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NORM in the Surface Sediments of Iraqi International Marine Waters

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Abstract

Most radioactivity studies focus on the activity concentration of ^{232}Th and ^{238}U which are associated with heavy minerals, as well as ^{40}K which associated with clay minerals. Within this study samples of surface sediments were collected from three sites 5, 7, and 11 along Khor Abdullah within Iraqi National Waters in order to document activity concentrations of radionuclides, ^{226}Ra, ^{232}Th, ^{238}U, and ^{40}K. Core sediments were investigated as well for determination of TOC. For studied radionuclides, levels recorded were 3.22 ± 0.59 – 37.6 ± 9.9 Bq/kg for ^{226}Ra, 0.1 ± 0.1 – 6 ± 1.6 Bq/kg for ^{232}Th, 1.02 ± 0.2-- 2.7 ±1.2 Bq/kg for ^{238}U, and 124.09 ± 10 – 548.36 ± 44 Bq/kg for ^{40}K. which are comparable to word wide levels. For TOC values recorded in bottom surface sediments from the three sites were 0.919% in site No. 2 to 1.904 % in site No. 1 Which were higher than levels reported in the same sites earlier averaged as 0.53 % . Radium equivalent activity was calculated for the sediments of Khor Abdullah and recorded as 51 Bq/kg which is quite low compared to the world wide level of 350 Bq/kg.

Keywords: Radionuclides; %TOC; Sediments; North west Arabian Gulf.

1.Introduction

During the 2nd and 3rd Gulf war (1991, and 2003), Basrah as a part of Southern Iraq exposed to a radiation attacks due to the usage of DU weapon against military troops and vehicles. Radiological results were detected within Basrah Governorate as reported by Subber , and his colleaques [1]. Moreover, Soils and Sediments from Southern part of Iraq is characterized by the existence of natural occurring radioactive materials (NORM), mostly are ^{40}K, ^{212}Pb,^{214}Pb, ^{214}Bi, and ^{228}Ac as mentioned by Bashar and his colleaques [2].

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Radioactive pollutants were spread over all Basrah Land as reported by Ali and his colleague [3].The radioactive materials are transferred from land towards waterways from where they move towards oceans and seas and accumulated in the water column, then they undergoes sedimentation to the bottom of the waterways, once they reach bottom bed they became a part of the ecosystem, water, sediments, and biota as maintioned by Akram [4].

The Northern west Arabian Gulf represented by Khor Abdullah and Shatt Al-Arab estuary delta, within Basra Governorate, is subjected to man made pollution due to oil production and transportation as well as a result of military events especially during the 2^{nd} Gulf ware(1991). Pollutants detected by Al-Hassan and his colleagues were Total Aromatic Hydrocarbons (TAH), Trace Metals (TM) [5] in addition to radionuclide released from using depleted uranium during 2^{nd} (1991) and 3^{rd} (2003) Gulf Wars as detected by Ali and his colleague [6], as well as Vartanian [7] , which were detected in water, soil, and sediments samples from upstream area of Khor Abdullah which described by Energy – Solutions, [8]. This area was continuously polluted by radionuclides related to Depleted Uranium.

Khor Abdullah is an important area in the Northern west Arabian Gulf which represented by estuarian delta of Iraqi upstream Shatt Al-Arab River and Shatt Al-Basrah canal.

Sediments are important sinks for various pollutants such as chemical elements with different origin, heavy, trace, or radionuclides. Sediments with high contents of silt and clay have high surface area can adsorb more chemical elements than sediments with compounds of large grain size such as sand as investigated by Ahmed and his colleagues [9]. More over their investigation was extended to the vertical distribution of natural as well as anthropogenic radionuclides in soil samples and studied their correlation with physicochemical characteristics of soil samples, pH, grain size, carbonate contents, and organic matter. They found that total organic compounds was positively correlated to activity concentrations of ^{226}R and ^{137}Cs.

Sedimentation rate in the bottom sediment cores from Kuwaiti Bay were calculated and the range was 0.2 – 0.6 cm/year and in the range of 0.1 – 0.39 cm/year as determined by Al-Zamel and his colleague's [10] and [11] respectively.

