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Radiotoxicity risk assessments of ceramic tiles used in Nigeria: The Monte Carlo approach

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ABSTRACT

This work reported the activity concentrations of ^{40}K, ^{238}U, ^{232}Th, radiological impact assessment, the concentration of Cd, Cr, Zn, Cu, Pb, As, Mn, Ni, Co, and human risk assessment of the potentially toxic elements (PTEs) of commonly used tiles in Nigeria. The gamma-ray spectrometry was carried out by means of High Purity Germanium gamma detector, and the whole content of the PTEs in the sampled tiles were analyzed using ICP-MS instrument. The gamma-ray analysis reveals varying results that are higher than their corresponding global values in most cases. Similarly, the analysis of the PTEs reveals concentrations that are in some cases 100 times higher than the recommended limits. Surprisingly, for gamma dose rates, all the tiles (100%) have values greater than the recommended limit of 84.00 nGy/h provided by UNSCEAR. The radiological impact assessment reveals that Nigerians are at high risk of overexposure to indoor ionizing radiation for using these tiles for their building and construction purposes. The P 95% and mean cumulative probabilities from the Monte Carlo Simulation (MCS) indicates that the lifetime cancer risks for most of the sampled tiles exceed the recommended limit of 3.75 $\times$ 10^{-3} by UNSCEA i.e. the P 95% ranges between 5.00 $\times$ 10^{-3} and 20.60 $\times$ 10^{-3}, and the mean cumulative probabilities ranges between 2.53 $\times$ 10^{-3} and 11.90 $\times$ 10^{-3}. This high risk was confirmed by the MCS, which reveals that inhabitants using NISPRO verified tiles, Goodwill Verified, Virony Glazed, IDDRIS tiles, PNT Verified, and PNT Ceramic tiles are most likely to experience over-exposure to indoor ionizing radiation because even their lowest probable exposure risk, P 5% exceeds the limit recommended by UNSCEAR for indoor exposure. However, the Hazard Index (HI) and the Incremental Lifetime Cancer hazard (ILCR) of the PTEs reveals low cancer and non-cancer risks for all the tiles investigated.

1. Introduction

Building and construction materials such as tiles, granites, quartz, marble resulting from mineral rocks generally contain varying quantities of contaminants such as heavy metals (PTEs) and naturally occurring radionuclides like *^{238}U, ^{232}Th,* their products, and the non-series *^{40}K* (Omeje et al., 2018; Joel et al., 2018a,b,c; Orosun et al., 2020a; Orosun et al., 2020b, Orosun et al., 2021a). These toxic metals and the primordial radionuclides are typically inherited from the mother rock (such as granitic rocks) during the pedogenic process (Orosun et al., 2020a) . Since the presence of these toxic elements and the naturally occurring radionuclides in the building and construction materials, which contributes to biotoxic effects and radiation exposure, information about the source, quantity, and assessment of the associated health risks becomes very important in assessing the likely radiological hazards and biotoxic effects to individual health. The knowledge about the

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Table 1
Levels and values of assessment standards according to Haqueet al. (2018) and Li et al. (2017).

Risk Levels	Range of risk value	Acceptability
Level I (Extremely low risk)	$< 10^{-6}$	Completely accept
Level II (Low risk)	$10^{-6}, -10^{-5}$	Not concerned about the possible risk
Level III (Low-medium risk)	$10^{-5}, -5 \times 10^{-5}$	Not to be mindful about the risk
Level IV (Medium risk)	5×10^{-5},- 10^{-4}	Worry about the probable risk
Level V (Medium-high risk)	$10^{-4}, -5 \times 10^{-4}$	Mind the risk and eager to invest
Level VI (High risk)	5×10^{-4},- 10^{-3}	Give thought and take step to solve it
Level VII (Extremely high risk)	$>10^{-3}$	Required to solve it

Table 2
Exposure factors used in solving the human health risks (Isinkaye, 2018; Orosun, 2020a, 2021).

S/N	Exposure Parameters	Values	S.I Unit
2.	Inhalation rate (InhR)	20	m^3/day
3.	Exposure frequency (EF)	365	day/year
4.	Exposure duration (ED)	55	Years
5.	Body mass (BW)	70	Kg
6.	Time period of exposure (AT)	ED × 365	Days
7.	Particle emission factor (PEF)	1.36×10^9	m^3/kg
8.	Exposed skin surface area (SA)	5700 for soil	cm^2
9.	Adherence factor (AF)	0.07	mg/cm^2-day
10.	Dermal absorption factor (ABS)	0.03 for As and 0.001 for others	
11.	Chronic reference dose (RfD)	Inhalation RfD:Mn (1.43×10^{-5}), Zn (3.00×10^{-1}), Cu (4.02×10^{-2}), Cr (2.86×10^{-5}), Ni (2.06×10^{-2}), Co (5.71×10^{-6}), Pb (3.25×10^{-3}), As (3.01×10^{-4}), Cd (5.70×10^{-5}), Dermal RfD: Mn (1.84×10^{-3}), Zn (6.00×10^{-2}), Cu (1.20×10^{-2}), Cr (6.00×10^{-5}), Cd (5.00×10^{-4}), Ni (5.40×10^{-3}), Co (1.60×10^{-2}), Pb (5.25×10^{-4}), As (1.23×10^{-4})	mg/kg/day
12.	Carcinogenic slope factor (SF)	Inhalation SF: Cr (6.30) , Cd (4.10), Ni (0.84) , As (15.1), Pb (0.042). Dermal SF: As (3.66), (0.042)	$(mg/kg/day)^{-1}$
13.	Permeability constant (KP)	Pb, As, Cu (0.0001), Cr (0.002), Zn (0.006)	cm/hour

concentration of these toxic nuclides is also crucial in developing guidelines and principles for the exploitation and running of these construction materials since 80–90% of most residents days are spent indoors (Turhan, 2009; Janković et al., 2013; Joel, 2018a; Orosun, 2020b). Tiles are one of the frequently used building materials that contain a mixture of diverse raw materials like clays, feldspar, and quartz. Almost all the commercial tiles in Nigeria are glazed with zircon. The presence of the zircon in these commercial tiles can significantly enhance the concentrations of these heavy metals and the naturally occurring radionuclides beyond limits acceptable for building materials (Dizman and Keser, 2019; Janković et al., 2013).

Exposure of humans to these heavy metals and the ionizing radiation emanating from the radionuclides and their progenies is a leading source of cancer and other health challenges from radiation, harmful to vital organs of the human body that can lead to death in some cases (Orosun, 2020c; United State Environmental Protection Agency "EPA", 2018; USEPA, 1997; Orosun, 2018, 2020b–e, 2021; Orosun et al., 2021b). However, tiles used for building purposes, the inhabitants are exposed to these dangerous radiations and heavy metals continuously over the lifetime of occupying such buildings.

Table 3
Mean (n = 6) activity concentrations of *^{40}K, ^{238}U, ^{232}Th* in the sampled tiles

Sample	Type	Country	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
BN Ceramic Floor tile	Pulp	Nigeria	670.00 ± 14.00	37.50 ± 7.00	101.50 ± 11.00
BN Ceromic	Pulp	Spain	570.00 ± 42.00	55.00 ± 3.00	104.50 ± 9.00
Golden Crown Ceramics	Pulp	Nigeria	460.00 ± 18.00	49.50 ± 3.00	57.50 ± 5.00
Golden Crown Floor tiles	Pulp	Nigeria	390.00 ± 18.00	27.00 ± 4.00	113.00 ± 18.00
Goodwill ceramics	Pulp	Nigeria	530.00 ± 23.00	62.00 ± 3.00	74.50 ± 15.00
Goodwill super polish	Pulp	Nigeria	270.00 ± 12.00	44.00 ± 3.00	51.50 ± 11.00
Gordwill Vitrified	Pulp	Nigeria	540.00 ± 28.00	70.50 ± 5.00	445.50 ± 18.00
IDDRIS tiles	Pulp	China	740.00 ± 46.00	65.00 ± 3.00	337.00 ± 11.00
IRIS Ceramic	Pulp	Italy	940.00 ± 92.00	59.50 ± 3.00	79.00 ± 12.00
NISPRO Vitrified tiles	Pulp	Nigeria	860.00 ± 78.00	59.50 ± 6.00	461.00 ± 29.00
Pamesa	Pulp	Spain	650.00 ± 13.00	30.50 ± 3.00	64.00 ± 3.00
PNT Ceramic tiles	Pulp	Nigeria	510.00 ± 18.00	241.00 ± 9.00	77.50 ± 3.00
PNT Vitrified	Pulp	Nigeria	370.00 ± 12.00	35.50 ± 3.00	346.50 ± 24.00
Rose bite	Pulp	India	940.00 ± 37.00	55.50 ± 3.00	95.50 ± 12.00
Royal	Pulp	Nigeria	630.00 ± 29.00	58.00 ± 4.00	76.00 ± 6.00
Royal Classic ceramic	Pulp	Nigeria	390.00 ± 14.00	65.50 ± 6.00	44.00 ± 3.00
Royal Crown	Pulp	Nigeria	440.00 ± 16.00	51.50 ± 3.00	41.00 ± 3.00
Time ceramics	Pulp	Nigeria	510.00 ± 18.00	27.00 ± 3.00	96.00 ± 9.00
Virony	Pulp	China	530.00 ± 21.00	55.50 ± 5.00	126.50 ± 12.00
Virony Glazed	Pulp	China	290.00 ± 23.00	75.00 ± 5.00	405.50 ± 23.00
Virony Rustic glass	Pulp	China	390.00 ± 22.00	42.50 ± 3.00	63.00 ± 21.00
Virony unglazed	Pulp	China	440.00 ± 32.00	55.00 ± 5.00	52.00 ± 9.00
Global average			420.00	32.00	45.00

Global average (UNSCEAR, 2000)

Hence, the aims of this study is to examine the concentration of heavy metals and the activity levels of ^{238}U, ^{232}Th, and ^{40}K in the sampled commercial tiles used in Nigeria and assess the human carcinogenic and non-carcinogenic risks connected with the heavy metals and radiological hazards associated with the primordial radionuclides in the tiles. This is necessary to ascertain the level of human exposure to the toxic chemical and radiological risks from the natural radionuclides. Additionally, since the estimation of the radiological risk values using the risk assessment model provided by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) either overestimates or underestimates the actual radiological risk, it is impossible to determine the likelihood (either above or below the 95th percentile) that the inhabitants will be at risk without simulation. Consequently, in this work, a probabilistic approach using Monte Carlo simulation (MCS) that has the advantage of minimizing uncertainty, has been employed to inspect the probable cancer risk risks associated with primordial radionuclides (^{238}U, ^{232}Th and ^{40}K) in the ceramic tiles.

Table 4
Mean radiological hazard parameters for the selected tiles samples.

