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LIFETIME CANCER RISK OF GAMMA RADIOACTIVITY RESULTS FROM SMOKING

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ABSTRACT

Smoking is one of the causes of heart attack, lung cancer and cancers of mouth and larynx. The latter could be arises from exposure of those sensitive organs to a combination of both chemical carcinogenic and radiological exposure results from naturally occurring radionuclides in tobacco leaves. Coal is also used in smoking some types of tobacco products that could be carcinogenic due to presence of a high percentage of an organic matter with inorganic matter such as minerals and trace elements. Varieties of commonly available tobacco products as well as coal samples were examined for their radioactivity content using gamma ray spectroscopy and calculate associated radiological hazards. Results shows that the average concentrations of ²⁹⁰Ra, ²⁹²Th, and ⁴⁰K in tobacco samples vary from 2.6 ± 0.2 to 8.9 ± 0.7 (average 5.4), 1.9 ± 0.1 to 9.5 ± 0.8 (average 4.5), and 517.4 ± 15.5 to 2401.2 ± 72 (average 1360.4) Bqkg^{c1}. Measured activity concentration for coal samples ranged from 10.8 ± 1.1 to 64.4 ± 2.1 (average 40.2), 3.5 ± 0.1 to 28.3 ± 0.3 (average 15.7), and 49.2 ± 0.4 to 301.2 ± 9.5 (average 215) Bqkg^{c1} for ²⁹⁶Ra, ²⁹⁰Th, and ⁴⁰K respectively. ¹⁵⁷Cs activity concentrations in cigarettes and coal samples were ranged from 0.1 ± 0.01 to 1.3 ± 0.02 (average 0.5) and 2 ± 0.01 to 5.8 ± 0.8 (average 3.1) Bqkg^{c1} respectively. Radium equivalent, total annual effective dose and excess lifetime cancer risk were calculated. ELCR was higher than world's average of $1.45x10^{-5}$ for tobacco and coal samples. In Egypt no special and clear regulations for monitoring radioactivity content in imported coal and tobacco leafs or its products, which appear to be necessary.

Keywords: Tobacco, Smoking, Activity concentration 226Ra, 232Th, 40K and 137Cs, Excess lifetime cancer risk, Dose assessment, Radiation indices, Annual effective dose.

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1. INTRODUCTION

Living organisms are continuously exposed to background ionizing radiation that emits from either natural or man-made radiation sources which causes radiation health effects. Natural radiation is mainly due to the activity concentration of primordial radionuclides ²³⁸U, ²³²Th and their product of decay, in addition to the other natural radionuclide ⁴⁰K present in the earth's crust [1].

²²⁶Ra (²³⁸U-series), ²²⁸Ra (²³²Th-series) and ⁴⁰K of the naturally occurring primordial radionuclides which are abundant in soil and in most fertilizers, ²³⁸U is associated with phosphate fertilizers [2] follow root uptake. ²²⁶Ra occurs in all ²³⁸U ores but it is more widely distributed because it forms water soluble compounds and its have life time is about (1600 y) for this reason ²³⁸U is sometimes called uranium or radium series. These radionuclides were incorporated metabolically into plants, and ultimately find their way into food chains and passed on to human. Many studies on radionuclides food contamination in the environment and their transfer or pathway mechanism to plant, animals, and human population have been reported [3-5].

In addition, manmade radionuclides such as ¹³⁷Cs, fission product radionuclides that could be received from fallout radioactivity in the past, behave in a similar manner and causes of food chains contamination by radionuclides.

Early studies showed the relation between alpha, beta and gamma-radiation dose, particularly that from alpha radiation, and lung cancer in smokers. This could be due to the presence of ²¹⁰Pb and ²¹⁰Po, decay of ²²⁶Ra, in tobacco, which greatly increases the intake of these radionuclides during smoking [6].

Coal and tobacco contain many known carcinogens [7]. Coal is composed from a high percentage of an organic matter with inorganic matter such as minerals and trace elements, which have been cited as a possible cause of health and environmental problems associated with the use of coal. Some trace elements in coal are naturally radioactive such as ²³⁸U, ²³²Th, and their progenies, including ²²⁶Ra and radon ²²²Rn [8].

