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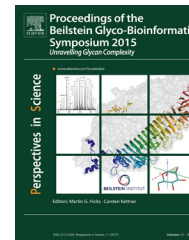
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Annual effective dose and excess life time cancer risk assessment from tobacco plants[☆]

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Tobacco plants;
Activity concentration;
Annual effective dose;
Lifetime cancer risk

Abstract Cigarette smoking is one of the pathways that might cause radiation exposure due to the presence of radioactive elements in tobacco leaves used in the manufacture of cigarettes. From the health point of view, the knowledge of radioactivity content of the various radionuclides in tobacco leaves and their derivative products is important to assess the radiological effects associated with tobacco smoking for smokers. In this work, naturally occurring radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K and man-made ¹³⁷Cs were measured in tobacco using gamma-ray spectrometry. Results show that the average concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K vary from 4.30 ± 0.2 to 11.85 ± 0.7 (average 8.17), 1.35 ± 0.1 to 16.12 ± 0.9 (average 6.08), and 565.6 ± 15.5 to 1235.81 ± 35 (average 908.09) Bq kg⁻¹, respectively. ¹³⁷Cs activity concentrations were ranged from 0.15 ± 0.01 to 0.67 ± 0.9 (average 0.44) Bq kg⁻¹. The most important radiological parameters, including radium equivalent (Ra_{eq}), total annual effective dose (AED) and excess lifetime cancer risk (ELCR) were calculated. For smokers, the total AED resulting from activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in tobacco samples ranged from 0.44 to 2.29 (average 1.39) mSv y⁻¹, while of ¹³⁷Cs varied from 0.01 to 0.04 (average 0.025) μ Sv y⁻¹ and so very little is contributing to the total dose due to inhalation to smokers. ELCR was higher than the world's average of 1.45×10^{-3} for tobacco and coal samples. The excess lifetime cancer risk ranged from 1.55×10^{-3} to 8.01×10^{-3} with an average value of 4.88×10^{-3} for all tobacco samples under investigation.

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Introduction

Living organisms are continuously exposed to background ionizing radiation emitted from either natural or man-made radiation sources. Effects from radiation usually fall under the category of stochastic effects (Flynn and Theodore,

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2002). Cancer and genetic effects are the two most common possible negative effects linked with exposure to radiation.

The great interest expressed worldwide for the study of the relation of smoking with cancer, particularly lung cancer, has led to extensive surveys and investigations in many countries. In general, the toxicity in tobacco was considered earliest mainly due to the presence of chemi-toxins like nicotine, tars, aromatic hydrocarbons, sterols and many other materials leading to mutagenicity and carcinogenicity (Mahabir et al., 2010). Although, there may be other cancer inducing mechanisms in humans, one of the causative factors is radioactive elements present in tobacco leaves used for the manufacture of cigarettes.

Like all soils, the soil in which tobacco plants are grown is radioactive (Gruppen, 2010). In addition, tobacco plants are frequently fertilized with phosphate fertilizers to fulfil their requirement of nutrients, which may significantly increase the radionuclide content in soil. This is because the phosphates used for the manufacturing of phosphate fertilizers are derived from a rock mineral named 'apatite' containing high levels of uranium and other radioactive elements (Abbady, 2005). The most important radionuclides are radium, thorium and their decay products, which can easily transfer from soil to plants through root uptake. In the radium decay chain, the radioactive gas ^{222}Rn occurs as well as the relatively long-lived radioisotopes ^{210}Pb and ^{210}Po . These radionuclides can accumulate on the surfaces of tobacco leaves as the plant grows, and can remain their throughout the cigarette-manufacturing process.

Due to the presence of the natural radionuclides in fresh tobacco leaves, in an amount exceeding that normally found in general foods for human usage, cigarette smoking is one practice of radiological concern (Kilthau, 1996). The contents of naturally occurring radioisotopes in manufactured tobacco vary with the source. The difference may result from production locality, culture and curing (Shousha and Ahmad, 2012). Therefore, continuous measurement of natural radioactivity concentration and calculating gamma dose rates in tobacco leaves and their derivative products are very important issues in order to evaluate accompanying radiological hazards.

