Environmental Science and Pollution Research (2020) 27:5387–5397
https://doi.org/10.1007/s11356-019-07222-3

RESEARCH ARTICLE

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Radiological and toxicity risk exposures of oil based mud: health implication on drilling crew in Niger Delta

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Received: 11 September 2019 / Accepted: 29 November 2019 / Published online: 17 December 2019

Abstract

Naturally occurring radioactive materials (NORMs) and the presence of toxic metals in drilling fluids/their additives have raised research interests in recent times owing to the risks associated with the exposure times for drillers of petroleum wells. In this study, two drilling fluids A and B were formulated, while two other Mud Samples C and D were obtained from drilled shale and shale-sand formation zones. All four fluids were collected and analyzed for the presence of radioactive and heavy metals. Lead (Pb), mercury (Hg), cadmium Cd), zinc (Zn), chromium (Cr), aluminum (Al), arsenic (As), nickel (Ni), and copper (Cu) were detected in the mud samples. The heavy metal contents of the mud samples are in the following decreasing order of magnitude Hg > Pb > Cd > Cr. In Samples A–D, Hg, Pb, Cr, and Cd were found to have significant concentrations, and the concentrations of these metals increased in the mud samples after they were used for drilling. The concentration of Hg was above the permissible limit. Also, the concentrations of Pb, Cu, As, and Al found in Mud Samples A and B can cause skin irritations over long-term exposures, while Cd, Hg, Zn, and Ni present in the samples were within levels that can cause lung infections or immune breakdown when ingested over long periods. The quantities of Cd, Hg, and Cu detected in Mud Samples C and D can cause skin irritations over long-term exposures, while those of As, Zn, Ni, and Al were seen to have the potential to cause dermal infections/diseases. Based on the results obtained, the cancer risk for the drilling crew lies within 1.1×10^{-3} - 7.7×10^{-3} HQ. The highest dose rate, radium release, and external hazard index were obtained for Mud Sample C whose radium equivalent was judged to be far below the critical safe limit for the drillers. The radium equivalent activity for the two field mud samples (C and D) were estimated to be 27.467 and 22.978 Bq kg^{-1}, respectively, which is the maximum activity obtained for the analyzed samples. The maximum radium equivalent activity for Mud Sample C was estimated as 27.48 Bq kg^{-1} with a corresponding external hazard index of 0.7. Based on the analysis, there is a significant correlation between the concentration of heavy metals and the radionuclides found in the mud samples.

Keywords NORM activity · Toxicity · Heavy metal · Human health risk assessment · Drilling mud systems

Responsible Editor: Philippe Garrigues

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s11356-019-07222-3) contains supplementary material, which is available to authorized users.

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Introduction

Naturally occurring radionuclides are often present in raw materials and minerals encountered during drilling. According to Vaasma et al. (2019), member states of the European Union (EU) have been obliged to capture dangers arising from natural radiation sources in their national legislatures. In order to fully capture these sources, more research works involving materials' processing are encouraged in order to possibly control and contain these naturally occurring radionuclides. Though the concerns related to naturally occurring radioactive materials (NORMs) have been deliberated among the majority of the European Union Member States, some of the member states are still seeking an extended time for the full implementation of some standards. However, there has not been any detailed legislature and policy for Africa with Nigeria as a case study, because of the need to establish optimal limits for these NORMs. Over the past five decades, Nigeria has been involved in the extraction of hydrocarbons from the subsurface. However, till date, there is no awareness on the risks associated with exposures to NORM. Taheri et al. (2019) identified the presence of NORMs on the Earth's crust during the extraction of hydrocarbons from the subsurface.

During drilling operations, formation cuttings are transported from the subsurface to the surface through drilling fluids, thus creating an avenue for the transfer of NORMs from the cuttings to the carrying medium (drilling fluid). Smith (1992) recorded that these radioactive materials can be transferred, precipitated, as well as accumulated on the circulating system during drilling. Khodashenas et al. (2012) reported that short-/long-term exposures of these radionuclides may cause possible increase in the risks of cancer and kidney diseases to oil workers. Therefore, drilling crews, mud loggers, and mud engineers are likely to be exposed to high concentrations of hazardous radioactive materials because of their daily contacts with these mud systems. Several geological surveys of these oil and gas wells may have different concentrations of natural radionuclides. Thus, it is important to investigate their possibilities and establish regulations in the areas of health, safety, and environment in order to reduce the employee and environmental risks to a bearable minimum.