According to the latest WHO data published in April 2011 Lung Cancers Deaths in Egypt reached 3,184 or 0.88% of total deaths. The age adjusted Death Rate is 5.98 per 100,000 of population ranks Egypt #142 in the world [9]. The long-term exposure to uranium and radium through inhalation has several health effects as chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth. Radium causes bone, cranial, and nasal tumors. Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukemia [10-12].

Chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth are known health effects due to ionizing radiation. Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukemia [13]. Therefore, continuous measurement of natural radioactivity concentration and calculating gamma dose rates are very important issues in order to evaluate accompanying radiological hazards.

The aim of this work is first, to assess the natural radioactivity content and man-made ¹³⁷Cs in different tobacco products (cigarettes) as well as smoky coal imported to Egyptian market for estimation of possible radiological health effects that could be resulted from smoking. Second, for monitoring the radioactivity concentration levels in such stuffs in Egyptian market and investigate to what extent that smoking gives rise to hazards radiological doses.

2. MATERIAL AND METHODS

2.1. Sampling and Samples Preparation

14 types of imported cigarettes were collected from the local market from several places, 5 packs for each type of each source were dried, grinded and mixed to obtain a homogeneous mixture. 6 types of smoking coal were collected, 4 of them imported and the rest were locally made from dried tree trunks burned to be coal. Samples then transferred to polyethylene containers of 150 cm³ capacity and sealed at least for 4 weeks to reach secular equilibrium between radium and thorium, and their progenies.

Because of the importance of background radiation in low level natural radioactivity measurements, it was measured prior each sample measurement and subtracted from its spectra. The ²²⁶Ra (²³⁸U) series, ²³²Th series, ⁴⁰K, and ¹³⁷Cs activity concentrations were measured using high resolution Canberra coaxial hyper pure germanium (HPGe) detector. A model 747 Canberra lead shield is used with the detector and has a 0.040 inch tin and 0.062 inch copper graded liner to prevent background interference by lead X-rays. The energy calibration and efficiency determination were performed using 138G Marinelly beakers containing mixed sources of ¹⁰⁹Cd, ⁵⁷Co, ^{123m}Tc, ¹¹³Sn, ¹³⁷Cs, ⁸⁸Y and ⁶⁰Co, allowing for some peaks in the measurement range. The system has a resolution of 1.9 keV full-widths at half-maximum for the 1332 keV gamma-ray line of ⁶⁰Co and a photo peak efficiency of 37 %. The spectrum of each sample was taken for 24 hrs., stored in a PC-based multichannel analyzer computer, and processed using a dedicated software program (GENIE-2000).

The Dray photo peaks corresponding to 351.9 keV of ²¹⁴Pb and 609.3, 1120.3, 1728.6 and 1760 keV of ²¹⁴Bi were considered for identifying the ²³⁸U. The Dray photo peak of 186 was considered for identifying the ²²⁶Ra. The Dray photo peaks of 238.6 of ²¹²Pb, 583.1 keV of ²⁰⁸Tl and 911.2 keV of ²²⁸Ac were used to identify ²³²Th in the samples [14]. ⁴⁰K and ¹³⁷Cs were recognized from their single peaks of 1460 and 662 keV respectively.

