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Radiological Exposure of Naturally Occurring Radioactive Materials (NORMs) in Soils at Gold Mining Areas: A Meta-Analysis

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ABSTRACT

Naturally Occurring Radioactive Materials (NORMs), although naturally occurring in the Earth's crust, become sources of radiological exposure through mining activities. This work provides a meta-analytical assessment of the radiological exposure of NORMs including ^{232}Th, ^{40}K and ^{238}U or ^{226}Ra in soils of gold mining areas. The analysis collates data from various studies on measurement of radiological parameters including Activity Concentrations, Absorbed Dose Rates, Annual Effective Doses, and Radium Equivalent Activities, as well as Internal and External Hazard Indices of different gold mining sites from Nigeria, Kenya, Tanzania and Laos. The studies employed gamma spectrometry using NaI (Tl) detector and decomposition of measured gamma spectra to analyse these parameters from soil samples previously dug out from gold mines. The results showed that while most areas remained within internationally established safety limits, certain sites demonstrated increased radiological risks; especially with regards to ^{40}K and ^{232}Th radionuclides. Sites with high radium equivalent activities approached critical thresholds of safety, and will, thus, require continuous monitoring and perhaps mitigation strategies in order to avoid hazardous levels of radiological exposure to workers and nearby residents. This meta-analysis gives a complete assessment of radiological hazards and provides information on environmental radiation protection in gold mining areas.

Keywords: Gold Mining, Gamma Spectrometry, Naturally Occurring Radioactive Materials, Radiological Exposure, Radiological Parameters

INTRODUCTION

Naturally Occurring Radioactive Materials (NORMs) are considered a key component of the Earth's geosphere and atmosphere [1]. The long-lived radioactive nature of NORMs contributes to radiation exposure in soils [1]. NORMs; such as potassium-40 and rubidium, as well as the radionuclides in the decay chains of thorium-232 and uranium-238; such as radon-222 and radium-226, especially at high geological concentrations, emit radiation from soils at mining areas, of which gold mines are no exception [2]. Mining activities expose embedded NORMs and their associated radioactivity from the earth's crust unto the surface of the earth and the atmosphere, which increases radiation exposure of NORMs in the environment [3]. Long-term radiation exposure to NORMs in high concentrations can be hazardous to the health of mining workers, the ecosystem around the mines, as well as surrounding residents [4].

NORMs emit various types of radiations; such as alpha, beta, and gamma. Alpha radiations, especially when internalized in the human body, cause damage to cells, which enhances the risk of cancer development. For instance, the alpha emitter, Radon-222 is a gaseous decay product of uranium, and may accumulate in poorly ventilated areas, especially underground mines. When inhaled, this contributes to the risk of lung cancer [5].

Without necessarily being internalized in the human body, beta and gamma radiation can readily affect living tissues and the environment due to their higher penetration power [6]. Whereas skin burns arise from large doses of beta radiation, long-term exposure increases the risk of organ damage. Being highly penetrating, gamma radiation arising from NORMs constitutes an external radiation hazard that could cause whole-body exposure leading to systemic health effects such as immune system suppression, genetic mutations and cancers [7].

The environmental impacts of NORMs are equally disturbing. Long-term radionuclide accumulation in ecosystems arising from radioactive contamination of soils, water, and air [8] bioaccumulates in plants and animals. This phenomenon causes biomagnification, which eventually puts humans at the risk of the various possible harmful effects of NORMs [9]. Moreover, long-lived radionuclides; such as thorium-232, when deposited in the environment, continue to emit radiation over several thousand years. In that respect, they may present an ecological hazard over a very long time [10].

Various studies have been conducted on soil from mining areas regarding NORM levels, with variable concentrations depending on local geology, mining methods, and environmental management methods. Most data, however, have remained fragmented, which has made it quite difficult to assess risks to overall exposures on a wider scale. This research therefore seeks to fill this gap through a meta-analysis of existing studies on NORM levels in soils from gold mining areas. This meta-analytical approach combines various studies towards a more wholesome assessment of radiological exposure of NORMs and its associated risks at gold mining sites across different countries. This study is therefore targeted at synthesizing evidence available on the rate and extent of radiological exposure at gold mining sites. It also attempts to provide a more transparent view of the exposure along with suggestions on the safety concern in mining areas.