Bottom surface and core sediments of Northern West Arabian Gulf is found by Al-Zamel and his colleague's [10] to accumulate natural radionuclides, ^{210}Pb, as well as artificial ones, ^{137}Cs, ^{235}U in a certain activity rates . On the other hand, concentrations of ^{40}K, total ^{210}Pb, ^{137}Cs, ^{90}Sr, ^{226}Ra, ^{235}U, ^{234}U, $^{239+240}$Pu, and ^{238}Pu were determined in sediment and ranged between 353 - 445, 23.6–44.3, 1.0–3.1, 4.8–5.29, 15–16.4, 1.26–1.30, and 0.028–0.03 Bq/kg dry weight, respectively. Significantly higher ^{137}Cs values have been reported from the Iranian coast compared to the western coast of the Gulf as investigated by Uddin and his colleague's [12].

2.Methods of Study

Study area: The study area is represented by the international water way along the Khor Abdullah within the Iraqi marine waters for the sites 1, 2, and 3 as shown in figure 1.

Figure 1: Map of Northern West Arabian Gulf Showing the Southern Iraqi.

3. water way (Khor Abdullah) and sampling sites

Sampling Sediment samples were collected by means of grape sampler for surface bottom sediments and core sampler for core sediment samples, Figure 2, fitted on a boat. These sampler tools have the ability to collect samples at a depth commensurate with the nature of the land in terms of porosity and composition, whether it is sandy or clay soil. Samples were kept in plastic bages and transferred to the lab in Marine Science Centre / Basra University for analysis.

A)Grape sampler B)Core Sampler

Figure 2: Sampling tools used in this study , A) for bottom surface sediments, and B) for core sediments.

In the lab samples were spread on plastic trays and left under the natural air temperature for about two days to dry, then crushed and ground by a grinder so that the size of the granules is about 0.2 mm. Then it is placed

in a marline beaker, closed tightly and left for 28 days for the purpose of reaching the equilibrium state, and then placed in a gamma Spectroscopy device for the purpose of analysis and measurement to find the effectiveness of the radioactive isotopes. The TOC for bottom surface sediments from Khor Abdullah were determined according to the method described by El-Wakeel and his colleaque [13] expressed as % organic carbon. The texture of the sediments were analyzed by distribution of particles according to their size in which sand, silt, and clay were separated by wet sieving according to Al-Mansory and his colleaque[14]; Page and his colleaque's [15], as large, medium, and small particles respectively.

4. Calculation of Radium equivalent activity

Radium equivalent activity (Ra_eq) is used to assess the hazards associated with material that contains ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg.

The Ra_eq of a sample in (Bq/kg) was achieved by using the following relation set by UNSCEAR [16]:

$$Ra\_eq = A\_Ra + 1.43\ A\_Th + 0.077\ A\_K$$

. where ARa, ATh, and AK are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg of the soil sample, respectively.

5. Results

Levels of %TOC in core sediments from Khor Abdullah are listed in table 1. The Activity concentrations for radionuclides recorded in surface layers 1-5 cm for bottom sediments are recorded and listed in table 2.

Table 1: Levels of TOC% in core sediments from sites 1, 2, and 3.

S. No.	Depth in cm	Core 1	Core 2	Core 3
1	**5**	**1.365**	**1.412**	**1.806**
2	**10**	**1.650**	**1.018**	**1.871**
3	**15**	**1.652**	**0.919**	**1.379**
4	**20**	**1.904**	**1.366**	**1.642**
5	**25**	**1.206**	**…..**	**……**
6	**30**	**1.840**	**…..**	**……**
7	**35**	**1.365**	**…..**	**……**

Table 2: Radionuclides activity and radium equivalent activity (Ra_eq) in (Bq/kg) in upper layer,1-5 cm, for bottom sediments from sites 1, 2, and 3 along Khor Abdullah Channal.

