Radioprotection 2005 DOI: 10.1051/radiopro:2005027
Vol. 40, n° 4, pages 489 à 501

Article

Application of *in-situ* gamma-ray spectrometry in the determination of activity concentrations of ^{40}K, ^{238}U and ^{232}Th and mean annual effective dose rate levels in southeastern cities in Nigeria

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(Manuscript received 27 July 2005, accepted 13 September 2005)

ABSTRACT **A low cost *in-situ* gamma-ray spectrometric method for rapid assessment of radiation exposure, identification of radionuclides and detection of changes in environmental radioactivity has been employed in the determination of the natural gamma radiation dose levels in six major cities of the southeastern region of Nigeria. The soil radioactivity concentrations of the primordial radionuclides; ^{40}K, ^{238}U and ^{232}Th were determined and have been related to the γ-dose rates in the cities. The activity concentration of ^{40}K across the region ranged between 0.021 and 0.200 kBq kg^{-1}; ^{238}U was between 0.001 and 0.066 kBq kg^{-1} while that of ^{232}Th ranged between 0.015 and 0.103 kBq kg^{-1}. The total γ-absorbed dose rates in air in the region ranged between 0.016 and 0.090 μGy h^{-1} with a mean of 0.039 ± 0.018 μGy h^{-1}. The mean of the annual outdoor effective dose due to the terrestrial gamma radiation for the region was calculated as 50 μSv y^{-1} representing 66% of the world average value.**

Key words: Natural radionuclides / Activity / Effective dose / Gamma-radiation

RÉSUMÉ **Application de la spectrométrie gamma *in situ* pour la détermination des concentrations de ^{40}K, ^{238}U et ^{232}Th. Niveaux moyens des débits de dose efficace annuels dans les villes du sud-est du Nigéria.**

Une méthode de spectrométrie gamma *in-situ* à prix réduit a été utilisée pour l'évaluation rapide de l'exposition à la radioactivité γ dans l'environnement et l'identification des radionucléides dans six villes principales de la région du sud-est du Nigéria. Les concentrations en radioactivité du sol et les débits de doses ont été déterminés pour les radionucléides primordiaux ^{40}K, ^{238}U et ^{232}Th. La concentration en activité du ^{40}K à travers la région allait de 0,021 et 0,200 kBq kg^{-1}, de 0,001 et 0,066 kBq kg^{-1} pour ^{238}U et de 0,015 et 0,103 kBq kg^{-1} pour ^{232}Th. Le débit de dose absorbée dans l'air varie de 0,016 et 0,090 μGy h^{-1} dans la région. La valeur moyenne étant de 0,039 ± 0,018 μGy h^{-1}. La valeur moyenne de la dose efficace extérieure annuelle due au rayonnement gamma terrestre pour la région est estimée à 50 μSv y^{-1} ce qui représente 66 % de la valeur moyenne mondiale.

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1. Introduction

A survey meter working with a GM tube or other probes is a rapid method of assessing radiation intensity in an environment but will give only the total dose rate or exposure rate in a mixed field. Samples of soil or vegetation taken from the field and analyzed in a standard laboratory spectrometry system will reveal the concentrations of the various components of the mixed radiation field but this method has the disadvantage of being strongly inhibited by the logistic problem of carrying samples from different locations to the laboratory. An *in-situ* gamma spectrometric assembly which is accurately calibrated will yield the same information as soil samples in a much shorter time and in a much more economical manner. *In-situ* field gamma ray spectrometry means the collection of a spectrum of the ambient gamma ray flux at a given site for analysis principally to identify and quantify the radionuclides present at the site. One of the principal advantages of in-situ monitoring is that the detector samples the photon flux from a large volume of soil which averages out inhomogenities. It is the most suitable method for rapid assessment of radiation exposure, identification of radionuclides and detection of changes in environmental radioactivity.

