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Activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K and gamma radiation absorbed dose rate levels in farm soil for the production of different brands of cigarette tobacco smoked in Nigeria

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Background: Presently, the British-American Tobacco Company (BAT) located in Ibadan, Oyo State is the only tobacco producing company in Nigeria accounting for about nine different brands of cigarette tobacco smoked in the country. The tobacco leaves are produced from some farmlands located in Oke-Ogun area of Ibadan where special fertilizers are used for the growing of the tobacco leaves. The use of this fertilizer may enhance natural radionuclide contents in the farm soils and through root uptake be accumulated in the leaves. In this study, soil samples from three farmlands used for the production of the tobacco leaves were collected in order to determine the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the farm soil. ***Materials and Methods:*** Measurements for the determination of the activity concentrations were carried out using gamma-ray spectrometry comprising a lead-shielded 76 mm × 76 mm NaI(Tl) detector crystal (Model 802 series, Bicron NaI) coupled to a Canberra series 10 plus multichannel analyzer (MCA) (model 1104) through an preamplifier base. ***Results:*** Results of measurements showed that the average activity concentrations of the natural radionuclides in the soil samples across the three farms varied from 2550.4± 154.6 to 3208.9 ± 188.7 Bq kg^{-1} for ^{40}K, 33.1±11.9 to 39.9±9.3 Bq kg^{-1} for ^{226}Ra, while for ^{232}Th it varied between 51.98± 8.4 and 56.08±17.51 Bq kg^{-1}. The activity concentrations of these radionuclides and gamma absorbed dose rates due to the concentration of these radionuclides were found to be higher than world average values while those of the control farm values were lower to world averages and the tobacco leaf farms. ***Conclusion:*** From the results obtained the fertilizer type used on the farmlands for the tobacco leaf production was very rich in ^{40}K with typical values greater 2000 Bq kg^{-1}. Also there may be possible high signatures of ^{210}Pb and ^{210}Po in the tobacco leaves and the cigarette samples smoked in the country. This is a subject for future research consideration given the fact that alpha radiation plays a major role in the etiology of lung cancer. **Iran. J. Radiat. Res., 2011; 8 (4): 201-206**

Keywords: *Activity concentration, natural radionuclides, absorbed dose rate, tobacco farm soil, smoked cigarette, Nigeria.*

INTRODUCTION

In recent times cancer is assuming greater importance in health agenda throughout the world, in developing as well as in the developed countries. In Nigeria, it has assumed prominence as a major cause of mortality in the last few decades. A rough estimate of incidence for malignant neoplasm in Ibadan was 33.7 and 45.1 per 100,000 for males and females, respectively in the 1960s and presently about 100,000 new cases of cancer are reported every year. Given the current population projections, it is expected that by the year 2010 about 500,000 cases will occur annually [1]. The common malignant tumours in Nigeria are those of; (i) breast, (ii) cervix, (iii) liver, (iv) skin, (v) lymphoid tissue except Burkitt's lymphoma, (vi) prostate, (vii) connective and soft tissues, (viii) Burkitt's lymphoma and (ix) stomach. The effects of low-level radiation (~10^{-3} mGy) on human carcinogenesis have been investigated on a large scale during recent years [1, 2]. Even though most of these studies could not agree that an increase of cancer mortality is correlated with increasing exposure, some of them showed an excess of mortality for some specific malignant diseases. Public interest in the long-term effects of ionizing radiation on humans has started to gain prominence following the establishment of a nuclear

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regulatory body in Nigeria and has been largely focused on carcinogenic effects from protracted exposure to low doses because the cancer risk of this kind of exposure has major implications to public health and radiation standards [3]. While some regions of the country especially the eastern and western parts of the country showed more prevalence of cancer cases of different types other regions do not. The cause of these incidences has not actually been related to any specific agent though cancer incidence is multifactoral, but to the extent each causative agent contributes to the overall cancer burden in a population is very important.

