

Natural Radioactivity Levels and Environmental Significance at Ngaka Coal Mine

Lazaro H. Meza¹, Mahmoud H. Shalaby² & Nader M. A. Mohamed³

¹Department of Nuclear and Radiation Engineering of Alexandria University, Alexandria, Egypt

²Nuclear Materials Authority, Cairo, Egypt,

³Atomic Energy Authority (ETRR-2), Cairo, Egypt

Abstract

ABSTRACT: This study aims to determine the activity concentrations of naturally occurring radionuclides and technically enhanced levels of radiation in fifteen (15) coal samples collected across the overlying and underlying rock beds from surface and sub-surface sections of northwest part of Ngaka Coal Mine Mbinga, Tanzania. Ngaka (Mbalawala project), is an open cast coal mining project located in the Ngaka coalfields area, Mbinga District in Ruvuma Region, southwest Tanzania.

Being the first of its own kind, this work aims to establish the first baseline measurements for radioactivity concentration at Ngaka Coal Mine. Representative samples from various locations across the mine have been collected and analyzed in order to establish the activity concentrations associated with the ²³⁸U/²²⁶Ra and ²³²Th natural decay chains, the long-lived naturally occurring radionuclide ⁴⁰K and the artificial man-made radionuclide ¹³⁷Cs. The activity concentration was measured via high resolution gamma ray spectroscopy using a hyper-pure germanium detector situated in a low-background environment with a copper inner-plated passive lead shield.

The average specific activity of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs (Bq kg⁻¹) in coal samples were 28.3± 1.6, 13.4±0.8, 52.8±2.7, and 5.7±0.4 Bq/kg respectively. These values lie within the expected range relative to the world average values in coal samples of 30, 35 and 400 Bq/kg respectively. The mean value of radium equivalent (51.56 Bq/kg), the external hazard index (0.14), internal hazard index (0.22), gamma dose rate (22.61 nGy/h) and annual effective dose equivalent (0.03 mSv/Y) for the complete set of samples as shown in brackets, are below the published maximal admissible values and indicate that the Ngaka Coal Mine can be regarded as having normal levels of natural background radiation. The value of gamma dose rate obtained for sample no. M3C was found to be higher than the permissible value which is 51 nGy/h. The obtained dose rate at M3C was 83.5 nGy/h. However, the value of the radium equivalent (Raeq) in the said sample was found to be just below the permissible value of Raeq of 370 Bq/kg, which corresponds to an annual effective dose of 1 mSv. It can be concluded that the measured level of the natural radiation background in the present study show that the studied area of the mine have normal levels of background radiation, with exception of sample M3C from Location MC.

Index Terms — Natural Radioactivity, Activity Concentrations, Absorbed Dose Rate, Outdoor Annual Effective Dose, Radium Equivalent Activity, External Hazard indices.

INTRODUCTION

OAL is largely composed of organic matter, but some inorganic matter occur in coal – minerals and trace elements that have been cited as possible causes of health, environmental and technological problems associated with the use of coal. Some trace elements in coal are naturally radioactive. These radioactive elements include Uranium (U), Thorium (Th) and their numerous decay products, including radium (Ra) and radon (Rn). Although these elements are less chemically toxic than other coal constituents such as Arsenic, Selenium and Mercury, but questions have been raised concerning possible risks from radiation [IAEA, (2003)].

Naturally occurring radionuclides are present in many natural resources. Elevated concentrations of these radionuclides are often found in certain geological materials, namely igneous rocks and ores. Human activities that exploit these resources may lead to enhanced concentrations of radionuclides (often referred to as technologically enhanced naturally occurring radioactive material (TE-NORM) and (or) enhanced potential for exposure to naturally occurring radioactive materials in products, by-products, residues and wastes.

