

Radionuclide Contamination in Soil and Radiological Hazard Assessment from Industrial Areas: A Systematic Review

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ABSTRACT

Background: The production of radionuclides as industrial by-products such as radium (²²⁶Ra), thorium (²³²Th), potassium (⁴⁰K), and uranium (²³⁸U) might contaminate the soil and harm the health of nearby populations for long-term. Due to the limited evidence of the associated relation and lack of public awareness of the potential risk, people tend to ignore this concerning issue. Therefore, this study aims to review the activity concentrations of the aforementioned radionuclides in the soil's nearest industrial vicinity and to assess their radiological hazard presented in the existing literature. **Method:** This systematic review was conducted using Preferred Reporting Items for Systematic Review and Meta-Analyses (PRISMA 2009) on online databases such as PubMed, SpringerLink, Scopus, and ProQuest. The following criteria were included: full-text English journal, studies from 2014 onwards with search keywords of "radionuclide exposure" AND "radiological hazard" OR "health effect". **Results:** A total of 1025 articles were screened and only 7 full-text articles were evaluated. Based on the review, the types of industries that produce ²²⁶Ra, ²³²Th, ⁴⁰K, and ²³⁸U were petrochemical, chemical, rare-earth element (REE), and gold mining industries. The findings showed the elevated ²²⁶Ra activity, nearly three times the global average of 35 Bq/kg, was found at petrochemical sites in Rayong, Thailand. The ²³²Th and ⁴⁰K activity levels at Nigeria mining sites were higher than the global average. All studied areas exceeded world average for ²³⁸U. The highest absorbed dose (D) values were observed in artisanal mining, Anka, Nigeria (127.00 nGy/h) and in petrochemical sites in Rayong, Thailand (84.98 nGy/h), both exceeding the limit of 60 nGy/h. The annual outdoor effective dose (AED) from similar industrial areas was 2.2 and 1.4 times higher than the global average of 0.07 mSv/y. The highest gamma index (I_γr) value was at 2.08, recorded in Anka artisanal mining area, exceeding the safe limit of 1. Meanwhile, all values for excess lifetime cancer risk (ELCR) were below a safe limit of 1.16×10^{-3} . **Conclusion:** In conclusion, radiological risks at Anka artisanal mining sites and Rayong petrochemical sites, exceeded UNSCEAR limits, but cancer risks were minimal, suggesting a need for further research including in groundwater samples and clinical studies.

Keywords:

Radionuclides exposure; radiological hazard assessment; soil; industrial areas

INTRODUCTION

A radionuclide, known as a radioisotope, radioactive isotope, or radioactive nuclide, is an unstable atom containing excess energy (Ansobarlo and Adam-Guillermín, 2012). According to the Centers for Disease Control and Prevention (2015), unstable radionuclides spontaneously emit radiation in the form of energetic particles of alpha, beta, or gamma radiation to other radioisotopes. This process is called radionuclide decay which can be measured by its half-life (Choppin, 2012).

The natural sources of radionuclides are commonly known as naturally occurring radioactive materials (NORMs) and previous studies have shown that most of the radionuclides can be found naturally in the environment (Almayahi, Tajuddin, & Jaafar, 2012). Sources of radionuclide contamination also could be generated from nuclear weapons programs, nuclear weapons testing, nuclear power plants, uranium mining and milling, commercial fuel reprocessing, nuclear accidents, and

radionuclides contained at the geological repository (Hu et al., 2010). These industries process the desired resource by releasing the radionuclides and generating technologically enhanced naturally occurring radioactive material (TENORMs) by-products.

These by-products of radionuclides may contaminate the air, soil, surface, and groundwater if not disposed of properly. Hence, human exposure to TENORMs and NORMs of earth gamma-emitting radionuclides such as radium, thorium, potassium, and uranium is inevitable. Radiation exposure to humans has been increasing since decades ago and might go unnoticed due to its diverse usage. According to the American Institute of Physics (2014), the Malaysian Rare Earth Corporation Plant (MAREC) at Papan, Perak has been operating until 1992 and stopped due to abundant radionuclide waste such as thorium and uranium found in soil. A previous study conducted in Malaysia by Almayahi, Tajuddin, & Jaafar (2012), from 2004 until 2008 revealed an increase in cancer cases in Penang, which recorded up to 9692 cases

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due to exposure to high concentrations of natural radioactivity.

