

Physico-chemical Properties, Heavy-metal Contents and Radioactivity Levels in Soils around a Cement-production Facility in Ogun State, Nigeria

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Doi: <https://doi.org/10.47011/15.5.9>

Received on: 22/04/2021;

Accepted on: 29/06/2021

Abstract: The physico-chemical properties, heavy metal content and the primordial radionuclide activities in soil samples from four locations around a cement production facility were studied. The physico-chemical properties of the soil samples were determined after aqua regia (HNO₃:HCl) digestion and analysis with atomic absorption spectrophotometer (AAS) Perkin Elmer Analyst 200, while the activity concentration of the radionuclides was determined by a non-destructive analysis using a computerized gamma-ray spectrometry system with high-purity germanium (HPGe). The pH and electrical conductivity in the soil were in the range of 6.57 to 7.39 and 25.5 to 462 $\mu\text{s cm}^{-1}$, respectively. The cation-exchange capacity (meq/100 g) for Na⁺, K⁺, Ca²⁺ and Mg²⁺ were within the ranges 0.142 to 0.622, 0.145 to 1.725, 2.112 to 4.279 and 0.092 to 0.271, respectively. Lead and cadmium were each detected in only one sample. The average activity for Uranium -238, Thorium - 232 and Potassium - 40 in soil was between $3.177 \pm 0.330 \text{ Bq kg}^{-1}$ - $7.934 \pm 1.190 \text{ Bq kg}^{-1}$, $4.201 \pm 0.429 \text{ Bq kg}^{-1}$ - $10.702 \pm 1.504 \text{ Bq kg}^{-1}$ and $97.733 \pm 2.268 \text{ Bq kg}^{-1}$ - $144.926 \pm 21.738 \text{ Bq kg}^{-1}$, respectively. The physico-chemical properties obtained were within the range of values observed by an earlier study conducted in the area; the concentrations of the heavy metals in the soils as well as the radiological hazard indices due to the presence of naturally -occurring radionuclide materials (NORMs) in the soils were very low and are not expected to constitute any environmental hazard.

Keywords: Physico-chemical properties, Radioactivity, Cement, Ewekoro, Effective dose.

1. Introduction

Environmental quality deterioration resulting from the increase in the level of potentially toxic substances is becoming more pronounced, thus raising the question on the safety status of the environment. Industrial activities rank higher among contributors to environmental degradation [1]. Non-nuclear industries, such as cement industry, use materials containing significant contents of naturally-occurring radioactive materials (NORMs), which may expose workers or the people living near such plants to radiation above the natural background.

Cement manufacturing is known to emit about 500-1700 kg of particulate matter daily [2].

Studies have shown that soils within the vicinity of cement factories are seriously degraded [3, 4]. These studies reported an elevated deposit of alkali earth metals and heavy metals, such as lead, nickel, chromium, zinc, copper and phosphorus from cement dust and other particles with attendant environment risk. Cement dust is also known to consist of significant amounts of primordial radionuclides of Uranium-238, Thorium-232 and Potassium-40

[5]. Emission of gases and dust particulates by the cement industry can thus affect human health through the release of NORMs and metals into the air, settlements and farmlands close to the plant. The metal contents in cement dust have peculiar characteristics, one of which is their non-deterioration decay with time. Some can be necessary or beneficial to plants at certain levels, but can become toxic when specific thresholds are exceeded. They often occur as cations which strongly interact with the soil matrix and consequently move into the food chains. According to United States Department of Agriculture (USDA) [6], acute poisoning from heavy metals is rare though ingestion or dermal contact is possible. Chronic problems associated with long-term heavy metal exposures are mental lapses, allergic dermatitis (lead) toxicological effect of the kidney, liver and gastro-intestinal tract (cadmium), skin poisoning and harmful effects on kidney and the central nervous system [7]. In a similar vein, long-term exposure to radionuclides through inhalation has severe health effects, such as chronic lung disease, acute leucopenia, anaemia and necrosis of the mouth [8]. Other diseases caused by radioactivity exposure include lung cancer, pancreas, hepatic, bone, skin and kidney cancers, cataracts, sterility, atrophy of the kidney and leukaemia. Exposure to excess levels of background ionization radiation causes somatic and genetic effects that lead to critical damage of

radiosensitive organs of the body, which ultimately can lead to death. [9]