The distribution of radionuclides in the geosphere depends on the distribution of the geological media from which they are derived and the processes which concentrate them at a specific location in specific media. The key to understand these distributions, therefore, is to understand the distribution of the source materials and the physical and geochemical processes that lead to elevated concentrations of radionuclides under specific conditions. The biggest producers of coal are China, USA and India, which together produce more than two thirds of the world total [IAEA, 2003].

Fossil fuels such as coal contain naturally occurring radioactivity emanated from the uranium (²³⁸U) and thorium (²³²Th) decay series

and the long lived naturally occurring radionuclide, Potassium (⁴⁰K). The concentration of Naturally Occurring Radioactive Material (NORM) in the coal, waste rocks and waste waters depends on the geological formation of coal seams. The concentration of NORM in the ash and other residues is closely related to the NORM concentrations in the coal used as fuel. Some examples of NORM concentrations in coal from selected countries are provided in Table I. Note that the concentrations can be significantly higher in some areas. For example, concentrations of NORM greater than 1000 Bq/kg in coal have been reported in Croatia [SUBASIC, 1996].

Coal is excavated in open pits (mainly in the USA and Australia) or in underground mines (mainly in China, Europe and Brazil). The excavation of coal is accompanied by the production of comparable quantities of waste rocks and large volumes of drainage water that can contain enhanced radioactivity. The mining activity can affect the overlying geological strata, creating new pathways for radon exhalation and, hence, its entry to dwellings. In such cases, elevated radon concentrations in buildings can be observed. A flow chart of the materials in a typical coal mining process is shown in Figure 01. The radioactivity of coal in seams is generally not enhanced compared with the average values for earth crust [UNSCEAR, 1993].

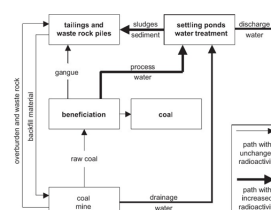


Figure 01: Flow chart of the materials in the coal mining process [IAEA, 2003]

The coal mining process itself, however, may lead to the dispersal of radionuclides from brines associated with either the coal or the host rock. Nevertheless, high radium concentrations in the outflows were reported, for example, in Poland and Germany [WYSOCKA, 2002; WIEGAND, 1999; MICHALIK, 2002].

As highlighted above that the radioactivity of coal in seams is generally not enhanced compared with the average values for earth crust and so the same is true for most of the overlying and underlying rocks excavated together with coal and deposited on the surface as spoils (Table 2, Appendix -I). Most of the residues from coal mining are deposited onto the land surface. Only a fraction of the waste rocks is used for backfilling and for other purposes. The radioactive sediments generated underground are emplaced mainly in mined-out voids. In mines, where radium co-precipitates in settling ponds, the sediments are mixed with material used for backfilling and transported back underground [ANNANMÄKI, 2000]

The objective of the present study is to establish radiological significance and or baseline measurements of radioactivity concentration at Ngaka coal mine. Activity concentration of ^{226}Ra (being in secular equilibrium with ^{238}U), ^{232}Th and ^{40}K was quantified prior to the assessment of the radiation hazard based on the findings and its impact to the mine workers and population residing at Ngaka was ascertained.

SITE INFORMATION

Site Location and Background

The Mbalawala open cast coal mining project is located in the Ngaka coalfields area, Mbinga District in Ruvuma Region, southwest Tanzania. This project, in addition to providing critical coal resources for industrial use and for power generation, it intends to contribute further towards economic and infrastructural improvement to the Tanzanians [EIS, 2011]. Figure 02 shows the location Map of the Ngaka (Mbalawala) Coal Mining Project area. The project is traversed by the mighty Ruhuhu River on the northern side of the license and this river pours its waters into Lake Nyasa.