Chen (2005) stated human health may be affected by prolonged exposure to low levels of radionuclides following contamination through the water, air, or soil, the radionuclides can be deposited in blood, brain, and bones by ingestion, inhalation, absorbed from skin, and wound contamination (Hao et al., 2015). Radiation risk among industrial workers is controlled by the International Atomic Energy Agency (IAEA) safety standard using personal radiation dosimeters to detect radionuclides exposure. However, the population living near the industrial area is also vulnerable to low doses of radionuclides from prolonged exposure but there is no radiation assessment available to them (Rana et al., 2010).

Zhe Hao et al. (2015) mentioned that there is limited evidence about the relationship between radionuclide exposure and potential health effects on residents living near industrial areas for a long period. It is important to determine the radiological hazard assessment (e.g.: annual effective dose, excess cancer risk, lifetime average daily dose, and hazard quotient) from exposed radionuclides in industrial areas. Thus, this study provides an opportunity to systematically review the radiological hazard at low levels of radionuclide exposure among the population living near industrial areas from previous literature.

Despite extensive research, limited studies were found on the associated link between the levels of radionuclides in soil and their radiological hazard assessment on human health. Therefore, this study aims to review the activity concentrations of radium (^{226}Ra), thorium (^{232}Th), potassium (^{40}K), and uranium (^{238}U) in soil and assess the radiological hazard of the above-mentioned radionuclides measured from industrial areas based on the published literature.

MATERIALS AND METHODS

Systematic Review Process

This systematic review applied the Preferred Reporting Items for Systematic Review and Meta-Analyses (PRISMA 2009) method to aid in reporting the findings. The PRISMA guidelines include identification, screening, eligibility, and included criteria.

Search Strategy

The articles were sought from Scopus, SpringerLink, PubMed, and ProQuest. The keywords (“uranium” OR “radium” OR “thorium” OR “potassium”) AND (“nuclear

plant” OR “petrochemical” OR “rare earth element (REE)” OR “industry”) AND (“health risk assessment”) were used. Boolean terms (AND and OR) were used to separate each keyword; "AND" is used to restrict the search, whereas the search is extended by "OR".

The search technique in this study aims to identify a comprehensive range of relevant papers on the topic, ensuring both high sensitivity and accuracy in the results. The PICOS elements, namely population (P), intervention (I), comparator (C), outcome (O), and study design (S)- were crucial in identifying the specific criteria to be included in this review as shown in Table 1.

Table 1: PICOS framework to determine the eligibility of studies

Criteria	Determinants
Problem	Residents living near the industrial area are at pose risk of exposure to radionuclides
Intervention	Exposure level of ^{226}Ra , ^{232}Th , ^{40}K and ^{238}U
Comparator	Radiological hazard assessment
Outcomes	Health effects on the population
Study design	Cross-sectional studies

Inclusion and Exclusion Criteria

The articles were screened by their title, especially those mentioned radium (^{226}Ra), thorium (^{232}Th), potassium (^{40}K), and uranium (^{238}U). The inclusion criteria such as full-text English language or English-translated literature that were published in 2014 onwards, with research articles must include exposure levels to public and radiological hazards of ^{226}Ra , ^{232}Th , ^{40}K , and ^{238}U in determining the health effects. The exclusion criteria such as review study, incomplete literature, unrelated topic, non-English language, and no available author were removed from this study.

Review Method

Articles were evaluated and assessed for eligibility according to the inclusion and exclusion criteria based on their title and abstract. Articles that fulfill the requirements were included to be reviewed. The quality of the studies was evaluated using the National Heart, Lung, and Blood Institute's (NHLBI) quality risk assessment method for cohort and cross-sectional studies from the National Institute of Health (NIH).

RESULTS

The article selection process was simplified in the PRISMA flow diagram as shown in Figure 1. A total of 1025 articles were derived from the online databases namely PubMed (n=6), Scopus (n=18), ProQuest (n=537), and SpringerLink (464). However, 49 studies were excluded due to duplication. Then, the articles were screened by title and abstract, resulting in the removal of 968 articles. The remaining eight articles were evaluated according to the inclusion and exclusion criteria. Next, three of them were removed due to the inaccessibility of the full text. After the full text was reviewed, two articles were excluded as it does not have any radiological risk assessment in their

study leaving only three articles available for the review. From the three articles, snowball techniques were conducted, and the addition of four articles was able to be retrieved from the reference list.

The quality of the included studies was evaluated using the NHLBI quality risk assessment tool. Only one out of seven articles were determined as good quality while others were fair quality. Those six articles were considered as fair as they did not give enough information on numerous checklist criteria such as sample size justification and participation rate, making it impossible to assess their quality. Despite that, all seven articles were found as eligible according to the inclusion criteria.