Studies have shown that tobacco contains minute quantities of isotopes of ^{210}Pb, ^{210}Po and ^{226}Ra from uranium and thorium-decay series which are radioactive and carcinogenic. The smoking of tobacco and its products increase the internal intake and radiation dose due to these radioisotopes [4]. Also in a number of studies, inhalation of some naturally occurring radionuclides via smoking has been considered to be one of the mort significant causes of lung cancer [5, 6]. The major source of the polonium is phosphate fertilizer which is used in growing tobacco and the trichomes of the leaves concentrate the polonium which could persist even when tobacco leaves are dried and processed [7-9]. Presently, the British-American Tobacco Company (BAT) located in Ibadan, Oyo State southwestern Nigeria is the only tobacco producing company in the country accounting for about nine different brands of cigarette tobacco smoked in the country. The tobacco leaves are produced from some farmlands located in Oke-Ogun (Ilua and Jobele areas) of Ibadan where special fertilizers are used for the growing of the tobacco leaves. These farms are major suppliers of tobacco leaves to British America Tobacco (BAT) Company, the major tobacco company in Nigeria. The detrimental effects of tobacco have been considerably under estimated making it less likely that chemical carcinogens alone are responsible for the observed incidences of tobacco-related carcinoma. The contribution of cigarette tobacco to overall cancer incidences in the country is not known or non existent. It is therefore imperative that basic study like this be carried out in order to investigate radioactivity levels as a first step in the farm soil used in the production of different brands of tobacco products smoked in Nigeria. It is also known that tobacco companies use chemical phosphate fertilizers, which is high in radioactive metals year after year on the same soil. These metals build up in the soil, attach themselves to resinous tobacco leaf and ride tobacco trichomes in tobacco smoke [10]. It is therefore expected that the radioactivity level in tobacco would vary widely depending on where and how it is grown. This work is aimed at investigating the radioactivity of the soil in farmlands where tobacco leaves are grown in order to determine:

1. activity concentration of the natural radionuclides in the farm soil used for the production of the tobacco leaves
2. estimate the radiation absorbed dose due to the concentration of natural radionuclides in the tobacco soil farms
3. useful radiometric signature data for ^{210}Po and ^{210}Pb likely contents in different brands of Nigerian cigarette tobacco samples.

MATERIALS AND METHODS

Sample collection

The tobacco leaf growing towns are Ilua and Jobele in Oke Ogun part of Oyo State Nigeria. Three tobacco leaf growing farmlands were chosen from the two areas. Two farms were chosen from Ilua while one farm was chosen from Jobele. The control farm was chosen from a farm where tobacco leaf is not grown but a cultivated farm. The choice was based on the fact that they are the major supplier of tobacco leaves to British-America Tobacco (BAT) Company, the only tobacco company in the country. In order to ensure a good representative sampling at the three chosen tobacco leaf

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growing farmlands and the control farmland, the farms were divided into several grids of sampling points of about 20 m^2 each. The numbers of grids depend on the size of each farm. Samples of soil were collected across the grids to a depth of 150 mm below the ground surface from about four points in each grid. They were thoroughly mixed together to represent a sample for that point.

Preparation of samples

The soil samples were dried at 110°C in a temperature controlled oven until there was no detectable change in the mass of the sample. The dried samples were thoroughly crushed, grounded and pulverized to powder. The powder was passed through a 2 mm sieve. Due to the limited space of the detector shield only 200 g of the soil samples (dry weight) were used for analysis. The samples after weighing were transferred to radon-impermeable cylindrical plastic containers of uniform size (60 mm height by 65 mm diameter) and were sealed for a period of about 30 days. This was done in order to allow for Radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy. The reference soil was also transferred to a container of the same material and dimensions as were used for the soil samples. This is to ensure that the geometry configuration remained the same. The standard reference soil sample used was prepared from Rocketdyne Laboratories California; USA which is traceable to a mixed standard gamma source (Ref. No. 48722-356) by Analytic Inc. Atlanta, Georgia.

Method of Measurement

The detector used for the radioactivity measurements is a lead-shielded 76 mm × 76 mm NaI(TI) detector crystal (Model No. 802 series, Canberra Inc.) coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model No.1104) through a preamplifier. It has a resolution (FWHM) of about 8% at energy of 0.662 MeV (^{137}Cs) which is considered adequate to distinguish the gamma ray energies of interest in the present study. The choice of radionuclides to be detected was predicated on the fact that the NaI(Tl) detector used in this study had a modest energy resolution. Hence the photons emitted by them would only be sufficiently discriminated if their emission probability and their energy were high enough, and the surrounding background continuum low enough. Therefore, the activity concentration of ^{214}Bi (determined from its 1.760 MeV γ-ray peak) was chosen to provide an estimate of ^{226}Ra (^{238}U) in the samples, while that of the daughter radionuclide ^{208}Tl (determined from its 2.615 MeV γ-ray peak) was chosen as an indicator of ^{232}Th. Potassium-40 was determined by measuring the 1.460 MeV γ-rays emitted during its decay. The samples were placed symmetrically on top of the detector and measured for a period of 10 hours. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks. From the net area, the activity concentrations in the samples were obtained using [11-14]:

$$C\left(Bq\ .kg^{-1}\right) = kC_n \qquad (1)$$

Where $k = 1/\varepsilon P_\gamma M_s$, C is the activity concentration of the radionuclide in the sample given in Bq kg^{-1}, C_n is the count rate under the corresponding peak, e 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 detection limit of a measuring system describes its operating capability without the influence of the sample. The detection limit (DL) given in Bq kg^{-1}, which is required to estimate the minimum detectable activity in a sample, was obtained:

$$DL\left(Bq\ kg^{-1}\right) = 4.65\frac{\sqrt{C_b}}{t}.k \qquad (2)$$

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where C_b is the net background count in the corresponding peak, t_b is the background counting time (s), k is the factor that converts cps (counts per second) to activity concentration ($Bq\ kg^{-1}$) as given in equation 1. With the measurement system used in the present work, detection limits obtained were 17.3 Bq kg^{-1}, 4.2 Bq kg^{-1}, and 5.1 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th, respectively. Values below these numbers were taken in this work as being below the detection limit (BDL) of the detector.

RESULTS AND DISCUSSION

Radionuclide concentrations

Using equation 1 the activity concentrations of the soil samples were calculated. The results are presented in tables 1, 2 and 3 for farms A, B and C respectively, while the activity concentration of the soil in control farm is presented in table 4. The error terms in the measured values in the tables represent the counting error. As could be observed from the tables, the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th for farm A ranged from 2340.99±195.24 to 2783.89±231.34 Bq kg^{-1}, 9.53±7.09 to 43.67±15.33 Bq kg^{-1} and 34.05±4.88 to 64.24±8.61 Bq kg^{-1}, respectively. For farm B, it ranged from 2868.04±238.05 to 3394.91±283.10 Bq kg^{-1}, 29.44±12.41 to 56.43±17.38 Bq kg^{-1} and 39.84±5.67 to 65.14±8.72 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. For farm C, activity concentrations varied from 2926.63±242.91 to 3394.81±281.43 Bq kg^{-1}, 27.20±11.60 to 46.20±17.38 Bq kg^{-1} and BDL to 71.64±9.61 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. For the control farm it ranged from 88.45±8.25 to 201.93±20.46 Bq kg^{-1}, 26.78±11.01 to 73.46±20.56 Bq kg^{-1} and 6.64±1.12 to 34.23±4.74 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. Results indicate that averagely, the concentration of the radionuclides is higher in farms A, B and C in comparison to the control farm. Most pronounced in the values of the activity concentrations of the radionuclides is that of ^{40}K in farms A, B and C in which values were typically greater than 2000 Bq kg^{-1}. The ^{40}K concentrations in these farms are between 2 to 5 orders of magnitude higher than those reported in normal and high radiation background areas of the country [12-16]. This is a clear indication of the effect of the preferential fertilizer type used in tobacco leaf production.

Table 1. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil samples from tobacco farm A.

S/No	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	Total absorbed dose rate (nGy h^{-1})
1	2403.69±200.47	9.53±7.09	54.61±7.38	141.46
2	2733.04±227.12	16.54±9.49	56.38±7.63	159.43
3	2393.43±199.61	42.76±14.99	64.24±8.61	161.65
4	2340.99±195.24	42.13±14.80	48.49±6.62	148.69
5	2783.89±231.34	35.40±13.81	60.04±8.11	172.10
6	2382.32±198.45	43.67±15.33	58.44±7.88	157.72
7	2587.02±215.24	36.73±13.97	54.19±7.38	160.50
8	2494.19±207.52	39.82±14.39	49.03±6.70	154.49
9	2635.73±219.29	20.89±10.58	47.93±6.60	151.58
10	2625.65±218.45	34.56±13.34	34.05±4.88	147.78
11	2673.98±222.48	41.92±14.87	44.29±6.14	159.79

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Table 2. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil samples from tobacco farm B.