MATERIALS AND METHODS

Scope of Study

This meta-analytical study followed the guidelines from 'Preferred Reporting Items for Systematic Reviews and Meta-Analyses' (PRISMA) [11]. The studies included were selected based on original studies that measured radiological parameters of ^{232}Th, ^{40}K, and ^{238}U or ^{226}Ra with respect to Activity Concentration, Absorbed Dose Rate (ADR), Annual Effective Dose (AED), Radium Equivalent Activity (Ra_{Eq}) and Internal and External Radiation Exposure (H_{in} and H_{ex}) determined quantitatively at gold mines in African countries including Nigeria [13, 14, 15, 16, 18, 19] Kenya [17], Tanzania [21], and the Asian country of Laos [20].

Selection of Articles

An extensive search based on peer reviewed publications was carried out through the following databases: Pubmed (197 articles), Web of Science (240 articles), Scopus (213 articles), Google Scholar (393 articles), and other sources (11 articles) such that the total number of articles were 1054. The use of keywords such as 'Radiological exposure', 'Naturally Occurring Radioactive Materials (NORMs)', 'Soils', 'Gold mining', 'Environmental Radioactivity', 'Radiological assessment', 'Mining sites', 'Radiation hazards', along with Boolean Operators [12] were used in the search. Duplicates (271 in number) were removed. Articles published in any other language apart from English (34 in number) were removed. The remaining articles (720 in number) were screened and reduced to 29; based on alignment of their titles and abstracts with the scope of the study. A further screening was done for the 29 articles with critical consideration of their methodology and the various radiological parameters such that only the articles that used the same or closely similar methodology in sampling and measurement of the radiological parameters of ^{232}Th, ^{40}K, and ^{238}U or ^{226}Ra with respect to Activity Concentration, Absorbed Dose Rate (ADR), Annual Effective Dose (AED), Radium Equivalent Activity (Ra_{Eq}) and Internal and External Radiation Exposure (H_{in} and H_{ex}) were considered for selection (19 in total). These articles were further screened to remove articles (10 in number) which were published 12 or more years ago. The remaining 9 articles were found to have satisfied all the given criteria, and were finally selected for this study. These 9 studies have been labelled in this article as R1 [13], R2 [14], R3 [15], R4 [16] R5 [17] R6 [18] R7 [19] R8 [20] R9 [21].

Figure 2.1 shows a scheme that summarizes the selection of these nine (9) studies.

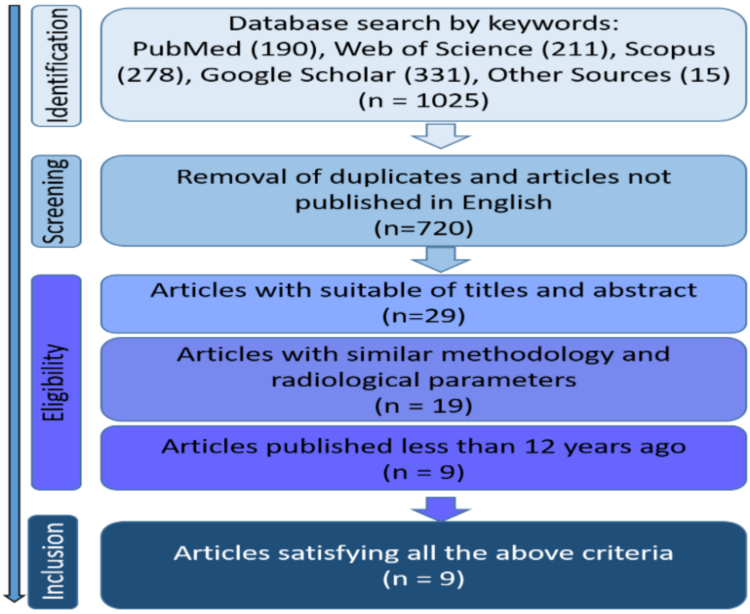

Figure 2.1: Article Selection Scheme

Sample Collection and Preparation

Soil samples for each study were randomly collected within an average radius of 2 km from a gold mine site at a depth averaging 8 to 12 cm. Samples collected in each of the studies were oven dried at an average of 110°C to remove the moisture content and ground to ensure homogeneity. The dried samples were sealed in thick walled water and air tight plastic containers and kept for 28 days to achieve radioactive equilibrium between ^{226}Ra and ^{232}Th and their daughter radionuclides [21].