Site No.	TOC %	^{226}Ra	^{232}Th	^{238}U	^{40}K	Ra_{eq}
1	1.365	37.6±9.9	0.10±0.1	1.5±0.1	548.36±44	79.97±54.2
2	1.412	27.5±11.2	6.0±1.6	2.7±1.2	293.4±24	58.67±37.1
3	1.806	3.22±0.59	0.89±0.24	1.02±0.2	124.09±10	14.05±5.2

For %TOC measurements in the sediments at sites 1, 2, and 3 within khor Abdullah , the results revealed

clear inverse relationship between activity concentrations and %TOC for ^{226}Ra and 40K, while for ^{232}Th and ^{238}U showed extreme levels at siteNo.2 with moderate %TOC of 1.412, then decreased in site No. 3 with high %TOC of 1.806 (Ahmed, and his colleagues 2019)[9]. On the other hand ^{226}Ra and ^{40}K activity concentration are decrease with increasing % sand from site 1 to site 3 while ^{238}U activity concentration is decrease with increasing % sand from site2 to site 3 which reported by Baba-Ahmed [17].

Radionuclides enter the aquatic and marine ecosystems through different sources including river and rain water transport. The main source of ^{137}Cs in the environment are atmospheric nuclear weapon as well as releases from nuclear reactor accidents (IAEA, 2003)[18].

There were no data available on environmental radioactivity in Southern Iraqi sediments. Therefore this study is practically important and interesting to radiometric study identical to the study by Tzortzis and his colleque[19].

The determination of the radioactive materials distributed in the study area enable us to estimate any probable hazard to humans reported by Issa, and his colleagues [20]. Radioactivity released in the study area which is regarded as sensitive area because it is an important source for fisheries reported by Amin and his colleagues [21].

This area might be effected by military weapon containing Depleted Uranium (DU) used during the 1991 Gulf war reported by Al-Zamel and his colloquies[10]

Higher contaminant burdens of metals are associated with sediments containing higher total organic carbon and finer grain size distribution.

For studied radionuclides, levels recorded were 3.22 ± 0.59 – 37.6 ± 9.9 Bq/kg for ^{226}Ra, 0.1 ± 0.1 – 6 ± 1.6 Bq/kg for ^{232}Th, 1.02 ± 0.2-- 2.7 ±1.2 Bq/kg for ^{238}U, and 124.09 ± 10 – 548.36 ± 44 Bq/kg for ^{40}K, as they illustrated in figure 3, which are comparable to levels reported for neibouring and word wide areas, as seen in table 3.

On comparison, the average concentrations for ^{232}Th, and ^{40}K in the soil depth 0-16 cm from Qatar were 10 and 210 Bq/kg, which were low compared to world wide levels of 45 and 412Bq/kg which has been recorded by Ahmed and his colloquies, [9], while measured gamma activity concentrations in sediments from around Barakah Nuclear Power Plant , UAE, were 4.73± 0.47, 1.83± 0.24, and 105± 10.03 Bq/kg for ^{238}U, ^{232}Th, and ^{40}K respectively as reported by Al-Rashdi and his colleague, [22].

On the other hand Radioactivity measurements were correlated to TOC of the sediments as shown in figure 4. In core sediments from Amvrakikor Gulf (Western Greece) activity concentrations for ^{232}Th, ^{238}U, ^{40}K, and ^{210}Pb were assessed and recorded as 10 - 20 Bq/kg for ^{226}Ra, 6 - 20 Bq/kg for ^{222}Rn, 20 –28 Bq/kg for ^{238}Ac, 400—830 Bq/kg for ^{40}K, and 11—360 Bq/kg for ^{210}Pb were recorded by Tsabaris and his colleague's [22]. For TOC values recorded in core sediments from the three sites were 0.919% in site No. 2 to 1.904 % in site No. 1. which were higher than levels reported in the same sites earlier averaged as 0.53 % by Al-Mansory and his colleague

[14]. Levels of % TOC and texture of the sediments from the same sits 1 and 2 at Khor Abdullah were reported as shown in table 4 below:

Table 4: Levels of %TOC and texture of the Bottom surface sediments from sites 1 and 2 at Khor Abdullah adopted from Al-Mansory and his colleaque[14].

Site	1	2
TOC%	0.53	0.53
Clay%	23.00	31.36
Silt%	29.30	47.94
Sand%	47.69	20.70

Mean Radium equivalent activity in soil from Qater was recorded to be 47 Bq/kg which was far below the minimum recommended international values reported by Ahmed and his colleague's [9].

