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NATURAL RADIOACTIVITY AND ESTIMATED DOSE IN BRAZILIAN TOBACCO PRODUCTS

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ABSTRACT

Tobacco products contain significant concentrations of natural radionuclides from ^{238}U and ^{232}Th series. The consumption of these products increases the internal dose of radiation due to the inhalation of the natural radionuclides. Studies from literature emphasize that tobacco products have measurable concentrations of ^{210}Po and ^{210}Pb, and may contribute significantly to the increase of internal radiation dose and a large number of lung cancer in smokers. The objectives of this work were to determine the concentrations (Bq/kg) of the radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po and calculate the internal doses of radiation due to the consumption of these products. In the present work 71 samples were analyzed, consisting of cigars, unflavored and flavored cigarettes, straw cigarettes, cigars and roll smoke. The samples were purchased in Brazilian popular commercial establishments. The analytical techniques employed were the gross alpha and beta measurement after radiochemical separation for the radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and alpha spectrometry for ^{210}Po. The internal radiation doses were calculated with the activity concentrations determined and using the ICRP Publication 119 dose coefficients. An annual consumption of 3,650 kg of tobacco products was considered. The inhalation rates of each radionuclide followed the rates of the current literature. The estimated mean annual dose varied from 76 to 263μSv/y for the tobacco product studied in this work.

1. INTRODUCTION

Human being is continuously exposed to ionizing radiation of natural occurrence, coming from cosmic rays and natural radionuclides from the ^{238}U and ^{232}Th series that are present in the terrestrial crust, building materials, water, air and food. Some habits of the human being may increase the radiation dose received, as the smoking habit [1].

Low concentrations of the natural radionuclides from the ^{232}Th and ^{238}U series can be found in several agricultural products coming from soil or by foliar deposition [2, 3, 4] and among them is the tobacco plant that presents a significant concentration of these natural radionuclides [3]. Tobacco plants absorb the radionuclides from the soil during their growth and mainly for ^{210}Pb by foliar deposition, which is a decay product of ^{222}Rn of natural occurrence in the atmosphere [5, 3, 4].

Concentration of natural radionuclides present in tobacco plants vary according to the region of the planting and the cultivation methods. For example, tobacco plants from India present five times less ^{210}Po than North American plants. These differences in the radionuclide

concentrations is due to different amount of rainfall, natural radioactivity of the soil, differences of the cultivated soil pH and the type of fertilization used [5, 4].

^{222}Rn, an inert gas, can be found in the atmosphere by the decay of ^{226}Ra, both from the ^{238}U decay series, which is present in the soil and rocks. This gas binds to dust particles that accumulate in tobacco leaves and these one present characteristics that facilitate the impregnation of ^{222}Rn. Tobacco leaves have a water-insoluble resin on both sides of the leaf which causes the leaves to exhibit concentrations of ^{222}Rn up to 10,000 times greater than the concentration in the other parts of the plant [6].

^{222}Rn decays to ^{210}Pb, that decays to ^{210}Po, the latter responsible for most of the radioactivity of cigarettes. ^{210}Pb and ^{210}Po form insoluble complexes with the resin of tobacco leafs, which are highly resistant to washing by precipitation or curing process of the tobacco leaves [6, 4].

The alpha radioactivity of ^{210}Po in tobacco smoke depends on several variables: geographical region of tobacco growth, storage time, presence of a filter, its length and composition and the way of smoking. About 50% of ^{210}Po of a cigarette is transferred with smoke, 35% remain in the butt and 15% are found in the ashes [4].

The absorption of ^{210}Pb and ^{210}Po in smokers is due to the fact that they present a degraded mucosal-ciliary. The radioactive particles spread throughout the body via the systemic and pulmonary circulation causing mutations, aging and cell death, generating a wide range of diseases [4].

Fertilizers, mainly phosphates used in the tobacco plantations, can change the plants physiology and modifying the minerals absorption from soil by the roots and may increase the concentration of the absorbed radionuclides. ^{226}Ra presents higher potential of translocation and accumulation in the plant and hence, can increase the concentrations of ^{210}Po e ^{210}Pb in the tobacco leaves [5, 6]. The cure process of the tobacco leaves does not decrease the radioactivity [6].

Experiments performed with bean plants show that more than 90% of the ^{210}Pb and about 30% of the ^{210}Po deposited in the leaves are retained in the plant, and low concentrations of ^{210}Po are displaced to other parts of the plant [7].