Sample	D_{indoor} (nGy/h)	AED_{indoor} (mSv/y)	Ra_{eq} (Bq/kg)	H_{ext}	H_{int}	RLI	ELCR (X 10^{-3})	AGED (mSv/y)
BN Ceramic Floor tile	199.75	0.98	234.24	0.64	0.74	1.72	3.43	0.75
BN Ceromic	211.15	1.04	248.33	0.68	0.82	1.80	3.63	0.79
Golden Crown Ceramics	145.59	0.71	167.15	0.45	0.59	1.21	2.50	0.54
Golden Crown Floor tiles	180.34	0.88	218.62	0.60	0.67	1.57	3.10	0.68
Goodwill ceramics	181.39	0.89	209.35	0.57	0.74	1.52	3.11	0.67
Goodwill super polish	118.73	0.58	138.44	0.38	0.50	0.99	2.04	0.44
Gordwill Vitrified	598.11	2.93	749.15	2.04	2.23	5.29	10.27	2.25
IDDRIS tiles	489.70	2.40	603.89	1.65	1.82	4.30	8.41	1.84
IRIS Ceramic	216.84	1.06	244.85	0.67	0.83	1.82	3.72	0.81
NISPRO Vitrified tiles	630.64	3.09	784.95	2.14	2.30	5.58	10.83	2.38
Pamesa	150.46	0.74	172.07	0.47	0.55	1.28	2.58	0.57
PNT Ceramic tiles	347.77	1.71	391.10	1.06	1.71	2.73	5.97	1.23
PNT Vitrified	443.41	2.18	559.49	1.52	1.62	3.95	7.61	1.67
Rose bite	231.31	1.13	264.45	0.72	0.87	1.96	3.97	0.87
Royal	187.36	0.92	215.19	0.59	0.74	1.57	3.22	0.69
Royal Classic ceramic	139.86	0.69	158.45	0.43	0.61	1.14	2.40	0.51
Royal Crown	127.68	0.63	144.01	0.39	0.53	1.05	2.19	0.47
Time ceramics	171.24	0.84	203.55	0.55	0.63	1.48	2.94	0.64
Virony	232.61	1.14	277.21	0.75	0.90	1.99	3.99	0.87
Virony Glazed	538.25	2.64	677.20	1.84	2.05	4.75	9.24	2.02
Virony Rustic glass	139.60	0.68	162.62	0.44	0.56	1.18	2.40	0.52
Virony unglazed	143.00	0.70	163.24	0.44	0.59	1.18	2.46	0.53
Recommended Limit	84.00	0.41	370.00	≤1	≤1	≤1	3.75	0.30

Recommended Limit (UNSCEAR, 2000).

2. Material and methods

2.1. Sample preparation and Gamma-Ray spectrometry

Six (6) samples of each of the twenty-two (22) selected ceramic tiles commonly used in Nigeria (both imported and Nigerian made) were gotten from diverse suppliers and were arranged based on IAEA TRS-295 (IAEA, 1996). This put the total number of samples at 132. Each sample was placed in a synthetic beaker and preserved for four (4) weeks secular balance. Analysis of the samples was carried out in Canada (Activation Laboratory System) employing High Purity Germanium detector, Canberra Lynx™ Digital Signal Analyzer (DSA), a 32 K control incorporated signal analyzer and a top-opening lead safeguard (4″lead, copper/tin liner) to avert increase environment counts with 50% relative effectiveness and resolution of 2.1 keV at 1.33 MeV gamma energy of 60 Co. The Genie-2 K V3.2 software sites and evaluates the climaxes, take away the background and recognizes the nuclides. The competence curves meant for this study are perfect for the reduction and self-absorption effects of the emanated gamma photons. CAMET and IAEA standards (DL-1a, UTS-2, UTS-4, IAEA-372, and IAEA-447) were utilized for examining the efficiency calibration of the system. In measuring the activity, the samples were calculated for 86,400 s with the background counts deduct from the net count (Joel et al., 2018b). The lowest noticeable activity of the detector was resolute with a confidence level of 95%. The uncertainty faults were predicted keeping into report the coupled faults from gamma courting emission possibility and effectiveness calibration average of the system. The progeny of radium, ^{214}Bi, and ^{214}Pb releases gamma line 609 keV, 934 keV, 2204 keV, 1764 keV, and 351 keV, 295 keV were used, however; the resolution of radium was from the release of 1764 keV because it has small self-attenuation consequence at high energy. Given that ^{232}Th cannot be honestly identified, the expected action through its progeny ^{208}Tl and ^{228}Ac by means of 2614.53 keV, (35.63%) 583 keV (30.3%) and 911 keV, 338 keV, 463 keV. The gamma line of 1461 keV (10.7%) was utilized to determine ^{40}K.

2.2. Materials and method for toxic metals contents in the samples

The diverse building material samples used in this study were acquired from the Orile market in Lagos State, Nigeria. The samples were compressed, pounded, and put through a sieve of 75 μm mesh for homogeneity, position in a plastic vial, and tagged in permanent marker for simple recognition prior to transferring it to Bureau Veritas Laboratory Ltd, Canada, for investigation. About 0.2 g of the samples were correctly weighed into a container perfluoroalkoxy polymer, that was subsequently positioned in a microwave pressure vessel (Ethos Plus Microwave Lactation, Milestone Inc., Shelton, CT, USA), using the standard of USEPA method 3052. Subsequent accumulation of concentrated nitric acid and concentrated hydrochloric acid at 4 ml and 0.5 ml and absorption of the samples using microwave power gradually raised to 400 W within 40 min. Following cooling, water was correctly added to the solutions to 100 ml. Nevertheless, open digestion in a beaker was performed with 0.5 g of sample, evaluated completely, by heating with 12 ml of aqua regia for 40 min, trailed via evaporation to drought. 25 ml of concentrated hydrochloric acid and 2.5 ml of hydrogen peroxide were added to the burning residue with a precise intensity of 50 ml of water. A duplicate per digestion technique was completed for every sample (Hoffmann et al., 1999). The whole content of heavy metals in the house materials was analyzed with an ICP-MS device attached to the intuitive WinLab32 software system encompasses the apparatus to examine, report as well as accomplished the calculated data (Bonta et al., 2016; Zhang & Hu, 2011; Hoffmann et al., 1999). Calibrating the samples, the standard resolutions (panreac) of 100 mgL^{-1} of all metals were used, and therefore, were calibrated from 10 to 100 ppb.

2.3. Quality control for the analysis of toxic metals in the samples

In this research, the quality control for the study of the samples with ICP-MS with the model, Perkin Elmer ICP-MS was accomplished through the standard operation procedures (SOPs) following the manufacturer guidelines. All equipment used in this work was regulated according to (Hoffmann et al., 1999). A calibration curve close to 1 was acquired for ICP-MS prior to the investigation was carried out on the samples so that the absorption of the atom of all components to be calculated will be more precise according to Bonta et al., 2016; Zhang & Hu, 2011.

2.4. Estimation of the radiological impact parameters

2.4.1. Absorbed dose rate

The indoor absorbed dose rate (*D*) in the air due to the average specific activities of ^{40}K, ^{238}U, and ^{232}Th (Bq/kg) in the ceramic tile samples was estimated using equation (1),

Table 5
Mean (n = 6) concentration of the selected heavy metals in the sampled tiles in ppm.

Sample	Cu	Pb	Zn	Ni	Co	Mn	As	Cd	Cr
BN Ceramic Floor tile	22.33 ± 1.1	67.62 ± 2.3	448.20 ± 42.1	71.80 ± 4.1	9.30 ± 1.1	191.00 ± 11.0	6.50 ± 1.1	1.24 ± 0.3	58.00 ± 2.7
BN Ceromic	21.19 ± 1.5	68.41 ± 3.6	425.20 ± 23.1	54.70 ± 2.2	15.40 ± 1.3	220.00 ± 21.0	10.80 ± 1.8	0.49 ± 0.1	69.00 ± 0.3
Golden Crown Ceramics	95.21 ± 3.1	57.44 ± 2.1	644.00 ± 43.9	333.60 ± 13.2	18.00 ± 1.3	309.00 ± 23.1	3.90 ± 0.3	0.22 ± 0.0	163.00 ± 9.1
Golden Crown Floor tiles	59.99 ± 2.3	52.36 ± 2.1	259.60 ± 21.1	268.70 ± 33.1	25.70 ± 1.7	283.00 ± 61.3	6.10 ± 0.6	0.22 ± 0.0	182.00 ± 11.2
Goodwill ceramics	25.41 ± 2.9	54.87 ± 2.3	733.70 ± 71.0	80.30 ± 1.5	9.70 ± 1.1	221.00 ± 22.0	5.30 ± 0.9	1.01 ± 0.0	98.00 ± 12.1
Goodwill super polish	29.39 ± 1.2	39.48 ± 1.3	696.60 ± 81.3	103.60 ± 1.7	17.60 ± 1.2	353.00 ± 21.8	3.40 ± 1.1	0.46 ± 0.1	95.00 ± 2.4
Gordwill Vitrified	24.46 ± 1.6	68.70 ± 5.2	429.60 ± 43.2	22.40 ± 1.3	6.00 ± 1.4	158.00 ± 15.1	4.80 ± 1.3	2.49 ± 0.7	34.00 ± 1.2
IDDRIS tiles	29.95 ± 1.9	98.41 ± 5.8	700.10 ± 47.1	26.30 ± 2.1	14.70 ± 1.8	159.00 ± 13.1	15.90 ± 0.9	0.42 ± 0.0	72.00 ± 2.5
IRIS Ceramic	2264.17 ± 243.0	26.86 ± 1.3	392.30 ± 51.2	4772.20 ± 244.0	16.70 ± 1.4	219.00 ± 12.4	14.60 ± 1.6	0.30 ± 0.1	35.00 ± 2.1
NISPRO Vitrified tiles	67.36 ± 1.8	43.29 ± 1.1	145.00 ± 11.3	118.30 ± 12.1	3.50 ± 1.1	88.00 ± 6.2	2.10 ± 0.3	0.11 ± 0.0	47.00 ± 2.1
Pamesa	1287.65 ± 103.0	73.80 ± 7.9	1353.00 ± 157.0	2621.70 ± 121.0	25.00 ± 2.3	390.00 ± 15.2	20.40 ± 0.6	0.68 ± 0.4	68.00 ± 2.8
PNT Ceramic tiles	22.33 ± 1.3	117.45 ± 7.1	443.50 ± 34.1	121.30 ± 2.7	27.70 ± 2.1	299.00 ± 29.1	4.10 ± 0.3	2.18 ± 1.1	131.00 ± 23.1
PNT Vitrified	23.67 ± 1.2	69.26 ± 4.3	527.60 ± 51.2	65.80 ± 3.2	24.60 ± 1.4	267.00 ± 21.8	7.40 ± 0.5	0.28 ± 0.0	85.00 ± 4.3
Rose bite	362.26 ± 23.9	24.41 ± 2.1	43.20 ± 1.3	694.60 ± 43.2	4.00 ± 1.2	732.00 ± 22.9	2.80 ± 0.3	0.02 ± 0.0	4.00 ± 1.1
Royal	1580.81 ± 133.0	44.56 ± 1.4	473.00 ± 43.3	3254.80 ± 134.0	15.10 ± 1.8	307.00 ± 23.1	3.30 ± 0.2	0.20 ± 0.0	56.00 ± 1.9
Royal Classic ceramic	22.98 ± 1.6	44.67 ± 1.3	425.60 ± 31.1	49.50 ± 1.7	6.50 ± 1.1	238.00 ± 21.6	2.30 ± 0.6	0.09 ± 0.0	49.00 ± 1.5
Royal Crown	18.80 ± 1.2	45.94 ± 1.9	2468.50 ± 95.0	40.40 ± 1.9	31.60 ± 1.1	231.00 ± 17.3	1.90 ± 0.1	0.23 ± 0.0	65.00 ± 3.1
Time ceramics	27.11 ± 1.2	49.67 ± 1.7	843.20 ± 21.2	134.90 ± 11.2	36.80 ± 1.3	183.00 ± 13.8	2.90 ± 0.1	1.49 ± 0.0	151.00 ± 13.1
Virony	325.14 ± 32.0	47.02 ± 1.2	135.30 ± 17.1	641.70 ± 32.1	5.70 ± 1.2	221.00 ± 12.9	25.10 ± 2.3	0.16 ± 0.0	34.00 ± 1.2
Virony Glazed	881.70 ± 67.0	68.68 ± 1.8	574.70 ± 51.4	1855.00 ± 91.3	383.70 ± 18.1	442.00 ± 32.7	14.00 ± 1.2	2.49 ± 0.1	315.00 ± 13.2
Virony Rustic glass	17.25 ± 1.3	54.42 ± 1.4	587.50 ± 61.3	9.70 ± 1.3	5.20 ± 1.1	214.00 ± 21.0	9.50 ± 1.3	0.34 ± 0.0	43.00 ± 2.1
Virony unglazed	16.08 ± 1.2	46.42 ± 2.1	139.40 ± 17.7	9.80 ± 1.3	3.70 ± 1.1	212.00 ± 21.1	26.00 ± 2.8	0.34 ± 0.0	33.00 ± 3.1
Global Average	**38.90**	**27.00**	**70.00**	**29.00**	**–**	**488.00**	**6.83**	**0.41**	**59.50**

Global average (Kabata-Pendias*et al.*, 2004 and Chen, 1999).

$$D_{indoor}(nGy/h) = 0.92C_u + 1.1C_{Th} + 0.08C_K \quad (1)$$

where C_K, C_{Ra}, and C_{Th} are the activities of ^{40}K, ^{226}Ra, and ^{232}Th in the tile samples respectively (UNSCEAR, 2000).