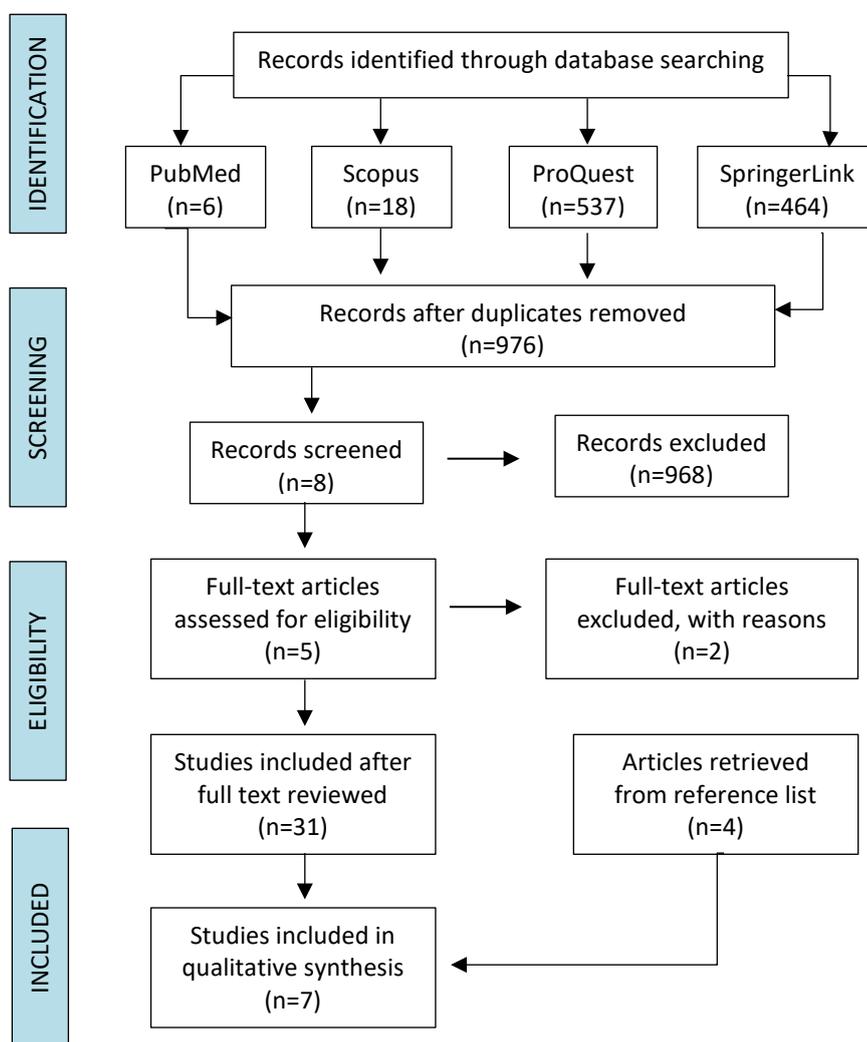


Figure 1: PRISMA flow diagram

DISCUSSION

Study Location and Types of Industrial Area

Seven reviewed studies highlighted five countries, namely Saudi Arabia, Nigeria, Thailand, Ghana, and Malaysia. The industrial city of the Arabian Gulf Coast in Saudi Arabia

hosts over a hundred petrochemical and chemical industries and has a population of approximately 100,000 residents, with residential districts located to the east, south, and north (Alshahri, 2019). Additionally, Ras Tanura, home to the largest and oldest oil refinery in the Middle East, spans an area of approximately 290 km² and includes residential zones with a population of around 74,000

inhabitants (Alshahri & El-Taher, 2018). The studies conducted in Anka and Itagunmodi, Nigeria, and Akyem, Ghana focused on gold mining activities (Akpanowo et al., 2020; Bekelesi, Darko & Andam, 2017; Ademola et al., 2014). The Anka gold mining area spans 2,940 km² and is home to an estimated population of 12,655, residing approximately 10 km from the mining site. Since 1980, various industries, including petrochemical, automotive, electronics, oil, and gas sectors, have been operating in Rayong, Thailand (Kessaratikoon et al., 2019). Nonetheless, the rare earth refinery industry in Kuantan, Malaysia is known as Lynas Advanced Material Plant (LAMP) is the largest, rare earth refinery project in the world with a study area between 0.9 km and 3 km from the LAMP (Kolo et al., 2015).