The purpose of this paper is first, to determine the activity concentrations of naturally occurring radionuclides ^{226}Ra (^{238}U), ^{232}Th and ^{40}K as well as the artificial radionuclide ^{137}Cs in different brands of tobacco leaves before cigarette production by means of gamma-ray spectrometry. Second, to estimate the annual effective dose received from cigarette tobacco and the excess lifetime cancer risk caused by gamma-radiation exposure.

Materials and methods

In this work, four brands of imported tobacco plants (leaves) commonly used in the manufacture of cigarettes in Algeria, commercially named: Basma (TAB1) type from Greece, Burley (TAB2) and Havane (TAB3) brands from Italy, and the last one Burley (TAB4), which is from Brazil, were examined for their radioactivity content using gamma-ray spectroscopy. At the Chrono-Environnement laboratory, the collected samples were dried at a temperature of 40°C , ground and mixed to obtain a homogeneous mixture, and then were packed and sealed in cylindrical polyethylene containers of 50cm^3 (SG50) for four weeks before measuring, in order to assure the secular equilibrium between radium and thorium, and their progenies.

For qualitative identification as well as quantitative determination of the specific activity of gamma-emitting radionuclides present in tobacco samples, a high-resolution gamma-ray spectrometer equipped with a planar BEGe (Broad Energy Germanium) detector (Canberra, model BE 3825) was used. The system has a relative efficiency of 20% and an energy resolution (FWHM) of 0.38 keV at 5.9 keV, 0.628 keV at 122 keV and 1.724 keV at 1332.5 keV. The detector was connected to a Digital Signal Analyser (DSA-LX) Unit (16K channel signal analyser, V1.0) from Canberra. It is shielded with 15 cm of lead (including 2 cm made of archaeological lead) to reduce the contribution of environmental radioactivity to its background. The lead shielding was surrounded with an inner layer of copper (0.2 cm thick) to reduce the contribution from Pb X-rays. The control of the acquisition parameters and the analysis of the collected spectra are carried out using GENIE-2000 computer software (Canberra, V3.3).

The mixed gamma emitting CRM IAEA-447 (Certified Reference Material) was used for the efficiency calibration of the detection system (Shakhashiro et al., 2012). It includes different radionuclides of the natural uranium and thorium series: ^{208}Tl , ^{210}Pb , ^{210}Po , ^{212}Pb , ^{214}Pb , ^{214}Bi , ^{226}Ra , ^{228}Ac , ^{234}Th , ^{234}U and ^{241}Pu associated with ^{40}K , ^{90}Sr , ^{137}Cs and ^{241}Am radionuclides. The following radionuclides with their respective peaks were used to determine the experimental full-energy peak efficiency (FEPE) curve with energies in the range 45–2615 keV.

Every prepared sample was counted for 24 h in the same counting geometry as the standard source. The activity concentrations of ^{226}Ra (^{238}U) and ^{232}Th in each sample can be estimated indirectly, via several gamma-ray lines from their decay products under the assumption of secular equilibrium. The gamma-ray lines at 295.2 keV (19.2%) and 351.9 keV (37.1%) from ^{214}Pb , and 609.3 keV (46.1%), 1120.3 keV (15%) and 1764.5 keV (15.4%) from ^{214}Bi were used to assess the ^{226}Ra activities. Also, the ^{232}Th activities were estimated using gamma-ray lines at 583.19 keV (86%) and 2614.51 keV (99.79%) from ^{208}Tl , 338.4 keV (12%) and 911.2 keV (29%) from ^{228}Ac and 238.6 keV (43.6%) from ^{212}Pb (Amrani and Tahtat, 2001). The ^{40}K activity was determined by measurement of the single gamma line at 1460.8 keV (10.6%). The background counting due to naturally occurring radionuclides in the environment around the detector was subtracted from the peak of each sample. The activity

Table 1 Activity concentrations in different types of tobacco leaves (Bq kg^{-1}).