Oil-based drilling fluids can be recycled after drilling using a de-sander, vibrating screen and de-silter before being reused in another field. Literature has shown that heavy metal concentration in oil-based mud can be harmful to the environment and drilling crews. Okoro et al. (2016) in their study identified the presence of heavy metals in the Earth's pit and highlighted some environmental concerns that may be associated with drilling operations. Kinigoma (2001) also reported the environmental impacts of drilling fluid additives. Pb, Cd, and Ni are some of the heavy metals that have adverse effects on humans, even at trace levels. Some of the additives used in drilling fluids contain heavy metals (Xu et al. 2018). There are lots of contaminants in drilling fluid systems, and Pozebon et al. (2005) reported in their study that barite ($BaSO_4$) used as weighting agent during the formulation of drilling muds is part of the main sources of heavy metals released while drilling a petroleum well. Other metals such as nickel, iron, cadmium, chromium, mercury, vanadium, arsenic, and barium have also been identified (Hedayatipour et al. 2017; Gonzalez-Munoz et al. 2006; Hu et al. 2012; Stynen et al. 2019; Chen et al. 2016).

Literature has shown that heart diseases, inflammations, DNA disorders, and lung diseases can be taken into the body via ingestion, dermal contact, and inhalation of some heavy metals (Xu et al. 2016; Gillis et al. 2012; Chen et al. 2016). Because of the nature of operations in upstream drilling, crew members often work and live at the production platforms, which increases their possibilities of ingesting and inhaling these heavy metals. Also, the oil-based drilling mud system which is often reused can possibly have some NORMs transferred to it from the formation cuttings that are being transported to the surface. Therefore, this study investigates the heavy metal pollution and NORM levels in oil-based drilling fluids after coming in contact with the subsurface formation cuttings. It also quantifies the health risks that result from the exposure of these substances to drillers, mud loggers, and mud engineers. Also, the compounds present in the mud samples were analyzed and identified using XRD analyzer.

Geographical location of the study area

The location of the study area

The study area is Field X, which is located in Bayelsa State, Nigeria. A lowland state characterized by coastal beaches, beach ridge barriers, tidal flats, and flood plains. The height decreases downstream. It is bound by latitudes 4° 44′ 59.99″ and longitudes N 6° 04′ 60.00″ E; the town where the oil and gas field is located is confidential and protected by Nigerian Law. Literature results necessitated this study, and there is a need to identify the possible concentration of naturally occurring radionuclides in the drill cuttings that personnel are exposed to in the region during drilling operations (Davies et al. 2013). The map of the study area and its environment is attached as supplementary file.

Climate condition of Bayelsa

The climate of Bayelsa State can be classified as tropical, and the location is classified under the Köppen and Geiger standard. The state experiences three weather conditions which are the humid/rainy, warm, and dry seasons. The state climate can be seen as an equatorial type toward the southern parts, while the northern parts experience tropical rain. These parts experience heavy downpours every month, whereas the downpours are lower in parts of the south down the north. The temperature is said to be uniform in the state, and it is in the range of 26 °C–31 °C (Okiongbo and Ogobiri 2011).

Geology of the area

The geology of Niger Delta covers about 256,000 km. Initially, it was the older transgressive Paleocene prodelta which was built and owned by Delta Construction Company; it proceeds in discreet mini basins (Adegoke, 2016). These mini basins exhibit several tectonic configurations ranging from extensional and translational to the compressional toe-thrust region. The Niger Delta outcropping units comprise of the Imo Formation and the Ameki Group (Adebiyi 2015).

Methods

Sample collection and preparation

Drilling fluid is one of the major drilling materials that drillers cannot avoid being exposed to. This fluid interacts with different soil and rock formations of reasonable depths of about 12 to 20 km below ground level. As such, the water-rock interaction, mud-water-rock interactions, and mud filtrate formation interactions require urgent assessments for adequate quantification of human risk exposure levels. In this study, two major types of drilling fluids were used, and they are: two samples formulated in the mud laboratory before they were used in the oil and gas field (Samples A and B). Two other mud samples (samples C and D) were obtained from shale and shale-sand formation zones, respectively. A total of four (4) mud samples were collected and analyzed for this study. Two (2) liters of each mud sample was collected and transported to the lab for analysis. The drilling crew for this study is classified under the adult weight and age as stipulated in the US EPA (2001), with an average body weight of 70 kg and age range of 21–70 years. According to the EPA Exposure Factors Handbook (1997), "Exhibit 6" illustrates and estimates exposure values that can be used when the entire body is exposed, and this was adopted in this study.

Radio-elemental analysis by gamma spectroscopy

The mud samples were kept under ambient temperature of about 28 °C and sieved. The recovered solutes were oven-dried at 105 °C. The samples were also pulverized and passed through a sieve mesh of 250 μm size so as to ensure homogeneity in particle size. The homogenized samples in powder form were put in high-density polyethylene bottles which were well labeled and sent to the Radiation Centre Laboratory, Ibadan, Nigeria. The samples were put in Marinelli beakers and sealed for 4 weeks in order to allow for secular equilibrium and, so that the daughter nuclide will be equivalent to the parent's. The samples were ready for gamma counts within 21,600 s. All conversions and concentrations of the measured radionuclides as well as their activity concentrations were determined according to the procedure discussed by Omeje et al. (2013a, b) and IAEA (1989).