Instrument Calibration

Whiles the technique used for the radiological analysis in each study was gamma spectrometry, the instrument used in each of the studies was gamma spectrometer with a NaI(Tl) detector. NaI(Tl) detector was calibrated, and the measured spectrum was decomposed into its components using three standard reference materials: RGK-1, RGTH-1 and RGU-1 or RGRa-1; for potassium, thorium and uranium or radium respectively [22]. Energy calibration of the spectrometer was done using the gamma lines at 352 keV (^{214}Pb), 1460 keV (^{40}K), 609 keV (^{214}Bi) and 2615 keV (^{208}Tl). Distilled water was measured in the same geometry as samples to get background radiation. Data of the background spectra were subtracted from the counts of the samples before further analysis. For the sediment samples, the average data acquisition time was approximately 36,000 seconds.

Sample Analysis

Activity Concentration

The determination of the radionuclide activity concentrations of the samples in each study were done with respect to the net area under the photo peaks. Equation 2.1 represents the analytical equation used to determine the particular radionuclide activity concentrations in Bqkg^{-1} [23].

$$A = \frac{N_c}{\gamma_p \cdot T \cdot \eta(E) \cdot m_s} \qquad (2.1)$$

Where A is the activity concentration of the radionuclide in the sample in Bqkg^{-1}, m_s is the mass of the sample in kg, η(E) is the detector efficiency at the specific γ-ray energy, T is the counting time, γ_p is the absolute transition probability of the specific γ-ray, and N_c is the net count rate under the corresponding peak.

Absorbed Dose Rate

The studies sought to determine the mean dose rates absorbed in air from gamma emission. The calculations were based only on ^{232}Th, ^{40}K and ^{235}U or ^{226}Ra; neglecting the concentrations of other elements of NORMs; such as ^{90}Sr, ^{137}Cs and the decay series of ^{235}U, due to insignificant concentration contributions to the whole environmental background doses [24]. As established by the Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [25] Absorbed Dose Rates were calculated using equations 2.2 or 2.3:

$$D = (0.427C_{Ra} + 0.662C_{Th} + 0.043C_{K})\ nGyh^{-1} \quad (2.2)$$

$$D = (0.427C_{U} + 0.662C_{Th} + 0.043C_{K})\ nGyh^{-1} \quad (2.3)$$

Where D is the dose rate ($nGyh^{-1}$) at 1 m above the ground due to ^{232}Th, ^{40}K and ^{226}Ra or ^{235}U in the soil samples, A_K, A_{Th} and A_{Ra} are the activity levels of ^{40}K, ^{232}Th and ^{226}Ra or ^{235}U in $Bqkg^{-1}$, respectively.

The dose conversion coefficient (0.7 Sv Gy^{-1}) was used in the calculation of the annual effective dose due to radiation emission from NORMs. Also, the occupancy of 0.2 for outdoor (an average of 4.8 hours spent in the area every day for a year) as suggested by [24] was used. The effective dose rate was calculated using equation 2.4 [26]. This parameter indicates the amount of radiation dose uptake in living tissues of humans.

$$E_{eff} = T \cdot O_f \cdot Q \cdot D \cdot X \quad (2.4)$$

Where Eeff is the effective dose rate in mSv y^{-1}, T is the time in seconds in a year, O_f is the occupancy term which fixes the mean time spent outside in the area, Q is the proportion of the effective and absorbed dose rate in air, X is the factor converting units to the micro from nano scales, and D is the absorbed dose rate in air in $nGyh^{-1}$.

Annual Effective Dose Rate

To determine the Annual Effective Dose (AED), the studies took into account the conversion coefficient, (0.7 $SvGy^{-1}$) from the absorbed dose in air to effective dose and the outdoor occupancy factor (~ 20 %) [27]. The occupancy factors; the average fraction of time spent indoor (T_{in}), and outdoor (T_{out}), in the respective countries where the various studies were conducted were also taken into account together with the world average indoor and outdoor occupancy factors: 0.8 and 0.2, for indoor and outdoor respectively [28]. The effective dose rate in units of $mSvy^{-1}$ were estimated using equation 4.1 and 4.2 [27];

$E_{in}\ (mSvy^{-1})$

$$= D(nGyh^{-1}) \cdot 8760(hy^{-1}) \cdot T_{in} \cdot 0.7(SvGy^{-1}) \cdot 10^{6} \quad (4.1)$$

$E_{out}\ (mSvy^{-1})$

$$= D(nGyh^{-1}) \cdot 8760(hy^{-1}) \cdot T_{out} \cdot 0.7(SvGy^{-1}) \cdot 10^{-6} \quad (4.2)$$

Where; E_{in} and E_{out} are Annual Effective Doses for indoor and outdoor respectively, D ($nGyh^{-1}$) is the absorbed dose rate in air, 8760(h y^{-1}) is the time in hours for one year, 0.7($SvGy^{-1}$) is the conversion factor which converts the absorbed dose rate in the air to effective dose, T_{in} is the indoor occupancy factor and T_{out} is the outdoor occupancy factor [28].

