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A STUDY OF NATURAL RADIOACTIVITY IN SOME BUILDING MATERIALS IN NIGERIA

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Building materials of different brands were assessed for the concentrations of ^{226}Ra, ^{232}Th and ^{40}K using HPGe detector. The activity concentrations in the measured samples ranged from 27 ± 8 to 82 ± 8 Bq kg^{-1} for ^{226}Ra, 41 ± 4 to 101 ± 8 Bq kg^{-1} for ^{232}Th and 140 ± 8 to 940 ± 19 Bq kg^{-1} for ^{40}K, respectively. The Radium equivalent (Ra$_{eq}$) activity from the samples was found to be <370 Bq kg^{-1} as the recommended value for construction materials. This study will set a baseline data for significant standards on radiation exposure of the measured radionuclides in the selected building materials used in Nigeria

INTRODUCTION

It is really of importance to understand better the risk accompanied with the exposure of a population to the radiations emitted from building materials(1). This exposure occurs on a daily basis and the ability of some radionuclides to move rapidly in air allows them to be easily transported into or within the environment in which humans come in contact with(2). There are two major aspects of radionuclides that should be considered when describing the release of radiation being exposed to by the populace, which are cosmic and terrestrial radiation(3). The terrestrial's radiation is associated with the naturally occurring radioactive materials (NORM) which are ^{238}U, ^{232}Th and ^{40}K(4). These radioactive elements present themselves as the major contributors to radiation in the environment having several effects on the general public(5). The indoor radiation exposure experienced by 80% of populace is due to these radioactive elements in the building materials as well as the ground where the construction is to be carried out(6, 7). Therefore, the aim of this present study is to establish baseline data for radiation exposure from the measured natural series radionuclides, to assess the radiological health risks, and to promote the setting standards for future references

MATERIALS AND METHODS

Sample collection and preparation for gamma analysis

Different construction material samples for this study were purchased from the Nigerian commercial markets and the river sand was scooped from a nearby river in Ota, Ogun state, Nigeria. Initial labeling and cataloging was done for easy identification. The ceramic tiles and the marbles were broken into smaller pieces so as to allow further processing. All the samples were crushed using the Pascall Engineering Lab milling machine to pulverizable size. After each tile sample was crushed, the crusher or lab milling machine was thoroughly cleaned with high pressure blower (Wolf from Kango Wolf power tools, made in London, type 8793 and serial no: 978 A) before the next sample was crushed. This whole process was repeated until all the samples were completely crushed into powder. The pulverizer used is the disk 'grinder/pulverizer' by Christy & Norris Limited. After each pulverizing process, the machine was cleaned properly and blown with high pressure blower to avoid cross contamination of the samples. A very fine power was achieved from the pulverized samples, but for homogeneity, a 250 μm sieve size was used and 1 kg of the sieved sample was weighed out. It was then placed in polythene nylon and labeled accordingly. High-density polyethylene bottles (HDPB) were used to package the samples for radioactivity study. The bottles were washed with water and detergent and then rinsed six times with ordinary borehole water before making a final rinse with distilled water. The sieved samples of ceramic tiles, cement, river channel sand (sharp and plaster) and white cements (2 Nigerian made and 1 from UAE) that were contained in each bottle weighed 200 g; there was a total of 25 samples in all.

