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ORIGINAL ARTICLE

The activities of ^{210}Po and ^{210}Pb in cigarette smoked in Tunisia

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Abstract

In this study, the activity concentration of polonium 210 in cigarette for Tunisian consumers was investigated by alpha spectrometry. After chemical digestion of tobacco, ^{210}Po was extracted, auto-deposited on disc and measured. The activity of ^{210}Pb was assessed after radioactive equilibrium was achieved. The activity levels of ^{210}Po ranged between 7.8 ± 0.3 and 17 ± 0.5 mBq per cigarette with an average of 12.9 ± 0.4 mBq per cigarette. Effective doses per year due to cigarette smoking were calculated assuming that 22% of the ^{210}Pb and ^{210}Po in tobacco were retained in the lungs of the smokers. It is concluded that for a smoker in Tunisia, the average effective dose is about 90.6 ± 3.3 µSv per year for a cigarette consumption of one pack of cigarettes per day. This value is somewhat lower than 106.4 ± 5.3 µSv per year estimated as the mean global effective dose from smoking.

Keywords Polonium 210 · Lead 210 · Tobacco · Cigarette smoke · Radiation dose · Tunisia

Introduction

Nicotiana tabacum, commonly known as tobacco, is a leafy plant, which is used for cigarettes, cigars, pipes and narghiles. Tobacco and tobacco products contain several toxic compounds and, consequently, smoking these products can cause serious health damage (Sakoda et al. 2012; Funck-Brentano et al. 2006; Goodwin et al. 2010). Tobacco smoke is considered one of the potential causes of lung cancer (Zaga et al. 2011). Among the toxic compounds are natural radioactive isotopes of uranium and thorium (^{234}U, ^{238}U, ^{228}Th, ^{230}Th, ^{232}Th) as well as their daughter products (e.g., ^{226}Ra, ^{210}Po, ^{210}Pb) (Goodwin et al. 2010). Polonium-210 is the daughter of ^{210}Pb. These radioisotopes are present among the hazardous chemicals in cigarettes and represent a source of ionizing radiation in the smoke which is inhaled into the respiratory system (Kilthau 1996; Martell 1982; Jenkins et al. 1984; Burns 1991; Skwarzec et al. 2001a, b; Khater 2004).

The origin of polonium and lead in tobacco plants can be absorption from the soil (supported Po), direct deposition of fallout on the tobacco leaf (unsupported Po) and ^{222}Ra which is deposited on tobacco leafs. Polonium concentration can be enhanced by use of phosphate fertilizers (Desideri et al. 2007; Lopes dos Santos et al. 1970). Phosphate rocks, with high concentrations of uranium and the daughter nuclide ^{210}Po, are extensively mined to produce phosphate fertilizers (Mussealo-Rauhammaa et al. 1985; Hussein 1994; Alam et al. 1997).

The distribution of radionuclides in area tissues, which are exposed to air as for example leaves, follows an acropetal gradient suggesting that Po-210 and Pb-210 are more concentrated in older leaves of tobacco (Athalye 1972). A smoker inhales such natural radionuclides into the respiratory system by smoking a cigarette (Zaga and Mistry 2011). Polonium-210 is the radioactive isotope most often investigated in the literature, and its half-life is 138.376 ± 0.002 days (Singh and Nikelani 1976; Martell 1974). The alpha particle emitted after the decay of ^{210}Po has a high energy (5.305 MeV), it represents a local source of radiation and can damage the lung cells (Martell 1982).

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As compared to other radionuclides, ^{210}Pb and its daughter ^{210}Po contribute the largest radiation dose to the lungs (Peres and Hiromoto 2002; Skwarzec et al. 2001a, b). Martell estimated that the cumulative alpha dose is about 16 Sv in bifurcations of smokers who died with lung cancer (Martell 1982).

The objective of the present work was to measure the ^{210}Po activity concentrations in selected brands of the most frequently purchased cigarettes in Tunisia. In this study, the activity concentrations of ^{210}Pb in cigarettes were assessed; the effective dose received by a smoker per year was calculated and compared with those reported by other studies.