Total heavy metal content test

A 2-mm mesh sieve was used to sieve the dry solute until a constant weight was obtained; this helped to eliminate particles of larger sizes from the process. About 0.5 g of the sieved samples was measured after homogenization, and the particles were ground with a laboratory pestle and mortar. The processed samples were later sieved again through a 250-μm mesh and transferred into a 250-ml beaker. Ten ml of 1:1 sample to nitric acid was measured into the beaker and covered with a glassware. The beaker was placed in a water bath at 99 °C and allowed for 1 h. The samples were further processed and then analyzed for lead, cadmium, copper, iron, manganese, and nickel using atomic absorption spectrophotometer (AAS).

Determination of radioactivity and radiological parameters on drilling crews

The dose rate of the NORM radionuclides was calculated using the formula for estimating external gamma dose rate (D_c), (Omeje et al. 2015) as shown in Eq. (1).

$$D_c = 0.462\,A\,(U{-}238) + 0.604\,A(Th{-}232) + 0.0417\,A(K{-}40) \quad (1)$$

where D_c = absorbed dose rate at 1 m above the ground surface, $A(^{238}U)$, $A(^{40}K)$, and $A(^{232}Th)$ = NORM radionuclides concentrations in Bq kg^{-1} for mud system sediments.

The radium equivalent activity for the sediments was calculated according to Eq. (2),

$$A_{Ra} = 1.43A_{Th} + 0.077A_K \quad (2)$$

The gamma ray radiation hazards due to specific radionuclides were determined using Eq. (3) for external radiation.

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4,810 \leq 1 \quad (3)$$

where, A_{Ra}~A_U, A_K, and A_{Th} = NORM radionuclides average concentrations in Bq kg^{-1}. The annual effective dose rate (AEDR) was calculated in Sv year^{-1}. Eqs. (1–4) were adopted from United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR (2000).

Heavy metal health risk assessment for drilling crew

The model that was used to assess the health risks posed by the drilling mud systems containing heavy metals was derived from US EPA (2001, 2011) and Xu et al. (2018). The exposure to heavy metals in the mud systems might take place in two possible ways. These are "ingestion and dermal contact" which are largely due to the nature of the drilling operation. The exposure dose of the heavy metals identified in the mud samples/sediments were calculated using Eqs. (4) and (5).

Ingestion exposure dose for the sediments is given as:

$$D = \frac{C \times IR \times EF \times CF}{BW} \quad (4)$$

Dermal contact dose for the sediments is given as:

$$D = \frac{C \times A \times AF \times EF \times CF}{BW} \quad (5)$$

where D = dose exposure (mg/kg/day), C = concentration of the contaminant (mg/kg), IR = contaminated sediments rate of intake (mg/day), EF = exposure factor (which is dimensionless), CF = conversion factor (10^{-6} kg/mg), BW = body weight (an average of 70 kg was used for this study), A is the total soil mass (326 mg was used for this study), and AF is the bioavailability factor; a value of 0.1 was adopted from literature (USEPA 2001; Xu et al. 2018). Bioavailability factor is the amount of a substance that is absorbed into a person's body. It is the percentage of the total amount of substances ingested, inhaled, or contacted which actually enter the bloodstream and can potentially harm a person. For screening purposes, the bioavailability factor was assumed to be 0.1.

Results and discussion

Heavy metal concentration in the drilling mud systems sediments

A total of nine (9) heavy metals were analyzed in the four (4) drilling fluid systems used in this study. These heavy metals were identified from literature to be significant environmental pollutants with significant toxicity impacts. Table 1 shows that lead (Pb), chromium (Cr), cadmium Cd), arsenic (As), aluminum (Al), mercury (Hg), nickel (Ni), zinc (Zn), and copper (Cu) are present in the additives used in the formulation of the drilling fluids (Samples A and B). Also, some naturally occurring elements were found in Samples C and D to exhibit increased concentrations after the drilling fluids were used in drilling petroleum wells. These results are similar to the observations made by Arif et al. (2015). The heavy metal contents in the mud systems are as follows Hg > Pb > Cd > Cr, in decreasing order of magnitude. Table 1 indicates the concentration levels of the aforementioned metals in the four (4) mud samples under consideration. The toxic-metal concentration difference in Samples A and B when compared to Samples C and D show that the naturally occurring heavy metals present in the formation cuttings were transferred into the drilling mud system during the transportation of formation cuttings from the subsurface to the surface circulation system. Hg, Pb, Cr, and Cd have a significant concentration, and their concentrations increased after the mud systems were used for drilling.