Gamma spectrometric analysis of the selected samples

The construction materials were prepared according to IAEA TRS-295[8]. The samples were put in a Marinelli beaker and sealed for 4 weeks to allow the radium-226 progeny to reach secular equilibrium. Analysis of the samples was conducted in Canada (Activation Analysis Laboratory System) using High-Resolution Germanium detector, Canberra Lynx™ Digital Signal Analyzer (DSA), a 32 K channel integrated signal analyzer and a top-opening lead shield (4′ lead, copper/tin liner) to prevent high background counts with 50% relative efficiency and resolution of 2.1 keV at 1.33 MeV gamma energy of ^{60}Co. The Genie-2K V3.2 software locates and analyzes the peaks, subtracts background, identifies the nuclides. The efficiency curves for this analysis were corrected for the attenuation and self-absorption effects of the emitted gamma photons. CAMET and IAEA standards (DL-1a, UTS-2, UTS-4, IAEA-372 and IAEA-447) were used for checking the efficiency calibration of the system. For the activity measurements, the samples were counted for 86 400 s with the background counts subtracted from the net count. The minimum detectable activity of the detector was determined with a confidence level of 95%[8]. The measurement uncertainties were taken into account in association with the overall uncertainties of the gamma counting system which included emission probability and calibration efficiency of the system. The progeny of radium, ^{214}Bi and ^{214}Pb emits gamma line 609, 934, 2204, 1764 and 351 keV, 295 keV were used but the resolution of radium was from the emission of 1764 keV since it has low self-attenuation effect at high energy. Since ^{232}Th cannot be directly detected, its activity concentration can be estimated via its progeny ^{208}Tl and ^{228}Act using 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 used to resolve ^{40}K. The activity concentrations were calculated according to the methods of Refs.[9, 10].

RESULTS AND DISCUSSION

Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in building material samples

Table 1 presents the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K measured in the selected samples of different brands of building materials. It can be observed that the activity concentrations of ^{226}Ra from the measured samples varied between 27 ± 8 and 82 ± 8 Bq kg^{-1} with a mean value of 52 ± 6 Bq kg^{-1} for all the samples. The highest activity value of 83 ± 8 Bq kg^{-1} noted in 450 × 450 mm^2 virony ceramic tile, whereas the lowest value of 26 ± 8 Bq kg^{-1} was found in Dangote Cement (Nigeria). The level of ^{232}Th activity concentrations varies from sample to sample with the highest value of 101 ± 8 Bq kg^{-1} found in JK White Cement (UAE) and the lowest value of 41 ± 4 Bq kg^{-1} was noted in Golden Crown. The ^{232}Th activity concentrations for all the samples ranges from 41 ± 4 to 101 ± 8 Bq kg^{-1} with a mean value of 73 ± 6 Bq kg^{-1}. The activity concentrations of ^{40}K presented in Table 1 for all the samples ranges from 140 ± 8 to 940 ± 19 Bq kg^{-1} with a mean value of 217 ± 13 Bq kg^{-1}. The highest value of 940 ± 19 Bq kg^{-1} was found in PNT ceramic tile, whereas the lowest ^{40}K activity value of 140 ± 8 Bq kg^{-1} was reported in Joy White Cement (Nigeria).

Comparison of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the present building material samples and other international organization and countries

In this present study, the activity concentrations measured in the building materials were compared with the international reference value. It can be observed that the mean concentrations of 52 ± 6, 73 ± 6 and 217 ± 13 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K from this present study for tile and marble ceramics are in good agreement with the previous study by Ademola[3] for ^{226}Ra, ^{232}Th and ^{40}K with values ranging between 52 ± 2 to 131 ± 4 Bq kg^{-1}, 59 ± 1 to 127 ± 2 Bq kg^{-1} and 491 ± 12 to 979 ± 16 Bq kg^{-1} respectively. On the other hand, comparing the highest activity concentrations of cement obtained in this study for ^{226}Ra, ^{232}Th and ^{40}K with values 66 ± 8 Bq kg^{-1} (IBETO Cement (Nigeria)), 101 ± 8 Bq kg^{-1} (JK White Cement (UAE)) and 850 ± 15.4 Bq kg^{-1} (JK White Cement (UAE)) with other values of 41.3–218.9, 18.8–60.1 and 160.9–248.1 Bq kg^{-1} (India)[11], 37 ± 3, 28 ± 3 and 200 ± 14 Bq kg^{-1} (Pakistan)[13], 54 ± 13, 65 ± 10 and 440 ± 91 Bq kg^{-1} (India)[14] 55.6 ± 0.75–86.71 ± 1.64, 7.19 ± 0.10 and 348.17 ± 10.00–265.75 ± 6.40 Bq kg^{-1} (Turkey)[15], 68.3 ± 3.6, 51.7 ± 5.4 and 173.8 ± 8.6 Bq kg^{-1} (China)[16], 134 ± 67, 88 ± 35 and 416 ± 162 Bq kg^{-1} (Egypt)[17], 20 ± 5, 13 ± 3 and 247 ± 68 Bq kg^{-1} (Greece)[18], 23.4 ± 0.6, 12.2 ± 0.2 and 158.8 ± 4.3 Bq kg^{-1} (Qatar)[19], respectively. The results of this work are in good agreement with the results of a previous study of building materials conducted in Nigeria[3]. Even the specific activities for cement are in line with results published elsewhere[11–19]. In contrast, the analyzed samples of sand in this study show a distinctly higher specific radioactivity than in other studies[19–23], except for ^{40}K that reported two values which are higher by factors of 1.18 (China) and 1.50 (South Korea), respectively.