Materials and methods

Sample preparation

All reagents used in the present study for chemical procedures were of analytical grade. Chemical recoveries were assessed using ^{209}Po as a radiotracer. Eleven brands of cigarettes mostly smoked in Tunisia were selected for analysis; 6 of them were local products (coded as L1-L6), while the others were imported (I1-I7) from other countries. Three packs of cigarettes of each brand were bought from the local market; the 60 cigarettes of each brand were dried at 60 °C overnight, ground and mixed well. A suitable quantity of a dry cigarette sample (i.e., 2.5 g) was transferred to beakers, treated with a mixture of acid concentrated solution [HNO_3, HCl], spiked with a known ^{209}Po activity (i.e., 37.4 mBq), and settled overnight. Pure sodium chloride was added to the sample which was then heated for 3 h at 70 °C near dryness. Then the residue was treated two times with concentrated HCl and H_2O_2 30% and evaporated. The dry residue was dissolved in 30 mL of a solution of 0.3 M HCl. For the reduction of Fe^{3+}, a solution of $NH_2OH.HCl$ was added to the mixture and polonium was auto-deposited on a stainless steel disc (diameter: 20 mm). It is noted that Pb and Bi could be co-deposited in a disc made of metal other than silver. To avoid any overestimation of ^{210}Po, the measurements were performed immediately after auto-deposition (Ehinger et al. 1986).

Measurements

For the measurements, a 7400 model alpha spectrometer (Canberra) was used including six vacuum chambers and a silicon detector (Canberra alpha PIPS detector); the active surface area was 450mm^2. A TRUMP 8 k-W3 multichannel analyzer connected the spectrometer with a computer. The distance between the source and the detector was 5 mm. The detection efficiency was of the order of 24% and the Canberra Genie-2000 software tool was applied for the acquisition of spectra. Depending on the sample, the counting time varied between 2 and 4 days. Consequently, the lowest limit of detection (LLD) calculated according to Currie (1968) was approximately 0.3 mBq $L^{-1}$for 2 days counting. For the local calibration, a multi alpha reference material was used for quality control (standard source with certificate with radionuclides (^{238}U, ^{235}U, ^{239}Pu, ^{241}Am)). Blank samples were used to check the purity of the chemicals.

Assessment of effective dose

Yearly effective dose was assessed from^{210}Po and ^{210}Pb in cigarette smoke according to Eqs. 1 and 2 (Sakoda et al. 2012):

$$E = A \times D, \tag{1}$$

where E is the effective dose (μSv/year), A is the activity of the radionuclide inhaled by smokers per year (Bq year^{-1}), and D is the effective dose per activity due to inhalation of ^{210}Po and ^{210}Pb (μSv Bq^{-1}). The effective dose conversion factor D was taken from Publication 119 of the International Commission of Radiological Protection (ICRP 2012) as 3.3 μSv Bq^{-1} for ^{210}Po and 1.1 μSv Bq^{-1} for ^{210}Pb.

$$A = F \times C \times T, \tag{2}$$

where F is the experimentally determined radionuclide activity per cigarette (Bq cigarette^{-1}), C is the number of cigarettes smoked per day (assumed to be 20 per day), and T is the smoking time per year (365 days per year).

Results and discussion

^{210}P concentrations

The results of the different concentrations of ^{210}Po in cigarettes (local and imported brands) in this study are summarized in Table 1. The data indicate that there are large differences of activities between the local and imported products. These differences depend essentially on the tobacco varieties and on differences in manufacturing procedures (Skwarzec et al. 2001a, b; Carvalho 1995). The average activity concentrations of ^{210}Po in cigarettes locally produced in Tunisia were between 11.7 ± 0.5 and 21.1 ± 0.8 mBq g^{-1} with a mean value of 16.3 ± 0.6 mBq g^{-1}. The differences observed are probably due to the quality of tobacco in the cigarettes: Athalye et al. assumed that the distribution of radionuclides in the tobacco plant follows an acropetal gradient and, consequently, radionuclides are more concentrated in older than in younger leaves (Athalye et al. 1972). For the imported brands, the average concentrations were between 18.8 ± 0.8 and 25.3 ± 0.8 mBq g^{-1} with a mean value of

Table 1 Activity concentrations of ^{210}Po in cigarette tobacco of commonly smoked brands in Tunisia. Concentrations are given per gram dry mass