2.4.2. Annual effective dose for external exposures (AED_{Ext})

The effective dose expected by a member of the general public annually was designed using the dose rates.

$$AED_{indoor}\,(mSv/y) = D_{indoor}\,(nGy/h) \times 8760\,h \times 0.7\,(Sv\,Gy^{-1}) \times 0.8 \times 10^{-6} \quad (2)$$

Dose conversion factor of 0.7 *Sv/G* and occupancy factor for indoor as 0.8 were adopted (Joel, 2018a–c, Orosun, 2018, 2019; UNSCEAR, 2000).

2.4.3. Radium equivalent activity index (Ra_{eq})

The radium equivalent (Ra_{eq}) was calculated using the formula:

$$Ra_{eq} = C_u + 1.43C_{Th} + 0.077C_K \quad (3)$$

whereC_u, C_{Th}, C_K are the radioactivity concentration in Bq/kg of ^{238}U, ^{232}Th and ^{40}K. Average value of the Radium Equivalent Activity Index (Ra_{eq}) is 370 Bq/kg.

2.4.4. Representative level index (RLI)

The *RLI* was estimated using equation (8) (UNSCEAR, 2000; Orosun et al., 2018; Adewoyin et al., 2019):

$$RLI = \frac{C_u}{150} + \frac{C_{Th}}{100} + \frac{C_k}{1500} \leq 1 \quad (4)$$

where, C_{Ra}, C_{Th}, and C_K maintain their us'ual definition.

RLI values = 1 corresponds to an *AED* ≤ 1 *mSv*. Thus, *RLI* is a radiological impact parameter for screening building materials containing significant amounts of these primordial radionuclides and assess the gamma radiation risk associated with them (UNSCEAR, 2000).

2.4.5. Radiation hazard indices

These indices were used to calculate the stage of Gamma- radiation hazard connected with the regular radionuclide in samples. The external radiation hazard (H_{ext}) and the internal radiation hazard (H_{int}) was estimated as follows:

$$H_{ext} = \left(\frac{C_U}{370}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad (5)$$

$$H_{int} = \left(\frac{C_U}{185}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad (6)$$

Table 6
Estimated $THQ_{inhalation}$, THQ_{Dermal} and the Hazard Index (HI) for the sampled tiles.

Sample	Mn (THQ_{inh} + THQ_{Derm})	Zn (THQ_{inh} + THQ_{Derm})	Cu (THQ_{inh} + THQ_{Derm})	Co (THQ_{inh} + THQ_{Derm})	Cd (THQ_{inh} + THQ_{Derm})	Pb (THQ_{inh} + THQ_{Derm})	As (THQ_{inh} + THQ_{Derm})	Cr (THQ_{inh} + THQ_{Derm})	Ni (THQ_{inh} + THQ_{Derm})	HI
BN Ceramic Floor tile	2.45E-05	8.83E-06	3.30E-06	9.30E-03	8.15E-04	7.38E-04	9.04E-03	5.94E-03	7.65E-05	2.59E-02
BN Ceromic	2.83E-05	8.38E-06	3.13E-06	1.54E-02	3.22E-04	7.47E-04	1.50E-02	7.06E-03	5.83E-05	3.87E-02
Golden Crown Ceramics	3.97E-05	1.27E-05	1.41E-05	1.80E-02	1.45E-04	6.27E-04	5.42E-03	1.67E-02	3.56E-04	4.13E-02
Golden Crown Floor tiles	3.64E-05	5.11E-06	8.86E-06	2.57E-02	1.45E-04	5.72E-04	8.48E-03	1.86E-02	2.86E-04	5.39E-02
Goodwill ceramics	2.84E-05	1.45E-05	3.75E-06	9.70E-03	6.64E-04	5.99E-04	7.37E-03	1.00E-02	8.56E-05	2.85E-02
Goodwill super polish	4.54E-05	1.37E-05	4.34E-06	1.76E-02	3.02E-04	4.31E-04	4.73E-03	9.72E-03	1.10E-04	3.30E-02
Gordwill Vitrified	2.03E-05	8.46E-06	3.61E-06	6.00E-03	1.64E-03	7.50E-04	6.68E-03	3.48E-03	2.39E-05	1.86E-02
IDDRIS tiles	2.04E-05	1.38E-05	4.43E-06	1.47E-02	2.76E-04	1.07E-03	2.21E-02	7.37E-03	2.80E-05	4.56E-02
IRIS Ceramic	2.81E-05	7.73E-06	3.35E-04	1.67E-02	1.97E-04	2.93E-04	2.03E-02	3.58E-03	5.09E-03	4.65E-02
NISPRO Vitrified tiles	1.13E-05	2.86E-06	9.95E-06	3.50E-03	7.23E-05	4.73E-04	2.92E-03	4.81E-03	1.26E-04	1.19E-02
Pamesa	5.01E-05	2.67E-05	1.90E-04	2.50E-02	4.47E-04	8.06E-04	2.84E-02	6.96E-03	2.79E-03	6.46E-02
PNT Ceramic tiles	3.84E-05	8.74E-06	3.30E-06	2.77E-02	1.43E-03	1.28E-03	5.70E-03	1.34E-02	1.29E-04	4.97E-02
PNT Vitrified	3.43E-05	1.04E-05	3.50E-06	2.46E-02	1.84E-04	7.56E-04	1.03E-02	8.70E-03	7.01E-05	4.47E-02
Rose bite	9.40E-05	8.51E-07	5.35E-05	4.00E-03	1.32E-05	2.66E-04	3.89E-03	4.09E-04	7.40E-04	9.47E-03
Royal	3.94E-05	9.32E-06	2.34E-04	1.51E-02	1.32E-04	4.86E-04	4.59E-03	5.73E-03	3.47E-03	2.98E-02
Royal Classic ceramic	3.06E-05	8.38E-06	3.40E-06	6.50E-03	5.92E-05	4.88E-04	3.20E-03	5.01E-03	5.28E-05	1.54E-02
Royal Crown	2.97E-05	4.86E-05	2.78E-06	3.16E-02	1.51E-04	5.02E-04	2.64E-03	6.65E-03	4.31E-05	4.17E-02
Time ceramics	2.35E-05	1.66E-05	4.01E-06	3.68E-02	9.80E-04	5.42E-04	4.03E-03	1.55E-02	1.44E-04	5.80E-02
Virony	2.84E-05	2.67E-06	4.80E-05	5.70E-03	1.05E-04	5.13E-04	3.49E-02	3.48E-03	6.84E-04	4.55E-02
Virony Glazed	5.68E-05	1.13E-05	1.30E-04	3.84E-01	1.64E-03	7.50E-04	1.95E-02	3.22E-02	1.98E-03	4.40E-01
Virony Rustic glass	2.75E-05	1.16E-05	2.55E-06	5.20E-03	2.24E-04	5.94E-04	1.32E-02	4.40E-03	1.03E-05	2.37E-02
Virony unglazed	2.72E-05	2.75E-06	2.38E-06	3.70E-03	2.24E-04	5.07E-04	3.62E-02	3.38E-03	1.04E-05	4.40E-02

H_{int} ought to be less than unity for the radiation hazard to be insignificant.

2.4.6. Excess Lifetime cancer risk (ELCR)

The Excess Lifetime Cancer Risk (*ELCR*) was estimated using equation (7):

$$ELCR = AED \times DL \times RF \tag{7}$$

where, *AED* is the Annual Effective Dose, *DL* is the average life interval (assuming 70 years) and RF is the fatal cancer risk per Sievert assumed to be 0.05 for stochastic effects for the populace ((UNSCEAR, 2000; Orosun et al., 2019; Adewoyin et al., 2019). The recommended limit for the ELCR is 3.75×10^{-3}.

2.4.7. Annual gonadal equivalent dose (AGED)

The AGED for the public using the tiles for building was calculated by the following equation (Omeje et al., 2020)

$$AGED\ (\mu Sv/y)\ C = 3.09C_U + 4.18C_{Th} + 0.314C_K \tag{8}$$

2.5. Human health risk assessment

Human health risk assessment of heavy metals involves the estimation and assessment of the form and likelihood of health effects in a person who might be uncovered to the potentially toxic substance in concentrations above the recommended threshold in a contaminated environment. The relationship between the concentration of these potentially toxic elements (heavy metals) and the probable risk to human health is generally evaluated by the human health risk assessment approach presented by the USEPA (2004), USEPA (2007), USEPA (2009) and UNC (2011). This procedure is available through a risk assessment information system (RAIS) (USEPA, 2004) and is reinforced by the toxicological profiles developed and assembled by the United State Environmental Protection Agency's Integrated Risk Information System (IRIS) (Orosun, 2020a, 2021; U.S. Environmental Protection Agency (EPA), 2007), and by the United State Agency for Toxic Substances and Disease Registry – Toxicological profiles (ATSDR, 2007). In this current research, the evaluation of exposure was completed by evaluating the chronic daily intake (CDI) of each of the selected heavy metals by inhalation and dermal contact.

Table 7
Estimated Incremental Life Cancer Risk (ILCR) values for the sampled tiles.