Smoking is considered a major risk factor for death worldwide [4, 8]. It is known that tobacco smoke has been harmful to health since the 1950s. Cigarette smoke is a mixture composed of 5% particulate phase and 95% gas phase, 5000 chemical compounds, some of which are toxic, irritant, inflammatory and co-carcinogenic, including 73 potentially carcinogenic compounds [4, 9, 10]. Smoking usually begins in adolescence, before fully developing knowledge about the addictive power, health issues and mortality associated with it [9]. It is the second drug most consumed among Brazilian students and according to the Brazilian government it is considered an important gateway to the use of illicit drugs [11].

Since the 1960s researchers have reported the presence of ^{210}Po and ^{210}Pb in both the gas and particulate phases of cigarette smoke [5, 6], alerting to the potential risk of cancer due to its radioactivity. However, the industries did not alerted or warned consumers about this potential risk and do not allow any publications related to tobacco's radioactivity; they are

resistant to researches about the removal of these radionuclides from tobacco leaves, since they do not believe in the veracity, reproducibility and reliability of the published data [6].

Radio and thorium isotopes are also found in cigarettes, though 99% of its radioactivity comes from ^{210}Po [4]. When the cigarette is burned, these elements are volatilized and inhaled directly into the lungs of smokers [5, 6] generating a dose of localized radiation higher than the natural doses of nonsmokers. This exposure for long periods increases significantly the risk of induction of cancerous lesions [5].

Some studies verified that the concentration of ^{210}Po in the urine of smokers is about six times greater than that of nonsmokers and that the rate of bladder cancer increases considerably compared to nonsmoker. Other studies show that concentrations of ^{210}Pb and ^{210}Po in the ribs and alveolar tissues of smokers are 100 times higher than in nonsmokers. ^{210}Po may also be found in the smokers' bone tissues because of its relatively high affinity for this tissue, increasing the risk of leukemia in smokers and making it a tumor marker [5, 4].

Zaga, et al. [4] mentions that until the second half of the 20th century lung cancer was considered an extremely rare disease. After this period the disease rose so rapidly that it became an epidemic in men in developed countries and those that adopted some measure of control of tobacco dependence in the 1950s had the incidence of lung cancer reduced. The author also cites that levels of ^{210}Po found in blood, urine, liver, kidney, heart, lung and some muscles and bones are up to six times greater than that of a non-smoker.

Wünsch Filho [12] points out that the expansion of tobacco consumption in modern society occurred during the Industrial Revolution and that during the 60s and 70s with the women's freedom movements there was also a marked increase in the female smoking public.

The filters commonly found in commercial cigarettes exhibit negligible retention of these radionuclides, therefore almost all of these concentrations in cigarette smoke are inhaled directly into the lungs [5].

Brazil is the second largest producer of tobacco and the largest exporter in the world since 1993. The Brazilian production is concentrated in the South region of the country and in the crop year of 2015/2016 this production reached 607.010 ton, becoming an important agricultural activity. Of this total production, 85% is destined for the international market and clients from many countries purchase Brazilian tobacco. Thus it is very important the study of different tobacco products whereas Brazil is the largest exporter of tobacco in the world [13] and has a smoking rate of 15% of its population [8].

Therefore the objectives of this work were to determine the concentrations (Bq/kg) of the radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po and calculate the internal doses of radiation due to the consumption of one cigarette packet per day (approximately 10g of tobacco products per day).

2. METHODOLOGY

In the present work, 71 products manufactured with national tobacco acquired in public stores were analyzed: 16 samples of unflavored cigarette, 9 samples of flavored cigarette,

30 samples of roll smoke, 12 samples of straw cigarettes and 4 samples of cigars. For the cigarettes samples purchased were analyzed the products of the three main companies in the Brazilian market, the main and most consumed brands, price and intensity variations.

The radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po were determined by alpha and gross beta measurement after radiochemical separation. The samplers were solubilized in a hot plate with the addition of HNO_3 conc. and 30% H_2O_2 to eliminate the organic matter. After solubilization they were diluted to 1 L with super pure water and submitted to a sequential radiochemistry procedure to determine the concentration of the radionuclides. The alpha and beta particles of ^{226}Ra and ^{228}Ra, respectively, were measured 21 days after precipitation of $Ba(Ra)SO_4$ and the beta particle of ^{210}Pb after 10 days after precipitation of $PbCrO_4$. A low background gas flow proportional detector, Berthold model LB 770, was used for the measurement [14].