The presence of Pb in the drilling mud samples can be considered as a systemic toxicant because it does not degrade and can be present in the mud sample for thousands of years. The concentration of Hg is above the reference value and should be a concern because of its hazardous nature. The heavy metal concentrations in Table 1 are above the maximum permissible addition (MPA) according to the data of Dutch ecologists (Vodyanitskii 2016). The

Table 1 Heavy metal content in the drilling mud systems

Mud	Heavy metal content, mg/kg								
	Pb	Cd	Hg	Cu	As	Cr	Zn	Ni	Al
A	78.2	3.2	5.7	13.8	6.7	21.6	68.8	86.3	95.7
B	78.2	3.2	5.7	13.8	6.7	21.6	68.8	86.3	95.7
C	96.3	7.6	8.8	17.4	11.3	47.8	137.2	112.5	128.6
D	89.4	6.1	7.4	15.7	8.6	36.7	121.8	98.6	113.8
EU guidelines	300	3	NA	140	NA	150	300	75	NA
WHO guidelines	100	3	NA	100	20	100	200	50	NA
GOST	55	0.76	1.9	3.5	4.5	3.8	1.6	NA	NA

**NA* not available

concentration of Hg for the lab and field used mud samples were all above the reference concentration. The concentration of Pb in the mud samples relative to the reference dose were very close, which was followed by Cd. This then implies that the impact of these three heavy metals will be more significant on the drilling crew and the immediate environment.

Table 1 shows that Cd and Ni values were found to be highest in the present study when compared to the European Union (EU) and World Health Organization (WHO) standards. The measured heavy metal values from the present study were all above the available Russian standard (GOST 17.4.102–83 1983).

Health risk assessment of heavy metals for drilling crew

Ingestion and dermal contact implications were considered in this study. The level of risk exposure for the drilling crew was tabulated in Table 2. The heavy metal risk exposure for the drilling crew considered in this study constitutes both noncarcinogenic and carcinogenic elements. Five of the identified heavy metals in this study are As, Cd, Ni, Al, Hg, Pb, and Zn which are classified as highly hazardous elements if found above the permissible level according to Russian sanitary hygienic standard (GOST 17.4.102–83).

Ingestion can ensue through the unintentional consumption of mud system sediments by the hands, since the crew often live and work within the oil and gas field. The dermal contact of crew members with the sediments depends on the chemicals involved, duration of contact, area of contact, and the ability of the sediments to penetrate the skin. Figures 1a, b show the ingestion exposure and dermal adsorption values of the heavy metals.

The plot shows the level of ingestion and dermal adsorption on the vertical axis and the mud systems on the horizontal axis. Figure 1a shows that Zn, Al, Ni, Pb, Cr, Cu, Hg, and Cd will pose health risks to the drilling crew for Mud Samples C and D after drilling, and this results from ingestion exposure. However, Mud Sample C had the highest risk factors for ingestion and dermal adsorption when compared with other mud samples. Figure 1b shows that Zn, Al, Ni, Pb, Cr, Cu, Hg, and Cd contained in Mud Samples C and D will also pose health risk to the drilling crew owing to the increased tendencies for dermal contact.

The ingestion and dermal adsorption/exposures of Zn, Al, Ni, Pb, Cr, Cu, Hg, and Cd can cause health risks, because the dose increased after the Mud Samples A and B were used for drilling operation (Samples C and D). Figure 1 shows the ingestion and dermal effects of these mud samples on the crew after oral or ingestion, with emphasis on four (4) most pronounced elements: Pb, Ni, Zn, and Al. Because the concentrations of Hg, Cd, As, Cu, and Cr were found distinctly lower than 0.00005 mg/kg/day exposure to the crew. The highest values for both Ni and Al were found to be 0.0002 mg/kg/day that each person can consume onsite through these two pathways. The ingestion rates of Zn, Al, Ni, and Pb are more significant than other heavy metals considered in this study. The potential for dermal contact was significant for Al, Zn, Ni, and Pb, and their doses were higher for the mud samples already used for drilling. Table 2 presents the dose intake and dermal contact potential of heavy metal exposure to the drillers. The ingestion and dermal hazards due to Pb in mud C is higher with a value of 1.321×10^{-4} mg/kg/day, whereas Hg and as noted in mud C were lower.

Table 2 Dose intake and dermal level of heavy metal health risk assessment

Mud	Heavy metal content health risk, 10^{-4} mg/kg/day									
	Pb		Cd		Hg		Cu		As	
	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact
A	1.072	0.350	0.044	0.014	0.078	0.025	0.189	0.062	0.092	0.030
B	1.072	0.350	0.044	0.014	0.078	0.025	0.189	0.062	0.092	0.030
C	1.321	0.431	0.104	0.034	0.121	0.039	0.239	0.078	0.155	0.051
D	1.226	0.400	0.084	0.027	0.101	0.033	0.215	0.070	0.118	0.038
	Cr		Zn		Ni		Al			
	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact	Ingestion exposure	Dermal contact	Ingestion exposure		Dermal contact	
A	0.296	0.097	0.944	0.308	1.184	0.386	1.312		0.428	
B	0.296	0.097	0.944	0.308	1.184	0.386	1.312		0.428	
C	0.656	0.214	1.882	0.613	1.543	0.503	1.764		0.575	
D	0.503	0.503	1.670	0.545	1.352	0.441	1.561		0.509	