Activity concentration (mBq g^{-1})			
Local brands		Imported brands	
L1	11.7 ± 0.5	I1	23.0 ± 0.9
L2	16.7 ± 0.6	I2	21.4 ± 0.8
L3	13.8 ± 0.5	I3	18.8 ± 0.6
L4	18.2 ± 0.6	I4	25.3 ± 0.8
L5	16.7 ± 0.9	I5	23.3 ± 0.7
L6	21.1 ± 0.8	I6	23.0 ± 0.7
		I7	18.8 ± 0.8
Min	11.7 ± 0.5		18.8 ± 0.8
Max	21.1 ± 0.8		25.3 ± 0.8
Mean	16.3 ± 0.6		22.9 ± 0.7

Mean cigarette mass was 0.67 g after drying

The uncertainty includes statistical uncertainty due to the number of measured alpha particles

22.9 ± 0.7 mBq g^{-1}. These high activities of polonium 210 in tobacco can be explained by the use of phosphate fertilizers during cultivation (Tso et al. 1968). The average of ^{210}Po activity concentration value in cigarettes sold in Tunisia, 19.6 mBq per gram, is not significantly different from the average of ^{210}Po concentrations reported in Finland, Cuba, Philippines, and Greece (Mussealo-Rauhammaa and Jaakkola 1985; Brigido Flores et al. 2015; Iwaoka et al. 2012; Savidou et al. 2006). On the other hand, Persson reported that the average activity of ^{210}Po was 13 ± 2 mBq g^{-1} in tobacco harvested in various countries and at different times (Persson and Holm 2011).

It takes about 24–30 months between the harvest of tobacco leaves and the final production of cigarettes. This represents six to seven half-lives of ^{210}Po. Therefore, secular equilibrium with ^{210}Pb is considered as completed (Sakoda et al. 2012), meaning that the activity concentration of both radionuclides should be in equilibrium (Tahir and Alaamer 2008; Papastefanou 2009; Nagamatsu et al. 2011). This was verified by several studies (Desideri et al. 2007; Godoy et al. 1992; Schayer et al. 2009). Consequently it was assumed in the present study that, when the measurements were conducted, secular equilibrium between ^{210}Pb and ^{210}Po was well accomplished and the activity concentrations of ^{210}Pb could be estimated from the activity concentration of ^{210}Po (Table 1).

Assessment of effective dose

In most studies, the transfer factor (F) assumed for ^{210}Pb and ^{210}Po from cigarettes to mainstream smoke varies between 10 and 35% (Schayer et al. 2009; Radford and Hunt 1964; Hill 1965; Ferri and Baratta 1966; Ferri and Christiansen 1967; Sakanoue et al. 1987; Carvalho and Olivira 2006). Peres and Hiromoto (2002), assuming that 10% of ^{210}Pb and 20% of ^{210}Po in the tobacco are inhaled. Schayer et al. (2009) indicated that an average of 13% of ^{210}Po and 8% of ^{210}Pb are transferred to the mainstream smoke and inhaled by a smoker. For calculation of the yearly effective dose, they used 3.7 μSv Bq^{-1} and 1.2 μSv Bq^{-1} as the conversion factors for ^{210}Po and ^{210}Pb (DCAL 2006), respectively. Jankovic-Mandic et al. (2016) used the arithmetic mean values as suggested by Iwaoka and Yonehara (2012), which were 12% for ^{210}Pb and 18% for ^{210}Po. M. Horvath et al. (2017) reported, after having performed a smoking experiment with a smoking machine, that the net intake of ^{210}Po from tobacco is 15 ± 10% (the range was between 5 and 25%). Kubalek et al. (2016) found in a smoking experiment with volunteer smokers that approximately 22% of ^{210}Po and ^{210}Pb are retained in the lungs of smokers. In the present study, this value was used to estimate the effective yearly dose from smoking of the investigated local and imported cigarettes (Table 2). The values calculated for the annual effective dose due to ^{210}Po in cigarette smoke varied between 41.5 ± 1.8 and 89.8 ± 2.8 μSv year^{-1} with a mean value

Table 2 Annual effective dose due to inhalation of ^{210}Po and ^{210}Pb for tobacco consumed in Tunisia (local and imported brands)

