Measurement of Hazard Indices due to Natural Radioactivity in Dust around Ririwai Artisanal Tin mine Kano State – Nigeria

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ABSTRACT
Instrument neutron Activation analysis (INAA) was used to determine the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K is soil dust inside a Tin mining pits around Ririwai artisanal tin mine. Hazards indicates such as Radium equivalent activity Ra_{eq}, Gamma index $I\gamma index H_{in}$ and external index H_{ex} were also estimated. The results show an elevated activity concentration in ^{226}Ra and ^{232}Th with mean concentration of 54.31±4.51Bq/kg and 146.64±0.91Bq/kg respectively as compared with world average value of 33Bq/kg and 45.Bq/kg in soil for ^{226}Ra and ^{232}Th respectively. The mean activity concentration of ^{40}K in the dust samples is 385.90±5.70 Bq/kg which is lower than 420Bq/kg for world average value in soil. The estimated hazard indices shows that Ra_{eq} has a mean value of 266.98 Bq/kg which is lower than the world average value of 370 Bq/kg, the gamma index has a mean value of 1.043 while the mean values of internal and external hazard indices are 0.972 and 0.793 respectively. The hazard indices estimated are within the tolerable limit.

Keywords: Radionuclides, Activity concentration, Gamma index, Hazard indices.

1.00 INTRODUCTION

Human are continuously exposed to radionuclides through ingestion and inhalation (internal exposure) and/or irradiation from external gamma rays emitted from the radionuclide (external exposure). Naturally occurring radioactive materials are present in several types of materials. Materials which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as radium, uranium, thorium, potassium, and their radioactive decay products, that are disturbed as a result of human activities. However the concentration of NORM in most natural substances is so low that the risk is generally regarded as negligible. Higher concentrations may arise as a result of human activities. In most NORM, several or all of the radioactive isotopes of the three primordial decay series (^{235}U, ^{238}U and ^{232}Th) are present in small concentrations in the natural matrix. The radionuclides in the decay series are more or less in radiological equilibrium however, this equilibrium becomes disturbed through human activities such as mining and mineral processing, resulting in either an enrichment or depletion of some of the radionuclides concentrations compared to the original matrix. This disequilibrium is as a result of differences in the properties of the radionuclides in the series, due to geochemical migration processes and differences in their half-lives [Cember, 1996; UNSCEAR, 2000; Sato and Endo, 2001].

Uranium like most heavy metals is chemically toxic and accumulates in kidneys (soluble) and also on bones. The dominant uranium valence states that are stable in geologic environments are uranous (U^{4+}) and uranyl (U^{6+}) states with uranyl being more soluble than the uranous [NRC, 1991]. In the near-surface environment, U and Th may both be mobilised but in different ways. Even though in a naturally undisturbed environment, uranium is generally more soluble than thorium. At low pH, such as in acid-leach uranium mill, thorium becomes more soluble. For instance acid-leach milling might dissolve 30-90 % of the thorium in the ore [NRC, 1991]. Thorium has extremely low solubility in natural waters and is entirely transported in particulate matter. Potassium has 24 known isotopes three of which occur naturally: ^{39}K (93.3%), ^{40}K which is the radioactive isotope of terrestrial importance (0.0117%) and ^{41}K (6.7%). Naturally occurring ^{40}K decays to stable ^{40}Ar (11.2%) by electron capture and by positron emission, and decays to stable ^{40}Ca (88.8%) by beta emission. During the decay process out of 100 disintegrations, 89 results in the release of beta particles with maximum energy of 1.33 MeV and 11 results in the release of gamma photons with maximum energy of 1.46 MeV. Potassium-40 (^{40}K) decays by beta (β-) emission to ^{40}Ca and by electron capture (E. C.) to ^{40}Ar. It has a half-life of 1.250×10^{9} years. In healthy animals and people, ^{40}K represents the largest source of radioactivity, greater even than ^{14}C. In a human body of 70 kg mass, about 4,400 nuclei of ^{40}K decay per second. The activity of natural potassium is 31 Bq/g [Knoll, 1989]