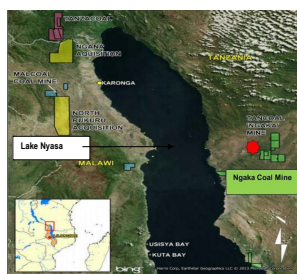


Figure 02: Location of Ngaka Coal Mine, Mbinga, Ruvuma, Tanzania [EIS, 2011]

Geology of the Mbalawala Project Area

Much of the geology is taken from the work completed by Colonial Development Company, CDC (1950's), and from the original definition of stratigraphy described by McKinley (Figure 03). No changes to stratigraphic nomenclature are interpreted or proposed as a result of the new drilling. Named lithostratigraphic units proved comparatively easy to identify in core, despite their often gradational contacts, and where drilled through in their entirety, were still observed to be surprisingly uniform in thickness. A number of units at different thicknesses were variously drilled. The total area covered by the open cast mine is approximately 4,000 m x 1,000 m excluding an area that will be covered by the initial waste dump. Open cast resources are approximately 16% of the total measured resources of 251 million tonnes. Preliminary geological study of the area indicated the presence of approximately 40.2 million tonnes of coal that can be mined by open cast method down to 600 m level.

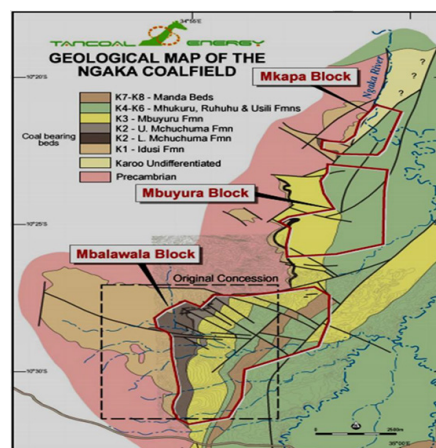


Figure 03: Schematic Geology showing the initial concession and the bounds of the Mbalawala Block as defined by the CDC during the 1950's [EIS, 2011]

MATERIAL AND METHODS

Experimental Techniques

A total of fifteen (15) samples of coal contained in overlying and underlying rock beds (Appendix – III, Figure 10) from surface and sub-surface sections of northwest of Ngaka Coal Mine Mbinga, Tanzania were collected and prepared for measurement by using gamma spectrometry analytical technique. The natural radioactive elements in the coal samples were determined using a gamma-ray spectroscopy facility at the Neutron Activation Analysis Laboratory, NAA, a property of the Egyptian Second Training Research Reactor, ETRR-2. These samples were dried, crushed and milled to 50 mesh. The mass of the milled samples varied from approximately within half a kilogram each. As a requirement for natural radioisotope analysis, samples were packed in plastic containers which were completely and air tightly sealed and kept for 4 weeks to reach secular equilibrium where the rate of decay of the daughters becomes equal to that of the parent [Mustonen, 1984]. This step is necessary to ensure that radon gas confined within the volume and the decay products will also remain in the sample.

Gamma Ray Spectrometry

Gamma ray Spectrometry technique was an experimental technique applied for determination of radioactivity concentration of natural radionuclides from the samples. In the present study, a high resolution gamma spectrometry system was used for measurement of the energy spectrum of the emitted gamma ray in the energy range of 200 to 2000 keV. The system consists of high purity germanium (HPGe) detector coupled to a signal processing units including a spectroscopic pulse amplifier and multichannel analyzer.

Two HPGe detector systems whose specifications are given below were used for sample measurements within a range of 7200 – 8640 seconds of counting live time. Detector 1 (D1): - a high resolution Gamma Spectrometry, it uses p-Type HPGe detector Model GEM-100210-Plus, Serial No. 38-P40884B, 100% efficiency and 2.3 keV resolutions at 1.33 MeV Co-60 line. Detector 3 (D3): - a high-resolution gamma ray spectrometry, it uses n-Type HPGe detector Model GMP-100 250-S and Serial No. 38-N31278A. The detector has a relative efficiency of 100% and resolution of 2.1 keV at 1.33 MeV of Co-60 line and a peak-to- Compton ratio (P/C) of 64:1 [Knoll, 2000].