Sample	Cd (Inh + Derm)	Pb (Inh + Derm)	As (Inh + Derm)	Cr (Inh + Derm)	Ni (Inh + Derm)	ILCR
BN Ceramic Floor tile	1.07E-09	6.00E-10	1.68E-05	7.68E-08	1.27E-08	1.69E-05
BN Ceromic	4.22E-10	6.07E-10	2.79E-05	9.13E-08	9.65E-09	2.80E-05
Golden Crown Ceramics	1.89E-10	5.10E-10	1.01E-05	2.16E-07	5.89E-08	1.03E-05
Golden Crown Floor tiles	1.89E-10	4.65E-10	1.58E-05	2.41E-07	4.74E-08	1.60E-05
Goodwill ceramics	8.70E-10	4.87E-10	1.37E-05	1.30E-07	1.42E-08	1.38E-05
Goodwill super polish	3.96E-10	3.50E-10	8.78E-06	1.26E-07	1.83E-08	8.92E-06
Gordwill Vitrified	2.14E-09	6.10E-10	1.24E-05	4.50E-08	3.95E-09	1.24E-05
IDDRIS tiles	3.62E-10	8.73E-10	4.11E-05	9.53E-08	4.64E-09	4.12E-05
IRIS Ceramic	2.58E-10	2.38E-10	3.77E-05	4.63E-08	8.42E-07	3.86E-05
NISPRO Vitrified tiles	9.47E-11	3.84E-10	5.42E-06	6.22E-08	2.09E-08	5.51E-06
Pamesa	5.86E-10	6.55E-10	5.27E-05	9.00E-08	4.63E-07	5.32E-05
PNT Ceramic tiles	1.88E-09	1.04E-09	1.06E-05	1.73E-07	2.14E-08	1.08E-05
PNT Vitrified	2.41E-10	6.14E-10	1.91E-05	1.13E-07	1.16E-08	1.92E-05
Rose bite	1.72E-11	2.17E-10	7.23E-06	5.29E-09	1.23E-07	7.36E-06
Royal	1.72E-10	3.95E-10	8.52E-06	7.41E-08	5.74E-07	9.17E-06
Royal Classic ceramic	7.75E-11	3.96E-10	5.94E-06	6.49E-08	8.74E-09	6.01E-06
Royal Crown	1.98E-10	4.08E-10	4.91E-06	8.60E-08	7.13E-09	5.00E-06
Time ceramics	1.28E-09	4.41E-10	7.49E-06	2.00E-07	2.38E-08	7.71E-06
Virony	1.38E-10	4.17E-10	6.48E-05	4.50E-08	1.13E-07	6.50E-05
Virony Glazed	2.14E-09	6.09E-10	3.62E-05	4.17E-07	3.27E-07	3.69E-05
Virony Rustic glass	2.93E-10	4.83E-10	2.45E-05	5.69E-08	1.71E-09	2.46E-05
Virony unglazed	2.93E-10	4.12E-10	6.71E-05	4.37E-08	1.73E-09	6.72E-05

For the inhalation and dermal contact exposure trailed, the chronic daily intake (CDI) (mg/kg/day) was predicted by the following equations set by USEPA (2007).

For inhalation pathway,

$$ADI_{inh-dust} = \frac{Cs \times InhRs \times EF \times ED}{PEF \times BW \times AT} \tag{9}$$

where Cs is the concentration of the given heavy metal in the sample tiles. BW is bodyweight of the exposed person, ED is the lifetime exposure period (year), EF is the exposure frequency (day/year), AT is the time period through which the dose is averaged (day) and InhRs is the inhalation rate of the tile dust (mg/day). PEF is the element emission factor (m^3/kg).

For Dermal pathway,

$$ADI_{derm} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \tag{10}$$

Table 8
Summary of the Monte Carlo simulation

	Excess Lifetime Cancer Risk (*ELCR*) ($\times 10^{-3}$)		
	P 5 %	Mean	P 95 %
NISPRO Vitrified tiles	5.55	11.90	20.60
Gordwill Vitrified	4.97	11.40	20.00
Virony Glazed	4.81	10.80	19.00
IDDRIS tiles	4.59	10.10	18.00
PNT Vitrified	4.40	9.72	17.30
PNT Ceramic tiles	3.67	7.35	12.50
Virony	3.08	6.11	10.60
Rose bite	3.08	6.06	10.40
IRIS Ceramic	2.87	5.75	10.00
BN Ceromic	2.53	5.46	9.82
BN Ceramic Floor tile	1.18	3.57	6.59
Royal	1.21	3.51	6.35
Goodwill ceramics	0.95	3.26	6.19
Golden Crown Floor tiles	0.95	3.25	6.20
Time ceramics	0.91	3.13	5.97
Pamesa	0.82	2.93	5.68
Golden Crown Ceramics	0.82	2.81	5.48
Virony unglazed	1.02	2.90	5.40
Royal Classic ceramic	0.83	2.80	5.44
Virony Rustic glass	0.98	2.87	5.41
Royal Crown	0.71	2.62	5.15
Goodwill super polish	0.72	2.53	5.00

where KP is the permeability constant of the skin, SA is the exposed skin surface area (cm^2), ET is the exposure time and ABS is the skin absorption factor.

2.5.1. The Non-Carcinogenic risk assessment

The expected Chronic daily intake (CDI) in proportion to reference dose (RfD) of an exact toxic element identified as target Hazard Quotient (HQ) (Orosun, 2020a, 2021; USEPA, 2007), is employed to highlight the non-carcinogenic risk measurement. The HQ employs the non-carcinogenic threshold (reference dose (RfD)), which is acknowledged as the daily absorption rate under which no major risk of unpleasant health effects is predicted above 70-years lifetime. The formula is given by USEPA as;

$$HQ = \frac{ADI}{RfD} \tag{11}$$

where CDI is the chronic daily intake of a given toxic constituent and RfD is the persistent reference dose for the element (mg/kg-day) (USEPA, 2007). If the HQ > 1, however, there is an increased probability of unfavorable health effects to the exposed populace. Conversely, if HQ < 1 subsequently there is no possibility of negative health effects (Saleh et al., 2019; Rinklebe et al., 2019).

The hazard index (HI) is described as the total addition of HQ and calculated for the different pathways using equation (11) (Rinklebe et al., 2019; USEPA, 2007). The significance of the hazard index (HI) is in the ability to calculate and predict the human health risks by more than a particular heavy metal (Orosun, 2021).

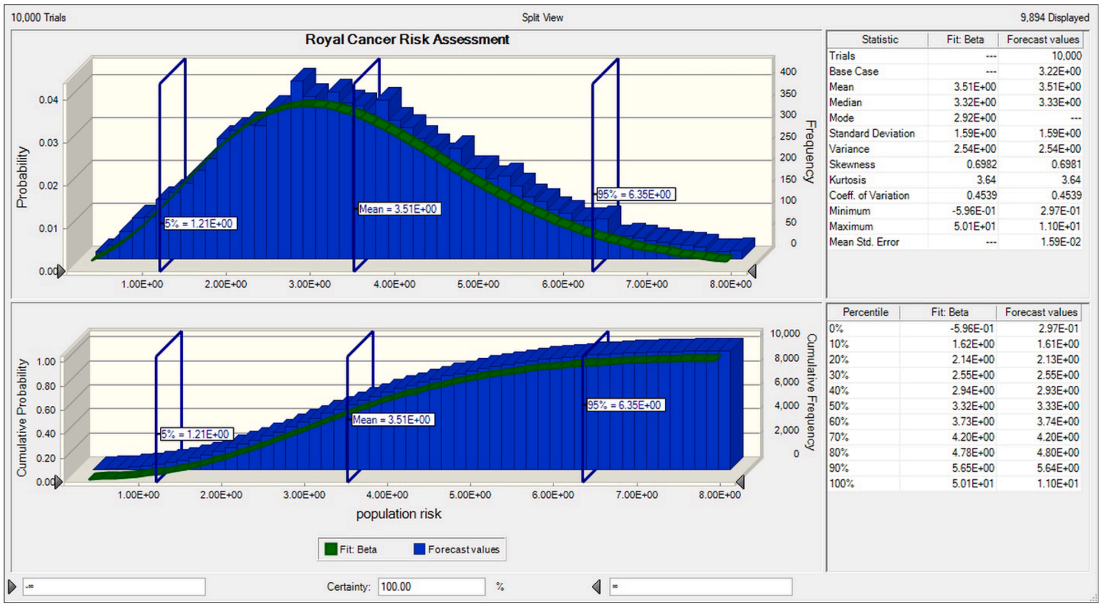
$$HI = \sum HQ \tag{12}$$

2.5.2. The carcinogenic risk assessment

The carcinogenic risk estimation gives an index of risk or possibility of an aimed people to grow cancer of several kinds following contact to carcinogen over a predictable lifetime (Isinkaye, 2018; Orosun, 2020a, 2021). Incremental Lifetime Cancer Risk (ILCR) that gives the incremental probability of an individual affected with cancer over a time because of exposure to the heavy metals is calculated using equation (13) (Isinkaye, 2018; Orosun, 2020a).

$$ILCR = ADI \times SF \tag{13}$$

where ADI (mg/kg/day) and SF (mg/kg/day) are the average everyday



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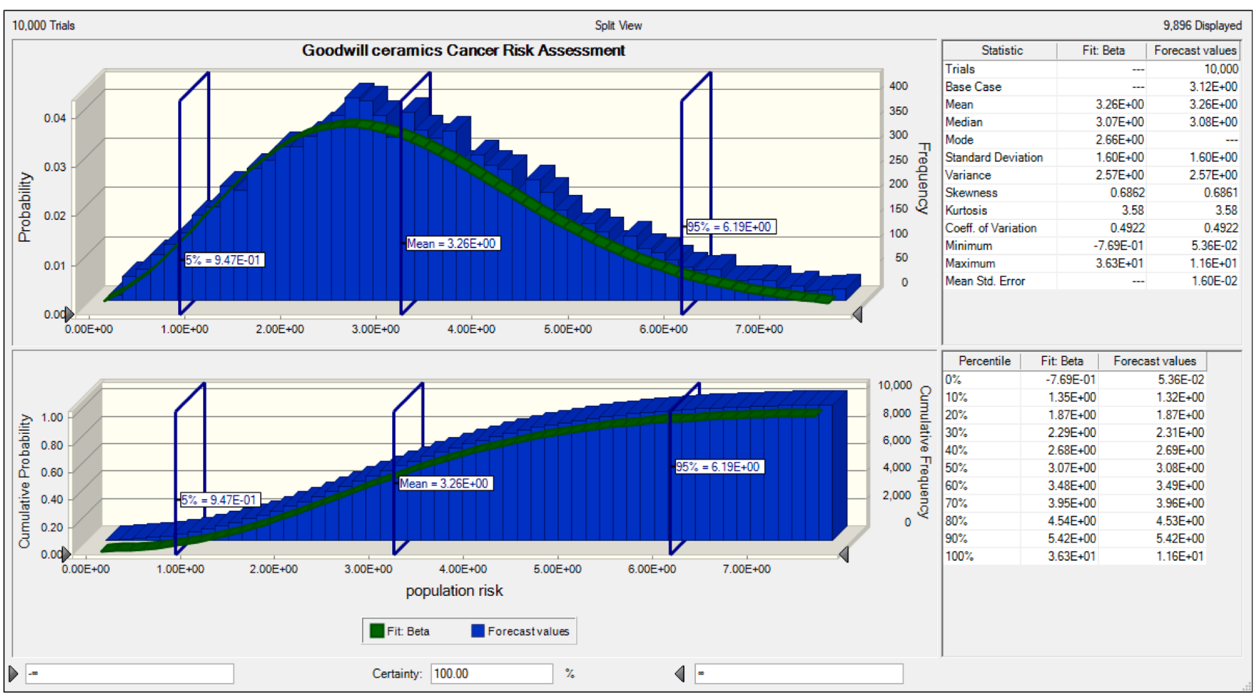

