediment especially 226Ra, while Farhan et al. (2020) measured radon gas (222Rn) concentration in the shallow groundwater of Abu-Jir village which stated the source of radon is U-rich hydrocarbons. Awadh et al. (2018) has recorded only gamma ray using the scintillometer (SSP-2-NF) device on both blocks (western and eastern) of the Abu-Jir Fault. Radioactive elements may be transferred from contaminated soil to the surface and groundwater by leaching and weathering processes, that cause deteriorates these systems if that presents at high concentrations and would become hazardous. This study aims to assess the concentration of radioactive elements (238U, 232Th, and 40K) in the study area to build up a database of the nature of radiation and to determine the geological materials emitted radiation.

2. Study area

The study area is located within the Stable Shelf in Iraq at the western side of Euphrates River. It is extended along the AJFZ from Al-Marj spring to the north, to Abu-Jir Village to the south (Fig. 1). It is restricted between latitudes (33°15ʹ33.30ʺ- 33°41ʹ46.40ʺ N) and longitudes (42°43ʹ57.33ʺ- 42°54ʹ11.0ʺ E) (Table 1). The stratigraphic succession of the area is represented by the Euphrates Formation (Early Miocene) composing of carbonates with little of clay minerals (Al-Dabbas et al., 2014 and Awadh, 2014), Fatha Formation (Middle Miocene) which is characterized by predominant evaporate facies comprised of anhydrite, gypsum, and salt, interbedded with limestone, marl, and relatively fine-grained clastics (Al-Obaidy, 2015), and Quaternary sediments which comprised from the Pleistocene and Holocene. The Pleistocene sediments are heterogeneous of fine pebbles consisting of quartz, chert, carbonate rock fragments, and clay (Awadh and Ahmed, 2013). The depositional environment of the Euphrates Formation was open to restricted platforms which indicated a lagoonal environment with warm and restricted open circulation (Al-Dabbas et al., 2013, and Al-Dabbas et al., 2014), while the depositional environment of the Fatha Formation is shallow marine water with normal to high saline water with clear oscillation in the sea level (Awadh and Al-Ankaz, 2016).

3. Materials and Methods

The field work was performed during 17-18 November 2020 focusing on radiometric survey to measure gamma ray using the scintillometer device (SSP-2-NF). A scale f-150 and 1500 of the scintillometers was chosen to record the whole concentrations of radionuclides $^{238}$U, $^{232}$Th, and $^{40}$K. A total of 88 measurements was measured and recorded in situ (Fig. 1).
Fig. 1. Geological map of the study area displays radiation reading sites along Abu-Jir Fault Zone.
The Surfer 13 program was used to design isorad map which display the variation in the radioactivity. Sixteen samples were collected from the Quaternary sediments, Fatha and Euphrates formations. Soil samples were collected at a depth of 10 cm, while samples from formations were collected randomly from the accessible areas. Samples (4M-S, 5M-S, 35F-B, 35F-BG, 39F-G, 68J-BS, 70J-BS, 82J-BS, 87J-B) were subjected to mineralogical identification using X-ray diffraction (XRD), geochemical analysis by inductively coupled plasma mass spectrometry (ICP-MS), and radioactivity detection via high purity germanium (HPGe) detector. Samples were prepared in the Geochemistry Laboratory at the Department of Geology, College of Science, University of Baghdad. Each sample was dried at room temperature and was crushed into fine powder using porcelain mortar and Fritsch pulverisette (Buhrke et al., 1998). XRD was carried out in Directorate of Materials Research, Ministry
of Science and Technology. Mineralogical composition was detected by semi-quantitative estimations, while the mineral abundance was evaluated according to normative calculation. Geochemical analysis was performed in the Mineral Division -ALS Chemex, Spain (Sevilla). The radioactive analysis was conducted in the Radiation Protection Center, Ministry of Health and Environment on 1kg weight of sample which was preserved in sealed bags for four weeks to achieve secular equilibrium between $^{226}$Ra, $^{232}$Th, and their progenies (Veiga et al., 2006).

4. Results

The results are presented in Table 2. The field measuring is represented by the field recording of the scintillometer (SSP-2-NF) for gamma ray. The data modified after calibration of the device with 137Cs to obtain the calibration constant, which equal to 3.24, thereafter, the field results multiplied by calibration constant and 3 or 30 for 150/F and 1500 scale of scintillometer respectively. The field measurements presented in Table 2 were plotted in an isorad map by Surfer 13 program which displays the distribution of radioactivity in the study area (Fig. 2).