For the ^{210}Po determination 3g of the sample, in duplicate, spiked with ^{209}Po tracer were dissolved with concentrated HNO_3 and 30% H_2O_2, to eliminate the organic matter, under heating at 80°C on a hot plate. A solution of 6.25 mol L^{-1} HCl was added to change the solution medium and evaporated to dryness three times. The final residue was filtered through a 0.45μm Millipore filter paper and ascorbic acid was added to the solution. The pH was adjusted to 1.5 with concentrated HCl or 25% ammonia solution. ^{210}Po was spontaneously plated on a cupper disc at 80°C for 4 hours, under agitation on a hot plate magnetic stirring. The alpha particle measurement was performed on a surface barrier detector, EG&G Ortec for 150,000 seconds [15].

3 Results and Discussion

Table 1 presents minimum, maximum, mean values and standard deviation in Bq/kg, and Fig. 1 the mean values of ^{226}Ra, ^{210}Pb, ^{210}Po and ^{228}Ra activity concentration determined in unflavored cigarette, flavored cigarette, straw cigarette, cigar and roll smoke samples analyzed in the present work. The detection limit of the ^{226}Ra, ^{210}Pb, ^{210}Po and ^{228}Ra determination are 2.2 ± 0.2 mBq/kg, 4.9 ± 0.4 mBq/kg, 3.7 ± 0.1 mBq/kg and 4.2 ± 0.4 mBq/kg [15, 16, 17].

^{228}Ra presented the highest activity concentrations in all samples analyzed, with a maximum value of 118 ± 6 Bq/kg in a sample of flavored cigarette and the lowest was for ^{210}Po in the majority of the samples, although ^{210}Pb presented a value below the detection limit also in a sample of flavored cigarette. It can be also observed that the natural radionuclides ^{210}Pb and ^{210}Po are in equilibrium in the majority of the samples, where the concentration ratio values for $^{210}Po/^{210}Pb$ is approximately 1. ^{210}Pb and ^{210}Pb tend to achieve radioactive equilibrium in tobacco leaves that are processed for cigarette production [3, 18]. This equilibrium is verified mainly in industrialized products, because since the harvest of the leaves until the tobacco product is commercialized approximately seven half-lives of the ^{210}Po are passed [18]. However, this equilibrium was not observed for roll smoke and straw cigarettes samples where the concentration of ^{210}Pb is larger than ^{210}Po; for cigar samples it was observed the opposite, concentration of ^{210}Po greater than ^{210}Pb.

The mean values obtained for ^{228}Ra, 48 ± 23 Bq/kg in roll smoke samples are lower than that determined by Silva [15], 78 ± 21 Bq/kg, which analyzed tobacco leaves, although roll smoke is a tobacco product less industrialized.

Table 1: ^{226}Ra, ^{210}Pb, ^{210}Po and ^{228}Ra mean activity concentrations in unflavored cigarette, flavored cigarette, straw cigarette, cigar and roll smoke

Activity concentrations in Bq/kg					
Tobacco products		^{226}Ra	^{210}Pb	^{210}Po	^{228}Ra
unflavored cigarette	**Min**	7 ± 1	15 ± 5	14 ± 1	34 ± 2
	Max	13 ± 2	26 ± 2	25 ± 1	45 ± 6
	Mean	10 ± 2	21 ± 4	21 ± 3	40 ± 3
flavored cigarette	**Min**	4,7 ± 0,2	< 4,9 ± 0,4	6,3 ± 0,5	34 ± 2
	Max	13 ± 2	25 ± 2	22 ± 1	118 ± 6
	Mean	7 ± 3	18 ± 8	16± 6	76 ± 31
straw cigarette	**Min**	6 ± 2	17 ± 2	11 ± 1	37 ± 3
	Max	22 ± 7	55 ± 5	35 ± 2	81 ± 4
	Mean	12 ± 5	30 ± 11	19 ± 7	57 ± 12
cigar	**Min**	18 ± 1	6,2 ± 0,5	15 ± 1	52 ± 3
	Max	28 ± 1	22 ± 2	22 ± 1	78 ± 4
	Mean	23 ± 5	13 ± 7	18 ± 3	63 ± 12
roll smoke	**Min**	6 ± 2	5 ± 1	10 ± 1	28 ± 6
	Max	40 ± 6	78 ± 2	28 ± 1	152 ± 1
	Mean	12 ± 6	35 ± 21	18 ± 5	48 ± 23