The method was first developed in 1964 (Beck *et al*., 1964), but a lot of modifications have been made to the procedure since then (Miller and Shebell, 1993). A comprehensive report by Beck *et al*. (1972) on in-situ gamma ray spectrometry has remained a very useful document. *In-situ* gamma ray spectrometry has been applied in a wide assortment of environmental radioactivity measurements. These include the measurements of residual (^{137}Cs) levels in soils (Miller and Beck, 1984), an analysis of power plant reactor plumes (Gogolak, 1984) and survey of indoor exposure rates (Miller and Beck, 1984). It has proved to be extremely valuable in making rapid measurements of fission products in the environment during the Three Mile Island incident (Miller *et al*., 1979) and the same success was equally recorded during the Chernobyl reactor accident (Gogolak *et al*., 1986; Andrasi *et al*., 1987).

Humans are exposed to different sources of radiation in the environment of which natural sources deliver the highest radiation dose that people normally receive. The average annual dose from natural sources is ~2.4 mSv which is a reference level representing the range 1–10 mSv y^{-1} and in extreme cases to 1 Sv or more (Marouf *et al*., 1993; UNSCEAR, 2000). Natural radioactivity in an uncontaminated environment is due mainly to the primordial radionuclides, ^{238}U and ^{232}Th and their progenies and ^{40}K which are distributed in varying amounts in the earth. The present survey was conducted in the major cities of Onitsha, Enugu, Owerri, Umuahia, Port-Harcourt and Warri spreading across the region and the geological formations of the southeastern Nigeria (Oshin and Rahaman, 1986).

As a result of the economic prospects of petroleum products and coal mining in Nigeria these cities have become major economic centers in the region where up to 60% of the population now reside (NPC, 1991). Presently, the petroleum industry is the highest importer and user of different radioactive substances in the country (Elegba, 1995). In this age of wide applications of nuclear technology, the risks of abuse and accidents are high with the consequence that the natural environment is becoming increasingly vulnerable to radiation pollution. In the last three decades cancer has assumed prominence as a major source of mortality in Nigeria (Obed, 2003). Data on cancer in Nigeria has revealed high incidence in this region. It has been suggested that the increase in cancer in this region could be due to with bombs containing radioactive materials being used during the Nigeria civil war (1967–1970). To determine the extent of possible radioactive contamination, measurements must be made in this region as very little data of this type exist in Nigeria. This work is aimed at determining the dose rate levels in the 6 major cities and also to detect any environmental radioactive contamination in this area.

2. Materials and methods

2.1. Selection of sites for measurements

The cities where measurements were made are shown in Figure 1. About 8 and 11 sites were selected for measurement across the cities surveyed and a total of 58 measurements were made. Over 90% of these sites were grass covered which had not been disturbed for sometime while less than 10% were not grass covered which probably had been disturbed during building and road constructions. Efforts were made to ensure that the sites were as evenly spaced as possible and the maximum distance between any two sites was between 400 and 500 m. In order to exclude the contributions from structures and road coatings, measurements were made at the center of empty spaces with areas of at least 20 m × 20 m. This was done in order to ensure that measurement at each location represents the true outdoor γ-radiation level at the sites.

2.2. Field measurements

In situ γ-spectroscopic method of radiation measurement was employed. The system used comprise of a 76 mm × 76 mm NaI(Tl) scintillation detector (Model No 802 series) by Canberra Inc. which was coupled through a preamplifier base to a Canberra series 10 plus multichannel analyser (MCA) (Model No 1104) for spectral analysis. The detector has a resolution of about 8% at 662 keV of ^{137}Cs. This was considered good enough especially for the identification of 1460 keV, 1760 keV and 2614 keV peak energies used in this study for the measurements of