Activity Concentrations of Radionuclides in Soil Samples

According to the seven included studies, the activity concentrations of radionuclides were used to determine the scientific evidence affecting the population's health effects from radium, thorium, potassium, and uranium exposure. The radionuclides activity concentrations were analysed using HPGe gamma-ray detector and gamma spectrometry analysis (Akpanowo et al., 2020; Kessaratikoon et al., 2019; Alshahri, 2019; Alshahri & El-Taher, 2018; Bekelesi, Darko & Andam, 2017; Kolo et al., 2015; Ademola et al., 2014). All extracted data is presented in Table 2.

Table 2: Mean Activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ²³⁸U in soil samples

Authors	Location	Type of industry	Mean Activity Concentrations (Bq/kg)			
			Radium (²²⁶ Ra)	Thorium (²³² Th)	Potassium (⁴⁰ K)	Uranium (²³⁸ U)
			World Average Concentrations (Bq/kg) (UNSCEAR, 2000)			
			35	30	400	35
Akpanowo et al. (2020)	Anka, Zamfara State, North-West, Nigeria	Artisanal mining and mine processing	37.94*	151.15*	380.34	41.60*
Alshahri (2019)	Northern Al Jubail, Arabian Gulf, Saudi Arabia	Petrochemical & Chemical Industries ^a	7.64	3.76	174.00	-
Kessaratikoon et al. (2019)	Rayong province, Thailand	Petrochemical	96.65*^b	36.73*^b	423.75*^b	-
Alshahri & El-Taher (2018)	Ras Tanura, Arabian Gulf, Saudi Arabia	Oil Refineries & Gas Plant	23.20	7.73	278.00	39.00*
Bekelesi, Darko & Andam (2017)	Akyem, Ghana	Gold mining	28.00	12.00	11.00	-
Kolo et al. (2015)	Gebeng Kuantan, Pahang, Malaysia	Rare Earth Oxides Processing Plant	6.56	10.62	41.02	-
Ademola et al. (2014)	Itagunmodi, South-Western, Nigeria	Gold mining	-	26.4	505.10*	55.30*

Note: *Indicate the value exceeds the world average concentrations; ^aIncluding industries of phosphate, iron, chemical, water treatment plant, gas plant, oil refinery, ethylene, and methanol Industries; ^bUsing median values due to asymmetrical distribution of data.

From Table 2, the activity concentrations of studied radionuclides in petrochemical and chemical industries, Al Jubail of Saudi Arabia, gold mining, Akyem of Ghana, and rare earth oxides processing, Kuantan, Malaysia were below the acceptable limits except for the Anka and Itaganmodi in Nigeria, Ras Tanura of Saudi Arabia, and Rayong province, Thailand. The highest level of ^{226}Ra was recorded in the petrochemical sites in Rayong, Thailand with a median activity concentration of 96.65 Bq/kg (mean values = 105.25 Bq/kg). This was followed by the artisanal mining industry in Anka, Nigeria, with a mean value of 37.94 Bq/kg, both exceeding the global average activity concentration of 35 Bq/kg by 2.8 and 1.1 times, respectively (Kessaratikoon et al., 2019; Akpanowo et al., 2020). In contrast, the lowest mean activity concentration, 6.56 Bq/kg, was observed in Kuantan, Malaysia's rare earth element (REE) industry (Kolo et al., 2015). The asymmetrical data observed in the study by Kessaratikoon et al. (2019) prompted the use of median values, which were selected for radiological hazard estimation.

Additionally, Anka, Nigeria, reported the highest mean activity concentration of ^{232}Th at 151.15 Bq/kg, significantly exceeding five times the global average of 30 Bq/kg (Akpanowo et al., 2020). Meanwhile, gold mining sites in Itaganmodi, Nigeria, and petrochemical sites in Rayong, Thailand, recorded the highest and second-highest activity concentrations of ^{40}K , with a mean value of 505.10 Bq/kg and a median value of 423.75 Bq/kg (mean value = 532.39 Bq/kg), respectively. Both exceeded the global average activity concentration of 400 Bq/kg.

The highest mean activity concentration of ^{238}U was detected at the Itaganmodi gold mining sites, with a mean value of 55.30 Bq/kg, followed by Anka gold mining (41.60 Bq/kg), oil refineries and gas plants at Ras Tanura, Saudi Arabia (39.0 Bq/kg). These three industrial areas exceeded the global average concentration of ^{238}U (30 Bq/kg).