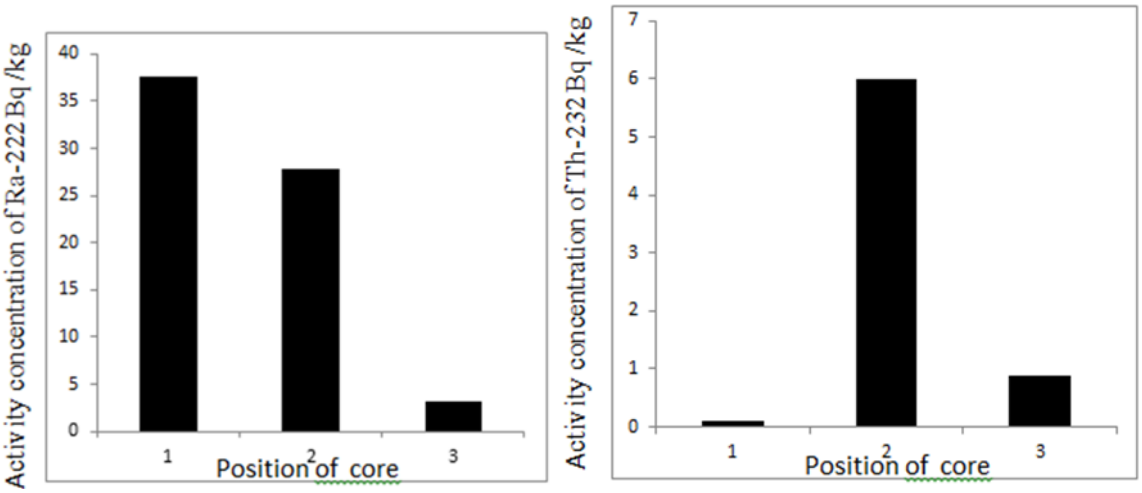
As it is shown in fig. 3, the radioactivity concentration of ^{226}Ra and ^{40}K were positively correlated to % TOC, while ^{232}Th and ^{238}U were negatively correlated to % TOC as reported by Ahmed and his colleague's [9].

Jaber and his colleagues' [23] reported that Ra eq were ranged between 80.0 Bq/kg to 184.2 Bq/kg with mean value =84.5 Bq/kg which was comparable to values reported for neighboring countries while it is less than world wide level.

In the last column of table 2 listed the values of Radium equivalent activity (Ra_eq) which is used to assess the hazards associated with material that contains ^{226}Ra, ^{232}Th, and ^{40}K in Bq.kg^{-1}.

Values of Ra-eq were ranged between 14.05±5.2 in sediments of site No. 3. and 79.97±54.2 in sediment of site No. 1 with a mean value of 47.01Bq/kg in the sediments from Khor Abdullah, which is strongly correlated to ^{40}K and ^{226}Ra.

Contaminated bottom surface sediments became a source of pollutants which are liable to release pollutants whether they are radioactive or nonradioactive materials as stated by Patton and his colleague [25].



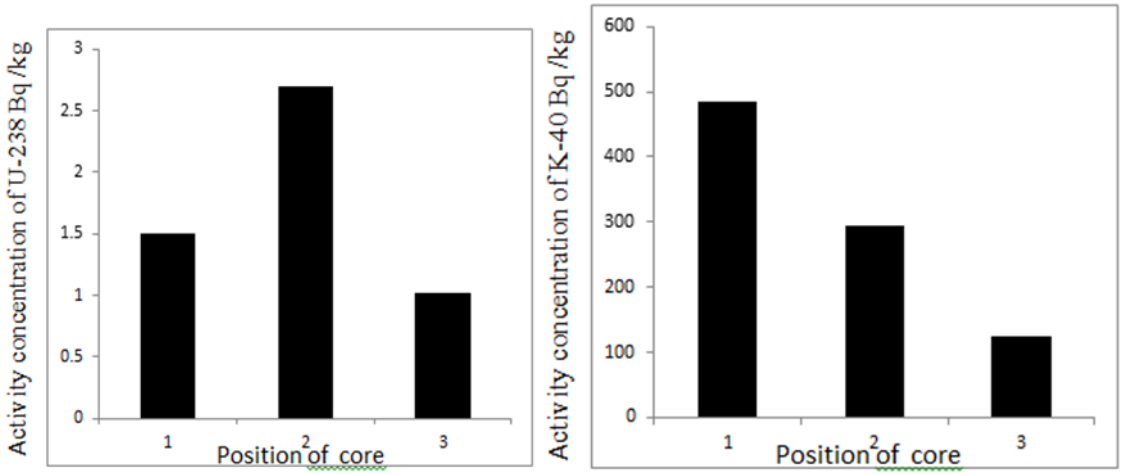


Figure 3: Variations in activity concentrations for studied radionuclides with the position of sampling sites at Khor Abdullah/Northern west Arabian Gulf.