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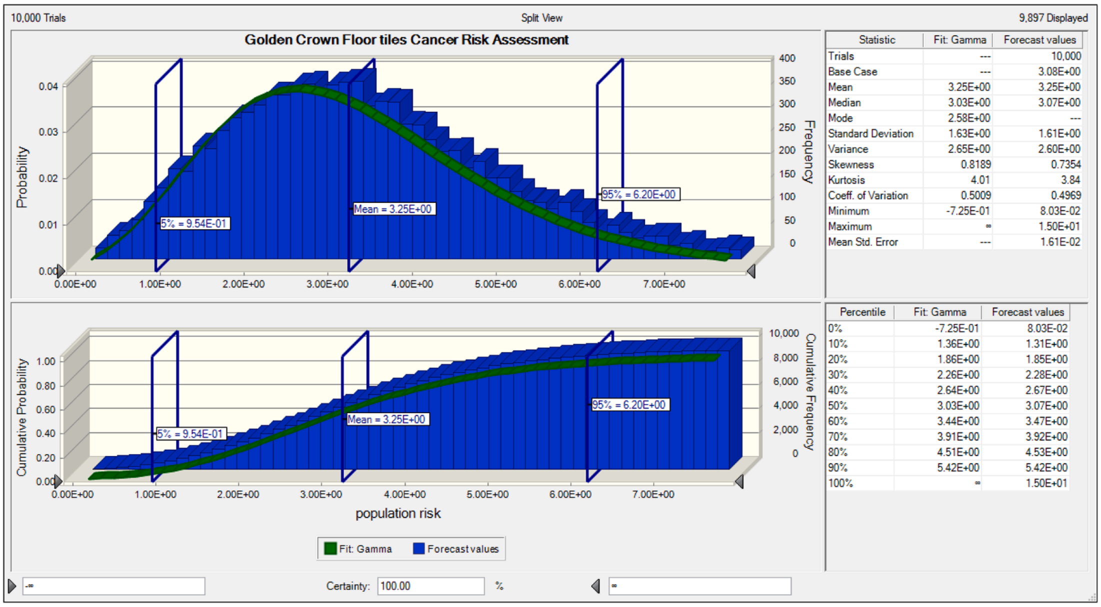
Fig. 1. Cumulative probability plot of the Excess Lifetime Cancer Risk associated with the measured radionuclides in the sampled tiles. (a) NISPRO Vitrified tiles, (b) Goodwill Vitrified, (c) Virony Glazed, (d) IDDRIS tiles, (e) PNT Vitrified, (f) PNT Ceramic tiles, (g) Virony, (h) Rose bite, (i) IRIS Ceramic, (j) BN Ceramic, (k) BN Ceramic Floor tile, (l) Royal, (m)Goodwill ceramics, (n) Golden Crown Floor tiles, (o) Time ceramics, (p) Pamesa, (q) Golden Crown Ceramics, (r) Virony unglazed, (s) Royal Classic ceramic, (t) Virony Rustic glass, (u) Royal Crown, and (v) Goodwill super polish.

intake and the carcinogenic gradient factor in that order. Cancer risk higher than 1×10^{-4} are measured high as they cause higher cancer danger while values below 1×10^{-6} are assumed not to cause any cancer danger to the populace; the suitable range is flanked by 1×10^{-4} and 1×10^{-6} (Saleh et al., 2019; Qasemi et al., 2019). The risk ideals are categorised into 7 levels as proposed by Delphii method according to Haque et al. (2018) and Li et al. (2017) and are set in Table 1. Table 2 provides the exposure parameters used in evaluating the human health risks.

3. Results and discussion

The outcomes of the activity application of the radionuclides, and heavy metal investigation conducted on the selected tile samples



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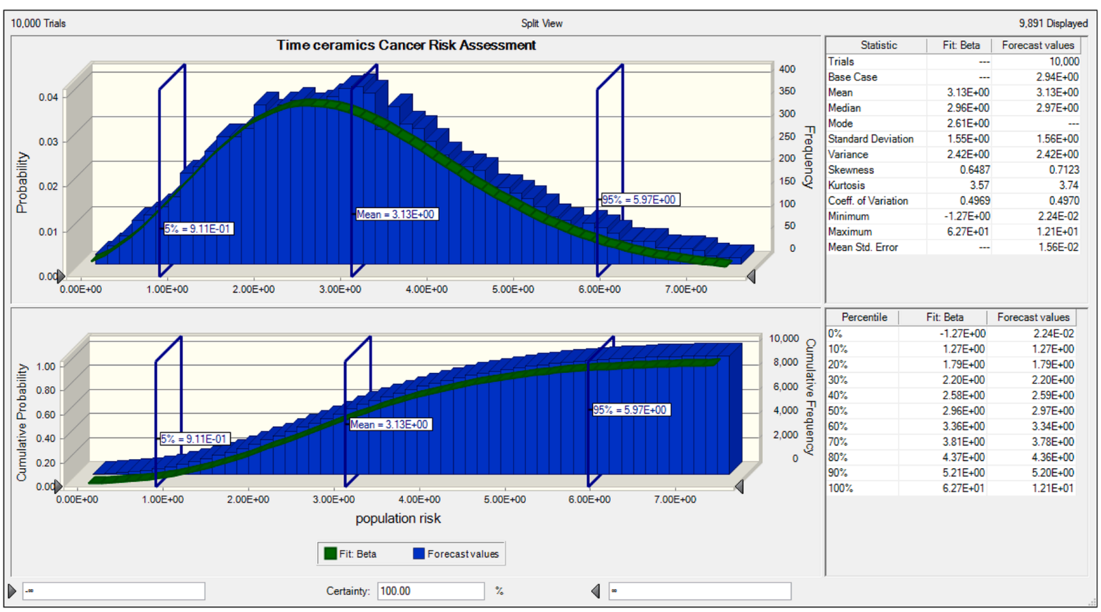

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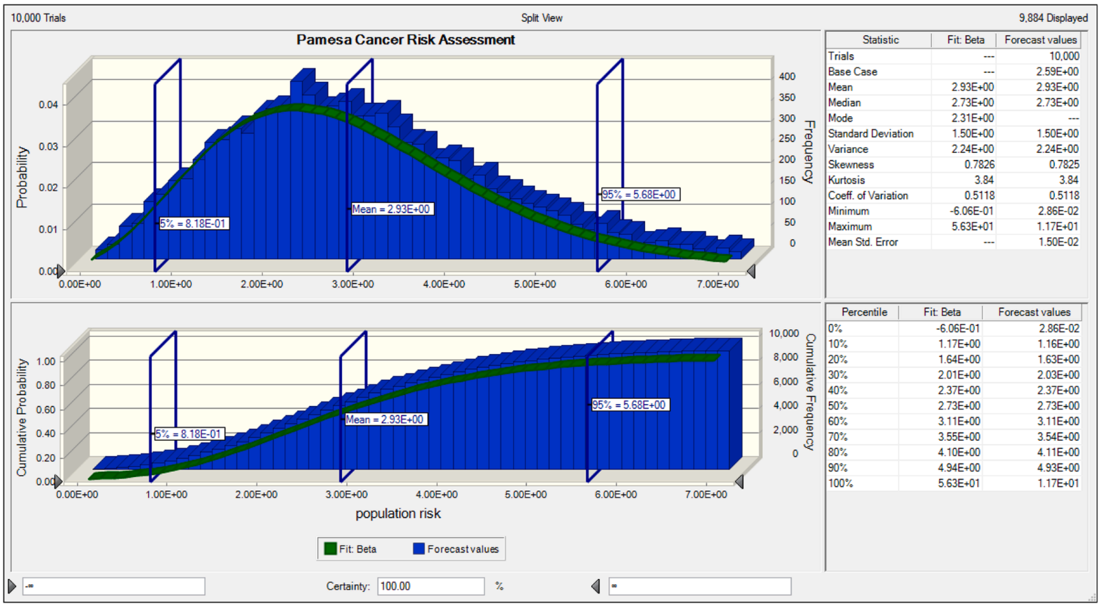
Fig. 1. (*continued*).

analyzed are given in Tables 3, and 5. The results are offered together with the present suggested thresholds available by numerous regulatory organizations, committees or agencies all over the world.

3.1. ^{238}U, ^{232}Th and ^{40}KActivity concentration in the sampled tiles

The average activity concentrations of ^{238}U, ^{232}Th, and $^{40}K$of the sampled tiles are available in Table 3. As expected, the activities of ^{40}K dominated that of ^{238}U and ^{232}Threspectively. The estimated average values of the activity concentration of ^{40}Kranges between 940 Bq/kg (Rose bite and IRIS Ceramic) and 270 (Goodwill super polish) Bq/kg. This follows that Rose bite (India) and IRIS Ceramic (Italy) have the highest ^{40}K activities and Goodwill super polish (Nigeria) have the lowest activities. Out of the 22 selected commonly used tiles in Nigeria, only 6(27%) have ^{40}K activities within the recommended level of 420 Bq/kg. The maximum and minimum activities of ^{238}U were confirmed in Virony Glazed (China) with 75 Bq/kg and Time ceramics (Nigeria) and Golden Crown Floor tiles (Nigeria) both with 27 Bq/kg. Only 3 (13%) of the sampled tiles have their activities within the recommended limits of 32 Bq/kg. From Table 3, the lowest ideals of the activity concentration of^{232}Th were recorded in Royal Crown (Nigeria) with the mean activity of 41 Bq/kg, while the corresponding highest value was recorded in NISPRO Vitrified tiles (Nigeria) with the mean value of 461 Bq/kg. It similarly reveals that only 2 (9%) of the sampled tiles have activities fewer than the recommended value of 45 Bq/kg. These high values observed in most of the sampled tiles call for serious concern since a



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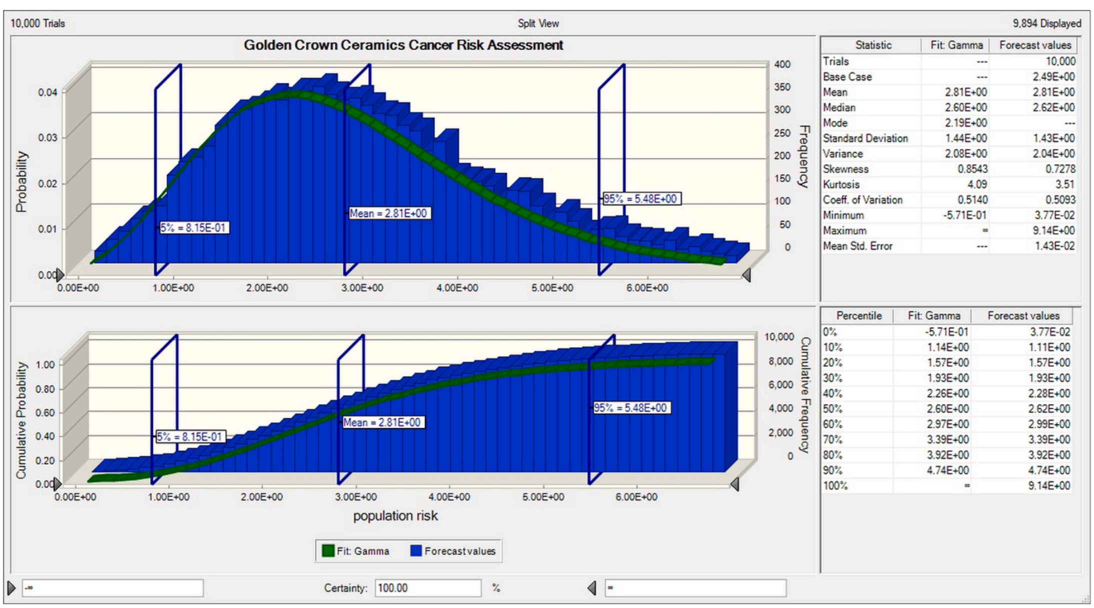