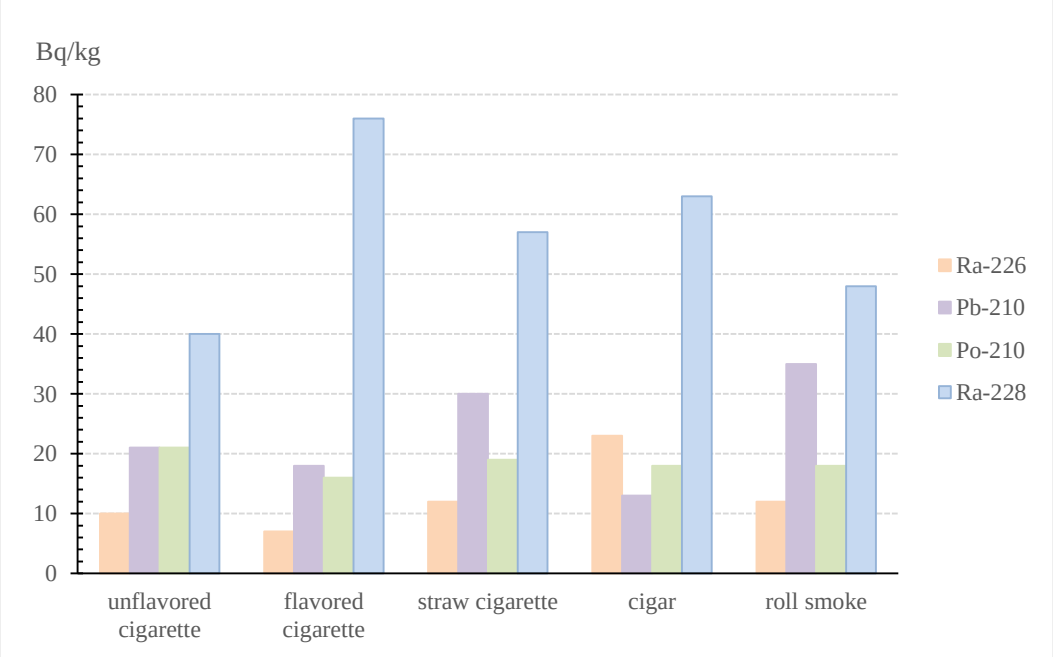

FIG.1: ^{226}Ra, ^{210}Pb, ^{210}Po and ^{228}Ra mean activity concentrations in unflavored cigarette, flavored cigarette, straw cigarette, cigar and roll smoke

Comparing the mean values obtained for ^{226}Ra of 12 ± 6 Bq/kg in both, straw cigarette and roll smoke samples, with values obtained by Silva [15], which analyzed Brazilian tobacco leaves and obtained an mean value of 10 ± 3 Bq/kg it was verified that the results obtained in

the present work are in the same order of magnitude; the same was also observed with the values obtained by Abd El-Aziz et al. that analyzed Moasel tobacco from Saudi Arabian [2].

Although ^{228}Ra had exhibited the highest activity concentrations in cigarette and other tobacco product samples of this work, attention should be directed to the radionuclides ^{210}Pb and ^{210}Po, since they are volatilized at the temperature of burning a cigarette and they are inhaled [3]. Polonium is a very volatile metal, of great toxicity and specific radioactivity; its alpha rays are highly ionizing, capable of causing considerable damage to the human organism. Approximately 1 milligram of polonium emits a quantity of alpha particles equivalent to 5 grams of radio [4].

The results obtained in the present work for flavored and unflavored cigarette were compared with data from the literature, Fig. 2. It is possible to observe that the results for ^{226}Ra are similar to those present by Kubalek [19], of the same magnitude for ^{210}Pb and smaller for ^{210}Po for the majority of the works.