Brands	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
TAB1	4.30	4.42	565.60	0.67
TAB2	11.85	10.45	744.44	0.40
TAB3	8.08	12.44	1086.49	0.56
TAB4	8.98	1.35	1235.81	0.15
Average	8.17	6.08	908.09	0.44

Table 2 Annual effective dose and ELCR of smokers due to smoking different brands of tobacco.

Brands	^{226}Ra (mSv y $^{-1}$)	^{232}Th (mSv y $^{-1}$)	^{40}K (mSv y $^{-1}$)	^{137}Cs ($\mu\text{Sv y}^{-1}$)	ELCR $\times 10^{-3}$
TAB1	0.74	0.085	0.02	0.04	2.96
TAB2	1.76	0.22	0.02	0.02	7.02
TAB3	2.10	0.16	0.03	0.03	8.01
TAB4	0.23	0.17	0.04	0.01	1.55
Average	1.20	0.16	0.03	0.025	4.88

concentration $A(E)$ in Bq kg $^{-1}$ of the radionuclides found in the samples was calculated using the following relation:

$$A(E) = \frac{C_{\text{net}}}{\varepsilon(E) \times I_{\gamma} \times t \times m} \quad (1)$$

where C_{net} is the counting rate for a specific gamma line (counts per second) corrected for background, $\varepsilon(E)$ is the absolute photopeak efficiency, I_{γ} is the intensity of the considered gamma-ray line, t is the counting time in s and m is the sample mass in kg.

Results and discussion

Activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs

The measured specific activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in tobacco samples vary from 4.30 ± 0.2 to 11.85 ± 0.7 (average 8.17), 1.35 ± 0.1 to 16.12 ± 0.9 (average 6.08), and 565.6 ± 15.5 to 1235.81 ± 35 (average 908.09) Bq kg $^{-1}$, respectively and shown in Table 1. It is clear that the concentrations of ^{226}Ra and ^{232}Th in tobacco leaves were comparable, which reflects their origin from the soil by root uptake, and lower than the world average concentrations of ^{238}U and ^{232}Th , which are 35 and 30 Bq kg $^{-1}$, respectively (UNSCEAR, 2000). Concerning ^{40}K , the average concentration was higher than the world average concentration, which is 400 Bq kg $^{-1}$, also proposed by UNSCEAR (2000). Results showed that the higher activity concentrations of ^{40}K were obtained in TAB3 and TAB4. The ^{40}K concentrations were also due to root uptake either from soil or from fertilizers.

^{137}Cs activity concentrations in the tobacco leaves ranged from 0.15 ± 0.01 to 0.67 ± 0.9 (average 0.44) Bq kg $^{-1}$. This concentration could be due to root uptake. Results showed that the highest activity concentration was in TAB1.

Assessment of radiological hazards

The knowledge of the radioactivity content of the various radionuclides in the investigated samples is important to assess the radiological hazard effects to human health through radiation hazard indexes.

The radium-equivalent activity (Ra_{eq}) index is used to compare the activity concentration levels of ^{226}Ra , ^{232}Th and ^{40}K in the materials and it takes into account the gamma

radiation hazards associated with them. It is calculated through the relation given by (Beretka and Mathew, 1985):

$$Ra_{\text{eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \quad (2)$$

where A_{Ra} , A_{Th} and A_{K} are the measured activities for the radionuclides ^{226}Ra , ^{232}Th and ^{40}K , respectively. The permissible maximum value of the radium equivalent activity should be less than 370 Bq kg $^{-1}$, in order to limit the annual effective dose to 1 mSv for the general public as reported by UNSCEAR (1982). The results for the calculation of Ra_{eq} show that Ra_{eq} values in the investigated tobacco samples ranged from 54.2 to 109.53 (average 88.34) Bq kg $^{-1}$ which is lower than the recommended limit of 370 Bq kg $^{-1}$.