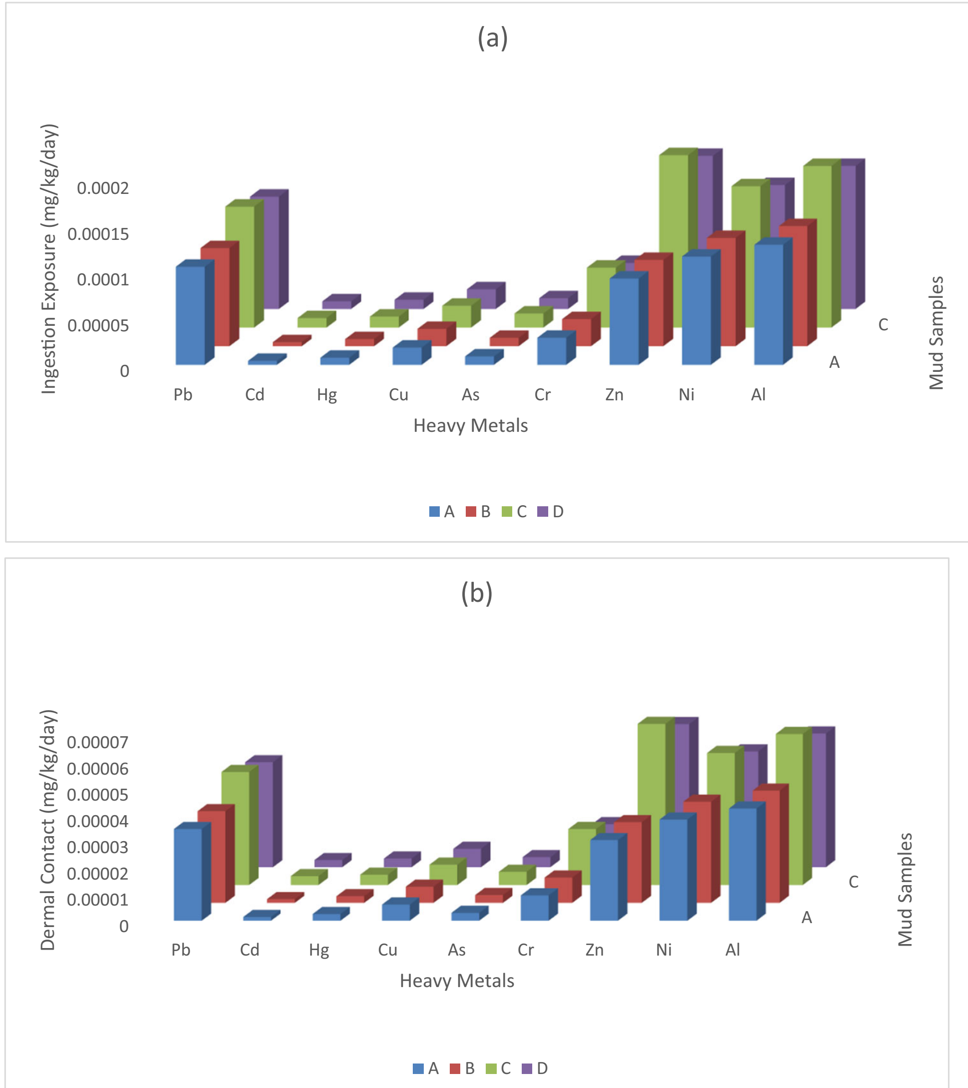

Fig. 1 **a** Ingestion and **b** dermal contact dose for the heavy metals present in the mud samples

Noncarcinogenic risk assessment of heavy metals for the drilling crew

Possible health effects of exposures to carcinogenic and noncarcinogenic heavy metals were examined. This study evaluated exposure levels by measuring the average daily intake (ADI) of heavy metals identified through dermal/skin contact and ingestion by drilling crew. Figure 2 shows the noncancerous health risk (HQ) of the drilling crew working in close contact with these mud systems. The HQ expresses