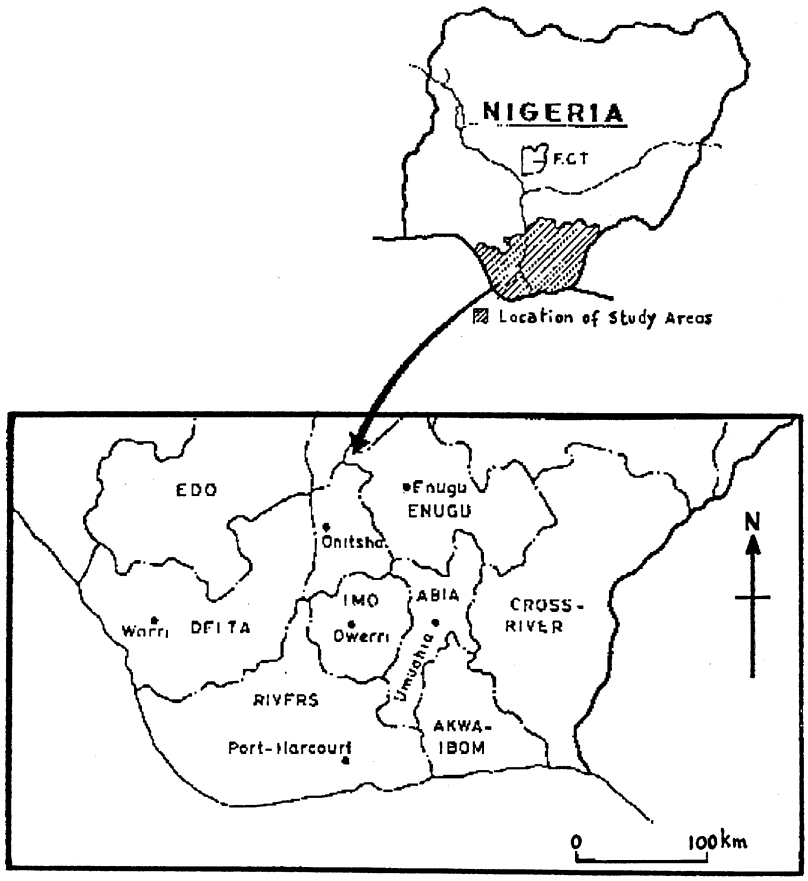

Figure 1 – ***Map of Nigeria showing the cities in the southeastern region where measurements were made. Carte du Nigéria montrant les villes où les mesures ont été faites dans la région du sud-est.***

^{40}K, ^{238}U and ^{232}Th respectively. ^{137}Cs ($T_{1/2}$ ~ 30 yrs) is very important component of fallout and its presence is often an indicator of environmental radioactive contamination from nuclear accidents and weapon tests. As a check for the presence of this radionuclide in all the surveyed locations as an indication for other artificial elements, a fourth region of interest was created around 662 keV line of ^{137}Cs.

At each measurement site, the detector was placed on a wooden stand at a height of 1m with the detector facing downward and its axis vertical, the gamma ray flux, both direct and scattered from the radionuclides in the soil were counted

for a preset time. The photopeak count rate N_f(cps) for each radionuclide at the end of each measurement time was computed from the memory of the MCA using an in built algorithm which subtracts counts due to Compton scattering of higher peaks and other background effects from total area. The counting errors for each peak determination were generally less than 25%. Measurement at each location was repeated about five times and the mean N_f determined. This was related to the soil radionuclide concentration and dose rate of each radionuclide using the calibration method described below.

2.3. Calibration for field spectrometry

The calibration process was the determination of the factors that relate the count rate under a photopeak to soil radioactivity concentration and the dose rate in air. According to Beck *et al*. (1972), the number of counts per second, N_f obtained under a photopeak due to a particular γ-energy, E, is related to the soil radioactivity concentration A of the radionuclide producing the peak by the equation:

$$\frac{N_f}{A}(E) = \frac{N_f}{N_o}\frac{N_o}{\phi}\frac{\phi}{A} \tag{1}$$

where,

$\frac{N_f}{A}$ = the photopeak count rate at the γ-energy per unit activity concentration of the radionuclide in the soil,

$\frac{N_o}{\phi}$ = the photopeak count rate per unit flux for a parallel beam of photons of energy E incident normally at the detector face,

$\frac{N_f}{N_o}$ = the correction factor for the detector response at γ-energy E to account for the fact that the flux from an extended source in the environment is not normal to the detector face but distributed across some range of angles,

$\frac{\phi}{A}$ = the gamma flux per unit activity concentrations of the soil arriving at the detector unscattered.