Radiation Exposure and Risk Analysis

The activity levels of natural radiations have been represented by a single quantity to estimate the radiological hazard and consequently the environmental impacts of radiations. Among several radioactive nuclides,

Ra-226 is often chosen in the majority of the published papers concerned with the environmental radiation studies. This is attributed to the fact that the external exposure to the population is mostly by gamma rays emitted from the two main daughters of radium, namely Pb-214 and Bi-214. About 98.5% of the radiological effects of the U-238 series are produced by radium and its daughters. Therefore, any disequilibrium between U-238 and Ra-226 has no effect on the dose estimation from the measurement of Ra-226, and the dose rates derived from Ra-226 are usually presented as that of U-238 [JIBIRI, (2006); BERETKA, (1985)].

Outdoor external absorbed dose rate in air (DR)

The outdoor external absorbed dose rate (DR) in air 1m above the ground surface to the population is calculated from the activities of the Ra, Th and K radionuclides. It is defined as:

$$DR = CRa \cdot ARa + CTh \cdot ATH + CK \cdot AK \quad 3.1$$

where DR is outdoor external absorbed dose rate in nGyh-1, ARa, ATH and AK are the activities (Bq kg-1) of Ra-226, Th-232 and K-40 respectively. CRa, CTh and CK are the conversion factors in nGyh-1 per Bq kg-1 for Ra-226, Th-232 and K-40, respectively. These factors were taken as means of those from Beck and Planque (1968), Saito and Jacob (1995) and Clouvas et al. (2000). Their mean values are CRa = 0.4368, CTh = 0.5993 and CK = 0.0417.

Annual effective dose rate (AEDR)

The annual affective dose rate (AEDR) is estimated by using conversion coefficient from absorbed dose in air to effective dose (0.7 SvGy-1) and the outdoor occupancy factor (0.2) proposed by UNSCEAR (1993-2000) [UNSCEAR, 2000]. Also, its estimation depends on the assumption that the annual average time for exposure to radiation is 8760 hours. Accordingly, this index is given in milli Sievert per year (mSvy-1) and calculated as:

$$AEDR \text{ (mSvy-1)} = \text{Dose rate (nGyh-1)} \cdot 8760 \cdot 0.2 \cdot 0.7 \cdot 10^{-6} \quad 3.2$$

The proposed indoor occupancy factor is 0.8, (which is mainly used to determine exposure in building materials). The recommended values of AEDR are 20 mSvy-1 for occupational members and 1 mSvy-1 for the public [ICRP, 1990; IAEA, 1996].

Radium equivalent activity (Raeq)

The distribution of Ra-226, Th-232 and K-40 in any concerned sample is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity in Bq kg-1 to compare the specific activity of materials containing different amount of Ra, Th and K. It is defined as:

$$Raeq = ARa + 1.43ATH + 0.077AK \quad 3.3$$

where ARa ATH and AK are activities (Bq kg-1) of Ra-226, Th-232 and K-40 respectively. The maximum value of this index must be <370 Bq kg-1 for keeping the external dose <1.5 m Gy y-1 [AKHTAR, 2005].

External hazard index (Hex)

The radium equivalent activity was modified by other quantity index named as the external hazard index. This index measures the external hazard due to gamma-radiation and is defined as:

$$H_{ex} = \frac{A_{Ra}}{370 \text{ Bq kg}^{-1}} + \frac{A_{Th}}{259 \text{ Bq kg}^{-1}} + \frac{A_K}{4810 \text{ Bq kg}^{-1}} \leq 1$$

where Hex is the external hazardous index and ARa, ATH as well as AK are the specific activities (Bq kg-1) of radium, thorium and potassium, respectively. The value of this index must be lower than unity in order to keep the radiation hazard insignificant. The maximum value of unity for this index corresponds to the limit of 370 Bq Kg-1 for Raeq.