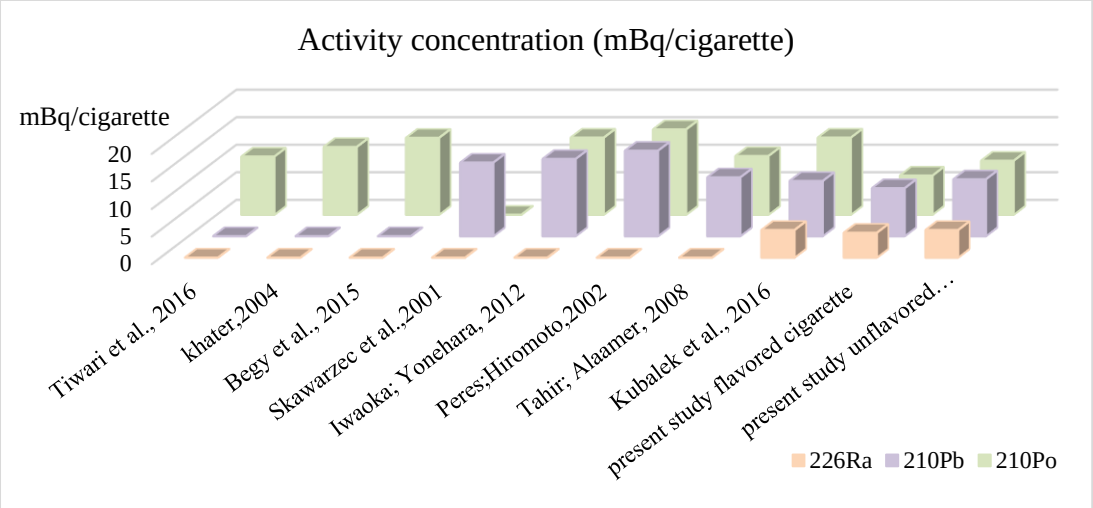

FIG.2: Comparison with literature of the activity concentrations in mBq/cigarette for ^{226}Ra, ^{210}Pb and ^{210}Po in cigarettes

The results obtained of ^{210}Pb and ^{210}Po in the present work for roll smoke were also compared with data from the literature of tobacco leaves and Moasel, Table 2. It is possible to observe that the results for ^{210}Pb are smaller to those present by Silva [15] and higher than the other works and for ^{210}Po the results are in the same magnitude of the works. [2, 15, 26, 27].

Table 2: Comparison with literature of the activity concentrations for ^{210}Pb and ^{210}Po in tobacco samples

Authors	Product	^{210}Pb (Bq/kg)	^{210}Po (Bq/kg)
Savidou et al., 2006	tobacco leaves	13	13
Yaprak and Uysal, 1998		16,6	16,6
Silva, 2015	Burley tobacco leaves	61	25
	Virginia tobacco leaves	69	29
Abd El-Aziz et al., 2005	Moasel tobacco	15,6	13,1
present study	roll smoke	35	18

The results obtained for ^{210}Pb and ^{210}Po in cigar samples in the present work are in the same magnitude than those obtained by Kelecon [20] that analyzed cigar samples from Brazil and other countries, Table 3.

Table 3: Activity concentratios for ^{210}Pb and ^{210}Po in cigar samples in the present work and Kelecon [20]

Authors	Origin	^{210}Po (Bq/kg)	^{210}Pb (Bq/kg)
Kelecon	Brazil	25,7 ± 4,6	15,2 ± 6,9
	Cuba	13,0 ± 4,7	6,0 ± 4,7
	Dominican Republic	24,5 ± 8,4	11,9 ± 5,3
	Europe	24,6 ± 8,7	5,1 ± 2,4
	Latin America	34,0 ± 14,4	12,3 ± 4,4
	USA	16,9 ± 7,3	5,4 ± 1,6
Present work	Brazil	18 ± 1	13 ± 1

3.2 Annual Dose Estimation

For the calculation of the annual dose estimation it was considered that a cigarette (unflavored and flavored) has approximately 0.5g of tobacco and that a smoker consumes a packet of 20 cigarettes per day, therefore a mass of 10g of tobacco per day or 3.65 kg per year. Table 3 presents the activity concentrations per cigarette, per day and per year,

Table 3 Range of the activity concentrations of ^{226}Ra, ^{210}Pb, ^{210}Po and ^{228}Ra in unflavored and flavored cigarettes per cigarette, per day and per year