The aim of this study is to examine the possible impact of cement dust on the soil's physico-chemical properties, heavy-metal contents and natural-radioactivity levels around Wapco - Lafarge Cement Facility, Ewekoro, Ogun state. The radiological health indices following the activity levels of radionuclides in the soil will also be determined to know what health risks people living or working around the cement facility may be exposed to.

2. Materials and Methods

2.1 Study Area

The study area comprises Ewekoro, Papalanto, Arigbajo and Ifo locations in Ogun state, South-west of Nigeria within latitudes 06.9355 °N, 06.9500 °N and longitudes 03.2167 °E and 03.5000 °E. The area is predominantly a tropical rain forest with average annual rainfall of over 200 cm [10]. These areas experience alternate dry and raining seasons annually with rainy season being between March and early October and dry season running from late October to late February. The vegetation has tall trees with buttress roots and evergreen leaves. The land use of this area is mainly agricultural and residential. The approximate distances (km) of the cement factory (situated at Ewekoro) from Papalanto, Arigbajo and Ifo locations are, respectively, 1.80, 6.58 and 8.84.

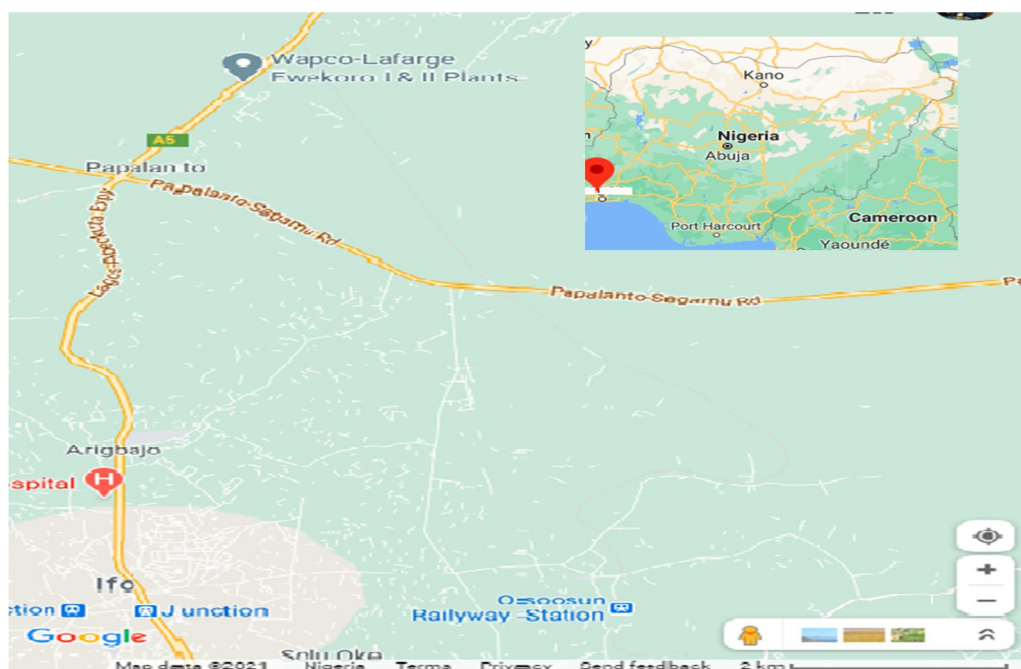


FIG. 1. Map of study locations (inset: map of Nigeria) (Google maps, 2021).