According to recommended maximum value of Ra-eq which equal 370 Bq/kg, the calculated value for the sediments of Khor Abdullah is 51 Bq/kg which is quite low compared to the world wide level reported by Bajoga and his colleagues' [26]. As expected, a small quantities of radioactive materials are discharges from land of battle zone upstream Shatt Al-Basrah canal, then they transferred with water direction towards the South reaching Khor Al-Zubair and then to Khor Abdullah and finally to the Northern Arabian Gulf in which they will deposit on the sediments over their and accumulate in the core sediments during the last three decades reported by Jaber and his colleaque's [24]; Jaber [27]; and Ali and his colleague [6;28].

6. Conclusion

Naturally occurring radioisotops such as ^{232}Th, ^{238}U, and ^{40}K are emiting Gamma radiation even they exist at trace levels and they became an external source of irradiation to human body especially the fisheries who use this area for fishing.

The levels of activity concentrations in the sediments of Khor Abdullal at the Northwest Arabian Gulf are close to their levels for sediments at nearby areas which concluded that the whole area of Northern Arabian Gulf starting from the Iranian coast passing through the Iraqi coasts reaching the Kuwaiti coast are contaminated to a certain extent with radioactive nuclides but exert no harmful risk for living organisms, animals, plants, and human beings. Even if there is an external radiation dose, the calculated value of Ra-eq, in the sediments of Khor Abdullah mean value is 51Bq/kg is very low compared with word wide level of 370 Bq/kg.

Table 3: Concentration activity and Radium equivalent activity for radionuclides in this study, compared with neighboring areas and word wide .

Sites	^{40}K	^{226}Ra	^{210}Pb	^{235}U	^{238}U	^{234}Th	Ra-eq	Reference's
Tigris River		4.06-7.52						Amin and his colleaque's [21]
Marsh land (Iraq)	147.7	13.8-25.28				5.3-6.5		Salman and his colleaque's [98]
Khor Abdulla	293.9	44.7		7.2	5.6		84.5	Jaber and his colleaque's [24]
Kuwait Bay	381	19.08	34	1.03		22.89		Al-Zamel and his colleaque's [10]
Arabian Gulf	700-740		^{212}Pb 10.6-11.1					Bashar and his colleaque's [30]
Malaysia	259-571		43		34.6-61.	28.16-71.77		Abdullah and his colleaque's [31]
East Malaysian Marine sediments	462	30						Yii and his colleagues 2009[32]
Red Sea	374.9 ±32	23.8± 3.8				^{232}Th 19.6±3.2	81.2	Zakaly and his colleagues (2019)[33]
World	850	60				64		UNSCEAR,[16].
World							370	Bajoga and his colleaque's [26]
Mediterranean Coast, Turkey	590	25.5				27.9		Özmen and his colleaque's [34]
Inani Beach, Bangladesh	560	28.67				49		Ahmed and his colleaque's [35]
Borcka Black Lake	473.67				14.99	13.85		Dizman and his colleaque's [36]
Western Greece	400-830	10-20						Tsabaris and his colleaque's [23]
Western Coastal of South Sulawesi	744	47.29			52.73			Prihatiningsih, and his colleaque [37]
Tamilnadu / India	360.3				<2.21	14.29		Ravisankara and his colleaque's [38]
Eastern, Saudi Arabia	561	26.4				16.3		Alshahri, [39]
Khor Abdullah	334	40.4				3.05	51	This Study