Radium Equivalent Dose

Radium equivalent (Ra_{Eq}) doses were determined in the various studies. Calculations were done to assess the gamma radiation hazards to human associated with the use of soil from mining sites in construction of houses (for example: filling and local brick making). This gives a single index which describes the gamma output from different mixture of ^{232}Th, ^{40}K and ^{238}U or ^{226}Ra, in the samples. Radium equivalent activity (Ra_{Eq}) was determined mathematically in accordance with [29] in equations 2.5 or 2.6.

$$Ra_{eq}\,(Bq\ kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.007A_K \qquad (2.5)$$

$$Ra_{eq}\,(Bq\ kg^{-1}) = A_U + 1.43A_{Th} + 0.007A_K \qquad (2.6)$$

Where A_{Th}, A_K and A_{Ra} or A_U are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. It may be noted that ^{238}U is replaced by the decay product ^{226}Ra, although there may be disequilibrium between ^{238}U and ^{226}Ra.

External Hazard Index

The various studies sought to determine the levels of external gamma-radiation dose from building materials using the External Hazard Index (H_{ex}) parameter. According to [29], this is calculated from equation 2.7 or 2.8.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (2.7)$$

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (2.8)$$

Where H_{ex} is the external hazard index and A_{Ra}, A_{Th} and A_K or A_U are the activity concentrations of ^{232}Th, ^{40}K and ^{226}Ra or ^{235}U respectively.

Internal Hazard Index

Internal exposure of gaseous NORMs; specifically, radon and its short-lived decay products, that may be trapped indoors, were also quantified using the internal hazard index. This was determined mathematically using equations 2.9 or 2.10 [29].

$$H_{in} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (2.9)$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (2.10)$$

Where H_{in} is the Internal Hazard Index and A_{Ra}, A_{Th} and A_K or A_U are the activity concentrations of ^{232}Th, ^{40}K and ^{226}Ra or ^{235}U respectively.

RESULTS AND DISCUSSION

Activity Concentrations of Radionuclides (Bqkg^{-1})

Activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the various soil samples collected from the gold mining areas are presented in Figure 3.1. The concentrations of ^{238}U varied from 11.8 Bqkg^{-1} to 108.6 Bqkg^{-1}, while the highest was recorded in the study R6, with a value of 108.6 Bqkg^{-1}, indicating that the uranium-bearing minerals have been highly disturbed in that area. The lowest value was recorded in study R5, with a concentration of 11.8 Bqkg^{-1}. In the measurements of ^{232}Th, minimum and maximum values recorded ranged between 4.8 Bqkg^{-1} and 110.3 Bqkg^{-1}, with the highest concentration revealed in study R3. Amongst the analyzed nuclides, ^{40}K displayed the highest range of variation, with values ranging from a minimum of 28.79 Bqkg^{-1} to a maximum of 1985 Bqkg^{-1}, both in studies R4 and R5 respectively, reflecting natural heterogeneity in the potassium content of the soils. Consequently, the obtained activity concentrations were compared with the global average values of the Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), which are 35 Bqkg^{-1} for ^{238}U, 30 Bqkg^{-1} for ^{232}Th and 400 Bqkg^{-1} for ^{40}K [29]. Figure 3.1 below gives a graphical illustration of Activity concentration of ^{40}K, ^{232}Th and ^{238}U or ^{236}Ra Radionuclides for the various studies (R1 – R9).

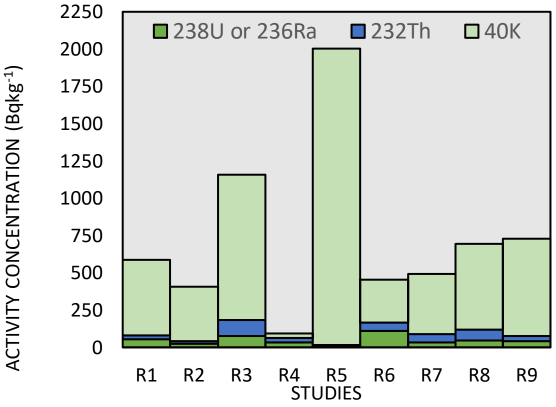

Figure 3.1: Comparison of Activity concentration of ^{40}K, ^{232}Th and ^{238}U or ^{236}Ra Radionuclides

From the obtained results, a couple of studies; R5 and R3, show higher values than the global averages. This may indicate specific geologic factors influencing potassium accumulation within these gold mining areas.