2.2 Sample Collection and Preparation

Soil samples were collected randomly from farmlands within the four locations of interest. The surface soil was taken using a clean plastic hand trowel. Composite samples of such randomly collected soil from each location were formed, from which large stones, particles of dirt or plant parts were removed. The samples were thereafter sun-dried and sieved with a 2-mm mesh to get very fine grains free of lumps. The dried fine soil particles were transferred into plastic containers, labelled and conveyed to the laboratory for analysis.

One part of the prepared soil samples was set aside for gamma analysis, while the other was set apart for physico-chemical and heavy-metal analyses. The samples meant for gamma analysis were stored in weighed Marinelli containers, sealed and afterwards allowed to stand for about four weeks prior to measurement to ensure that secular equilibrium is established between long-lived parent and their short-lived daughter radionuclides.

2.3 Sample Analysis

2.3.1 Determination of the Physico-chemical Properties of Soil and Heavy-metal Analysis

The pH value was determined using a pH meter as described by Jackson and Barak [11]. For this, 5 g of sieved soil sample was mixed with 10 ml distilled water in 1:2 ratio. The suspension was stirred intermittently with a glass rod for 30 minutes and was left for one hour. The combined electrode was inserted into the supernatant and the pH was recorded. The Electrical Conductivity (EC) was determined with a digital electrical-conductivity meter for which 5 g of sieved sample was weighed and mixed with 20 ml of distilled water (1:4) and stirred intermittently with a glass rod for 30 minutes. Thereafter, it was allowed to settle down and then the conductivity probe was inserted into the solution to take the reading.

For heavy-metal analysis, about 5 g of soil sample was weighed into a clean porcelain crucible and heated over a hot plate. The residue was then heated in a muffle furnace at 550 °C until the carbon content (organic matter) was carefully and completely oxidized (about 1 hour). The residue left was dissolved in a few drops of aqua regia (3 parts concentrated HCl + 1 part concentrated HNO₃) and then diluted with

distilled water. The solution was then filtered, rinsed very well and the filtrate made up to 100 ml in a standard volumetric flask. The resulting solution from digestion was then aspirated into the flame of the atomic absorption spectrophotometer (AAS), Perkin Elmer Analyst 200, using air acetylene flame for the metal analysis against standard metal solutions. Each metal was determined using a specific hollow cathode lamp at a specific wavelength. Total metal concentrations of heavy metals, such as Co, Cu, Cd, Pb and Zn, were determined.

The same procedure used in detecting the heavy metals was employed for determining the cation-exchange capacity (CEC).

2.3.2 Determination of Activity Concentrations

The measurements to determine the activity in the samples were carried out at the Ghana Atomic Energy Commission, Accra. The activity in the soil samples was determined by a non-destructive analysis using a computerized gamma-ray spectrometry system with high-purity germanium (HPGe). The relative efficiency of the detector system was 25%, with a resolution of 1.8 keV at 1.33 MeV of ⁶⁰Co. The gamma spectrometer is coupled to conventional electronics connected to a multichannel analyzer card (MCA) installed in a desk-top computer. A software program called MAESTRO- 32 was used to accumulate and analyze the data manually using a spread sheet (Microsoft Excel) to calculate the natural-radioactivity concentrations in the samples. The detector is located inside a cylindrical lead shield of 5 cm thickness with an internal diameter of 24 cm and a height of 60 cm. The lead shield is lined with various layers of copper, cadmium and plexiglas, each 3 mm thick.

A counting time of 36,000 seconds (10 hrs) was used to acquire spectral data for each sample. The activity concentrations of the U-238 series were determined using γ -ray emissions of ²¹⁴Pb at 351.9 keV (35.8%) and ²¹⁴Bi at 609.3 keV (44.8%) for Ra -226, and for the Th-232 series, the emissions of ²²⁸Ac at 911 keV (26.6%), ²¹²Pb at 238.6 keV (43.3%) and ²⁰⁸Th at 583 keV (30.1%) were used. The K-40 activity concentration was determined directly from its emission line at 1460.8 keV (10.7%).