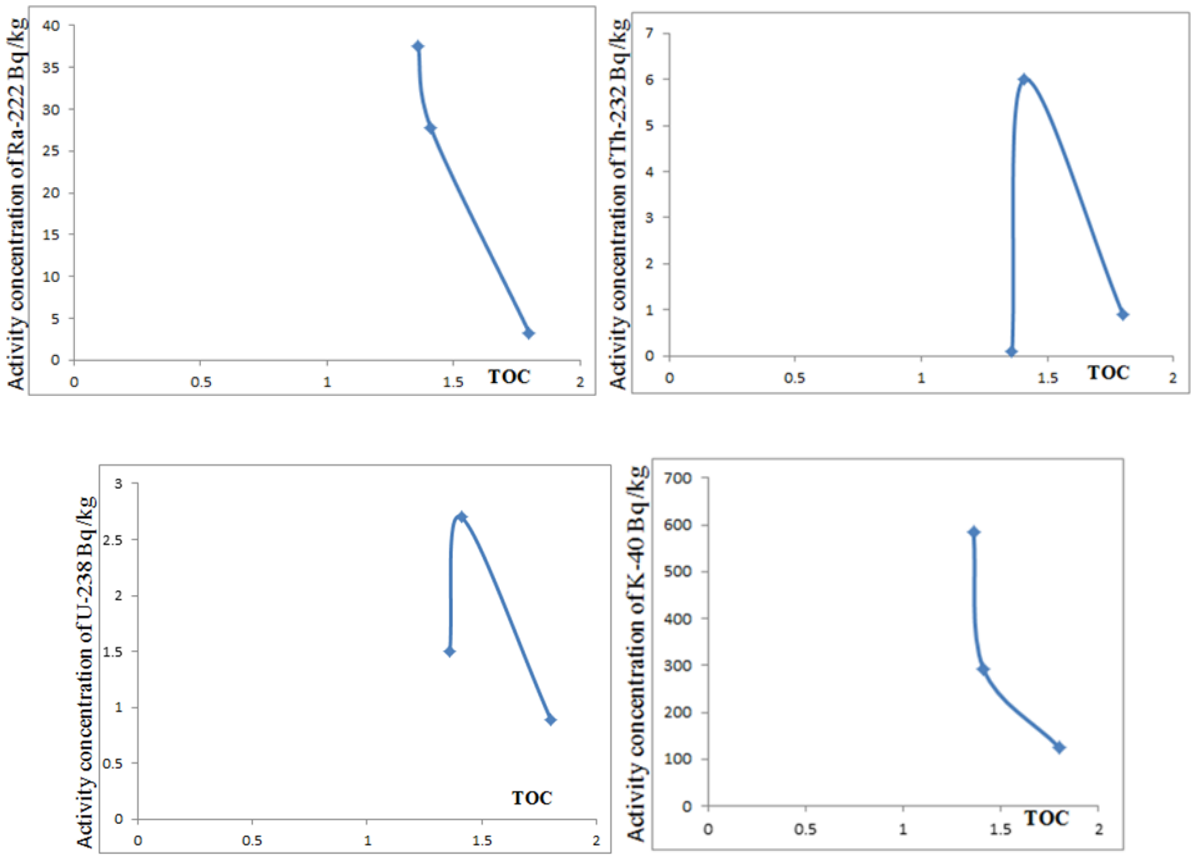


Figure 4: Relationship of activity concentrations for radioactive nuclides and TOC of the sediments from Khor Abdullah.

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Biography of author(s)

Figure 5: Experience Prof. Dr. Faris Jasim Mohammad Al-Imarah,

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Born on the 4th of September 1951 in Basra Governorate of Iraq, studied in primary (1963), and secondary (1966). Higher preparatory (1969) schools in Basrah province, he obtained his B.Sc. in Chemistry at Basra University in 1973, M.Sc. in Physical Chemistry at Baghdad University in 1976, and finally received his D. Phil. As a laser spectroscopic at Sussex University/The United Kingdom in 1984, Back to Basra joined the staff of Basra University as a lecturer. Dr. Imarah received his first promotion to Ass. Prof. at Marine Science Centre/Basra University in 1990, then he received his second promotion to Prof. in 1995 at the same Centre. Prof Imarah supervised 20 students as M.Sc. and Ph. D., and he was involved in different fields of research mainly Chemistry, and Environment and Pollution of different media, air, water, soil, sediments, and biota of Southern Iraqi Marshlands, Southern Iraqi waterways, and Northern West Arabian Gulf. He has 190 research papers published in local, national, and international journals.

Prof. Imarah received his title as an experienced prof. from Basrah University in 2022.