Activity Concentration						
	unflavored cigarette			flavored cigarette		
	mBq/cigarette	mBq/day	Bq/year	mBq/cigarette	mBq/day	Bq/year
^{226}Ra	3,5 - 6,5	70 - 130	26 - 47	2,4 - 6,5	48 - 130	18 - 47
^{210}Pb	7,5 - 13,0	150 - 260	55 - 95	<4,9 - 12,5	98 - 250	36 - 91
^{210}Po	7,0 - 12,5	140 - 250	51 - 91	3,1 - 11,0	62 - 220	23 - 80
^{228}Ra	17 - 23,5	340 - 450	124 - 164	17 - 59	340 - 1180	124 - 431

The ingested fractions of the studied radionuclides from the smoke to the lungs followed the values of 0.5 for ^{210}Po [18], and 0.2 for ^{210}Pb [23]. Equation (1) was used for the calculations:

$$D = Ma \times A \times DCF \times if \qquad (1)$$

In equation (1) D is the annual dose estimation (Sv/year); Ma is the annual mass of tobacco consumed (kg); A is the activity concentration of the radionuclide (Bq/kg); DCF is the dose conversion factor for the radionuclide (Sv/Bq) from ICRP Publication 119 (1,0 x 10^{-6} Sv/Bq for ^{210}Pb; 3,3 x 10^{-6} Sv/Bq for ^{210}Po and 3,5 x 10^{-6} Sv/Bq for ^{226}Ra [28]; and if is the ingested fraction of the radionuclide [18; 23, 29]

Fig. 3 presents the results of mean annual dose estimation for unflavored and flavored cigarette, straw cigarette, cigar and roll smoke for the radionuclides ^{226}Ra, ^{210}Pb and ^{210}Po. The highest values obtained were for ^{226}Ra in cigars samples and for ^{210}Po in straw cigarette, cigar and roll smoke samples and the lowest for ^{210}Pb in unflavored and flavored cigarette and cigars.

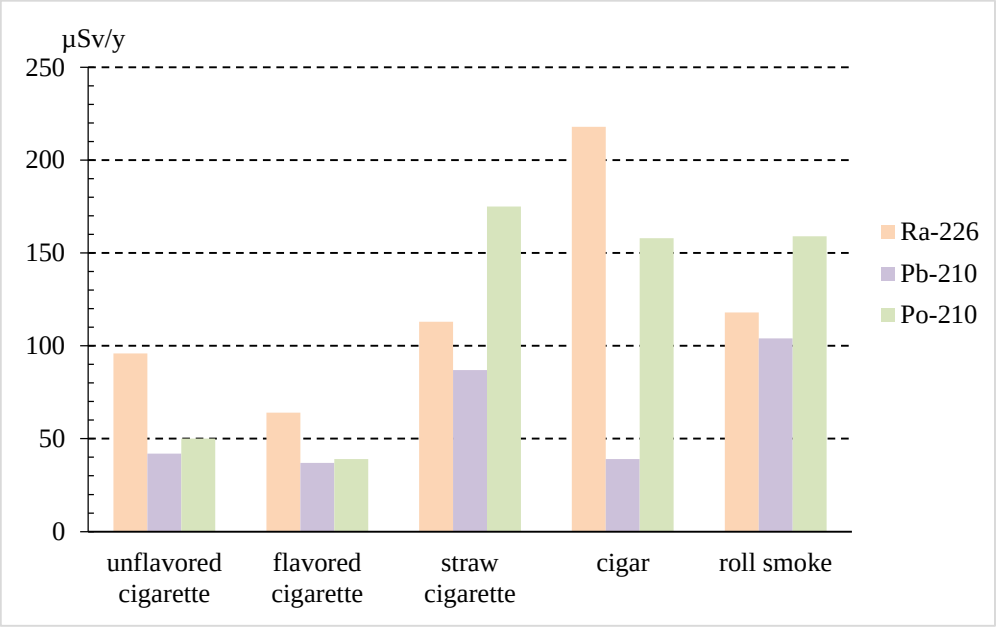

FIG.3: Mean Annual Dose Estimation for unflavored and flavored cigarette, straw cigarette, cigar and roll smoke for ^{226}Ra, ^{210}Pb and ^{210}Po

Tabel 4 presents a comparison of the results of annual dose estimation, µSv/y among the present work and results from the literature for ^{210}Pb and ^{210}Po. The results obtained in the present work for unflavored cigarette samples are in the same order of magnitude of the works of Skawarzec [24] and Tahir and Allamer [18] that analyzed cigarette samples from Poland and Pakistan, respectively; the same can be observed for roll smoke samples and the results of Savidou [26].