N_f is also related to the dose rate contribution D of the radionuclide to the environmental radiation level by:

$$\frac{N_f}{D}(E) = \frac{N_f}{N_o}\frac{N_o}{\phi}\frac{\phi}{D} \tag{2}$$

where,

$\frac{\phi}{D}$ = the gamma flux per unit absorbed dose rate in air,

$\frac{N_f}{D}$ = the photopeak count rate per unit absorbed dose rate in air.

The terms ϕ/A and ϕ/D are characteristics of the source distribution in the soil and γ-energies and do not depend on the detector. These terms can be obtained from earlier reported works (Beck *et al.*, 1972; Helfer and Miller, 1988; Miller and Shebell, 1993). The terms N_o/ϕ and N_f/N_o are detector dependent and they are essential parameters to the spectrometric assembly.

The factor N_o/ϕ represents the response of the detector to photons at normal incidence. Its value at a particular γ-energy was determined by a standard ^{152}Eu point source of known strength at a distance of 1 m from the detector face. The source was chosen because of its many γ-lines (0.122–1.408 MeV) to observe N_o/ϕ at different energies without altering the source. The flux $\phi_o(E)$ of a γ–ray energy from ^{152}Eu at the detector was determined using the expression:

$$\phi_o(E) = \frac{A \times y}{4\pi R^2} \tag{3}$$

where A is the activity, y is the γ-yield at a particular γ-energy and R is the source-detector distance (1 m). Corrections to the flux $\phi_o(E)$ in equation (3) were made in order to account for attenuation effects due to air and source encapsulation. For the air attenuation, the correction was made to $\phi_o(E)$ using the expression:

$$\phi_k(E) = \phi_o(E) \exp(-\sigma R \rho_a) \tag{4}$$

where σ is the mass attenuation coefficient for air at a given γ-energy, E and ρ_a is the density of air. The flux $\phi_k(E)$ was further corrected to account for attenuation due to source encapsulation by:

$$\phi(E) = \phi_k(E) \times q \tag{5}$$

where q is the percentage attenuation for a given gamma-ray energy due to source encapsulation. The values $\phi(E)$ and N_o/ϕ obtained for different γ-ray energies are also presented in Table I.

To determine the detector response to angular incidence of photons, the angle of incidence was varied by moving the source from 0° to 90° in steps of 10°. The count rate at various angles per unit flux at a particular γ-energy, $N_{f(E,\theta)}/\phi$ was then determined. The response of the detector to angular incidence of photons as shown

TABLE I.

Conversion ratios of natural gamma ray intensities at 1 m above the ground level.
Taux de conversion des intensités de rayonnement gamma naturel à 1 m au-dessus du niveau du sol.

Energy (MeV)	[a]Attenuation due to air ($m^{-2}kg^{-1}$)	[b]Attenuation due to source capsule	Flux ϕ_o ($\gamma m^{-2}s^{-1}$)	Corrected flux ϕ ($\gamma m^{-2}s^{-1}$)	$\frac{N_o}{\phi}\left(\frac{cps}{\gamma m^{-2}s^{-1}}\right)$	$\frac{\phi}{A}\left(\frac{\gamma m^{-2}s^{-1}}{Bqkg^{-1}}\right)$ [c]	$\frac{N_f}{N_o}$	$\frac{N_f}{A}\left(\frac{cps}{Bqkg^{-1}}\right)$
0.122	0.0143	2.5%	4760	4679	0.0175	8.50	0.70	0.104
0.244	0.0110	1.8%	1260	1243	0.0117	3.31	0.68	0.025
0.344	0.0101	1.7%	4460	4406	0.0084	1.22	0.73	0.007
0.778	0.0072	1.3%	2170	2151	0.0056	8.61	0.81	0.039
0.964	0.0065	1.0%	2450	2431	0.0046	1.08	0.86	0.004
1.408	0.0054	0.98%	3450	3428	0.0030	3.38	0.91	0.009