Factors Influence the Mean Activity Concentrations of Radionuclides

The mean activity concentrations for ^{226}Ra , ^{232}Th , and ^{238}U in the artisanal mining areas exceeded global averages, primarily due to the geological characteristics and activity concentrations of geology in the mining region and mineral processing activities, further contributing to elevated radioactivity levels in the soil (Moshupya et al., 2022; Akpanowo et al., 2020). Variations in geological structures and dust generated during mining activities can contribute to exposure to naturally occurring radioactive materials (NORMs) and radon gas (Ademola et al., 2014).

Other factors, such as ongoing construction activities and the physicochemical and geochemical properties of specific radionuclides, can also influence soil turnover concentrations. Meteorological factors, such as wind direction and rainfall distribution, can also influence the movement and deposition of radionuclides (Alshahri and El-Taher, 2018).

Radiological Hazard Assessment and Comparison

Industrial by-products containing radionuclides pose a risk to nearby populations, as the waste can accumulate in the soil, potentially leading to adverse health effects (Alshahri & El-Taher, 2018; Kolo et al, 2014). To assess the radiological hazard effects in soil samples for specific activities of ^{226}Ra , ^{232}Th , ^{40}K , and ^{238}U , the radium equivalent activity (R_{eq}), air absorbed gamma radiation dose rate (D), annual effective dose equivalent (E), external hazard (Hex), gamma representative level index (I_γ), excess lifetime cancer risk (ELCR) and geoaccumulation index (I_{geo}) and pollution load index (PLI) was calculated and presented in Table 3.

As shown in Table 3, the highest mean value of R_{eq} is documented at the artisanal mining site in Anka, Nigeria, with 288.51 Bq/kg, followed by the petrochemical industries in Ras Tanura, Saudi Arabia (62.10 Bq/kg), the gold mining site in Itaganmodi, Nigeria (31.75 Bq/kg), the petrochemical and chemical industries in Northern Al Jubail, Saudi Arabia (26.40 Bq/kg), and the lowest value at the rare earth oxides processing plant in Kuantan, Pahang (24.92 Bq/kg). All reviewed studies recorded R_{eq} was below the world average of 370 Bq/kg (UNSCEAR, 2000), indicating that the gamma output and the radiation hazards mixture of ^{232}Th , ^{40}K , and ^{238}U in analysed soils samples are within safe limits for human health and environment.

The International Commission on Radiological Protection (ICRP) recommends an absorbed dose value of 55 nGy/h (Alshahri and El-Taher, 2018; Kessaratikoon et al., 2019), while UNSCEAR (2000) sets the threshold at 60 nGy/h. In the studies reviewed, the highest absorbed dose values were observed in the artisanal mining area in Anka and the petrochemical industry in Rayong, Thailand, with reported values of 127.00 nGy/h and 84.98 nGy/h, respectively. This absorbed dose rate shows an elevated of ^{226}Ra , ^{232}Th , and ^{40}K from terrestrial gamma radiation sources.

The annual outdoor effective dose (AED) from artisanal mining in Anka, Nigeria, and the petrochemical industry in Rayong, Thailand, is 2.2 and 1.4 times higher, respectively, than the global average of 0.07 mSv/y (Kessaratikoon et al., 2019; Akpanowo et al., 2020). This indicates that global

Table 3: Estimated radiological hazard in soil samples (mean values)

Reference	Location	Radiological Hazard					
		Ra _{eq} (Bq/kg)	D (nGy/h)	AED (mSv/y)	Hex	I _{yr}	ELCR (x10 ⁻³)
Akpanowo et al. (2020)	Anka, Zamfara State, North-West, Nigeria	288.51	127.00*	0.156*	0.780	2.06*	0.550
Alshahri (2019)	Northern Al Jubail, Arabian Gulf, Saudi Arabia	26.40	13.00	0.016	-	-	-
Kessaratikoon et al. (2019)	Rayong province, Thailand	181.80	84.98*	0.100*	0.490	-	0.390
Alshahri & El-Taher (2018)	Ras Tanura, Arabian Gulf, Saudi Arabia	62.10	29.30	0.038	0.160	0.45	-
Bekelesi, Darko & Andam (2017)	Akyem, Ghana	37.53	-	0.044	0.101	-	-
Kolo et al. (2015)	Gebeng Kuantan, Pahang	24.92	11.16	0.010	0.070	0.18	0.050
Ademola et al. (2014)	Itaganmodi, South-Western, Nigeria	31.75	20.40	0.025	0.110	0.33	-
Global Average Limit		370 ^a	60 ^a	0.07 ^a	1 ^c	1 ^c	1.16 × 10 ^{-3a}