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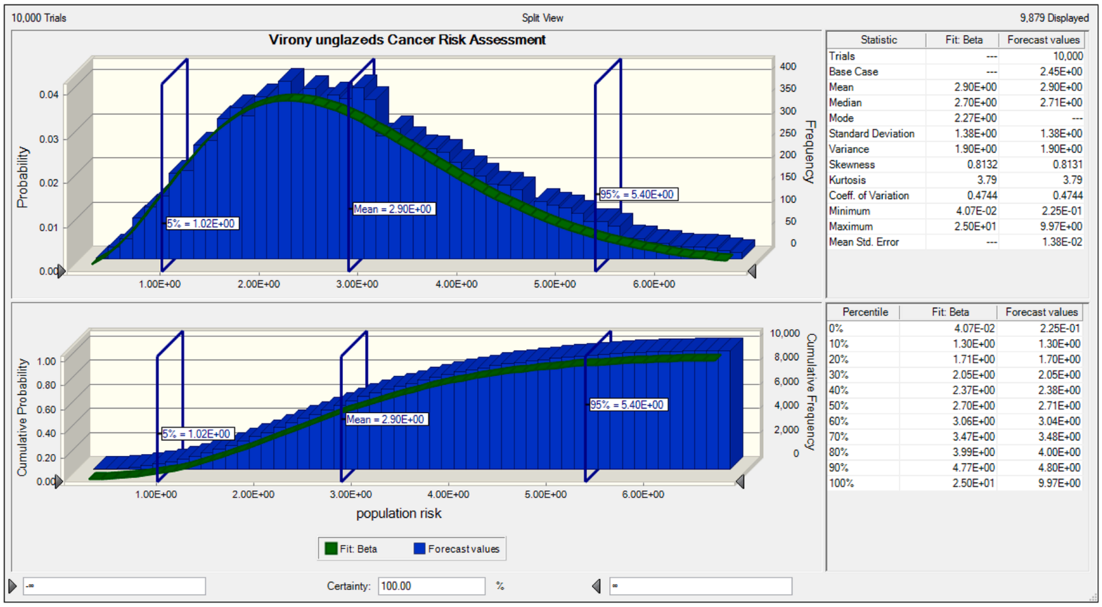
Fig. 1. (*continued*).

considerable increase in the concentration of the radionuclides usually leads to an increase in the level of background radiation that is very harmful to human health.

3.2. *Evaluation of the radiological hazard parameters for the selected tiles samples*

The radiological impact parameters were estimated to assess the radiological hazards that relate to the use of the sampled tiles and assess their suitability for building and construction purposes. Table 4 presents the estimated hazards indices. The absorbed dose rate (D) was estimated using equation (1) and the ensuing values were used to evaluate the annual effective doses. The maximum and minimum mean values of absorbed dose rate were observed in NISPRO Vitrified tiles (Nigeria) with 630.64 nGy/h and Goodwill super polish (Nigeria) with 118.73 nGy/h respectively. Surprisingly, all the tiles (100%) have values greater than the recommended limit of 84.00 nGy/h provided by UNSCEAR. This trails that the risk of interior γ- radiation exposure is higher for all the sampled tiles (both Nigerian-made and imported). Similarly, the highest and lowest mean annual effective dose values were observed in NISPRO Vitrified tiles (Nigeria) with 3.09 mSv/y and Goodwill super polish (Nigeria) with 0.58 mSv/y respectively. The samples tiles have their AED 100% above the recommended level of 0.41 mSv/y for indoor exposures, revealing that Nigerians are at risk of overexposure to indoor ionizing radiation for using these tiles for their building and construction purposes.



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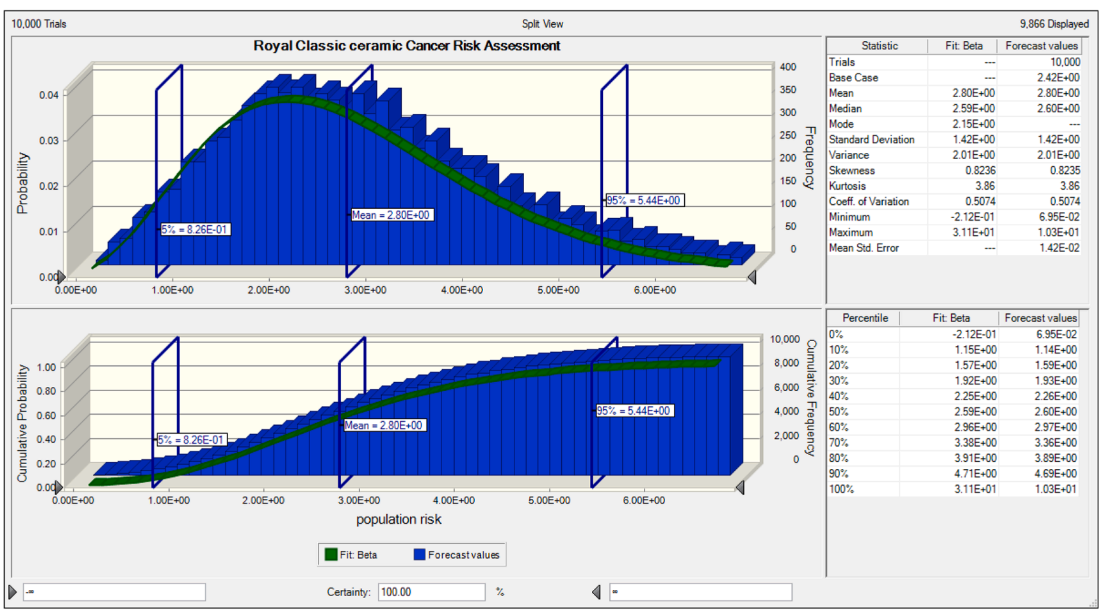

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Fig. 1. (*continued*).

The estimated radium equivalent (Ra_{eq}), $H_{ext,}$ and H_{in} follow similar trends with maximum values observed in NISPRO Vitrified tiles (Nigeria) and minimum values recorded in Goodwill super polish (Nigeria) respectively. Only six (27%) out of the 22 products sampled have their Ra_{eq}, H_{ext} and H_{in} above the recommended limits provided by UNSCEAR and other regulatory agencies.

The results of the representative level index (*RLI*) otherwise referred to as gamma-index (Iγ), reveal that only Goodwill super polish tiles (Nigeria) whose only value is ≤ 1, is fairly suitable for use in structure and building purpose. The estimated values for the *ELCR* and *AGED* corroborated our earlier findings with NISPRO Vitrified tiles (Nigeria) and Goodwill super polish (Nigeria) recording the maximum and minimum mean values respectively. These high values of *ELCR*and *AGED* further amplified our concerns in the use of these tiles for building and construction purposes.

3.3. Measured concentration of the selected heavy metals in the sampled tiles

Table 5 presents the average concentration of the selected heavy metals in the sampled tiles. Varying results were observed that are in some cases 100 times higher than their corresponding global values. The maximum concentration recorded was Ni's 4772.20 ppm (IRIS Ceramic) and the minimum was found in Cd with0.02 ppm (Rose bite). The concentration of Cu ranges between 2264.17 ppm (IRIS Ceramic) and 16.08 ppm (Virony unglazed), the concentration of Pb ranges between

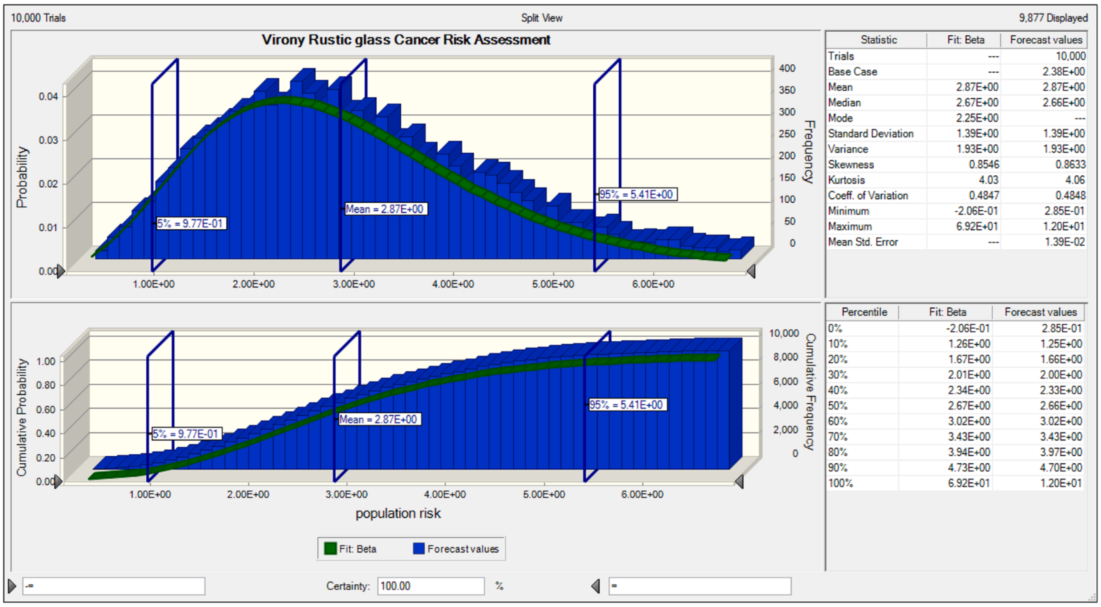

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Fig. 1. (*continued*).

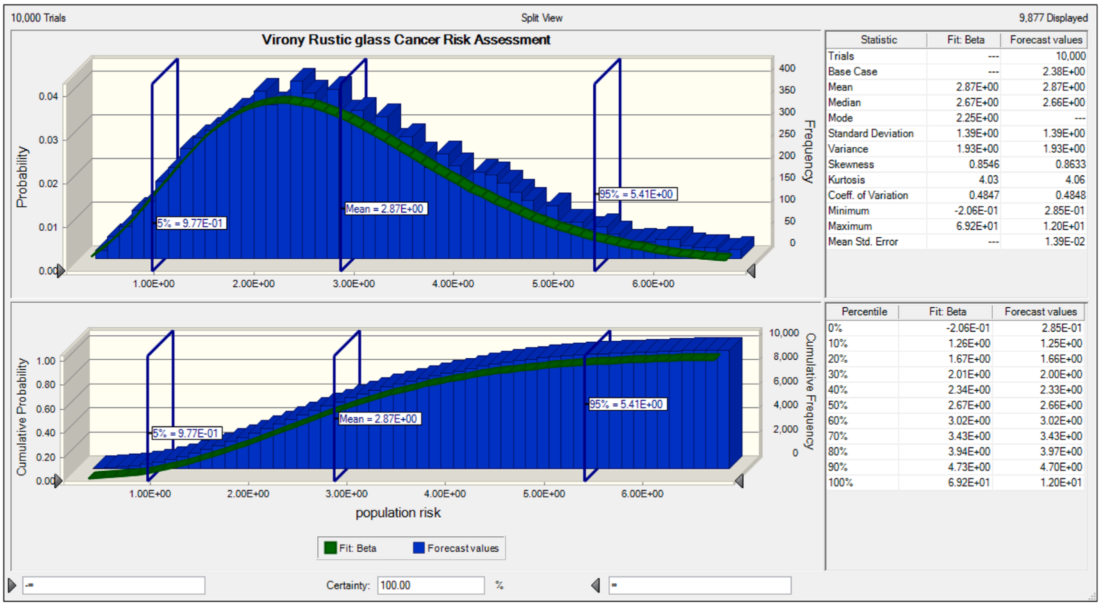
117.45 ppm (PNT Ceramic tiles) and 24.41 ppm (Rose bite). The concentration of Zn ranges between 2468.50 ppm (Royal Crown) and 43.20 ppm (Rose bite), Ni ranges between 4772.20 ppm (IRIS Ceramic) and 9.70 (Virony Rustic glass), Co ranges between 383.70 ppm (Virony Glazed) and 3.50 ppm (NISPRO Vitrified tiles). The concentration of Mn ranges between 732.00 ppm (Rose bite) and 88.00 ppm (NISPRO Vitrified tiles), the concentration of As ranges between 26.00 ppm (Virony unglazed) and 1.90 ppm (Royal Crown). The concertation of Cd ranges between 2.49 ppm (Gordwill Vitrified and Virony Glazed) and 0.02 ppm (Rose bite), and finally, the concentration of Cr ranges between 315.00 ppm (Virony Glazed) and 4.00 ppm (Rose bite). The high concentrations of these potential toxic elements call for serious worries due to the very detrimental biotoxic effects on human health. For instance, it was observed to cause "dermal lesions, skin cancer, peripheral neuropathy, and peripheral vascular disease" (Orosun, 2021). These heavy metals can get into the human system via inhalation of the tile dust and dermal contact.