Internal hazard index (Hin)

In addition to the external irradiation, radon and its short-lived products are also hazardous to respiratory organs. The internal exposure to radon

and its daughter products is quantified by the internal hazard index (Hin). This index is given by the following equation:

$$H_{in} = \frac{A_{Ra}}{185 \text{ Bq kg}^{-1}} + \frac{A_{Th}}{259 \text{ Bq kg}^{-1}} + \frac{A_K}{4810 \text{ Bq kg}^{-1}} \leq 1$$

The value of this index must be lower than unity to keep the radiation hazard insignificant.

3.3.6 Radioactivity level Index (I_γ)

This index is used to estimate the level of radiation risk, especially γ-rays, associated with natural radionuclides in specific materials. It is defined as:

$$I_{\gamma} \text{ (Bq Kg}^{-1}) = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}$$

where ARa, ATH, AK are the activity concentration of Ra-226, Th-232 and K-40 in Bqkg-1 respectively. According to the European committee (EC), this index is derived for identifying whether a dose criterion is met. The index is correlated with the annual dose due to the excess external gamma radiation caused by superficial material. The value of I_γ ≤ 2 corresponds to 0.3 mSv/y, while the I_γ ≤ 6 corresponds to 1mSv/y. According to this dose criterion, the material with I_γ ≥ 6 should be avoided, since this value corresponds to dose rate higher than 1 mSv-1.

RESULTS AND DISCUSSIONS

Tables (1-3) under Appendix – II summarizes results of terrestrial radionuclides concentration and estimation of radiation hazard indices of Ngaka Coal mine. The summary shows that the average specific activity of 226Ra, 232Th, 40K and the man- made 137Cs (Bq/kg) in coal samples are 28.3± 1.6, 13.4±0.8, 52.8±2.7, and 5.7±0.4 Bq/kg respectively. These values lie within the expected range relative to the world average values in coal samples of 30, 35 and 400 Bq/kg respectively. With exception of sample M3C which measures external dose rate of 83.5 nGy/h, exceeding the World average of 51 nGy/h, other radioactive indices measures within the permissible maximal limits as recommended by ICRP 1990 [ICRP, (1990), IAEA, 1996] and related World Organizations. Figures (04 – 09) shows different graphical representation of Activity concentrations of Terrestrial radionuclides and profiles of radiation hazard indices. With exception of the influence of sample M3C in respective radiation hazard quantities, all other indices/quantities lie within acceptable limits.

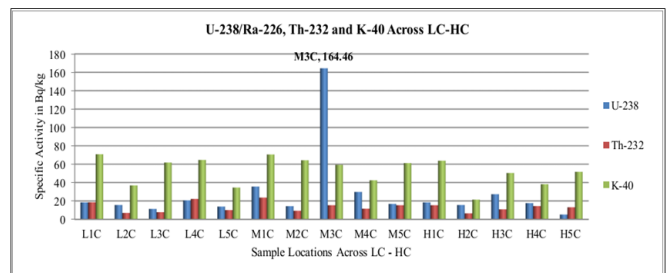


Figure 04: Terrestrial Radionuclides concentration for samples in vertical orientation Across LC-HC

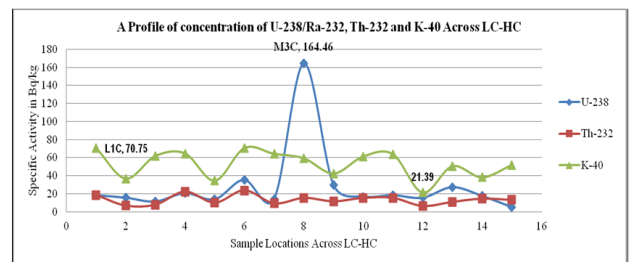


Figure 05: A Profile of Terrestrial Radionuclides concentration with Depth (across LC – HC)