[a] Miller and Shebell, 1993; [b] IAEA, 1983; [c] Beck *et al.*, 1972.

in Figure 2 can be observed to be more pronounced at low γ–energies than at higher energies where response appears less sensitive to angular variation. This may be due to the fact that at low energies the photons are highly attenuated and the penetration of the photons into the detector is minimal than at higher energies where penetration is high and primary and secondary absorption occur throughout the detector (Andrasi *et al.*, 1987; Miller and Schebell, 1993).

The hypothetical situation of normal incidence is related to the real situation on the field through the determination of N_f/N_o. This quantity represents the ratio of the actual count rate under a photopeak due to an extended source in the field to the count rate that would be obtained if the source were producing only parallel flux that incident normally on the detector. If $N_f(E, \theta)/N_o$ is the ratio of the detector response at $\theta = 0°$ for γ-ray energy, E, then, N_f/N_o can be determined using the equation (Miller and Shebell, 1993):

$$\frac{N_f}{N_o} = \frac{1}{\phi}\int_0^{\pi/2} \phi(\theta)\frac{N(E,\theta)}{N_o}d\theta\,. \qquad (6)$$

The determination of N_f/N_o can be achieved by numerically integrating equation (6). This was performed using the experimental values of $N_f(E, \theta)/N_o$ and the values of $\phi(\theta)$ for different γ-ray energies from documented report (Miller and Shebell, 1993). The determined values N_f/N_o of for different γ-ray energies are also presented in Table I. By substituting the values of N_o/ϕ, N_f/N_o and ϕ/A in equation (1), the desired conversion factor, N_f/A for each γ-ray energy was obtained. The values of these ratios obtained for 76 mm × 76 mm NaI (Tl) are presented in Table I. However, due to lack of suitable gamma sources, the highest gamma energy used was for the calibration 1.408 MeV and this energy is below the gamma lines of the energy of the natural radionuclides of interest in this study.