Determination of radium equivalent activity (Ra_{eq})

The level of radionuclides from ^{226}Ra, ^{232}Th and ^{40}K in the analyzed building materials is non-uniformly distributed. The Ra_{eq} activity of the measured radionuclides is used to compare the activity

Table 1. The mean values and standard deviation of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) for different types of building materials.

Sample ID	Sample origin	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Marbles				
Marble (India)	India	61 ± 4	60 ± 5	330 ± 11
Rose Marble (India)	India	56 ± 5	96 ± 7	140 ± 15
Ceramics and Tiles				
Royal Ceramics	Nigeria	58 ± 5	76 ± 6	630 ± 21
Goodwill Ceramics	Nigeria	54 ± 6	57 ± 8	240 ± 10
Royal Ceramics	Nigeria	41 ± 7	68 ± 7	380 ± 12
NISPRO	Nigeria	61 ± 3	79 ± 4	860 ± 16
Virony Glazed	China	30 ± 4	77 ± 3	290 ± 9
Time Ceramics	Nigeria	27 ± 8	96 ± 8	510 ± 14
Goodwill Vitrified	Nigeria	71 ± 3	81 ± 6	540 ± 14
PNT Vitrified Tiles	Nigeria	53 ± 4	68 ± 6	420 ± 12
PNT Ceramics	Nigeria	36 ± 7	68 ± 5	370 ± 11
IDDRIS Floor Tiles (China)	China	65 ± 11	90 ± 4	740 ± 14
Royal Ceramics	Nigeria	60 ± 2	54 ± 3	240 ± 10
Golden Crown	Nigeria	27 ± 10	41 ± 4	390 ± 12
Pumise	India	52 ± 4	51 ± 3	820 ± 15
Virony Ceramics	China	82 ± 8	42 ± 8	570 ± 13
PNT Ceramic Tile	Nigeria	56 ± 8	96 ± 9	940 ± 19
Cements				
Elephant Portland Cement (Nigeria)	Nigeria	65 ± 9	73 ± 4	170 ± 8
Perfect Superfix White Cement	Nigeria	38 ± 3	51 ± 10	360 ± 11
JK White Cement (UAE)	UAE	28 ± 2	101 ± 8	850 ± 15
Joy White Cement (Nigeria)	Nigeria	54 ± 8	92 ± 10	140 ± 8
IBETO Cement (Nigeria)	Nigeria	66 ± 8	71 ± 6	380 ± 10
Dangote Cement (Nigeria)	Nigeria	26 ± 8	68 ± 6	430 ± 13
Sand				
Sharp Sand Igboloye village, Ota (Nigeria)	Nigeria	77 ± 3	87 ± 9	670 ± 14
Mean value (s)		52 ± 6	73 ± 6	217 ± 13

of each of ^{226}Ra, ^{232}Th and ^{40}K contents in the building materials. Ra_{eq} with unit as BqKg^{-1} was calculated using the following equation[23–27]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K measured in Bq kg^{-1}, respectively. This radium equivalent activity defines the weighted sum of the individual activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The same external and internal gamma dose rate is produced from the radium equivalent activity. The maximum value of Ra_{eq} in building materials must be <370 Bq kg^{-1} as recommended by Refs.[28, 29]. This amount is equivalent to 1.5 nGyh^{-1}[30, 31]. The radium equivalent activity values obtained from the present study varies from 116 to 274 Bq kg^{-1}. The highest value was measured in Perfect Superfix White Cement (Nigeria) and the lowest value in Royal Ceramics tile. It can be observed that none of the Ra_{eq} values in all the measured samples exceeds the recommended limit of 370 Bq kg^{-1}[29].