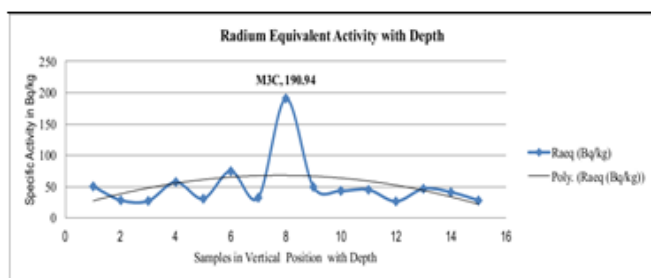


Figure 06: Radium Equivalent Activity with Depth

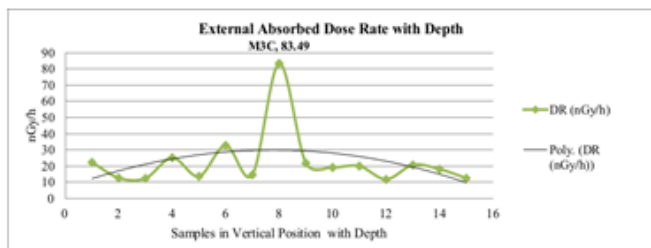


Figure 07: External Absorbed Dose Rate with Depth

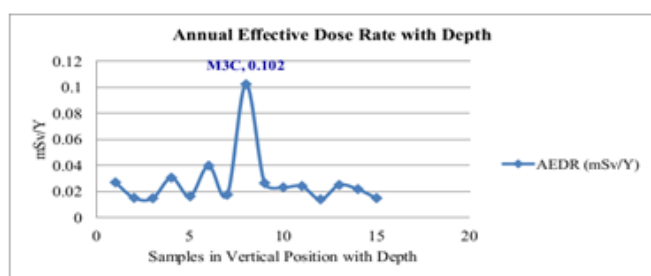


Figure 08: Annual Effective Dose Rate with Depth

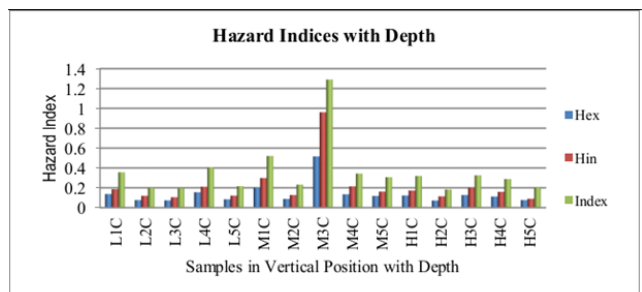


Figure 09: External (Hex), Internal (Hin) hazard & Radioactivity level (I) Indices with Depth

CONCLUSION

The baseline radioactivity measurement at Ngaka coal mine has been established. At least for the time being there is no any serious radiological threat to workers and to the members of the public however, as the mine activity continues further studies are strongly recommended in order to monitor any possible changes in radioactivity levels and so establish future radioactivity working levels at the mine and the region around Ngaka Coal mine.

ACKNOWLEDGMENT

The Author(s) are exceedingly humbled to express deep appreciations to the International Atomic Energy Agency, IAEA, for sponsoring AFRA postgraduate programmes in Africa. It was during the course of one of the IAEA/AFRA programmes at the Department of Nuclear & Radiation Engineering of Alexandria University when this paper was published. An outstanding support from Prof. Alya A. Badawi, the Coordinator AFRA

Postgraduate Programme (Egypt) & Head of Department of Nuclear & Radiation Engineering of Alexandria University is highly appreciated. Also, the author is grateful to Prof. Dr. Eng. Iddi Mkilaha, the Director General & CEO of the Tanzania Atomic Energy Commission, TAEC, for his encouragement and support to this work. Author's unreserved thanks go to the management of Egyptian Second Training Research Reactor, ETRR-2 (Inshaas) for the permission to use NAA-Lab (HPGe) for sample measurement and analysis. The kind support and commitment of NAA – Lab's group is exceptionally appreciated.