2.3.2.1 Calibration of Gamma Spectrometry System

Prior to the measurements, the detector and measuring assembly were calibrated for energy and efficiency to enable both qualitative and quantitative analysis of the samples to be performed. The energy and efficiency calibrations were performed using mixed radionuclide calibration standard homogeneously distributed in the form of solid water, serial number NW 146 with approximate volume 1000 mL and density 1.0 g cm^{-3} in a 1.0 L Marinelli beaker. The standard was supplied by Deutscher Kalibrierdienst (DKD-3), QSA Global GmbH, Germany and contains radionuclides with known energies (^{241}Am (59.54 keV), ^{109}Cd (88.03 keV), ^{57}Co (122.06 keV), ^{139}Ce (165.86 keV), ^{203}Hg (279.20 keV), ^{113}Sn (391.69 keV), ^{85}Sr (514.01 keV), ^{137}Cs (661.66 keV), ^{60}Co (1173.2 keV and 1332.5 keV) and ^{88}Y (898.04 keV and 1836.1 keV) and a 1000-ml Marinelli beaker was used.

2.3.2.2 Calculation of Radionuclide Activity

The specific activity, C_i of U-238, Th-232 and K-40 for the soil samples was determined using the following expression [12]:

$$C_i (\text{Bq kg}^{-1}) = K C_n \quad (1)$$

where $K = \frac{1}{\varepsilon P_\gamma M_s}$, C is the activity concentration of the radionuclide in the sample, C_n is the count rate under the corresponding peak, ε is the detector efficiency at the specific γ -ray energy, P_γ is the absolute transition probability of the specific γ -ray and M_s is the mass of the sample (kg).

The minimum detectable activity (MDA) of the γ -ray measurements was calculated according to the formula:

$$MDA = \frac{\sigma\sqrt{B}}{\varepsilon P_\gamma W} (\text{Bq kg}^{-1}) \quad (2)$$

where σ is the statistical coverage factor equal to 1.645 at a confidence level 95%, B is the background counts for the region of interest of a certain radionuclide, T is the counting time in seconds, P is the gamma yield for any particular element, W is the weight of the empty Marinelli beaker and ε is the efficiency of the detector.

The minimum detectable activity (MDA) derived from background measurements was approximately 0.11 Bq kg^{-1} for ^{238}U , 0.10 Bq kg^{-1} for ^{232}Th and 0.15 Bq kg^{-1} for ^{40}K . Concentration

values below these detection limits have been taken in this work to be below the minimum detection limit (MDL).

2.3.3 Estimation of Radiological Health Risks

Workers at the cement factory as well as dwellers in the vicinity of Wapco Lafarge Cement Factory are at risk through the exposure to radioactive materials *via* inhalation of dust particles. The radiological health risks to these categories of people were estimated using some radiological parameters. These include absorbed dose rate, annual outdoor effective dose, excess lifetime cancer risk, external hazard, radium equivalent, gamma index and annual gonadal equivalent dose.

The absorbed dose rate in air (nGy hr^{-1}) at 1m above the ground surface due to the activity concentrations of U-238, Th-232 and K-40 was calculated using Eq. 3 [13]:

$$D (\text{nGy hr}^{-1}) = \sum(DCF_i * C_i) \quad (3)$$

where C_i is the concentration of the radionuclide of interest and DCF_i is the corresponding dose conversion factor for the radionuclide. The dose conversion factors of 0.462, 0.604 and 0.042 were respectively used for Uranium-238, Thorium -232 and Potassium -40.

The annual outdoor effective dose D_{out} , was determined (Eq. 4) using a conversion coefficient of 0.7 Sv/Gy for an absorbed dose in air to effective dose in the human body.

$$D_{\text{out}} (\text{mSv yr}^{-1}) = D (\text{nGy hr}^{-1}) \times 24 \text{ hours} \times 365.25 \text{ days} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (4)$$

where 0.2 represents the outdoor occupancy factor (assuming people spend about 20% of their time outdoors) [13].