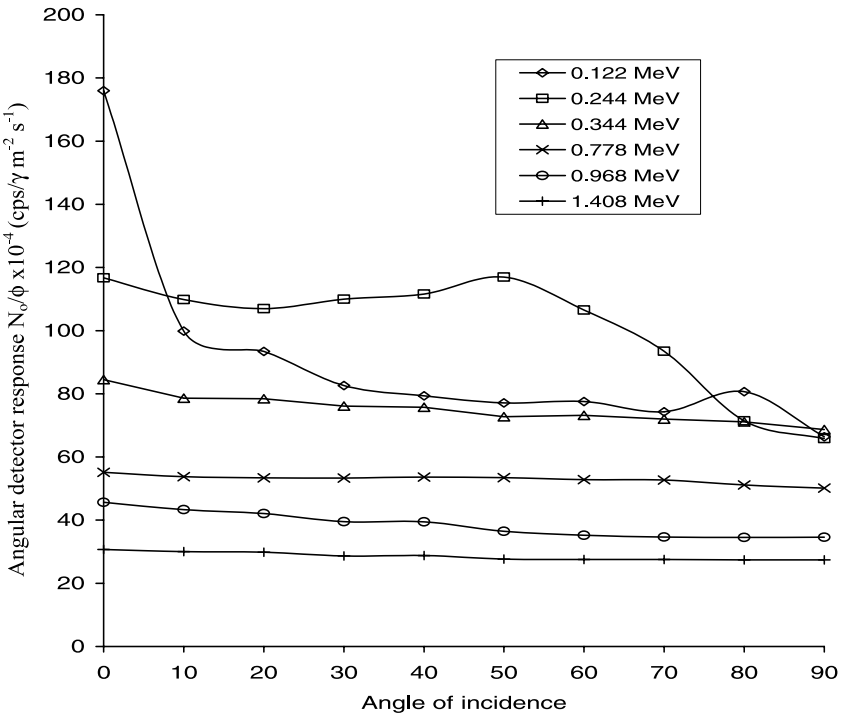

Figure 2 – ***Angular dependence of the detector response.***
Dépendance angulaire de la réponse de détecteur.

TABLE II.

Relevant conversion ratios of natural gamma ray intensities at 1 m above the ground level (Zombori *et al.*, 1983).

Taux de conversion appropriés des intensités gamma naturelles à 1 m au-dessus du niveau du sol (Zombori *et al*., 1983).

Radionuclide	N_p/D (cps/nGy h^{-1})	N_p/A (cps/Bq kg^{-1})	D/A (nGy h^{-1}/Bq kg^{-1})
^{40}K (1460 keV)	0.240 ± 0.007	0.0101 ± 0.0005	0.042
^{238}U (1760 keV)	0.0245 ± 0.0012	0.0105 ± 0.0005	0.429
^{232}Th (2614 keV)	0.0308 ± 0.0008	0.0205 ± 0.0010	0.666

In a similar calibration of 76 mm × 76 mm NaI (Tl) by Zombori *et al*. (1983) for an identical application obtained the values of the ratios as summarized in Table II. By comparing the value of N_f/A at 1.408 MeV there is an agreement in the trend. Based on this agreement the Zombori factors were, however, adopted as

TABLE III.

The range and mean of the activity concentrations of the radionuclides and outdoor absorbed dose rates in the cities.

Moyennes et variations des concentrations en activité des radionucléides et des débits de dose absorbés à l'extérieur dans les villes étudiées.

City	^{40}K (kBq kg^{-1})		^{238}U (kBq kg^{-1})		^{232}Th (kBq kg^{-1})		Total dose rate (μ Gy h^{-1})	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Enugu	0.052-0.114	0.079±0.015	0.038-0.066	0.046±0.028	0.075-0.103	0.088±0.009	0.074-0.090	0.081±0.013
Onitsha	0.085-0.200	0.128±0.033	0.007-0.028	0.015±0.006	0.031-0.054	0.036±0.009	0.023-0.060	0.038±0.006
Owerri	0.021-0.124	0.056±0.034	0.001-0.016	0.007±0.004	0.019-0.039	0.029±0.005	0.016-0.034	0.025±0.004
Umuahia	0.040-0.078	0.058±0.010	0.008-0.027	0.013±0.005	0.029-0.047	0.039±0.005	0.025-0.040	0.033±0.004
P.Harcourt	0.031-0.048	0.039±0.006	0.012-0.037	0.019±0.007	0.028-0.042	0.031±0.005	0.025-0.045	0.031±0.004
Warri	0.044-0.155	0.088±0.029	0.008-0.033	0.016±0.003	0.015-0.036	0.027±0.006	0.017-0.035	0.029±0.005

projections of the result of the calibration done in this work for the analysis. Reliability of these factors has been established through laboratory gamma-ray spectrometric measurements of activity concentration of the natural radionuclides of soil samples collected at the same sites where *in-situ* measurements were made. Both the *in-situ* and laboratory radionuclide concentrations were determined using the same detector and the values obtained were then compared. A comparison of these two sets of results were in good agreement and not significantly different at 95% confidence level (Jibiri, 2000; Farai and Jibiri, 2000). The factor *D*/*A* in the fourth column of Table II is calculated from the second and third column for each radionuclide. Its value is independent of the detector type and those given in the table are in agreement with those obtained by Beck *et al*. (1972) expressed in equation below:

$$D = 0.042\, A_k + 0.429\, A_U + 0.666\, A_{Th} \tag{7}$$

where, D is the total absorbed dose rate in nGy h^{-1}, A_K, A_U and A_{Th} are the activity concentrations of ^{40}K, ^{238}U and ^{232}Th in Bq kg^{-1}, respectively.