There are several pathways by which the radioactive material can reach humans. The pathway largely depends on the processes involved and can be broadly categorised into; on-site, off-site, airborne, waterborne, food products, etc [O'Brien et al, 1998]. For on-site pathways, the 38 exposures tend to be direct from external gamma radiation or internal exposure resulting from inhalation of radioactive dust or radon progeny. Due to the presence of NORM in most soils and rocks, underground mining activities can lead to enhanced levels of radioactive dust, and radon isotopes and other radioactive isotopes [O'Brien and Cooper, 1998]. In open-pit mining, ventilation cannot be controlled and work practices have to be carefully controlled to minimize the radiological risk to the on-site workers [O'Brien et al., 1998].

Off-site exposure situation assessments also involve analysis of the potential exposures to humans living within or near the site where NORM is likely to be produced. Exposure to NORM under this situation will normally result from the transfer through environmental pathways or the use of industrial wastes containing NORM. Off-site exposure pathways are normally more indirect and complex and members of the public become the target of exposure. For instance transfer of radionuclides through the food chain [Dahlgaard, 1996] by river and oceanic transport [McDonald et al., 1996], by atmospheric deposition, by re-suspension

of radioactive dust, etc are some of the possible pathways through which members of the public may be exposed to radiation.

On-site external exposures in industrial or mining situations could be due to the presence of NORM in stockpiles, waste piles, storage tanks, build-up of surface contamination on equipment, in pipes and storage tanks. External exposures to members of the public (off-site) can result from exposure to gamma radiation from passage of cloud shine or exposure to gamma radiation from material deposited on the ground (ground shine) [O'Brien et al., 1998]. The dominant exposure pathways in most situations are external gamma radiation, inhalation of radon gas and its decay products, ingestion of contaminated food and/or water [O'Brien et al., 1998]. The guiding principle in controlling the radiological impact of NORM in all these situations is the ALARA principle [ICRP, 1977] which states that all exposures should be kept as low as reasonably achievable (ALARA), social and economic factors taken into account. In this work instrumental neutron activation analysis using Nigeria's Research Reactor 1 (NRR1) was used to determine the hazard indices due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil dust collected from active mining pits around Ririwai Artisanal Tin Mining areas in Kano State Nigeria.

2.00 MATERIALS AND METHODS

The study area is Ririwai town headquarter of Doguwa Local Government Area in the extreme south of Kano State, Nigeria. It has an area of 1,473 km² and a population of 151,181 at the 2006 census. Figures 1 shows the Map of Kano State showing the study area. Table 1 shows the locations where samples were collected.

Dust samples were collected mainly within the mines inside active pits using air sampler. The air sampler was powered using a power generator continuously, dust samples were collected on filter papers attached to the suction part of the air sampler. In order to prevent dust being collected on the filter paper emanating from other sources the air sampler was placed inside the active pits. The sampling was carried out for about 3 hours where dust of average weight of 0.20 mg was collected.

The preparation started from the time of sample collection where the weight of the filter paper was measured before placing it at the suction part of the air sampler. After a period of 3 hrs the weight of the filter paper and dust content were measured, the actual weight of the dust content were then recorded and the filter paper containing the dust were then sealed in a polyethylene bag. The filter paper containing the samples were wrapped in a polyethylene and then placed in 7 cm^3 rabbit capsules. The polyethylene and rabbit capsules containing the samples were cleaned by soaking in 1:1 HNO_3 (Nitric acid) and then washed with de-ionised water in order to eliminate every contamination prior to sample irradiation in addition two blank filter papers were also analyzed using Nigerian Research Reactor 1 (NRR1) at the Centre for Energy Research and Training, Ahmadu Bello University Zaria, Nigeria.

Table 2 below was used to convert radio element concentration to specific activity while equations 1,2,3 and 4 were used to calculate the hazard indices.