CONCLUSION

In this present study, the activity concentrations of naturally occurring radionuclides in building materials were measured. The calculated radium equivalent was found to be lower than the recommended value of 370 Bq kg^{-1}. The mean values of the radiological parameter obtained in this present study are less than the unity, and lie within the acceptable level; thus, it can be assumed that the analyzed materials do not present radiological hazards. Currently, there is no established standard or guideline prescribing the acceptable levels of radioactivity in decorative or other construction materials in Nigeria. Significantly, there is need to introduce in both international and national levels environmentally safe reasonably standard regulations which are based on justified radiological and economical concepts.

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REFERENCES

1. Joshua, E. O., Ademola, J. A., Akpanowo, M. A., Oyebanjo, O. A. and Olorode, D. O. *Natural radionuclides and hazards of rock samples collected from Southeastern Nigeria*. Radiat. Meas. **44**(4), 401–404 (2009).
2. Gupta, M. and Chauhan, R. P. *Estimation of low-level radiation dose from some building materials using gamma spectroscopy*. Indoor Built Environ. **21**(3), 465–473 (2012).
3. Ademola, J. A. *Natural radioactivity and hazard assessment of imported ceramic tiles in Nigeria*. Afr. J. Biomed. Res. **12**(3), 161–165 (2009).
4. Kant, K., Upadhyay, S. B., Sonkawade, R. G. and Chakarvarti, S. K. *Radiological risk assessment of use of phosphate fertilizers in soil*. Iranian J. Radiat. Res. **4** (2), 63–70 (2006).
5. World Health Organization. *WHO Handbook on Indoor Radon: A Public Health Perspective* (Geneva, Switzerland: World Health Organization) (2009).
6. Sahu, S. K., Bhangare, R. C., Ajmal, P. Y. and Pandit, G. G. *Evaluation of the radiation dose due to the use of fly ash from thermal power plants as a building material*. Radioprotection **51**(2), 135–140 (2016).
7. Kendall, G. M. and Smith, T. J. *Doses to organs and tissues from radon and its decay products*. J. Radiol. Prot. **22**(4), 389 (2002).
8. Holm, E. and Ballestra, S. *Measurement of radionuclides in food and the environment, a guidebook*. IAEA Tech. Rept. Vienna, Ser; 1989.
9. Currie, L. A. *Limits for qualitative detection and quantitative determination. Application to radiochemistry*. Anal. Chem. **40**(3), 586–593 (1968).
10. Debertin, K. and Helmer, R. G. *Gamma- and X-ray Spectrometry With Semiconductor Detectors* (Amsterdam: North-Holland) (2001).
11. Davisson, C. M. and Evans, R. D. *Gamma-ray absorption coefficients*. Rev. Mod. Phys. **24**(2), 79 (1952).
12. Sharma, N., Singh, J., Esakki, S. C. and Tripathi, R. M. *A study of the natural radioactivity and radon exhalation rate in some cements used in India and its radiological significance*. J. Radiat. Res. Appl. Sci. **9**(1), 47–56 (2016).
13. Rahman, S. U., Rafique, M. and Jabbar, A. *Radiological hazards due to naturally occurring radionuclides in the selected building materials used for the construction of dwellings in four districts of the Punjab Province, Pakistan*. Radiat. Prot. Dosim. **153**(3), 352–360 (2012).
14. Erees, F. S., Dayanıklı, S. A. and Çam, S. *Natural radionuclides in the building materials used in Manisa city, Turkey*. Indoor Built Environ. **15**(5), 495–498 (2006).