3. RESULTS AND DISCUSSION

3.1. Activity Concentrations of 226 Ra, 232 Th, 40K and 137 Cs

The measured activity concentration of 226 Ra, 232 Th, and 40 K in tobacco samples vary from 2.6±0.2 to 8.9±0.7 (average 5.4), 1.9±0.1 to 9.5±0.8 (average 4.5), and 517.4±15.5 to 2401.2±72 (average 1360.4) Bqkg⁻¹ respectively and shown in table 1. It is clear that the concentrations of 226 Ra and 232 Th were comparable, which reflect their origin in soil by root uptake, and lower than the world average concentration of 238 U and 232 Th, which is 35 and 30 Bqkg⁻¹ respectively. For ⁴⁰K, the average concentration was higher than the world average concentration, which is 400 Bqkg⁻¹ [1]. Also, measured activity concentration for coal samples ranged from 10.8±1.1 to 64.4±2.1 (average 40.2), 3.5±0.1 to 28.3±0.3 (average 15.7), and 49.2±0.4 to 301.2±9.5 (average 215) Bqkg⁻¹ for 226 Ra, 232 Th, and 40 K respectively. This was higher than the average world concentration of 20 and 50 Bqkg⁻¹ for 226 Ra and 40 K respectively, while lower than world average concentration of 20 Bqkg⁻¹ for 232 Th [15]. Figures 1 and 2 shows the activity concentration of various radionuclides in cigarettes and smoking coal with reference to sample Nos., average and world's averages.

Sample	Statistics	²²⁶ Ra	²³² Th	40 K	¹³⁷ Cs
Cigarettes	Minimum	2.6	1.9	517.4	0.1
	Maximum	8.9	9.5	2401.2	1.3
	Mean	5.4	4.5	1360.4	0.5
	SD	2.0	2.7	622.5	0.4
	Skewness	0.173	0.919	0.31	0.708
	World average	*35	*30	*400	-
Coal	Minimum	10.8	3.5	49.2	2.0
	Maximum	64.4	28.3	301.2	5.8
	Mean	40.2	15.7	215.0	3.1
	SD	23.3	11.5	103.6	1.6
	Skewness	-0.622	0.029	-1.046	2.255
	World average	**20	**20	**50	-

Table-1. Summary of basic statistics of natural radionuclides Bqkg-1 for different samples.

* UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) [1] ** UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) [15]

¹³⁷Cs activity concentrations in cigarettes and coal samples were ranged from 0.1 ± 0.01 to 1.3 ± 0.02 (average 0.5) and 2 ± 0.01 to 5.8 ± 0.8 (average 3.1) Bqkg⁻¹ respectively.

In the present study the standard deviation of all measured radionuclides, table 1, were lower than the mean value. This indicates that the concentration of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in samples have high degree of uniformity [16]. Skewness characterizes the symmetry or asymmetry of a distribution around its mean. So, when skewness equal zero, i.e., data points have normal distribution. Therefore, an understanding of the skewness of the dataset indicates whether deviations from the mean are going to be positive or negative [17]. Table (1) showed positive skewness of activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in tobacco samples, which indicates a distribution with an asymmetric tail extending towards values that are more positive. Coal samples shows the same results except for ²²⁶Ra and ⁴⁰K that have a negative skewness of activity concentrations which indicates a distribution with an asymmetric tail extending towards values that are more negative. The frequency histogram and the associated distribution curves of activity concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in cigarette and coal samples were given

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in figures 3 and 4 respectively showing some degree of multi-modality. This multi-model feature of the radioelements demonstrates the complexity of minerals in soil samples.



Figure-2. Coal activity concentration of various radionuclides with reference to sample No.

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Table-2. Con	nparison between	the activity concen	trations of our st	tudied cigarette sa	nples with that	of other authors i	n the world.
				A			

	Cigarettes						
Radionuclide	Papastefanou [18]	Shousha and Ahmad [19]	This study				
226 Ra	1.8 - 8	-	2.6 - 8.9				
²³² Th	1.1 - 7	1.4 - 2.1	1.4 - 9.5				
40K	273 - 2080	990.6 - 1392.1	517.4 - 2401.2				
¹³⁷ Cs	-	0.3 - 0.4	0.1 - 1.3				







Figure-4. The frequency distribution of activity concentrations of 226 Ra, 232 Th, 40K and 137 Cs in coal samples

Table 2 shows previous studies for the concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in cigarettes. The variation between our result and the reported values could be attributed to the variation in radioactivity content in tobacco

leaves and emphasize to the importance of continuous monitoring of radioactivity levels in tobacco products newly imported to the local market.