<table>
<thead>
<tr>
<th>S. No.</th>
<th>$\gamma$-ray (c/s)</th>
<th>S. No.</th>
<th>$\gamma$-ray (c/s)</th>
<th>S. No.</th>
<th>$\gamma$-ray (c/s)</th>
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<td>82.52</td>
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<td>136.08</td>
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</table>

* scale of 1500 (c/s)
The semi quantitative estimation for XRD analysis of soil samples showed a prevalence calcite mineral, followed by dolomite, then quartz and clay minerals. The normative calculation of mineral composition is listed in Table 3 based on chemical analysis of major and minor oxides (CaO, MgO, SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, Na$_2$O, K$_2$O, TiO$_2$, MnO, P$_2$O$_5$, SO$_3$, and LOI) as well as the concentration of U and Th in ppm as listed in (Table 4).

Fig. 2. Isorad map displays the radioactivity in the study area
Table 3. Mineralogical composition of samples collected from Al-Marj valley and Abu-Jir village

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Formation</th>
<th>Mineralogical composition (%)</th>
<th>CM</th>
<th>Total</th>
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<td>Calcite</td>
<td>Dolomite</td>
<td>Gypsum</td>
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<td>Fatha</td>
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<td>39F-G</td>
<td>Formation</td>
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<td>82J-BS</td>
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<td>38.37</td>
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<td>6.92</td>
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</table>

Min: 22.08 2.91 5.01 12.24 5.6 1.8 4.8 53.74
Max: 57.75 28.09 10 35.17
Av: 37.36 15.97 7.12 17.46

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<th>S. No.</th>
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<th>Fe2O3</th>
<th>Na2O</th>
<th>K2O</th>
<th>TiO2</th>
<th>MnO</th>
<th>P2O5</th>
<th>SO3</th>
<th>LOI</th>
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<td>0.19</td>
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<td>0.08</td>
<td>18.6</td>
<td>40</td>
<td>-</td>
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</table>

* Loss on ignition

The radioactivity levels of 226Ra, 232Th, and 40K were measured in the samples that collected from the study area and the results in Bq/kg are presented in Table 5 and Fig. 3. The radionuclide of 238U-series yields 226Ra isotope, so it can be represented as a reference alternative of 238U.

Table 5. The specific activity of 226Ra, 232Th, and 40K in Bq/kg based on HPGe

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<td>70.1±10.9</td>
<td></td>
</tr>
<tr>
<td>Av</td>
<td>352.73±31.53</td>
<td>4.44±0.91</td>
<td>149.9±18.19</td>
</tr>
</tbody>
</table>

5. Discussion

The specific activity of 226Ra, 232Th, and 40K in the soil samples were found in the range of 100.6±18.1 to 1526±102, 0 to 8.4±1.4, and 127.8±23.1 to 328.2±73 respectively. There is a large variation in the concentration of 226Ra, especially in bituminous soil (BS) samples as shown in Fig. 3.
This might be due to variation in mineralogical content in soil which may be attributed mainly to clay minerals and organic matter constitutes. The clay minerals provide a favorable surface to attach radioactive elements by adsorption, but the organic matter is least able to attach these elements. Native bitumen has no capacity for attractive uranium as appearing in 35F-B and 87J-B samples (Table 5), but some of the organic acids that are consist of very minor contents of petroliferous matters may readily make uranium salts (Bell, 1963) which may interpret the high activity of radium in bituminous soil samples (70J-BS and 82J-BS) in Table 4. Mahdi and Al-Kadhimy (2007) attribute the radioactive anomaly in the study area to their association with spring and hydrocarbon deposits which spread along AJFZ in the area, as well as, the outcrops of the Euphrates Formation that proved to contain unordinary concentration of uranium in the upper part of the formation (Al-Atia and Mehdi, 2005). The location of hydrocarbon seepages was closely to the samples collected from Abu-Jir village that also explain the high activity of these samples and the higher value of gamma radiation in the samples from Al-Marj spring (from 18M-S to 26M-S readings), as well as, the seepages close to Al-Wafaa area (from 503-BS to 59J-BS readings) as shown in Table 2. In 5M-S sample, the radium activity may attribute to carbonate content which has fossiliferous constitutes that adsorb radioactive elements.

The concentration of uranium in soil samples in ppm does not cooperate with their specific activity of radium, that was belonged to circumstances of the host rocks. This can illustrate by the influence the oxidation state which yields to chelate of uranium thereafter establish disruption of radioactive equilibrium of U-series then tendency in favor of radium (Al-Atia and Mehdi, 2005). The specific activity of $^{40}$K is attributed to clay minerals content in samples, especially illite which has a strong ability to adsorption with radioactive elements (Guo et al., 2021), that is reflected in the activity of $^{40}$K of sample (4M-S) which associated with high clay mineral abundance comparing with other samples, this is show obviously in Table 3 and Table 5.