3.4. *Human health risk assessment due to concentration of the selected heavy metals in the sampled tiles*

The Human health risk assessment was carried out via estimation of the human carcinogenic and non-carcinogenic risk assessment of the selected heavy metals in the sampled tiles. The estimated mean values of the $THQ_{inhalation}$, THQ_{Dermal}, and the Hazard Index (HI) are given in Table 6. Table 7 presents the estimated Incremental Life Cancer Risk (ILCR) values for the sampled tiles. The Hazard Indices (HI) expected for all the tiles are generally lower than the recommended value of 1.00 given by USEPA (2007). The highest and the lowest assessments of the hazard index (HI) are 4.40E−01 (Virony Glazed) and 1.19E−02 (NISPRO Vitrified tiles) respectively. This hence, means that the likely non-cancerous risk is small for all the sampled tiles. The Incremental Lifetime Cancer Risk (ILCR) was calculated and the values range between 6.72E and 05 (Virony unglazed) and 5.00E-06(Royal Crown). Bearing in mind that cancer hazards higher than 1.00E−4 are said to be higher as they cause greater cancer risk and values<1.00E−6 are assumed not to cause any cancer risk to human beings. For that reason, trails that the cancer hazards are within the acceptable range. Similarly, based on the Delphi classification shown in Table 1, the risk levels fall within level II.

3.5. *Monte Carlo simulation (MCS)*

Decision-makers as regards human health and environmental safety often encounter obscurities, variabilities, and uncertainties in radiological risk assessments or analyses (Changshenget al., 2012, NRC, 1994; USEPA, 1997). Estimation of the excess lifetime cancer risk (*ELCR*) using the model presented by equation (7) generally underestimate or exaggerates the actual cancer risk (Orosun, 2021; Omeje et al., 2021; Changsheng et al., 2012). Whereas underestimation of the cancer risks will result in avoidable radiological health hazards to the residents, exaggerating the risks can result in expending resources on unneeded remediation. Consequently, Monte Carlo simulation (*MCS*), a probabilistic approach, has been employed appropriately in this study to evaluate more realistic cancer risks attributed to the concentration of natural radionuclides (^{238}U, ^{232}Th, and ^{40}K) present in the sampled tiles. This technique executes the hazard analysis through building models of probable consequences by exchanging an array of values (probability distribution) for any reason with intrinsic doubt (Changsheng, 2012; Ghaderpoori et al., 2020; Orosun, 2020a, 2021; NRC, 1994; USEPA, 1997). The Monte Carlo simulation then calculates the outcomes numerous times (10,000 trials were used in this work), expending several arbitrary values from the probability functions on each occasion. That is, the MCS would take in many computations involving rate of exposure to the primordial radionuclides rather than a singular computation; in each case (computation), the model uses an assessment for each randomly selected input factor from the probability density function for that variable (Ghaderpoori et al., 2020). It takes an array of values for the input parameters, which mirrors the probability density function of each input parameter. Therefore, the recurring estimations take many haphazardly chosen mixtures of the rate of exposures and the level of activity concentrations into account, thereby working out probability densities for the outputs. Thus, a level of risk signifying 95th percentile, mean, 5th percentile or any other level of probability of interest, can be determined based on the distribution of the output. The Monte Carlo Simulations were performed using the Oracle Crystal Ball package version 11.1.2.4.850.



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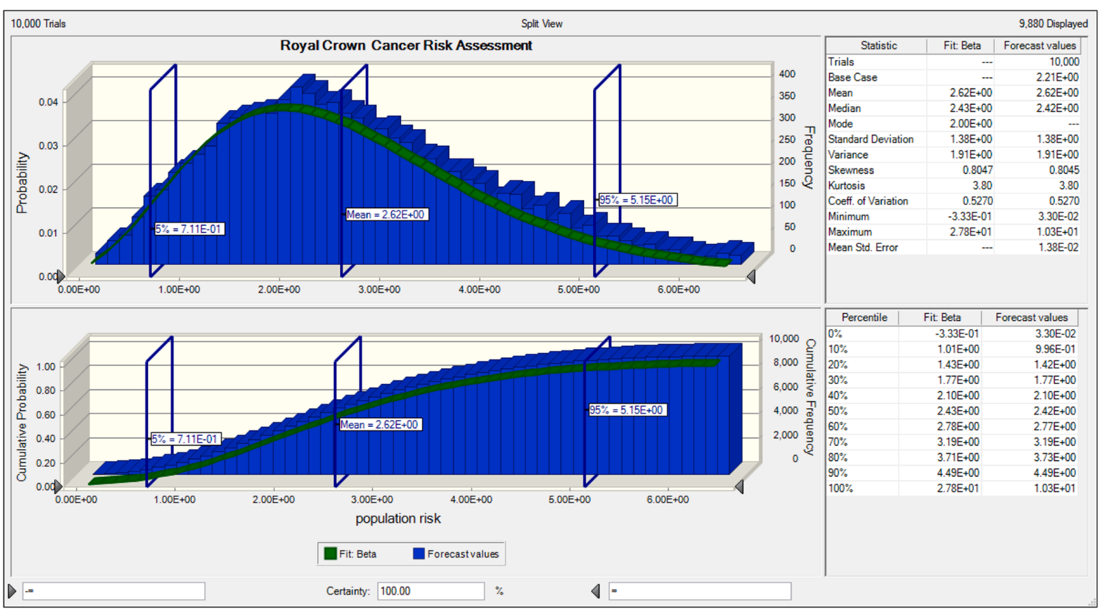

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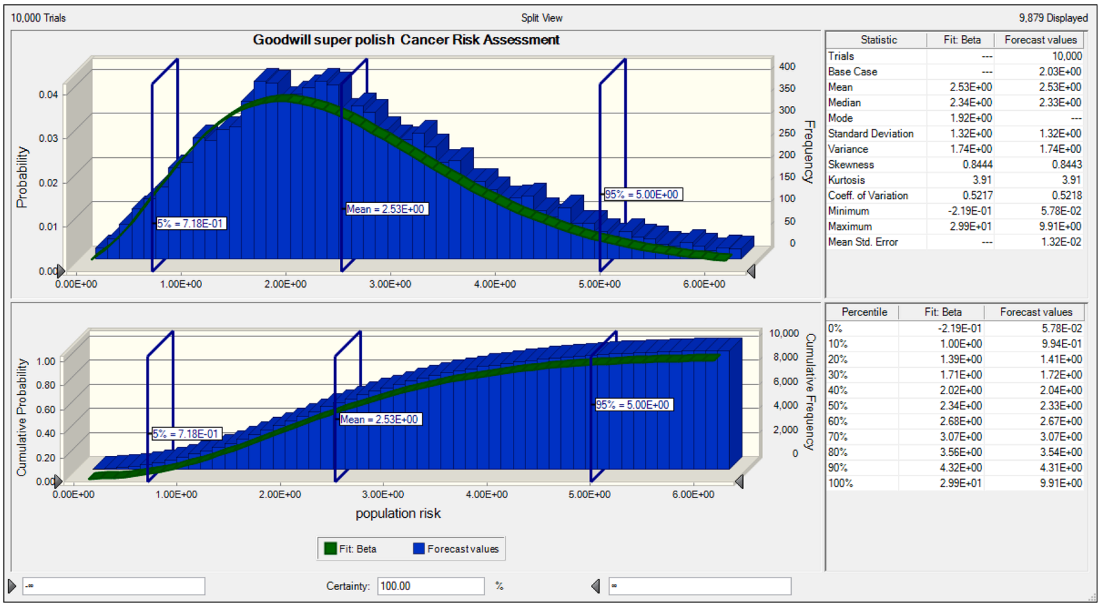
Fig. 1. (*continued*).

The summary of the simulation is presented in Table 8 and Fig. 1a-v. The P95% and mean cumulative probabilities for the lifetime cancer risks for most of the sampled tiles exceed the recommended limit of 3.75 $\times$ 10^{-3} (UNSCEAR, 2000). The results reveal that inhabitants using NISPRO Vitrified tiles, Goodwill Vitrified, Virony Glazed, IDDRIS tiles, PNT Vitrified, and PNT Ceramic tiles are most likely to experience overexposure to indoor ionizing radiation because even their P 5% cumulative probabilities(best case scenario i.e. lowest possible exposure risk) exceeds the limit recommended by UNSCEAR for indoor exposure.

4. Conclusion

This research reported the activity concentrations of ^{40}K, ^{238}U, ^{232}Th, radiological impact assessment, the concentration of Cd, Cr, Zn, Cu, Pb, As, Mn, Ni, Co, and human risk assessment of the potentially toxic elements of commonly used tiles in Nigeria. The gamma-ray analysis reveals varying results that are higher than their corresponding global values in most cases. Similarly, the analysis of the heavy metal reveals concentrations that are in some cases 100 times higher than the recommended limits. The radiological impact assessment reveals that Nigerians are at high risk of overexposure to indoor ionizing radiation for using these tiles for their building and construction purposes. This



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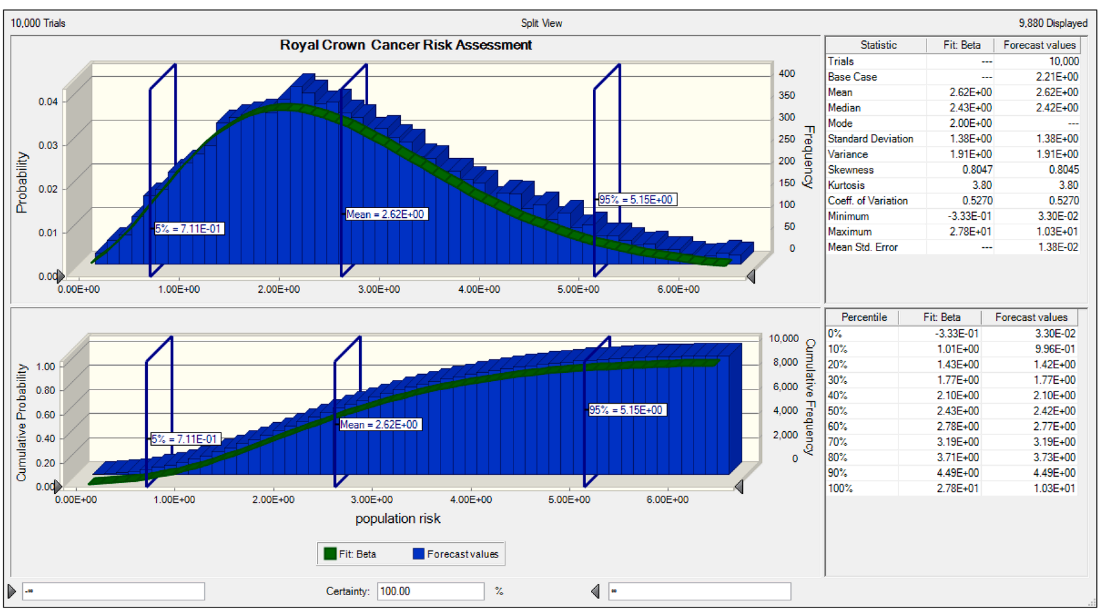

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Fig. 1. (*continued*).

high risk was confirmed by the Monte Carlo simulation, which reveals that inhabitants using NISPRO Vitrified tiles, Gordwill Vitrified, Virony Glazed, IDDRIS tiles, PNT Vitrified, and PNT Ceramic tiles are most likely to experience over-exposure to indoor ionizing radiation because even their lowest probable exposure risk exceeds the limit recommended by UNSCEAR for indoor exposure. However, the Hazard Index (HI) and the Incremental Lifetime Cancer Risk of the heavy metals reveal low cancer and non-cancer risks for all the tiles investigated.