REFERENCES

- [1] IAEA, (2003), (International Atomic Energy Agency), "Extent of Environmental Contamination by Naturally Occurring Radioactive Material (NORM) and Technological options for Mitigation" Technical Report Series 419, Vienna, (pp. 1-26).
- [2] SUBASIC, D. (1996) "Restoration of radioactively contaminated sites in the Republic of Croatia", Planning for Environmental Restoration of Radioactively Contaminated Sites in Central and Eastern Europe, Vol. 2, IAEA-TECDOC- 865, Vienna, pp 55–79.
- [3] USCEAR, (1993), (United Nations Scientific Committee On The Effects Of Atomic Radiation), "Sources and Effects of Ionizing Radiation", United Nations Sales Publication No. E.94.IX.2, UN, New York, pp 912.
- [4] WYSOCKA, M., (2002), "Assessment of Radon Risk in Upper Silesia - Influence of Geology and Mining Activity", PhD Thesis, Central Mining Institute, Katowice (2002) (in Polish).
- [5] WIEGAND, J.W., SEBASTIAN, F., (1999) "Origin of radium in high-mineralized waters", Technologically Enhanced Natural Radiation (TENR II) (Proc. Int. Symp. Rio de Janeiro, 1999), IAEA-TECDOC-1271, International Atomic Energy Agency, Vienna (2002) 107–111.
- [6] MICHALIK, B., CHALUPNIK, S. (2002) "Contamination of settling ponds of coal mines caused by natural radionuclides", Technologically Enhanced Natural Radiation (TENR II), (Proc. Int. Symp. Rio de Janeiro, 1999), IAEA- TECDOC-1271, International Atomic Energy Agency, Vienna, pp. 19–29.
- [7] ANNANMÄKI, M., TURTIAINEN, T., (2000), "Disposal of Radioactive Waste Arising from Water Treatment: Recommendations for the EC", Final Report of the WP8 of the TENAWA Project, Rep. STUK-4175, STUK, Helsinki, pp. 77.
- [8] EIS, (2011), "Environmental Impact Statement for Mbalawala (Ngaka Coal Fields) Coal Mine", Mbinga, Tanzania.
- [9] MICHALIK, B., (2002), "A Complex Method of Estimation of Risk due to the Natural Radioactivity of Mining Waste Tailings", PhD Thesis, Central Mining Institute, Katowice (in Polish).
- [10] Mustonen R, (1984). Radioactivity in and Radon Exhalation from Finnish Building Materials, Health Phys. 46, 1195–1203 1984.
- [11] UNSCEAR, (2000), (United Nations Scientific Committee Of The Effects Of Atomic Radiation). "Sources and Effects of Ionizing Radiation". Report on General Assembly, United Nations New York.
- [12] JIBIRI, N.N, BANKOLE, O.S, (2006), "Soil Radioactivity and Radiation Absorbed Dose Rates at Road sides in High- Traffic Density Areas in Ibadan Metropolis, Southwestern Nigeria", Journal Radiation Protection Dosimetry. 118 (4), pp453–458.
- [13] Knoll, F.G, (2000), "Radiation Detection and Measurement", Third Edition, John Wiley & Sons, Inc.
- [14] BERETKA, J. and P.J. MATHEW, (1985). "Natural radioactivity of Australian building materials, industrial wastes and by-product". Healthy Physics, 48: 87-95.
- [15] ICRP, (1990), (International Commission On Radiological Protection). "Recommendations of the International Commission on Radiological Protection". Annals of the ICRP 21(1-3). ICRP Publication 60. Pergamon Press, Oxford, 1991.
- [16] IAEA, (1996), (International Atomic Energy Agency), "International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources", Safety Series No. 115, IAEA, Vienna.
- [17] N. AKHTAR, M. TUFALL, M. Ashraf, A. Mohsin, M. Iqbal, (2005) "Measurements of environmental radioactivity for estimation of radiation exposure from saline soil of Lahore", Pakistan, Radiation. Measurements. 39, pp. 11–14.