For cigarettes, it was assumed that an adult person consumes about 30 cigarettes (one and a half packs) of 0.82 g of tobacco per cigarette. Then the annual consumption of tobacco will be 8.985 kg y $^{-1}$. According to Khater (2004), about 75% of the radionuclide activity concentration (Bq kg $^{-1}$) contained in cigarette smoke was partially inhaled and deposited in lung tissues and about 25% is retained in the cigarette filter and ash. The annual effective dose, H_E (Sv y $^{-1}$), due to inhalation for adult smokers, was calculated according to equation:

$$H_E = 0.75 \times M_T \times C_i \times F \quad (3)$$

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 (Sv Bq $^{-1}$) refers to the dose conversion factor of the radionuclides which is equal to 2.90×10^{-6} and 2.50×10^{-5} for ^{226}Ra and ^{232}Th , respectively (UNSCEAR, 2008), 4.66×10^{-9} for ^{40}K (Eckerman et al., 1989) and 8.7×10^{-9} for ^{137}Cs (UNSCEAR, 1993).

As shown in Table 2, the annual effective dose to ^{226}Ra varied from 0.23 to 2.10 (average 1.2) mSv y $^{-1}$, for ^{232}Th series from 0.085 to 0.22 (average 0.16) mSv y $^{-1}$ while for ^{40}K from 0.02 to 0.04 (average 0.03) $\mu\text{Sv y}^{-1}$. The total annual effective dose resulting from activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in tobacco samples ranged from 0.44 to 2.29 (average 1.39) 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 $^{-1}$ (UNSCEAR, 2008). The annual effective dose due to ^{137}Cs varied from 0.01 to 0.04 (average 0.025) $\mu\text{Sv y}^{-1}$ and so very little is contributing to the total dose due to inhalation by smokers.

Excess lifetime cancer risk (ELCR) is the risk that someone has of getting cancer if that person were exposed to cancer-causing materials for a long time. It is calculated assuming that there is a linear relationship between dose

and the stochastic effects. The ELCR factor can be estimated using the following equation (Taskin et al., 2009):

$$\text{ELCR} = \text{AED} \times \text{DL} \times \text{RF} \quad (4)$$

where AED is the annual effective dose equivalent, DL is the average life exposition expectancy (assumed to be 70 years) and RF is the fatal cancer risk factor per Sievert, which is stochastically determined by ICRP (1990) to be 0.05 for the public.

The ELCR calculated values from tobacco samples are listed in the last column of Table 2. As shown, the ELCR ranged from 1.55×10^{-3} to 8.01×10^{-3} . The average of the ELCR factor was 4.88×10^{-3} which is 5.5 times higher than the world's average of 1.45×10^{-3} (Shousha and Ahmad, 2016) for tobacco samples.

Conclusion

The radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K as well as the artificial radionuclide ^{137}Cs in tobacco leaves samples as investigated using gamma ray spectrometry showed that there are low level activities in the studied locations. The activity concentrations in the tobacco leaves would be either through root uptake or through the fertilizers used for cultivation of tobacco plants in the fields. In estimating the radiation dose induced from smoking, it was concluded that the annual effective dose to lungs due to inhalation for adults (smokers) averaged to 1.2 mSv y^{-1} for ^{226}Ra , 0.16 mSv y^{-1} for ^{232}Th and $0.03 \text{ } \mu\text{Sv y}^{-1}$ for ^{40}K giving total of 1.39 mSv y^{-1} . The annual effective dose due to ^{137}Cs averaged to about 25 nSv, which is very much lower than that of the naturally occurring radionuclides. The effective dose of 1.39 mSv y^{-1} can be compared with the average worldwide exposure to natural radiation sources due to inhalation of 1.26 mSv y^{-1} . Excess lifetime cancer risk was also averaged to the value 4.88×10^{-3} that is higher than world's average of 1.45×10^{-3} for tobacco and coal samples. This study may be taken as a basic to continuous measurement of naturally occurring radionuclides for imported tobacco leaves, processed cigarettes and other tobacco products in order to protect the population from risks caused by tobacco smoking.

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