Therefore, it is recommended that the Nigerian Environmental Protection Agency (NEPA) and other regulatory bodies should implement specific statutory requirements and laws to check and monitor all materials used for building and construction purposes. In addition, in accordance with international recommendations quoted in the Basic Safety Series No.115 from the IAEA, the use of building materials containing enhanced concentrations of these toxic elements and radionuclides should be controlled and restricted under the application of the radiation safety standards.

5. Declarations:

- *Consent for publication*: All the authors consented and approve the publication of the manuscript.
- *Consent to Participate*: All the authors consented to padticipate.
- *Availability of data and materials*: All the data and metrials are available.

6. Authors' contributions

O.M., M.M.O., A.O.O., E.O.E., and J.E.S. conceived and designed the research work, collect the data and compiled the work. M.M.O. performed the risks analysis, performed the Monte Carlo Simulations, and wrote the paper. M.R.U., O.O., and O.U.A supervised the work and final editing of the manuscript.

7. Ethics approval and consent to participate

Not applicable (No human or animal specimens are involved).

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Adewoyin, et al., 2019. Comparative assessment of natural radioactivity and radiological hazards in building tiles and sharp sand sourced locally and those imported from China and India. Int. J. Radiat. Res. 17 (3), 455–463.

ATSDR (2007). U.S. Agency for Toxic Substances and Disease Registry. http://www.atsdr.cdc.gov/mrls/ (accessed 25 November, 2020).

Bonta, M., Hegedus, B., Limbeck, A., 2016. Application of dried-droplets deposited on pre- cut filter paper disks for quantitative LA-ICP-MS imaging of biologically relevant minor and trace elements in tissue samples. Anal. Chim. Acta. 908, 54–62.

Changsheng, et al., 2012. Monte Carlo simulation-based health risk assessment of heavy metal soil pollution: a case study in the Qixia Mining Area, China. Hum. Ecol. Risk Assess.: Int. J. 18 (4), 733–750. https://doi.org/10.1080/10807039.2012.688697.

Dizman, S., Keser, R., 2019. Natural radioactivity in ceramic tiles and associated radiological hazards. Int. J. Radiat. Res. 17 (2), 245–252. https://doi.org/10.18869/acadpub.ijrr.17.2.245.

Ghaderpoori, M., Kamarehie, B., Jafari, A., et al., 2020. Health risk assessment of heavy metals in cosmetic products sold in Iran: the Monte Carlo simulation. Environ. Sci. Pollut. Res. 27, 7588–7595. https://doi.org/10.1007/s11356-019-07423-w.

Haque, et al., 2018. Carcinogenic and Non-carcinogenic Human Health Risk from Exposure to Heavy Metals in Surface Water of Padma River. Res. J. Environmental Toxicol. 12 (1), 18–23.

Hoffmann, E., Skole, J., Kriews, M., 1999. Determination of trace metals in size fractionated particles from arctic air by ETV–ICP-MS. J. Anal. At. Spectrom. 1685–1690.

IAEA, 1996. Radiation protection and the safety of Radiation sources. InternationalAtomic Energy Agency, Wagramerstrsse 5, P. O Box 100, A1400 Vienna, Austria. IAEA-RPSR-1 Rev 1.

Isinkaye, O.M., 2018. Distribution and multivariate pollution risks assessment of heavy metals and natural radionuclides around abandoned iron-ore mines in North Central Nigeria. Earth Syst. Environ. https://doi.org/10.1007/s41748-018-0035-0.

Janković, et al., 2013. Natural radioactivity in imported ceramic tiles used in Serbia. Process. Appl. Ceram. 7 (3), 123–127. https://doi.org/10.2298/PAC1303123J.

Joel, et al., 2018a. Comparative analysis of natural radioactivity content in tiles made in Nigeria and Imported Tiles from China. Sci. Rep. 8, 1842. https://doi.org/10.1038/s41598-018-20309-0.

Joel, et al., 2018b. Assessment of natural radionuclides and its radiological hazards from tiles made in Nigeria. Radiat. Phys. Chem. 144, 43–47. https://doi.org/10.1016/j.radphyschem.2017.11.003.

Joel, et al., 2018c. Assessment of natural radioactivity in various commercial tiles used for building purposes in Nigeria. MethodsX. 5, 8–19. https://doi.org/10.1016/j.mex.2017.12.002.

Li, et al., 2017. Spatial distribution and fuzzy health risk assessment of trace elements in surface water from Honghu Lake. Int. J. Environ. Res. Public Health. 14 https://doi.org/10.3390/ijerph14091011.

NRC, 1994. Science and Judgment in Risk Assessment. National Research Council. Washington, DC, USA Plum LM, Rink L.

Omeje, et al., 2018. Natural radioactivity concentrations of 226Ra, 232Th, and 40K in commercial building materials and their lifetime cancer risk assessment in Dwellers. Hum. Ecol. Risk Assess.: Int. J. 24 (8), 2036–2053.

Omeje, et al., 2020. Spatial distribution of gamma radiation dose rates from natural radionuclides and its radiological hazards in sediments along river Iju, Ogun state Nigeria. MethodsX 7, 101086.

Omeje et al., 2021. Measurements of Seasonal Variations of Radioactivity Distributions in Riverine Soil Sediment of Ado-Odo Ota, South-West Nigeria: Probabilistic Approach Using Monte Carlo. Radiation Protection Dosimetry. 2021: ncab027, doi: 10.1093/rpd/ncab027.

Orosun, et al., 2018. Radiological Safety of Water from Hadejia River. IOP Conf. Series: Earth Environ. Sci. 173, 012036 https://doi.org/10.1088/1755-1315/173/1/012036.

Orosun, et al., 2019. Natural Radionuclides and Radiological Risk Assessment of Granite Mining Field in Asa, North-central Nigeria. MethodsX. 6, 2504–2514. https://doi.org/10.1016/j.mex.2019.10.032.

Orosun, et al., 2020a. Monte Carlo approach to risks assessment of heavy metals at automobile spare part and recycling market in Ilorin, Nigeria. Sci. Rep. 10 (2020), 22084. https://doi.org/10.1038/s41598-020-79141-0.

Orosun, et al., 2020b. Radiological hazards assessment of laterite mining field in Ilorin, North-central Nigeria. Int. J. Radiat. Res. 18 (4), 895–906.

Orosun, et al., 2020c. Radioactivity levels and transfer factor for granite mining field in Asa, North-central Nigeria. Heliyon 6 (6), e04240. https://doi.org/10.1016/j.heliyon.2020.e04240.

Orosun, et al., 2020d. Dataset on radioactivity measurement of Beryllium mining field in Ifelodun and Gold mining field in Moro, Kwara State, North-central Nigeria. Data in Brief. 31, 105888 https://doi.org/10.1016/j.dib.2020.105888.

Orosun, et al., 2020e. Magnetic susceptibility measurement and heavy metal pollution at an automobile station in Ilorin, North-Central Nigeria. Environ. Res. Commun. 2 (2020), 015001 https://doi.org/10.1088/2515-7620/ab636a.

Orosun, M.M., 2021. Assessment of Arsenic and Its Associated Health Risks Due to Mining Activities in Parts of North-Central Nigeria: Probabilistic Approach Using Monte Carlo. Journal of Hazardous Materials 412 (2021), 125262. https://doi.org/10.1016/j.jhazmat.2021.125262.

Orosun, et al., 2021a. Radiological Hazard Assessment of Sharp-Sand from Ilorin-East, Kwara State, Nigeria. J. Phys.: Conf. Ser. 1734, 012040. https://doi.org/10.1088/1742-6596/1734/1/012040.

Orosun, M.M., Ajibola, T.B., Akinyose, F.C., et al., 2021b. Assessment of ambient gamma radiation dose and annual effective dose associated with radon in drinking water from gold and lead mining area of Moro, North-Central Nigeria. J. Radioanal. Nucl. Chem. 328, 129–136. https://doi.org/10.1007/s10967-021-07644-9.

Qasemi, M., Shams, M., Sajjadi, S.A., et al., 2019. Cadmium in Groundwater Consumed in the Rural Areas of Gonabad and Bajestan, Iran: Occurrence and Health Risk Assessment. Biol. Trace Elem. Res. 192, 106–115. https://doi.org/10.1007/s12011-019-1660-7.

Rinklebe, et al., 2019. Health risk assessment of potentially toxic elements in soils along the Central Elbe River, Germany. Environ. Int. 126, 76–88.

Saleh, H.N., Panahande, M., Yousefi, M., et al., 2019. Carcinogenic and Non-carcinogenic Risk Assessment of Heavy Metals in Groundwater Wells in Neyshabur Plain, Iran. Biol. Trace Elem. Res. 190, 251–261. https://doi.org/10.1007/s12011-018-1516-6.

Turhan, Ş., 2009. Radiological impacts of the usability of clay and kaolin as raw material in manufacturing of structural building materials in Turkey. J. Radiol. Prot. 29 (1), 75–83. https://doi.org/10.1088/0952-4746/29/1/005.

U.S. Environmental Protection Agency (EPA), 2004, Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment) Final; Office of Emergency and Remedial Response, EPA/540/R/99/005, OSWER 9285.7-02EP PB99-963312. July.

U.S. Environmental Protection Agency (EPA), 2007, ProUCL Version 4.00.02 User Guide: Prepared by A. Singh, R. Maichle, A. K. Singh, S. Lee, N. Armbya, EPA/600/R-07/038.April.

U.S. Environmental Protection Agency (EPA), 2009, Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part F, Supplemental Guidance for Inhalation Risk Assessment), Office of Superfund Remediation and Technology Innovation, EPA-540-R-070-002, OSWER 9285.7-82. January.

United Nuclear Corporation (UNC), 2011, Updated Baseline Human Health Risk Assessment, Church Rock Tailings Site, Church Rock, New Mexico.

United State Environmental Protection Agency "EPA" (2018). Granite-countertops-and-radiation. Updated on 3rd December, 2018 and accessed on 15th February, 2019. Available at: (https://www.epa.gov/radiation/granite-countertops-and-radiation).

UNSCEAR, 2000. Sources, effects and risks of ionization radiation, United Nations Scientific Committee on the Effects of Atomic Radiation. Report to The General Assembly, with Scientific Annexes B: Exposures from Natural Radiation Sources New York.

USEPA, 1997. Guiding Principles for Monte Carlo Analysis. Washington, DC, USA.

Zhang, Y., Hu, B., 2011. Determination of some refractory elements and Pb by fluorination assisted electrothermal vaporization inductively coupled plasma mass spectrometry with platform and wall vaporization. Spectrochim. Acta B. 66, 163–169.