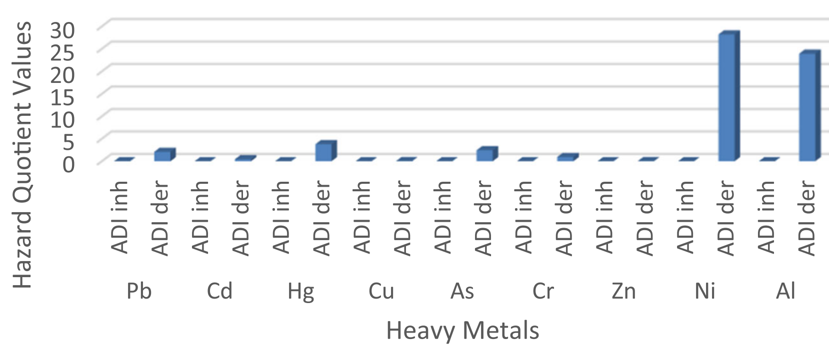

Fig. 2 Hazard quotient (HQ) values for heavy metals for the mud system samples

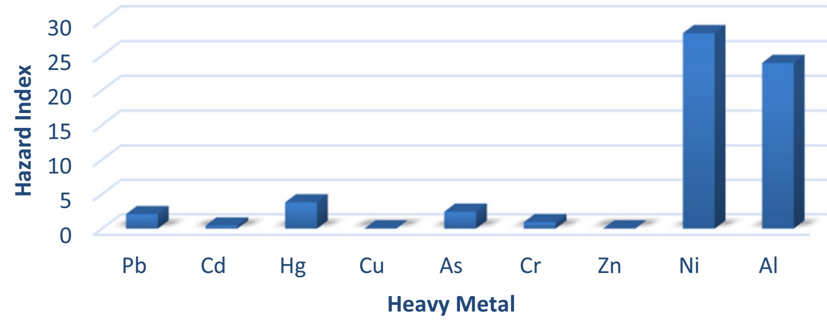

Fig. 3 Hazard indices (HI) for heavy metals for the mud system samples

the probability of the drilling crew suffering an adverse effect (Kamunda et al. 2016).

The HQ values of the nine (9) heavy metals and the ADI of heavy metals ingested from the mud sediments are less than 1. Furthermore, there is no obvious risk posed by these metals to the drilling crew. But, the average daily dermal exposure doses of Metals such as Ni, Al, Hg, As, and Pb values are greater than 1 and will be an obvious risk to the crew. The high concentrations indicated that these five (5) heavy metals will not pose any cancer risk to the drilling crews. However, their hazard indices (HI) are also greater than 1 which is indicative of other health consequences for the five (5) heavy metals (Fig. 3) thereby causing other health-related issues to the crew.

Carcinogenic risk assessment of heavy metals for the drilling crew

The lifelong risk of cancer for drillers was calculated separately from the average contribution of individual heavy metals in the mud samples for all the two pathways. Carcinogenic risk was calculated based on Pb, Cd, Hg, Cu, As, Cr, and Ni. Figure 7 shows that Ni has the highest risk tendency followed by Cd, Cr, and As, which is most conducive for the development of cancer. The US Environmental Protection Agency considers acceptable cancer risks in the range of 1×10^{6} to 1×10^{4} as acceptable. The risk of a crew member having stroke ranges from 1.1×10^{-3} to 7.7×10^{-3} (Fig. 4). Dermal/skin contact is a major cause of an excessive risk of developing cancer throughout the lifetime exposure.

Activity concentrations of ^{232}Th, ^{238}U, and ^{40}K in the mud system samples

The results of the analysis for activity concentrations of ^{40}K nuclides from ^{232}Th series (^{208}TI, ^{228}Ac) and ^{238}U series (^{214}Pb, ^{214} Bi) in the drilling mud samples are tabulated in Table 3. The average radionuclides concentration in the mud samples that were not used in drilling (Samples A and B) was 0.72 Bq kg^{-1} for ^{238}U, 7.20 Bq kg^{-1} for ^{232}Th, and 89.94 Bq kg^{-1} for ^{40}K radionuclides. The average radionuclide concentration in the mud samples (Samples C and D) used in drilling ranges between 0.80 and 0.98 Bq kg^{-1} for the ^{238}U, 8.10 and 10.30 Bq kg^{-1} for ^{232}Th, and 137.60 and 152.71 Bq kg^{-1} for ^{40}K series radionuclides. The ^{40}K series radionuclide was highest for the four mud systems under consideration (Fig. 5).

The highest radionuclide concentrations in the mud samples were found in Mud Sample C with values for uranium, thorium, and potassium noted in the samples as 0.98 Bq kg^{-1}, 10.30 Bq kg^{-1}, and 152.71 Bq kg^{-1}, respectively. The values are below the clearance and exemption levels as discussed by Xu et al. (2018), but the concern remains the radiological burden that these mud systems may impose on the health of

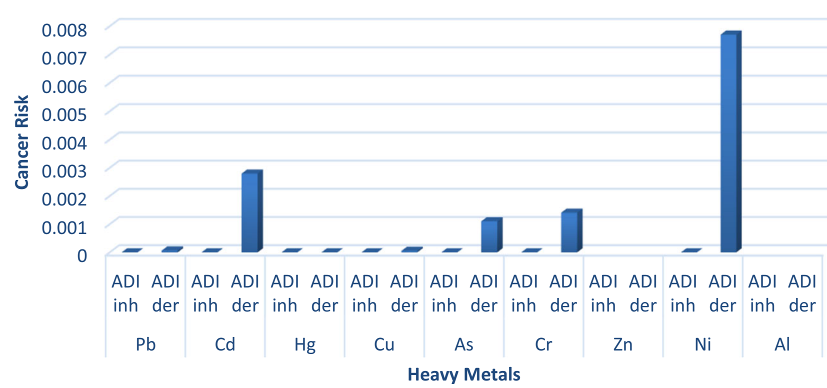

Fig. 4 Cancer risk values of heavy metals on the mud samples

Table 3 Radioactivity level in the drilling mud samples sediments

Sample	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
A	0.72	7.2	86.944
B	0.72	7.2	86.944
C	0.98	10.3	152.706
D	0.80	8.1	137.595

the drilling crew due to their long operation periods of about 25 years.