Absorbed Dose Rate (nGyh^{-1})

The absorbed dose rate variation was from 34.73 nGyh^{-1} in study R9 to 146.9 nGyh^{-1} in study R3 (Figure 3.2).

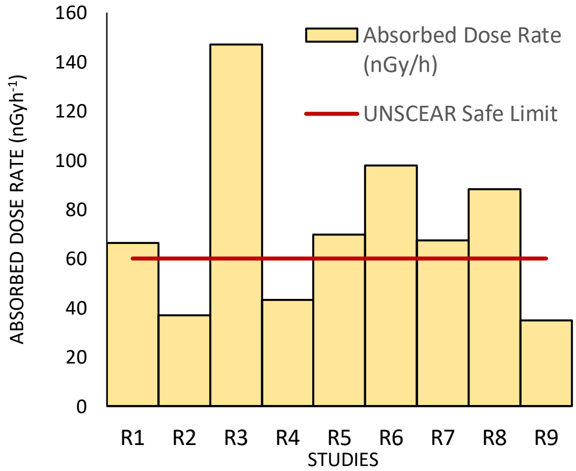

Figure 3.2: Absorbed Dose Rates (nGyh^{-1}) of the collected soil samples from the various studies

The Average Dose Rate for one of the studies; R3, is more than twice the global outdoors average of 59 nGyh^{-1} established by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [29]. This indicates that radiological hazards can be far higher in certain gold mining areas compared to other areas. The studies with high ^{232}Th and ^{238}U concentrations respectively; R3 and R6, contributed more toward the Absorbed Dose Rates. This increases the risk of radiation exposure to miners and local inhabitants. On the contrary, the studies with the lowest contribution to Absorbed Dose Rates: R9, R2 and R4, showed rates below or near the global average. In principle, such areas with relatively lower Absorbed Dose Rates compared with the global average, may present less radiological risk.

Annual Effective Dose Rate

The Annual Effective Dose Rates, as represented in Figure 3.3, ranged from 0.04 mSvy^{-1}; in R7, to 0.81 mSvy^{-1}; in R1. The limit recommended for the public exposure by the guidelines of the International Commission on Radiological Protection (ICRP) is 1 mSvy^{-1} [30].

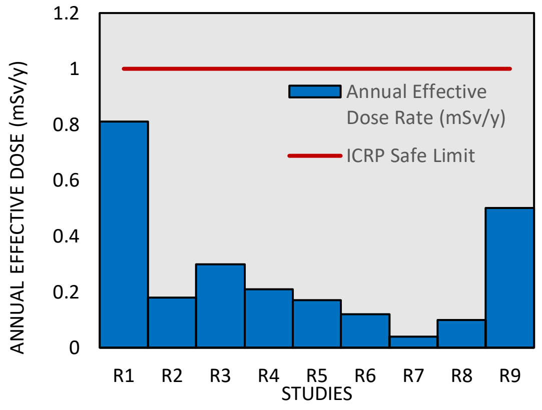

Figure 3.3: Annual Effective Dose Rates ($mSvy^{-1}$) of the collected soil samples from the various studies

Whiles R1, with 0.81 $mSvy^{-1}$, was the closest, the results show that none of the studies exceeded this limit. Although this is considered safe [30], the concern for a long-time radiological exposure risk to humans may be valid. Studies that recorded values of 0.18 $mSvy^{-1}$ for R2, 0.17 $mSvy^{-1}$ for R5, and 0.12 $mSvy^{-1}$ for R6, using this criterion present a minimal risk, although continuous monitoring is recommendable.

Radium Equivalent Activity ($Bqkg^{-1}$)

The radium equivalent activity, Ra_{Eq}, permits a single index to represent the radiation hazards of ^{232}Th, ^{40}K and ^{238}U or ^{236}Ra. The Ra_{Eq} varies from a minimum of 69.73 $Bqkg^{-1}$ for study R9 to 307.2 $Bqkg^{-1}$ for study R3 represented in Figure 3.4 below. The internationally accepted limit of safety for Ra_{Eq} is 370 $Bqkg^{-1}$ [31].