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.0077C_k \quad \ldots\ldots 1$$

$$I_Y = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_k}{3000} \quad \ldots\ldots 2$$

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_k}{4810} \quad \ldots\ldots 3$$

$$H_{ex} = \frac{C_{Ra}}{187} + \frac{C_{Th}}{259} + \frac{C_k}{4810} \quad \ldots\ldots 4$$

Fig. 1: Showing map of Kano State and the study area

Table 1: Sampling locations

S/No	North	East	Elevation
1	10^0 44′ 35.3″	008^0 45′ 16.4″	856m
2	10^0 44′ 36.7″	008^0 45′ 15.8″	856m
3	10^0 44′ 33.8″	008^0 45′ 17.8″	856m
4	10^0 44′ 32.3″	008^0 45′ 21.0″	858m
5	10^0 44′ 30.3″	008^0 45′ 27.0″	862m
6	10^0 43′ 48.2″	008^0 44′ 57.1″	896m
7	10^0 43′ 49.1″	008^0 44′ 53.4″	894m
8	10^0 43′ 48.5″	008^0 44′ 53.0″	895m
9	10^0 43′ 50.2″	008^0 44′ 58.7″	892m
10	10^0 43′ 49.5″	008^0 44′ 59.2″	894m

3.0 RESULTS AND DISCUSSIONS

The N.A.A analysis gives the elemental concentrations in part per million (ppm) from where the radioelement concentration of U, Th and K were extracted and the concentration of K in ppm was converted to percentage Table 3 gives the radioelement concentration in ppm U, Th and K%.

Activity Concentration

Using the conversion factors for radioelement concentration to specific activity in Table 2 the activity concentrations in Bq/kg for ^{226}Ra, ^{232}Th and ^{40}k in Dust were calculated and tabulated in Table 4.

Hazard indices

The radiological hazard indices calculated from the concentrations of natural radionuclides in Dust samples were radium equivalent activity (Ra_{eq}), gamma index **(Iɣ)** and external (H_{ex}) and internal hazards indices (H_{in}). These hazards indices were calculated using Equations 1 Radium equivalent activity (Ra_{eq}), gamma index **(Iɣ)** 2, external hazard (H_{ex}) equation 3 and internal hazard (H_{in})equation4and the results is summarsed in Table 5

Descriptive statistics and one way analysis of variance (ANOVA) at the 5% level of significance was employed and the results have shown that ^{226}Ra has a mean activity concentration of 54.31 ± 4.51 Bq/kg in range between 0 to 290.23 ± 8.60 Bq/kg, ^{232}Th has a mean value of 146.64 ± 0.91 Bq/kg with minimum and maximum values of 45.07 ± 0.41Bq/kg and 200.16 ± 1.22 Bq/kg respectively while ^{40}K has a mean value of 385.90 ± 5.70 Bq/kg with lowest and highest values of 140.85 ± 1.57 Bq/kg and 1039.16 ± 14.71 Bq/kg respectively.

Similarly the hazard indices calculated due to the activity concentration above shows that R eq activity has a mean value of 266.98 Bq/kg with minimum and maximum values of 120.82 and 475.89 Bq/kg respectively. The gamma index Iɣ has a mean value of 1.043 in range between 0.483 to1.934. The internal hazard index H_{in} has a mean value of 0.927 in range between 0.400 to 2.157 while the external hazard index H_{ex} has a mean value of 0.793 with minimum and maximum values of 0.364 and 1.486 respectively.

Table 2: Conversion of radio element concentration to specific activity (IAEA 2003).

1%k in rock/soil	=	313 Bq/kg ^{40}K
1 ppm of U in rock/soil	=	12.35 Bq/kg ^{238}U or ^{226}Ra
1 ppm of Th in rock/soil	=	4.06 Bq/kg ^{232}Th

Table 3: Neutron activation analysis for dust sample showing concentrations of U and Th in ppm and K in %