3. Result and discussion

The activity concentrations of the primordial radionuclides have been detected and measured in the 58 locations across the region. Ceasium-137 was not detected at any of the sites, thus indicating that the radioactivity in the region is still due to natural radioactive elements. The range and average values of the activity concentration of the natural radionuclides in each city are presented in Table III. Using equation (7), the total absorbed dose rates in air were obtained. The range and average total absorbed dose rates in the cities are also presented in Table III.

The quoted errors in Table III are the arithmetic standard deviations at the 95% confidence interval. This study shows that that variation in concentration of radionuclides in soils of this region is small with over 80% of the concentrations lying within $\pm 1\sigma$ of the average values. The specific activities of the radionuclides are generally low in this region when compared with the values obtained in a parallel study in some cities in the Northern region of the country where values are as much as 5 times higher (Jibiri, 2000). However, the generally low values obtained in the region are comparable to those reported (Jibiri and Farai, 1998) for Lagos city which has similar sedimentary formation as the southeastern region. These variations and similarities are in agreement with previous studies that terrestrial radioactivity to large extent depends on the local geology and other related factors in the environment (Wollenberg and Smith, 1990). The relatively higher value of absorbed dose rate of 0.081 ± 0.013 μGy h^{-1} obtained at Enugu in comparison to other cities in the region may be attributed to coal deposit and mining activities in the city. The frequency distribution of the average outdoor-absorbed dose rates in air in the cities is shown in Figure 3 with mode around 0.030 μGy h^{-1}. The average absorbed dose rates due to terrestrial gamma radiation dose in the region is calculated as 0.039 ± 0.018 μGy h^{-1} and it represents about 66% of the world average of 0.059 μGy h^{-1} of outdoor terrestrial gamma radiation exposure (UNSCEAR, 2000).

Applying the conversion factor of 0.7 Sv Gy^{-1} which converts absorbed dose in air to human effective dose and an outdoor occupancy factor of 0.2 as recommended by UNSCEAR (2000), the average annual effective dose due to γ-radiation from these terrestrial sources was determined for each city. This is presented in Table IV. As a further step to evaluating the collective detriment to the population due to outdoor terrestrial γ-radiation dose, the annual collective effective dose in the cities was assessed according to Publication 60 of ICRP (1991):

$$S_E = H_i N(H_i) \tag{8}$$

where, S_E is the collective effective dose, H_i average annual effective dose and $N(H_i)$ is the number of individuals in the population subgroup in the region. The results are also presented in Table IV for each city together with the population of the cities according to 1991 national census (NPC, 1991). Using the average annual outdoor effective dose calculated for the area, the collective effective dose is therefore 1.1×10^2 man-Sv y^{-1}.

4. Conclusion

A low cost *in-situ* gamma-ray spectrometric method for rapid assessment of radiation exposure, identification of radionuclides and detection of changes in