3.2. Assessment of Radiological Hazards

The knowledge of the specific activity in investigated samples is important for the assessment of the possible radiological hazards to human health. To evaluate the impact of radioactivity concentration of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs and radiological hazards associated with it, radium equivalent activity, annual effective dose and internal hazard index were also evaluated.

Radium equivalent activity (Ra_{eq}) was calculated to assess the hazard of radioactivity concentration in cigarettes and smoking coal using the formula

 $Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_{K}$

Where C_{Ra} , C_{Th} and C_K are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in Bqkg⁻¹ respectively. Table 3 shows that the Ra_{eq} in cigarettes and smoky coal were ranged from 45.2 to 204.4 (average 116.5) and 19.6 to 126.6 (average 79.2) Bqkg⁻¹ respectively which is lower than the recommended limit of 370 Bqkg⁻¹ [1].

Total annual dose resulting from activity concentration of 226 Ra, 232 Th, 40 K and 137 Cs in cigarettes and coal were calculated. For cigarettes, it was assumed that adult person consume about two packets per day each having 20 cigarettes of 0.85 g then the annual consumption of tobacco will be 12.702 Kg y⁻¹. Considering that 0.75 of radionuclide concentration contained in cigarette smoke which partially inhaled and deposited in lung and 0.25 retained in cigarette filter and ash [20] then the annual effective dose, H_E (Sv y⁻¹), due to inhalation for smokers, was calculated according to equation

 $H_E=0.75~X~M_T~X~C_i~X~F$

where M_T (kg y⁻¹) refers to the annual amount (in mass) of tobacco consumed, C_i (Bq kg⁻¹) refers to the concentration of the radionuclide and F (SvBq⁻¹) refers to the dose conversion factor which is equal to 2.90x10⁻⁶ and 2.5x10⁻⁵ for ²²⁶Ra, and ²³²Th respectively [6] 4.66x10⁻⁹ for ⁴⁰K [21] and 8.7x10⁻⁹ for ¹³⁷Cs [22].

As shown in table 3 total annual effective dose resulting from activity concentration of 226 Ra, 232 Th, and 40 K in cigarette samples ranged from 0.5 to 2.5 (average 1.27) mSv y⁻¹ and is comparable with the total average worldwide exposure to natural radiation sources of 2.4 mSv y⁻¹, especially the part due to inhalation which is 1.26 mSv y⁻¹ [6].

For coal samples used for smoking tobacco with hookah, the annual effective dose calculated considering that the person smoke two-tobacco bowls using about 12g each time, i.e 4.38 Kg y⁻¹. Total annual effective dose resulting from activity concentration of 226 Ra, 232 Th, and 40 K ranged from 0.39 to 2.9 (average 1.7) mSv y⁻¹, which is lower than the total average worldwide exposure to natural radiation sources of 2.4 mSv y⁻¹ and higher than the part due to inhalation which is 1.26 mSv y⁻¹ [6].

Sample	Statistics	\mathbf{Ra}_{eq}	HE	Hin	ELCR X10 ⁻³
Cigarettes	Minimum	45.2	0.5	0.1	1.97
	Maximum	204.4	2.5	0.6	9.25
	Mean	116.6	1.3	0.3	4.7
	SD	53.2	0.7	0.1	2.6
	World average	370	2.4	< 1	1.16
Coal	Minimum	19.6	0.4	0.082	1.425
	Maximum	126.6	2.9	0.5	10.7
	Mean	79.2	1.7	0.3	6.1
	SD	43.7	1.1	0.2	4.0
	World average	370	2.4	< 1	1.16

Table-3. Summary of radiological hazard parameters for different measured samples.