Brand	Activity	Effective dose (μSv.year^{-1})		Brand	Activity	Effective dose (μSv.year^{-1})	
Local	mBq.cig^{-1}	^{210}Po	^{210}Pb	Imported	mBq.cig^{-1}	^{210}Po	^{210}Pb
L1	7.83 ± 0.33	41.50 ± 1.75	13.85 ± 0.58	I1	15.41 ± 0.60	81.65 ± 3.18	27.22 ± 1.05
L2	11.20 ± 0.40	59.35 ± 2.12	19.79 ± 0.70	I2	14.34 ± 0.53	76.00 ± 2.80	25.33 ± 0.93
L3	9.20 ± 0.33	48.75 ± 1.75	16.25 ± 0.58	I3	12.60 ± 0.40	66.80 ± 2.12	22.26 ± 0.70
L4	12.20 ± 0.40	64.65 ± 2.12	21.55 ± 0.70	I4	16.95 ± 0.53	89.82 ± 2.80	29.95 ± 0.93
L5	11.20 ± 0.60	59.35 ± 3.18	19.79 ± 1.05	I5	15.62 ± 0.46	82.77 ± 2.43	27.60 ± 0.80
L6	14.20 ± 0.53	75.25 ± 2.80	25.09 ± 0.93	I6	15.41 ± 0.46	81.65 ± 2.43	27.23 ± 0.80
				I7	12.60 ± 0.53	66.80 ± 2.80	22.26 ± 0.93
Min	7.83 ± 0.33	41.50 ± 1.75	13.85 ± 0.58		12.60 ± 0.40	66.80 ± 2.80	22.26 ± 0.93
Max	14.2 ± 0.53	75.25 ± 2.80	25.09 ± 0.93		16.95 ± 0.53	89.82 ± 2.80	29.95 ± 0.93
Mean	10.97 ± 0.43	58.14 ± 2.28	19.38 ± 0.75		14.70 ± 0.50	77.93 ± 2.65	25.98 ± 0.88

Table 3 Literature data on ^{210}Pb and ^{210}Po activity concentrations in tobacco products, estimation of the re-calculated effective dose due to inhalation of ^{210}Po and ^{210}Pb, and the total effective dose per year. For details see text

Country	Activity ^{210}Po mBq/cig	Activity ^{210}Pb (mBq/cig)	Dose ^{210}Po per year (1)	Dose ^{210}Pb per year (2)	Total dose (1) + (2)	Authors
Brazil	15.45	15.55	81.8	27.4	109.2	Peres and Hiromoto (2002)
Bulgaria	14	14*	74.2	24.7	98.9	Parfenov (1974)
Canada	7.9	7.9*	41.8	13.9	55.7	Black and Brethauer (1968)
China	23	23	121.9	40.6	162.5	Schayer et al. (2009)
Cuba	11.8	11.8*	62.5	20.8	83.3	Brigido Flores et al. (2015)
Czech Republic	16.65	16.65*	88.2	29.4	117.6	Jandl and Petr (1988)
Egypt	16.3	16.3	86.4	28.9	115.3	Khater (2004)
England	17.3	17.3*	91.7	30.5	122.2	Black and Brethauer (1968)
Finland	11.1	11.1*	58.8	19.6	78.4	Mussealo-Rauhammaa et al. (1985)
France	23.2	23.2*	123	41	164	Black and Brethauer (1968)
Germany	19.2	19.2*	101.7	33.9	135.6	Black and Brethauer (1968)
Greece	10.5	10.7	55.6	18.9	74.5	Savidou et al. (2006)
Hungary	22	20.8	116.6	36.7	153.3	Kovacs et al. (2007)
India	13.2	11.9	70	21	91	Christobher et al. (2019)
Iran	18.1	18.1*	99.6	31.9	131.5	Horvath et al. (2017)
Italy	15.7	15.7	83.2	27.7	110.9	Taroni et al. (2014)
Japan	8*	8	42.4	14.1	56.5	Sakoda et al. (2012)
Norway	8.6	8.6*	45.5	15.2	60.7	Black and Brethauer (1968)
Pakistan	13*	13	68.9	22.9	91.8	Tahir and Alaamer (2008)
Philippines	12	12*	63.6	21.2	84.8	Iwaoka et al. (2019)
Poland	13.9	13.9	73.6	24.5	98.1	Skwarzec et al. (2001a, b)
Portugal	14	14*	74.2	24.7	98.9	Carvalho and Olivira (2006)
Romania	14	14*	74.2	24.7	98.9	Robert-Csaba et al. (2015)
Russia	14.1	14.1*	74.7	24.9	99.6	Black and Brethauer (1968)
Saudi Arabia	15.1	15.1*	80	26.7	106.7	Khater anf Al-Sewaidan (2006)
Serbia	26.25	25.76	139.1	45.5	184.6	Jankovic-Mandic et al. (2016)
Slovenia	14	10	74.2	17.6	91.8	Kubalek et al. (2016)
Syria	1.88	1.88*	9.9	3.3	13.2	Batarekh and Teherani (1987)
Turkey	14.3	14.3*	75.7	25.2	100.9	Karali et al. (1996)
USA	19.9	19.9*	105.4	35.1	140.5	Black and Bretthauer (1968)
Vietnam	26.4	25.8	139.9	45.5	185.4	Tran et al. (2014)
Tunisia	12.9 ± 0.4	12.9 ± 0.4*	68.3 ± 2.4	22.7 ± 0.8	90.6 ± 3.3	This study
Min	1.88	1.88	9.9	3.3	13.2	
Max	26.25	25.76	139.1	45.5	184.6	
Mean	15.11	14.88	80.2	26.2	106.4 ± 5.3	