The excess lifetime cancer risk, ELCR, was estimated with the aim of evaluating the chances of developing cancer by humans working and living around the cement factory. The ELCR is estimated using Eq. 5:

$$ELCR = D_{\text{Outdoor}} \times DL \times RF \quad (5)$$

where DL is the duration of lifetime, which is assumed to be 70 years; RF is the risk factor, which is given as 0.05 Sv^{-1} [14].

The external hazard index, H_{ex} , is estimated from Eq. 6. The implication of equation 6 is that the activity concentrations of U-238, Th-232 and K-40 are assumed to possess the same γ -radiation dose of 370, 259 and 4810 Bq kg^{-1} of

uranium, thorium and potassium, respectively [15].

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (6)$$

where C_U , C_{Th} and C_K are the activities of uranium, thorium and potassium, respectively.

The radium equivalent activity, Ra_{eq} , is a widely used radiation hazard index to estimate the suitability of any material to be utilized as a component of building construction. It is calculated based on the same assumption as that used in determining the external hazard index, H_{ex} [16]:

$$Ra_{eq} (Bq Kg^{-1}) = C_U + \left(\frac{C_{Th}}{0.699}\right) + \left(\frac{C_K}{12.99}\right) \leq 370 \quad (7)$$

The gamma index, I_γ , is one of the radiological indices used to assess human safety

when exposed to γ -radiation [13]. It is estimated based on Eq. 8.

$$I_\gamma = \left(\frac{C_U}{150}\right) + \left(\frac{C_{Th}}{100}\right) + \left(\frac{C_K}{1500}\right) \leq 1 \quad (8)$$

It is essential to estimate the annual gonadal equivalent dose (AGED), because it is the parameter that predicts whether the gonad, bone cells and marrow of humans are safe after exposure to γ -radiation or not [16]. The AGED as a result of contributions from the activity concentrations of uranium, thorium and potassium was determined using Eq. 9 [17].

$$AGED (\mu Sv y^{-1}) = (C_U \times 3.09) + (C_{Th} \times 4.18) + (C_K \times 0.314) \quad (9)$$

3. Result and Discussion

The pH, electrical conductivity (EC) ($\mu s/cm$) and the cation-exchange capacity (CEC) (meq/100 g) values are shown in Table 1.

3.1 Physico-chemical Properties

TABLE 1. Physico-chemical properties of soil samples.

Location	pH	EC($\mu s/cm$)	CEC (meq/100 g)			
			Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
Ewekoro	7.39	155.9	0.394	0.571	3.181	0.092
Papalanto	6.95	462.0	0.812	0.522	3.055	0.271
Arigbajo	6.71	91.9	0.622	0.145	4.279	0.133
Ifo	6.57	25.5	0.142	1.725	2.112	0.109

Table 1 shows the pH, EC and CEC values of soil samples from Ewekoro and its environs. The pH depicts the level of acidity or basicity of the soil samples and the values ranged from 6.57-7.39, which indicates that the soil samples are near neutral. The slightly alkaline nature in soil samples at Ewekoro could be a result of heavy cement dust (the major constituent of which is lime) deposit and percolation of the soil in the area. Comparatively, the cement-dust dispersal and deposition are greater at Ewekoro, where the cement facility is sited, than at the other locations.

The EC values ($\mu s/cm$) ranged from 25.5 in the Ifo samples to 462 in the Papalanto samples. Table 1 also shows that there is a correlation between pH and EC; the EC increased proportionally with the pH. According to Fullen *et al.* [18], the implication is that there are reasonable or significant presences of ions in the soil. However, the values are almost insignificant and this can be attributed to low formation of some soluble and ionizable inorganic salts in the soils.

The cation-exchange capacity (CEC) shows that Na⁺ ranged from 0.142 to 0.812 (meq/100 g). The soil samples from Papalanto have the highest exchangeable Na⁺, while the samples from Ifo have the lowest. K⁺ in the soil samples ranged from 0.145 in Arigbajo to 1.725 (meq/100 g) in Ifo soil. Ca⁺ was observed highest in Arigbajo samples (4.279 meq/100 g) and lowest in Ifo samples (2.112 meq/100 g). Mg⁺ increased from 0.092 in Ewekoro samples to 0.271 (meq/100 g) in Papalanto soil. The results of the CEC shows that there is no regular pattern of increase or decrease in the samples. The values of the physico-chemical properties fall within the range of values obtained by [19] in the same Ewekoro area. Noting that CEC determines the ability of soils to exchange cations, the Arigbajo samples have the lowest exchangeable cations.

