

## Determination of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in Brazilian Cigarette Tobacco

A.C. Peres and G. Hiromoto  
Instituto de Pesquisas Energéticas e Nucleares, Caixa Postal 11049, São Paulo, Brasil

### INTRODUCTION

Due to the presence of natural radionuclides of the uranium and thorium series, in an amount larger than that normally found in general foods for human consumption, the tobacco has been well studied under the radiological point of view; however, no consensus have yet been reached about the contribution of the radiation for the increment in the incidence of lung cancer among smokers (1).

Among the several toxicant agents found in cigarette, the  $^{210}\text{Po}$  is the element of major radiological concern because, due to its low volatilization temperature, it can be inhaled with the smoke of the cigarette; for being its precursor in the decay chain and for the characteristics of the processes of production of the cigarette, the  $^{210}\text{Pb}$  are another element of interest.

Related studies reported in the literature indicate activity concentrations of  $^{210}\text{Po}$  in cigarettes ranging from 6 to 31 mBq per gram of dry tobacco (2).

This work is part of a study that seeks to contribute for the increment of the database related to the natural radioactivity in Brazil, and to motivate the discussion on the legislative aspects of the commercial use of consumer products containing amount of natural radionuclides artificially enhanced by man. In this paper the radiochemical procedures adopted for the determination of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are described and presents the results of the activity concentration found in major brands of the cigarettes produced in Brazil, for these two elements.

### MATERIALS AND METHODS

#### Radiochemical procedure for $^{210}\text{Pb}$ determination

To a dry sample of 5 g of tobacco, 1 mL of lead loader ( $20 \text{ mg.mL}^{-1}$ ) and 1 mL of barium loader ( $20 \text{ mg.mL}^{-1}$ ) were added, in order to increase the concentration of these ions in the solution (3).

The sample was leached with 40 mL of concentrated nitric acid, under heating. At close to the dryness, it was added more 30 mL of nitric acid; this procedure was repeated once more.

After, 10 mL of hydrogen peroxide were added for the destruction of the organic matter (4,5) and 10 mL of nitric acid 8M were added, being taken to the dryness for twice. The sample was filtered and diluted for 1 L with deionized water.

To proceed, 10 mL of citric acid 3 M were added, the solution was heated up to ebullition and 70 mL of sulfuric acid 3M were added under agitation for the precipitation of the barium and lead sulphate.

After 24 h of settling, the aqueous phase was discarded and the precipitate transferred for a centrifuge tube with the aid of deionized water; it was centrifuged by 10 minutes at 2000 rpm and then washed with approximately 20 mL of sulfuric acid 0.1M and centrifuged again.

To the precipitate, 2 g of nitrilotriacetic acid (Titrplex I), 40 mL of deionized water and 7 mL of sodium hydroxide 6M were added, dissolving under heating. It was added 5 mL of ammonium sulphate ( $25 \text{ mg.mL}^{-1}$ ) for the Ra separation and 3 drops of methyl red; acetic acid was added until the precipitation and the sample was let to repose for 24 h; the lead remained in solution, complexed with the nitrilotriacetic acid.

The solution was centrifuged and the aqueous phase was transferred for another centrifuge tube; the precipitate was washed with ammonium acetate 20%, centrifuged again and the aqueous phase was added to the previous one. The solution was heated in steam bath until almost the ebullition; it was added 1 mL of sodium sulphide 1M for the lead sulphide precipitation and heated for more 10 minutes.

After 20 minutes centrifugation, the precipitate was washed with deionized water, centrifuged again and then dissolved with drops of concentrated nitric acid and 10 mL of deionized water. The solution was filtered in Whatman 44 paper. The pH of the filtrate was adjusted to 4.5-5.0 with ammonium acetate 40%. The filtrate was heated until the ebullition and 2.5 mL of sodium cromate 30% was added for the lead cromate precipitation. The solution was vacuum filtered in Millipore paper, washing with ethyl alcohol 50%.

The filter paper with the precipitate was covered with an aluminized foil, in order to avoid the interference of the low energy beta emission of  $^{210}\text{Pb}$  in the counting process.

After about 10 days, the  $^{210}\text{Pb}$  was determined by beta counting of the  $^{210}\text{Bi}$  grown in the precipitate, using a low background gas flow proportional counter.