S/N	Sample Code	U in ppm	Th in ppm	K in %
1	RP_1	2.53 ± 0.28	40.20 ± 0.20	0.98 ± 0.15
2	RP_2	1.19 ± 0.140	52.20 ± 0.20	0.54 ± 0.007
3	RP_3	1.10 ± 0.180	18.20 ± 0.20	0.65 ± 0.017
4	RP_4	1.15 ± 0.120	41.60 ± 0.20	0.45 ± 0.005
5	RP_5	1.88 ± 0.260	37.60 ± 0.23	1.27 ± 0.190
6	RP_6	BDL	49.30 ± 0.30	0.91 ± 0.010
7	RP_7	1.49 ± 0.150	31.90 ± 0.20	0.63 ± 0.008
8	RP_8	2.79 ± 0.360	48.50 ± 0.30	1.78 ± 0.027
9	RP_9	8.35 ± 0.44	11.10 ± 0.10	1.79 ± 0.032
10	RP_{10}	23.50± 0.70	30.60 ± 0.30	3.32 ± 0.047

Table 4: Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/Kg in dust samples

S/N	Sample Code	^{226}Ra(Bq/Kg)	^{232}Th(Bq/Kg)	^{40}K(Bq/Kg)
1	RP_1	31.25 ± 3.46	163.21 ± 0.81	306.74± 4.70
2	RP_2	14.70 ± 1.73	211.9 ± 0.81	169.02 ± 2.19
3	RP_3	13.5 ± 2.22	73.89 ± 0.81	203.45 ± 5.32
4	RP_4	14.20 ± 1.48	168.890 ± 0.81	140.85 ± 1.57
5	RP_5	23.22 ± 3.21	152.66 ± 0.93	397.51 ± 5.95
6	RP_6	BDL	200.16 ± 1.22	287.83 ± 3.13
7	RP_7	18.40 ± 1.85	129.51 ± 0.81	197.19 ± 2.50
8	RP_8	34.46 ± 4.45	196.91 ± 1.22	557.14 ± 8.45
9	RP_9	103.12 ± 5.43	45.07 ± 0.41	560.14 ± 8.45
10	RP_{10}	290.23 ± 0.86	124.24 ± 1.22	1039.16 ± 14.71
	Mean	**54.31±2.51**	**146.64±0.91**	**358.90±5.70**

Table 5: Radium equivalent, activity external and internal hazard indices

S/N	Sample Code	Ra $_{eq}$. Bq/kg	Gamma Index (I_γ)	External Hazard (H_{ex})	Internal Hazard (H_{in})
1	RP_1	267.00	1.002	0.777	0.860
2	RP_2	319.06	1.165	0.892	0.932
3	RP_3	120.82	0.483	0.364	0.400
4	RP_4	256.79	0.938	0.719	0.757
5	RP_5	244.58	0.973	0.735	0.796
6	RP_6	288.45	1.097	0.833	0.833
7	RP_7	205.12	0.775	0.591	0.639
8	RP_8	320.33	1.286	0.969	1.060
9	RP_9	171.78	0.756	0.569	0.841
10	RP_{10}	475.89	1.934	1.486	2.157
	Mean	**266.98**	**1.043**	**0.793**	**0.927**

CONCLUSSION

The mean activity concentration of ^{226}Ra and ^{232}Th obtained in this are 5 4.31 ± 2.5 and 146.64 ± 0.91 Bq/kg respectively. These values are higher than 33 Bq/kg and 45 Bq/kg world average values in soil for ^{226}Ra and ^{232}Th respectively while the activity concentration of 358.90 ± 5.70 Bq/kg for ^{40}K is less than 420 Bq/kg world average value in soil. The exemption levels of ^{226}Ra, ^{232}Th and ^{40}K recommended by the ICRP and adopted by the IAEA in any material are 10Bq/g, 1Bq/g and 100Bq/g for ^{226}Ra, ^{232}Th and ^{40}K respectively (ICRP 1991, IAEA 1996). The values obtained in this work are lower than the above exemption levels. Similarly the Radium equivalent activity Ra_{eq}. obtained with a mean value of 266.98 Bq/kg is lower than the internationally accepted value of 370 Bq/kg. The gamma index Iɣ obtained in this work with mean value of 1.043 is almost the same with $I\gamma \leq 1$ lower limit and less than $I\gamma \leq 6$ upper limit recommended by EC (1999). The internal and external hazard indices have mean values of 0.927 and 0.793 respectively both these value are lower than the internationally accepted recommended values of 1.00. The results obtained in this work fall within the safety levels and does not pose significant hazards to the miners and the surrounding communities.

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