15. Khandaker, M. U., Jojo, P. J., Kassim, H. A. and Amin, Y. M. *Radiometric analysis of construction materials using HPGe gamma-ray spectrometry*. Radiat. Prot. Dosim. **152**(1–3), 33–37 (2012).
16. Xinwei, L., Lingqing, W., Xiaodan, J., Leipeng, Y. and Gelian, D. *Specific activity and hazards of Archeozoic-Cambrian rock samples collected from the Weibei area of Shaanxi, China*. Radiat. Prot. Dosim. **118**(3), 352–359 (2005).
17. Ahmed, N. K. *Measurement of natural radioactivity in building materials in Qena city, Upper Egypt*. J. Environ. Radioact. **83**(1), 91–99 (2005).
18. Stoulos, S., Manolopoulou, M. and Papastefanou, C. *Assessment of natural radiation exposure and radon exhalation from building materials in Greece*. J. Environ. Radioact. **69**(3), 225–240 (2003).
19. Ding, X., Lu, X., Zhao, C., Yang, G. and Li, N. *Measurement of natural radioactivity in building materials used in Urumqi, China*. Radiat. Prot. Dosim. **155**(3), 374–379 (2013).
20. Villegas, G., Liao, W., Criollo, R. *et al*. *Detection of leaf structures in close-range hyperspectral images using morphological fusion J*. Geo-spat. Inf. Sci. **20**(4), 325–332 (2017).
21. Yang, G., Song, L., Lu, X. *et al*. *Effect of the exposure to suspended solids on the enzymatic activity in the bivalve Sinonovacula constricta J*. Aquac. Fish. **2**(1), 10–17 (2017).
22. Zhou, L. and Gui, J. *Natural and artificial polyploids in aquaculture[J]*. Aquac. Fish. **2**(3), 103–111 (2017).
23. Maxwell, O., Adewoyin, O. O., Joel, E. S., Ehi-Eromosele, C. O., Akinwumi, S. A., Usikalu, M. R., Emenike, C. P., Embong, Z. and Hassina, M. *Radiation exposure to dwellers due to naturally occurring radionuclides found in selected commercial building materials sold in Nigeria*. J. Radiat. Res. Appl. Sci. 1–7 (2018). doi:10.1016/j.jrras.2018.01.007.
24. Lee, S. C., Kim, C. K., Lee, D. M. and Kang, H. D. *Natural radionuclides contents and radon exhalation rates in building materials used in South Korea*. Radiat. Prot. Dosim. **94**(3), 269–274 (2001).
25. Ravisankar, R., Vanasundari, K., Chandrasekaran, A., Rajalakshmi, A., Suganya, M., Vijayagopal, P. and Meenakshisundaram, V. *Measurement of natural radioactivity in building materials of Namakkal, Tamil Nadu, India using gamma-ray spectrometry*. Appl. Radiat. Isot. **70**(4), 699–704 (2012).
26. UNSCEAR (United Nation Scientific Committee on the Effects of Atomic Radiation). *Sources, effects and risks of ionizing radiation*. Annex B, United Nations, New York, 1988.
27. Krisiuk, E. M., Tarasov, S. I., Shamov, V. P., Shalak, N. I., Lisachenko, E. P. and Gomelsky, L. G. *A study on radioactivity in building materials*. Res. Inst. Radiat. Hyg. Leningr. **144**, (1971).
28. Diao, S., Wang, Y., Ding, C. *et al*. *No consistent daily variation in DNA methylation detected in Populus nigra leaves by methylation-sensitive amplification polymorphism analysis*. J. Forest. Res. **28**(4), 653–660 (2017).
29. UNSCEAR (United Nation Scientific Committee on the Effects of Atomic Radiation). *Exposures from natural radiation sources*. Annex B, United Nations, New York, 2000.
30. Beck, H. L. *Exposure Rate Conversion Factors for Radionuclides Deposited on the Ground. Department of Energy* (New York (USA): Environmental Measurements Lab) (1980).
31. Akinloye, M. K., Isola, G. A. and Oladapo, O. O. *Investigation of natural gamma radioactivity levels and associated dose rates from surface soils in Ore Metropolis, Ondo State, Nigeria*. Environ. Nat. Resour. Res. **2**(1), 140 (2012).