Table 4: Comparison of present work results of annual dose estimation and data from literature for ^{210}Pb and ^{210}Po

Annual Dose Estimation (µSv/y)			
Authors	**Tobacco Product**	**Radionuclide**	
		^{210}Pb	**^{210}Po**
Tiwari et al., 2016	cigarette	-	300
Skawarzec, 2001		70	35
Khater, 2004		251	193
Kubalek et al., 2016		9	61
Begy et al., 2015		-	8
Iwaoka and Yonehara, 2012		270	270
Tahir and Allamer, 2008		64	64
Peres, 2002		160	160
Present work	unflavored cigarette	42	50
	flavored cigarette	37	39
Savidou et al., 2006	tobacco leaves	163	124
Present study	straw cigarette	87	175
	cigar	39	158
	roll smoke	104	118

- Not determined

3.3 Annual Effective Dose

Equation (2) was used for calculate the Annual Effective Dose (µSv/y) [4].

$$E_D = D_{Po} + D_{Pb} \qquad (2)$$

In equation (2) E_D is the Annual Effective Dose; D_{Po} is the Annual Dose Estimation for ^{210}Po and D_{Pb} is the Annual Dose Estimation for ^{210}Pb. Fig. 4 presents the results of the annual effective dose for the tobacco products analyzed in this work.

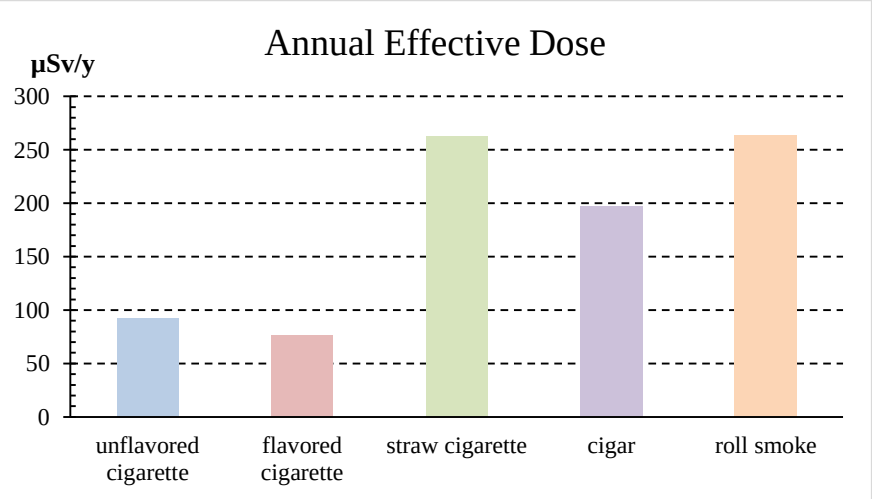

FIG.4: Annual Effective Dose for tobacco products

Straw cigarette and roll smoke were the samples that presented the highest annual effective dose. The values of annual effective dose ranged from 76 µSv/y to 263 µSv/y, however, these results are below the ICRP [30] stipulated dose limit of 1 mSv /y.

3. Conclusions

In this study, 71 samples of tobacco products produced and commercialized marketed in Brazil were analyzed for the activity concentration of radionuclides ^{226}Ra, ^{210}Pb and ^{210}Po from ^{238}U decay series and the radionuclide ^{228}Ra from the ^{232}Th decay series.

Mean activity concentrations varied from 7 ± 1 Bq/kg to 23 ± 1 Bq/kg for ^{226}Ra, from 13 ± 1 Bq/kg to 35 ± 3 Bq/kg for ^{210}Pb, from 16 ± 1 Bq/kg to 21 ± 1 Bq/kg for ^{210}Po and from 40 ± 5 Bq/kg to 76 ± 4 Bq/kg for ^{228}Ra. The estimated mean annual effective dose was 92µSv/y for unflavored cigarette, 76 µSv/y for flavored cigarette, 262 µSv/y for straw cigarette, 197 µSv/y for cigar and 263 µSv/y for roll smoke; these results are below the stipulated by the International Commission on Radiological Protection of 1mSv /y.

The data generated in this study contribute to a better knowledge of the levels of radioactivity and stipulates the effective dose of radiation inhaled during smoking, considering a daily consumption of 10g of tobacco.

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