Note: *Exceed the global average limit; Ra_{eq}: Radium equivalent; D: Absorbed dose rate; AED: Annual effective dose (outdoor); Hex: External hazard; I_{yr}: Gamma index; ELCR: Excess lifetime cancer risk (outdoor); I_{geo}: Geoaccumulation index; PLI: Pollution load index; ^aSource: UNSCEAR, 2000; ^bSource: Alshahri and El-Taher (2018); Kessaratikoon et al. (2019); ^cSource: Akpanowo et al. (2020).

average, highlighting elevated radiological exposure in the vicinity of these studied locations.

For the external hazard index (Hex), the highest value was recorded in Anka, Nigeria, at 0.78, while the lowest value was observed in the rare earth oxides industry in Kuantan, Pahang, at 0.07. None of the studied locations exceeded the recommended Hex limit of 1 (Akpanowo et al., 2020). Regarding the radioactivity level index (I_{yr}), the mean values for the petrochemical industry in Ras Tanura, Saudi Arabia, the rare earth processing industry in Kuantan, Pahang, and the gold mining industry in Itaganmodi, Nigeria, were 2.06, 0.45, 0.18, and 0.33, respectively. However, the highest I_{yr} value was recorded in the artisanal mining area of Anka, Nigeria, at 2.08, exceeding the recommended safe limit of 1 (Akpanowo et al., 2020). This indicates that gamma radiation exposure in the area is more than twice the recommended threshold, potentially posing health risks to the nearby population.

Three out of the seven articles assessed the excess lifetime cancer risk (ELCR), with all countries reporting values below the global safe limit of 1.16 × 10⁻³ for outdoor exposure (Akpanowo et al., 2020; Kessaratikoon et al.

2019; Kolo et al., 2015). This indicates that the populations in the studied areas are unlikely to develop cancer due to the levels of gamma radiation reported in these studies.

Artisanal mining in Anka, Nigeria and the petrochemical site in Rayong, Thailand both exhibited elevated values for absorbed dose (D), annual effective dose (AED), and radioactivity level index (I_{yr}). These findings suggest that individuals in these areas, particularly those working near these sites, are exposed to higher levels of environmental gamma radiation. This increased exposure could pose a potential radiological hazard to the nearby local populations. Akpanowo et al. (2020) highlighted those concerns regarding environmental radioactivity were more pronounced for artisanal workers in the mining industry, while nearby populations were not considered to be at significant risk. Despite the elevated radiation levels, the estimated cancer risk for all studied areas remains below the threshold, Kessaratikoon et al. (2019), however, argued that prolonged radiation exposure in the general population could increase the risk of cancer over time.

Previous reviewed studies have several limitations. The estimated excess lifetime cancer risk is more relevant to

artisanal miners and mineral processing workers, as the general population may not face an immediate radiological risk (Akpanowo et al., 2020). However, the scope of these studies is limited to the current investigations and analyzed samples. Expanding research to cover broader areas is recommended, particularly industrial zones near densely populated residential areas or water sources (Kolo et al., 2015; Alshahri, 2019).

CONCLUSION

In conclusion, radionuclide activity exceeding global average concentrations was observed in certain studied areas. Elevated levels of ^{226}Ra , almost three times higher than the global average, were detected at petrochemical sites in Rayong, Thailand. Meanwhile, ^{232}Th activity at artisanal mining sites in Anka, Nigeria, was five times greater than the global average. Additionally, elevated levels of ^{40}K were predominantly found at the Itagunmodi gold mining sites in Nigeria. Notably, ^{238}U activity surpassed the global average across all the studied areas. Geological factors appear to be a significant contributor to the elevated radionuclide concentrations, in addition to the by-products of industrial activities themselves. The radiological risks of absorbed dose (D) and annual outdoor effective dose (AED) were notably above the UNSCEAR safe limits at artisanal mining sites in Anka, Nigeria and petrochemicals sites in Rayong, Thailand, suggesting these areas may expose nearby populations and particularly those working in close proximity, to elevated levels of gamma radiation. All cancer risk values of studied radionuclides were below world safe limits of 1.16×10^{-3} , indicating the exposure to gamma radiation in the studied industrial areas is minimal. Assessing radiological hazards in other mediums, such as groundwater samples and through clinical studies, could yield different findings regarding the potential risks of radionuclide exposure to human health.

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