Health risk assessment of NORM radioactivity for drilling crew

It is possible to estimate the human health impacts caused by exposures to NORMs, and in this study, the dose rate values obtained from the health risk calculations are tabulated in Table 4. The health consequences of the mud samples were based on the radiological risks calculated. The effect of greater intensity is considered at the point where the three indicators considered for radiation risk measurements exceed standard limits. A general description of the results shows that the drilling fluid sample used for the drilling operation shows a high radioactivity for all NORM radionuclides. However, ^{238}U had the lowest activity among all samples, and ^{40}K had the highest activity among the drilling fluid samples shown in Table 4. The radium equivalent activity for the two field mud samples (C and D) reported the activity level of 27.467 and 22.978 Bq kg^{-1}, respectively, which indicates maximum activity for the analyzed samples (Table 4). The samples have an external risk index of less than 1.

Thus, one can deduce that naturally occurring radioactive substances in formation cuttings (Taheri et al. 2019) can be transferred to the drilling mud systems as it transports the cuttings to the surface during drilling operations. These recycled/ reused oil-based mud systems should be properly evaluated before redeployment to the next drilling site in order to reduce the possible health risks. The possible health risk is driven by long-term emissions from the mud system of radionuclides which in turn affects the crew working with the mud at the drilling site.

The toxic metal levels that these radionuclides can emit from each of the mud system samples were considered in this study. The calculated external radiation hazard, annual effective dose rate, radium equivalent, and dose rate of the Mud Sample C were higher than that of other samples. The highest calculated dose rate was 6.72 n Gy h^{-1} which can be credited to the type of subsurface formation geology (Omeje et al. 2013a, b). The lab mud samples (A and B) had the lowest dose rate because both samples were not used for drilling operations and have not had any contact with the subsurface formations. Mud Sample C also recorded the highest radium equivalent activity of 27.48 Bq kg^{-1} and highest external hazard index of 0.7. This radium equivalent value is far below the 370 Bq kg^{-1} level that may pose health risks.

The relationship between the heavy metal content and radionuclides were analyzed by determining their correlation coefficients. This is a measure of the strength of a linear association between the heavy metals and the radionuclides. Figures 6 and 7 show the impact and correlation coefficient for the heavy metal concentration and radionuclides, respectively. The ingestion rate of the heavy metals was compared with the annual effective dose rate of the radionuclides, while the external hazard index of the radionuclides was compared with the dermal exposure of the drilling crew to the heavy metals in the mud samples. This analysis gave a 0.91 correlation coefficient which indicate a strong association of the heavy metals and the radionuclides in the mud samples.

Even in cases where regulatory problems are absent, since the values of emissions and releases are not exceeded, calculating the cumulative dose with which the most affected drilling crew members are exposed can help to get a more quantitative picture of the impact on the population.

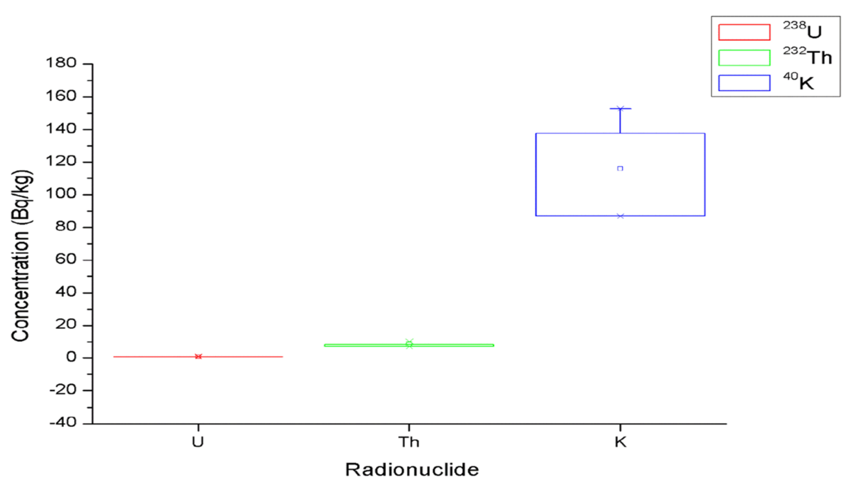

Fig. 5 Radioactivity level in the drilling mud sample sediments

Table 4 Radium equivalent, external hazard index, internal hazard index, dose rate, and annual effective dose for the mud samples sediments

Mud sample	Radium equivalent activity (Bq kg^{-1})	External hazard index	Dose rate (nGyh^{-1})	AEDR (mSvy^{-1})
A	17.710	0.0478	4.723	5.792
B	17.710	0.0478	4.723	5.792
C	27.467	0.0742	6.716	8.236
D	22.978	0.0620	5.304	6.504
Reference	127	0.35	60	74