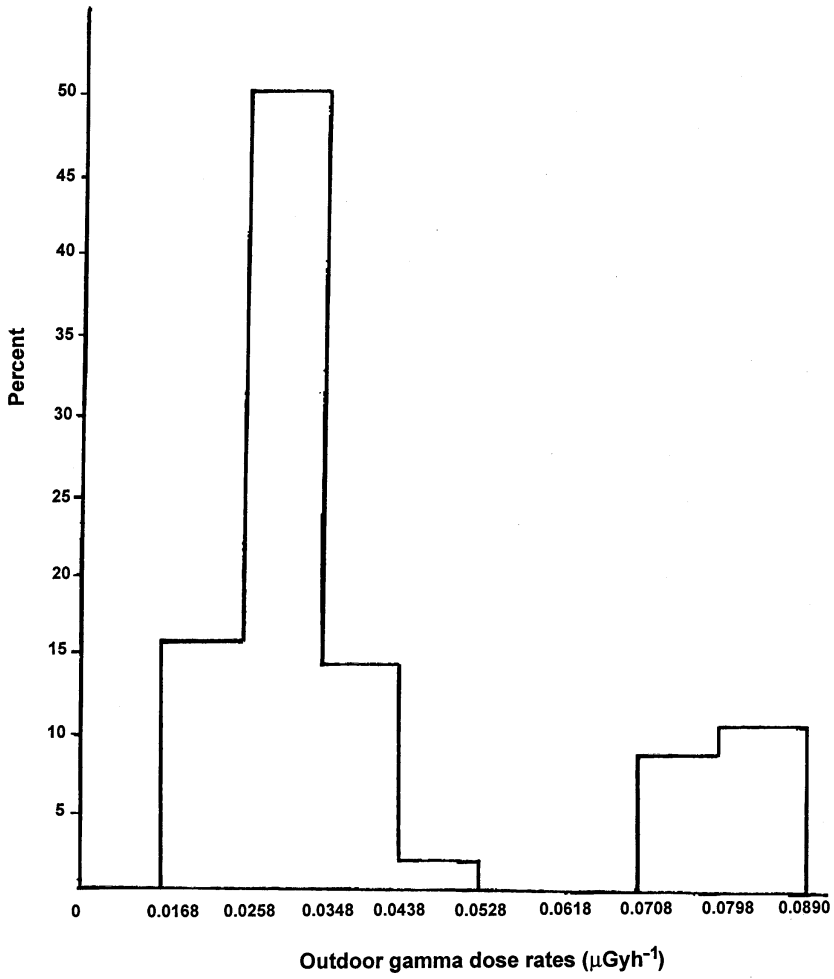

Figure 3 – ***The frequency distribution of the outdoor absorbed dose rate in air in the cities. Distribution du débit de dose absorbée dans l'air dans les villes.***

environmental radioactivity has been employed in the determination of the natural gamma radiation dose levels in six major cities of the southeastern region of Nigeria. The natural gamma radiation dose levels in the cities have been determined from the measurement of specific activities of the primordial radionuclides (^{40}K, ^{238}U and ^{232}Th). The average annual effective dose due to

TABLE IV.

The average annual outdoor effective dose, population and collective effective dose in the cities.
Dose efficace annuelle moyenne à l'extérieur, population, et dose efficace collective dans les villes.

City	Effective dose (μSv y^{-1})	Population ($\times 10^6$)	Collective effective dose (man–Sv y^{-1})
Enugu	103	0.520	54
Onitsha	49	0.287	14
Owerri	32	0.319	10
Umuahai	42	0.239	10
P.Harcourt	40	0.455	18
Warri	37	0.336	12

terrestrial gamma radiation in this part of the country was found to be 50 μSv y^{-1} representing about 66% of the world average value while the collective effective dose was calculated as 1.1×10^2 man-Sv y^{-1}. The dose levels obtained for the region suggest low and almost insignificant health burden on the population from radiation protection point of view. Results also indicate that radiation dose in the region is still due to natural radioactive sources. These results are expected to provide the background data for the evaluation of any future contamination in the environment due to local accidental releases or large scale international scale accident.

Acknowledgment. *The authors are grateful to Mr. Miller Kelvin of Environmental Measurement Laboratory (EML), US Department of Energy for sending useful materials to us. We thank the Association of African Universities (AAU) for granting a financial assistance to carry out the survey. We also wish to acknowledge International Atomic Energy (IAEA) for donating the equipment and the Federal Radiation Protection Service, University of Ibadan, Nigeria for the use of the equipment.*

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