The uncertainty of the estimated values for ^{210}Po and ^{210}Pb is 5%

*The activity concentration was estimated from ^{210}Po assuming secular equilibrium

68 ± 2.4 μSv year^{-1}, while for ^{210}Pb annual effective dose varied between 13.9 ± 0.6 and 30.0 ± 0.9 μSv year^{-1} with an average 22.6 ± 0.8 μSv year^{-1}.

It is known that, for a smoker, the effective dose depends on several parameters such as the conversion factor and the percentage of concentration of radionuclides retained in the lungs. Effective doses reported in different studies are often difficult to compare, because often different approaches and parameters were used to calculate them. For this reason, the doses estimated in the majority of studies were re-calculated in the present study on the basis of their reported activity concentrations of ^{210}Po and ^{210}Pb using the same parameters as used in the present study (i.e., using a 22% net intake and the conversion factors recommended by ICRP (2012) for ^{210}Po and ^{210}Pb of 3.3 nSv/Bq and 1.1 nSv/Bq, respectively). The new estimated effective doses due to inhalation of ^{210}Pb and ^{210}Po and the combined doses in different countries of the world are summarized in Table 3.

The data shown in Table 3 demonstrate that some countries such as Syria, Canada, Japan, and Norway have low total effective doses per year of 13.2, 55.7, 56.5, and 60.7 μSv year^{-1}, respectively, while Iran, Germany, Hungary, France, Serbia, and Vietnam have higher doses of 131.5, 135.6, 153.3, 164, 184.6, and 185.4 μSv year^{-1}, respectively. The mean value of the total effective dose due to inhalation of ^{210}Po and ^{210}Pb for a smoker in Tunisia is 90.6 ± 3.3 μSv year^{-1}. The mean value of the total effective dose from these radionuclides in the world is estimated to be 106.4 ± 5.3 μSv year^{-1}. Persson and Holm assumed that for a heavy smoker (30 cigarettes per day) and with the conversion factor of 1.1 μSv Bq^{-1}, the committed annual dose equivalent is in the order of 100 μSv year^{-1} (Persson and Holm 2011). Thus, for a smoker who consumes 20 cigarettes per day, the annual effective dose will be about 200 μSv if a conversion factor of 3.3 μSv Bq^{-1} for ^{210}Po is used (ICRP 2012). This value, without the contribution of ^{210}Pb, is rather high compared to the values calculated in the present study.

Conclusion

The annual effective doses due to inhalation of ^{210}Po and ^{210}Pb estimated for Tunisian tobacco smokers as obtained in the present study were 68 ± 2.4 μSv and 22.76 ± 0.8 μSv year^{-1}, respectively, which is lower than the corresponding global mean values. It is known that tobacco smoking is a global pandemic affecting over 1.1 billion people and causing nearly 8 million deaths annually (WHO 2019) by inhaling various chemical and radioactive materials. The radiological risk associated with cigarette smoking was assessed in the present study with focus on ^{210}Po and ^{210}Pb. This assessment must be supplemented by accurate estimation of the activity concentrations of thorium isotopes and their progeny. Consequently, further work will be focused on the investigation of the intake of thorium isotopes and progeny in cigarettes that are also retained for quite a long time in the lungs of smokers.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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