3.2 Heavy-metal Analysis

The concentrations of heavy metals (mg kg⁻¹) in the soil samples are shown in Table 2.

TABLE 2. The concentrations of heavy metals (mg kg⁻¹) in soil samples.

Location	Cu	Pb	Cd	Cr	Ni	Zn	Mn
Ewekoro	0.316	ND	0.004	0.006	0.016	2.012	0.098
Papalanto	0.018	ND	ND	0.007	0.004	0.304	0.094
Arigbajo	0.016	ND	ND	0.012	0.004	1.201	0.051
Ifo	0.214	0.182	ND	0.009	0.012	0.292	0.029

ND – Not detected.

The results of the heavy-metal analysis are presented in Table 2. The results reveal that the metals were in low concentrations in the soil samples. However, Zn was observed to be higher than the other elements in all the samples. Pb and Cd were not detected in most of the samples. Pb was only found in Ifo samples, while Cd was found in Ewekoro samples. The low levels of these metals could be a result of non-presence of materials containing these metals, such as batteries, discarded metal rails, machinery parts and wastes from welding works and spray paintings [20]. Some of these heavy metals are

of economic importance to land animals. For instance, Mn in trace amounts is essential to both man and animals. Higher concentration of manganese however results in kidney failure, liver and pancreas malfunctioning, but its optimum concentration is very essential for respiratory enzymes and connective tissues' development.

3.3 Activity Concentrations in Soil

The activity concentrations of the K-40, U-238 and Th-232 in the soil samples collected from the area of study are presented in Table 3.

TABLE 3. Mean activity (Bq kg⁻¹) of primordial radionuclides in soil samples.

Location	U-238	Th-232	K-40
Ewekoro	7.934±1.190	10.702±1.504	144.926±21.738
Papalanto	6.457±0.443	8.301±0.605	97.733±2.268
Arigbajo	3.946±0.603	8.347±0.531	132.421±19.863
Ifo	3.177±0.330	4.201±0.429	128.607±2.129

The mean activity (Bq kg⁻¹) ranged between 3.177 ± 0.330 - 7.934 ± 1.190, 4.201 ± 0.429 - 10.702 ± 1.504 and 97.733 ± 2.268 - 144.926 ± 21.738, respectively for U-238, Th-232 and K-40. Uranium -238 recorded the lowest concentration in all locations, whereas K-40 has the highest concentration. The highest activity recorded for K-40 could be as a result of the abundance of K-40 in the earth crust in addition to possible use of NPK fertilizer by farmers in

the area to improve the crop yield in the soil [21].

Generally, the activity of radionuclides decreased with increasing the distance from the cement factory, as seen in Fig. 2, thus suggesting that greater deposition of cement dust occurred in the immediate environment of the cement factory than at an appreciable distance away from it.