S/No	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	Total Absorbed Dose rate (nGy h^{-1})
1	3122.84±258.88	31.33 ±13.03	65.14 ±8.72	187.99
2	3394.91±281.10	56.43 ± 17.71	51.13 ±7.01	200.85
3	3370.18±279.39	34.21 ±13.68	53.96 ±7.40	192.16
4	3232.29±267.63	43.67 ±15.58	60.18 ±8.11	194.57
5	3354.23±277.73	41.85 ±14.78	51.10 ±6.99	192.86
6	3120.14±258.97	42.62 ±15.05	39.84 ±5.67	175.86
7	2868.04±238.05	29.44 ±12.41	51.37 ±7.03	167.30

Table 3. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil samples from tobacco farm C.

S/No	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	Total Absorbed Dose rate (nGy h^{-1})
1	3215.03 ±266.53	41.29±16.71	71.64±9.61	200.46
2	2935.78 ±243.67	46.20±17.38	55.38±7.59	180.01
3	3241.43 ±268.71	40.52±14.58	51.89±7.02	188.08
4	2926.63 ±242.91	27.20±11.60	34.64±4.84	157.66
5	3394.81 ±281.43	37.36±14.38	66.84±8.98	203.13
6	3134.69 ±260.18	26.71±12.11	BDL	143.11

Table 4. The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil samples from a Control farm.

S/No	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	Total Absorbed Dose rate (nGy h^{-1})
1	154.41±16.14	26.78±11.01	6.64±1.12	20.10
2	165.61±17.03	44.93±14.60	17.39±2.60	30.22
3	301.91±28.92	73.46±20.56	20.65±3.03	50.20
4	88.45±8.25	56.71±17.17	25.47±3.65	33.31
5	202.93±20.46	58.95±17.69	34.23±4.74	39.81

External gamma absorbed dose rates

The external absorbed dose rate, D (nGy.h^{-1}) in air at 1 m above the ground level for soils containing the concentrations of the radionuclides measured in the samples is calculated using the equation [1, 17]:

$$D_{ext} = \sum_{R} A_R DC_{ext,R} \qquad (3)$$

Where $DC_{ext,R}$ is the coefficient of dose rate per unit activity concentration of radionuclide R (nGy h^{-1}/ Bq kg^{-1}) and A_R is the concentration of the radionuclide R in the sample (Bq kg^{-1}). UNSCEAR 2000 [18] prescribes $DC_{ext,R}$ coefficient of ^{226}Ra as 4.27 × 10^{-10} Gy.h^{-1}/Bq. kg^{-1}, ^{232}Th as 6.62 × 10^{-10} Gy.h^{-1}/Bq.kg^{-1}), ^{40}K as 0.43 × 10^{-10} Gy h^{-1}/ Bq.kg^{-1} and ^{137}Cs as 0.30 × 10^{-10} Gy.h^{-1}/ Bq.kg^{-1}). Since ^{137}Cs was not detected in any of the samples its coefficient was taken as zero. Using equation (2) and the activity concentrations of the radionuclides in tables 1 to 4 for each farm, the total absorbed dose rates were calculated for each farm. The results are presented in the fourth column of tables 1, 2, 3 and 4 for farms A, B, C and control farm respectively. The total gamma absorbed dose rates due to the three radiounuclides as could be seen from the tables ranged between 141 and 200 nGy h^{-1} across farms A, B and C. These values are 2-4 orders of magnitude greater than world average value of 59 nGy h^{-1} [18]. For the

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control farm, total gamma absorbed dose rates varied from 20 to 50 nGy h^{-1}. It is therefore anticipated that the accumulation of these radionuclides in the tobacco leaves through root uptake may also present high internal radiation burden to smokers in Nigeria especially due to alpha-radiation from ^{226}Ra and its progenies in the tobacco leaves. Results suggest possible high signature of ^{210}Pb and ^{210}Po in the tobacco leaves and the cigarette samples in the country. This is a subject for future research consideration given the fact that alpha radiation plays a major role in the etiology of lung cancer.

CONCLUSION

Farm soil samples from three farmlands used in the production of tobacco leaves for different brands of cigarettes tobacco smoked in Nigeria by British-America Tobacco Company were collected and analyzed in order to determine the activity concentration levels of ^{40}K, ^{226}Ra and ^{232}Th. Results of measurement showed relatively high values of these naturally occurring radionuclides in the soil when compared with previous environmental radioactivity studies in the area. Apparently, the specialized fertilizer used in the tobacco leaf production may be responsible for the high values obtained in this study. Through root uptake these radionuclides may accumulate on the tobaccos plant in large quantities and will be a major source of internal alpha radiation dose burden to smokers especially from ^{210}Po; as the trichomes of the tobacco leaves is known to concentrate polonium, which persists even when tobacco is dried and processed.

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