The internal exposure by radon and its progeny from cigarettes and smoking coal were controlled by the internal hazard index (H_{in}) . H_{in} is given by the equation:-

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810$$

where C_{Ra} , C_{Th} and C_K are activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in Bqkg⁻¹. The value of this index should be less than 1 mSvy⁻¹ in order for the radiation hazard to have negligible hazardous effects to the respiratory organs [23]. The average internal hazard index ranged from for all the samples

Excess lifetime cancer risk (ELCR) is a human health risk assistance indicates that someone might have of getting cancer if that person was exposed to cancer-causing materials for a longer time. ELCR was calculated using equation

ELCR = AED XLEX RF

Where AED is the annual effective dose, LE is life expectancy at birth (72.7 year) $\lfloor 24 \rfloor$ and RF is risk factor (Sv⁻¹), fatal cancer risk per Sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public $\lfloor 25 \rfloor$.

The ELCR ranged from 1.97×10^{-3} to 9.25×10^{-3} (average 4.7×10^{-3}) and from 1.4×10^{-3} to 10.7×10^{-3} (average 6.1×10^{-3}) for cigarettes and coal samples respectively, table 3. The average ELCR factor assessed during present study was higher than the world's average of 1.16×10^{-3} [26] for both samples. Correlation analysis has been carried out, as a bivariation statistics in order to determine the mutual relationships and strength of association between radionuclides and associated radiological parameters through calculation of the linear Pearson correlation coefficient for tobacco and coal samples. Results of the Pearson correlation coefficients among all the studied radiological parameters are summarized in tables 4 and 5. From table 4, it is clear that positive correlation exist among the four radionuclides and all the radiation hazard parameters i.e. all them contributes significantly to gamma-ray emission from processed tobacco leafs.

Variables	²²⁶ Ra	²³² Th	40K	¹³⁷ Cs	Raeq	He	\mathbf{H}_{in}	ELCR
²²⁶ Ra	1							
²³² Th	0.847	1						
40K	0.929	0.838	1					
^{137}Cs	0.711	0.755	0.576	1				
Ra_{eq}	0.938	0.862	0.999	0.602	1			
H _e	0.876	0.998	0.865	0.757	0.887	1		
H _{in}	0.942	0.864	0.999	0.608	1.000	0.889	1	
ELCR	0.876	0.998	0.865	0.757	0.887	1.000	0.889	1

Table-4. Pearson correlation matrix among the variables for tobacco samples.

Table-5. Pearson correlation matrix among the variables for coal samples.

Variables	²²⁶ Ra	²³² Th	40K	¹³⁷ Cs	\mathbf{Ra}_{eq}	He	\mathbf{H}_{in}	ELCR
²²⁶ Ra	1							
²³² Th	0.704	1						
40K	0.896	0.630	1					
^{137}Cs	-0.448	-0.739	-0.492	1				
Ra_{eq}	0.960	0.866	0.896	-0.606	1			
H_{e}	0.798	0.990	0.714	-0.717	0.927	1		
H _{in}	0.983	0.818	0.904	-0.557	0.995	0.891	1	
ELCR	0.798	0.990	0.714	-0.717	0.927	1.000	0.891	1

For coal samples, table 5, strong correlations were observed between ²²⁶Ra and the other radionuclides ⁴⁰K and ²³²Th, while all of them are weakly correlated with ¹³⁷Cs. The strong relationship between the three radionuclides suggesting that their content in coal are mostly influenced and controlled by similar origin of sources [27, 28]. However, very weak negative correlation observed between ¹³⁷Cs and the other radionuclides indicates that ¹³⁷Cs has a different origin and behavior in coal samples. Also, positive correlation coefficient was observed between

²²⁶Ra, ²³²Th and ⁴⁰K with all radiological parameters which implies that very strong relationship between the radionuclides in coal and radiological parameters.

4. CONCLUSION

Results showed the wide range of activity concentrations of natural radionuclides in studied samples due to their different origin. In addition, the health risks of smoking are not uniform across all smokers that vary according to the amount of smoked tobacco. This study may be taken as a base to continuous measurement of naturally occurring radionuclides for imported tobacco leaves, processed cigarettes and other tobacco products in order to protect population from risks caused by tobacco smoking. Also it encourage government to control the consumption of tobacco through different ways such as awareness about the role of naturally occurring radionuclides and its radiological hazards to smoker and increasing taxes on tobacco products and hence increasing price.

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