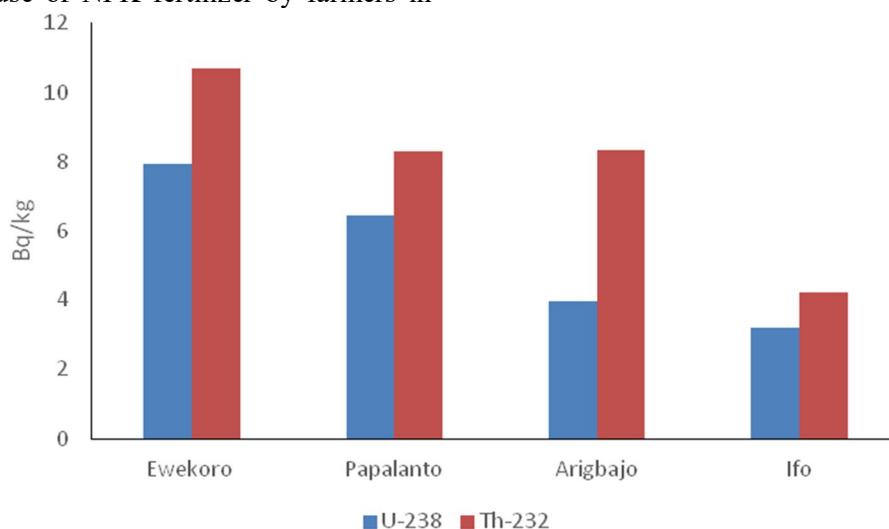


FIG. 2. Concentrations of radionuclides in soil in the locations (K-40 excluded).

The activities of U-238 and Th-232 in soils in this study compare favourably to those measured around Ewekoro by [22]. There was however a wide difference in the activity of K-40 measured. It is likely that improved farming activities which involve the use of inorganic fertilizers

(e.g. NPK) may have contributed to the enhanced level of potassium radionuclide presence in the soil. The results of the present study are also compared to those obtained in the vicinity of cement factories in other locations within and outside Nigeria (Table 4).

TABLE 4. Comparison of the mean activity of the primordial radionuclides in soils from locations around cement factories.

Location	U-238	Th-232	K-40	References
Ewekoro, Nigeria	7.92	8.62	17.45	[22]
Ewekoro, Nigeria	7.93	10.70	144.93	This study
Port Harcourt, Nigeria	49.9	5.51	473.95	[23]
Kogi, Nigeria	28.1	31.40	257.6	[24]
Pakistan	50.7	70.2	531.7	[25]
Egypt	24.8	35.3	202.5	[26]

3.4 Radiation Hazard Indices

The results of the calculated radiation hazard indices are shown in Table 5.

The absorbed dose rate was between 9.41 - 16.22 nGy h⁻¹, which is below the world's average of 57 nGy h⁻¹. The annual effective dose

was also below the world's average value and much lower than the maximum permissible limits of 1 mSv y⁻¹ (public) and 20 mSv y⁻¹ (occupational) and hence, may not have serious radiation health effects on communities.

TABLE 5. Radiation hazard indices in the locations.

Location	D (nGy hr ⁻¹)	D _{out} (mSv yr ⁻¹)	ELCR (x 10 ⁻³)	H _{ex}	Ra _{eq} (Bq kg ⁻¹)	I _γ	AGED (μSv yr ⁻¹)
Ewekoro	16.22	0.02	0.070	0.093	34.401	0.257	114.76
Papalanto	12.10	0.01	0.035	0.070	25.856	0.191	85.34
Arigbajo	12.42	0.02	0.070	0.070	26.081	0.198	88.66
Ifo	9.41	0.01	0.035	0.052	19.087	0.149	67.76
World's average [13]	57	0.07	0.29	1	370	1	300

Other radiation hazard indices computed were also below the world's average or the permissible limits and as such pose no adverse health effect to both the workers and dwellers in the vicinity of the cement factory.

4. Conclusion

The physico-chemical properties, heavy-metal concentrations and activity concentrations of soil samples from four locations situated in the vicinity of West African Portland Cement Wapco - Lafarge production facility in Ogun State were determined. The radiological hazard indices following continual exposure to the cement-contaminated soil were also estimated. The physico-chemical analysis of soil samples

under study showed variable values of the parameters. The pH of the soil indicates that the soil is near neutral. The concentrations of the heavy metals were found to be low. The cation-exchange capacity indicates that calcium ions are the most exchangeable cations. The radiological hazard indices were all below the global mean values and the permissible limits. It can thus be concluded that the concentrations of heavy metals and radioactive nuclides in the area do not constitute an environmental threat. Although the environment is radiologically safe, a community-based awareness programme on the issue of environmental contamination, its health impacts and possible prevention, is still recommended.

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