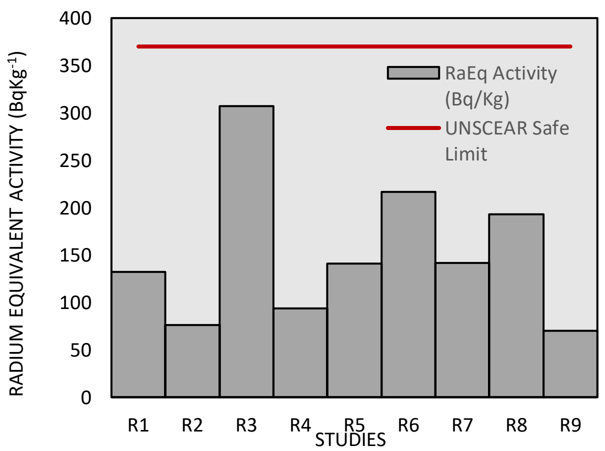

Figure 3.4: Radium Equivalent Activity measurements ($Bqkg^{-1}$) from various studies

All the values obtained for the various studies were less than is 370 $Bqkg^{-1}$, the internationally accepted limit of safety for Ra_{Eq} [31]. Hence, the overall radiation hazard due to these NORMs in the soils are within the acceptable limits of safety. However, the highest Ra_{Eq} value, 216.79 $Bqkg^{-1}$, recorded in studies R3 and R6, is considered a reason for further attention because their value approaches the safety limit, especially in the case of long-term exposure.

Internal and External Hazard Indices

Internal Hazard Index (H_{in}) and External Hazard Index (H_{ex}) is significant for the determination of the radiological health risk to humans whiles indoors and outdoors respectively. Trends in the Internal and the External Hazard Indices are shown in Figure 3.5.

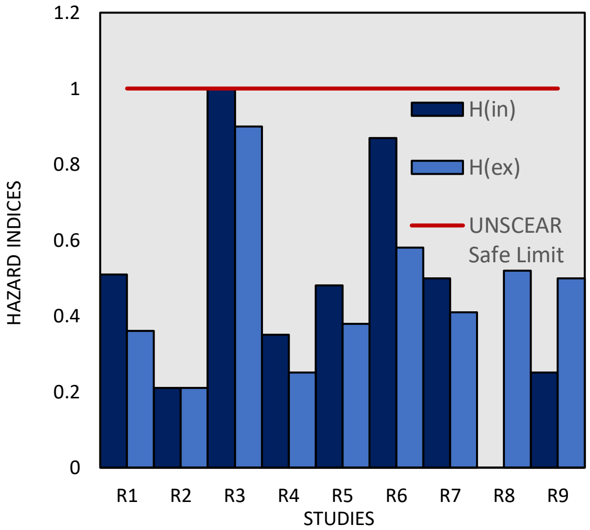

Figure 3.5: Internal and External Hazard Indices from various studies

Considering study R3, the critical H_{in} value of 1 presents a possibility of internal exposure hazard; particularly due to inhalation [32]. Similarly, the H_{in} value of R6; 0.87, approaches 1, and is also considered high even though it does not exceed the safety limit. Localized mitigation strategies should be pursued to reduce the radiological health risk in these gold mining areas where studies R3 and R6 were conducted whiles continuous monitoring is recommended for the other studies.

CONCLUSIONS

Comparing these findings with international standards and guidelines, some of the studies had high levels of radiological exposure. These studies include R3, R6 and R5. Although most of the studies showed that most parameters are within the internationally established safety limits, other studies, such as R3, R6 and R5, recorded critical thresholds of majority of the observed radiological parameters; including Absorbed Dose Rates, Radium Equivalent Activity and Internal and External Hazard Indices, which is indicative of the necessity to continue monitoring. In general, radiological risks across the various studies varied form one study to the other. Considering the specific conditions that may have facilitated deviations from the internationally established safety limits, it is recommended that thorough mitigation strategies be carried out to reduce the risk of exposure of NORMs to humans and the environment as a whole. It also is recommended that research be carried out to ascertain the impact of long-term radiological exposure of NORMs to the workers and residents at or near high risk mining sites. Additionally, control measures to significantly reduce radiological exposure of NORMs in high risk areas are to be put in place to reduce NORM exposure risks.

Conflict of Interest

The authors declare no conflict of interest regarding the publication of this article.

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