The correlation coefficient was done between the specific activity of $^{226}$Ra, $^{232}$Th, and $^{40}$K which appeared that they all have positive correlation with each other as shown in Fig. 4. $^{232}$Th has a good positive correlation with $^{40}$K ($r=0.89$), that be owing to the fact the radionuclides develop from the same rock formation (Munyao et al., 2020), which is illite due to potassium in its structure as the main constitute. While $^{226}$Ra has a poor positive correlation with $^{232}$Th and $^{40}$K, that means the source of $^{226}$Ra from ascending fluids as a result of its high mobility, that transport $^{226}$Ra from source rock and adsorption on clay minerals and organic matter. This is explained that the variation occurring between $^{226}$Ra with $^{232}$Th and $^{40}$K in different samples. Finally, by comparison of the average of specific activity of $^{226}$Ra, $^{232}$Th, and $^{40}$K in soil samples with their activity of other countries in Table 6, that reveals the average concentration of $^{226}$Ra in the study area is higher than other countries and the world average, hydrocarbon leakages are consider the source of uranium (radium) in the area (Farhan et al., 2020). The concentration of $^{232}$Th and $^{40}$K is lower than the world average value and other countries except for Jordan and Saudi Arabia (Asir) have a lower concentration of $^{40}$K than the present study. The variation
in the concentration of radionuclides in the soil of various regions of the world depends on the geological and geographical condition of each region of the world, as well as, using chemical fertilizers which applied to agricultural lands in some regions (El-Taher and Al-Zahrani, 2014) which provide radionuclides and heavy metals to soil, especially $^{40}$K.

![Pearson correlation plot showing the correlation between: (a) $^{226}$Ra and $^{232}$Th; (b) $^{226}$Ra and $^{40}$K; (c) $^{232}$Th and $^{40}$K.](image-url)}
Table 6. The specific activity of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in Bq/kg in soil samples in different countries and the present study

<table>
<thead>
<tr>
<th>Country</th>
<th>Specific activity (Bq/kg)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{226}\text{Ra}$</td>
<td>$^{232}\text{Th}$</td>
</tr>
<tr>
<td>Iraq (Nineveh)</td>
<td>33.55</td>
<td>21.52</td>
</tr>
<tr>
<td>Northern India</td>
<td>63.88</td>
<td>91.56</td>
</tr>
<tr>
<td>Turkey</td>
<td>32</td>
<td>22.86</td>
</tr>
<tr>
<td>Iraq (Baghdad-Karakh side)</td>
<td>16.47</td>
<td>9.72</td>
</tr>
<tr>
<td>Jordan</td>
<td>44.9</td>
<td>18.1</td>
</tr>
<tr>
<td>Yemen</td>
<td>30.41</td>
<td>36.26</td>
</tr>
<tr>
<td>Egypt</td>
<td>17</td>
<td>18</td>
</tr>
<tr>
<td>Malaysia</td>
<td>67</td>
<td>82</td>
</tr>
<tr>
<td>Iran</td>
<td>38.8</td>
<td>43.4</td>
</tr>
<tr>
<td>Egypt (Abou Zabal region)</td>
<td>31.12</td>
<td>10.96</td>
</tr>
<tr>
<td>Saudi Arabia (Asir)</td>
<td>38.7</td>
<td>23.5</td>
</tr>
<tr>
<td>Algeria</td>
<td>53.2</td>
<td>50.03</td>
</tr>
<tr>
<td>Brazil (Panama)</td>
<td>10.22</td>
<td>7.27</td>
</tr>
<tr>
<td>Saudi Arabia (Qassim)</td>
<td>9.5</td>
<td>12.3</td>
</tr>
<tr>
<td>World average</td>
<td>33</td>
<td>45</td>
</tr>
<tr>
<td>Iraq (Al-Marj spring to Abu-Jir village)</td>
<td>601</td>
<td>5.65</td>
</tr>
</tbody>
</table>

6. Conclusions

The presence the study area along Abu-Jir Fault Zone is a matter of worth, especially many phenomena are associated with it like springs and hydrocarbon seepages. This study is proved the presence of radiation along this fault which cannot separate the origin of this radiation from the phenomena related. The variation is obviously in soil samples and became more obvious in bituminous soil samples of Abu-Jir village, which due to clay minerals and organic materials constitutes in samples. This variation is appeared in $^{226}$Ra activity which increases in bituminous soil, while bitumen samples have lower activity than soil, as a result of high mobility of radium from rich-uranium hydrocarbon seeps. The correlation analysis proved the positive correlation among radioactive elements which mean that elements came from the same source. The comparison of the activity of naturally occurring radioactive elements with other countries showed the study area has higher concentration of $^{226}$Ra than over the world averages and other countries.

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