Conclusion

The results from gamma spectroscopy show that specific activities due to K-40 made a greater contribution to the total radionuclide activity. In this study, the concentrations of U-238 and Th-232 in the mud were low, and the concentrations of K-40 were high. In this context, it was found that the equivalent radioactivity of the radium emission and the external risk

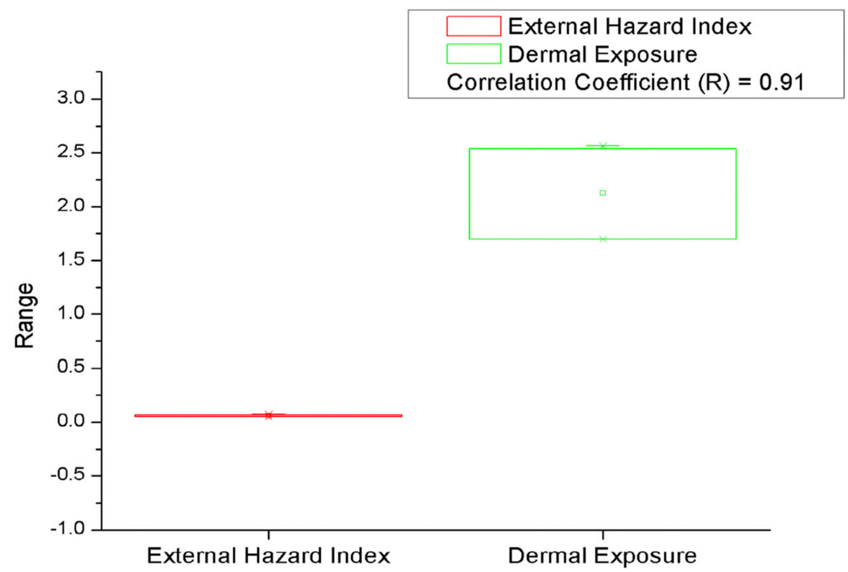

Fig. 6 Impact of heavy metals and the radionuclides exposure with the mud samples

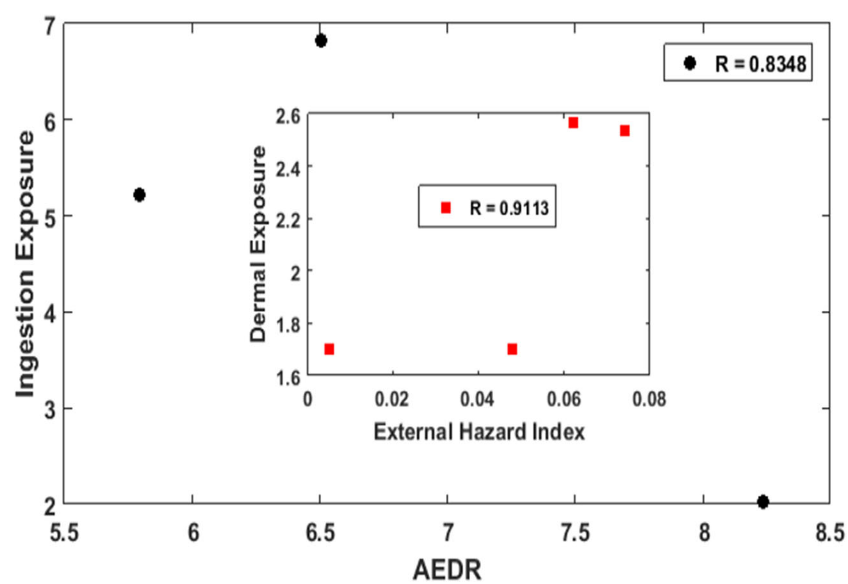

Fig. 7 Correlation coefficient of the heavy metals and the radionuclides in the mud samples

index in the mud sediments were below safe limits. The toxic effects of heavy metals and radioactive materials when exposed to levels obtained in this study can be fatal and cannot be ignored. The trend observed from the results is that the mud sample metal concentration of heavy metals and NORMs increase when being used for drilling. This is a concern because the more exposed these mud samples are to the formation cuttings with these NORM radionuclides, the more the possibility of transfer of these NORM radionuclides to humans.

The presence of trace amounts of radioactive elements in the mud sediments of the study area indicates the need to take adequate precautions in order to avoid prolonged exposure to these metals. The average specific activity of K-40, Th-232, and Ra-226 in the mud deposits, their corresponding equivalent radioactivity, as well as the external risk indicators were below the maximum permissible limits (370 Bq kg^{-1} and 1). It is known that the absorbed dose rate due to these natural nuclides is lower than the world's average standards. Thus, it is necessary to consider the possibility of the existence of NORMs during treatment of drill cuttings that these mud systems transport from the subsurface to the surface. Also, mud engineers should be conscious of the possibility of being exposed to radionuclides when reusing these oil-based mud systems.

Acknowledgements The authors appreciate Covenant University Centre for Research, Innovation and Discovery Ota, Nigeria for their support in making the publication of this research possible.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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