The counting efficiency was determined using a standard solution of  $^{210}\text{Pb}$ , prepared and measured following the same methodology adopted for the tobacco sample; the value obtained was about 35% for each

detector. The yield of the process ranged from 61% to 98% and the typical lower limit of detection of this method was  $4.5 \text{ mBq.g}^{-1}$ , for 400 minutes time counting.

### Radiochemical procedure for $^{210}\text{Po}$ determination (6,7)

To a dry sample of 5 g of tobacco, 1 mL of  $^{208}\text{Po}$  tracer was added. The sample was leached with 40 mL of concentrated nitric acid, under heating at temperature not exceeding  $70^\circ\text{C}$  in order to avoid losses of polonium by volatilization. At close to the dryness, it was added more 30 mL of nitric acid; this procedure was repeated once more.

Following, 10 mL of hydrogen peroxide was added for the destruction of the organic matter and 10 mL of nitric acid 8M was added under heating. At close to the dryness, it was added more 10 mL of nitric acid 8M. The sample was filtered directly into a separation funnel containing 5 mL of TBP (tributylphosphate). The mixture was shaken by 5 minutes. After 10 minutes in rest, the polonium is found in the aqueous phase. The extraction was repeated twice with 10 mL of nitric acid 8M.

Then, the aqueous phase was taken close to the dryness by heating, in order to eliminate the nitric acid. After, 20 mL of concentrated chloridric acid was added under heating; at close to the dryness, more 20 mL of chloridric acid 2M was added. The iron was complexed by adding L(+)-ascorbic acid, until the change of the coloration was observed, from yellow to colorless.

The solution was transferred to a plating cell, where the  $^{208}\text{Po}$  and the  $^{210}\text{Po}$  were spontaneously deposited onto a copper disc, after 4 h under continuous agitation and heating at temperature bellow  $70^\circ\text{C}$  (8). The disc was washed with deionized water and let to dry at room temperature. The alpha spectrum was obtained by counting in a surface barrier detector.

The counting efficiency was determined using an electrodeposited source of  $^{241}\text{Am}$ ; the value obtained was  $0.125 \pm 0.001$ . The yield of the process ranged from 23 to 67% and the typical lower limit of detection of this method was  $5 \times 10^{-5} \text{ mBq.g}^{-1}$ , for 1000 minutes time counting.

## RESULTS

Eight of the most commercialized Brazilian brands of cigarettes, representing about 80% of the total national market, were analyzed in this work; although each brand has several types of cigarettes, only the most popular one was chosen for analysis. The cigarettes were acquired randomly in the trade.

For each analysis, accomplished in triplicate, the content of three packages was homogenized; from the mixture, 15 g were reserved for  $^{210}\text{Pb}$  determination and another 15 g for  $^{210}\text{Po}$  determination. The results obtained are presented in the Table 1.

## CONCLUSION

The radiochemical procedures adopted in the present work, although requiring a large number of steps for Pb and Po extraction, presented a good reliability for routine measurements of environmental samples.

The values of activity concentration of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  found in Brazilian cigarettes are within of the range of values found in the literature. The  $^{210}\text{Pb}$  presented concentrations ranging from 11.9 to  $30.2 \text{ mBq}$  per gram of tobacco, with arithmetic mean of  $21.3 \pm 4.1 \text{ mBq.g}^{-1}$ , while the  $^{210}\text{Po}$  presented concentrations ranging from 10.9 to  $27.4 \text{ mBq.g}^{-1}$ , with arithmetic mean of  $21.2 \pm 3.7 \text{ mBq.g}^{-1}$ .

Considering the error measurements, radioactive equilibrium can be observed between those two radionuclides; this kind of result was expected, since the time elapsed between the harvest of the tobacco leaves and the placement of the cigarette in the market, is enough for  $^{210}\text{Po}$  growing until reaching radioactive equilibrium with  $^{210}\text{Pb}$ .

Assuming that 10% of the Pb and 20% of the Po are inhaled by the primary smokers, and considering an annual production of  $5 \times 10^8$  kg of cigarette in Brazil, the collective committed effective dose resulting from the consumption of cigarettes corresponding to one year of production is estimated to be  $1.5 \times 10^4 \text{ man.Sv}$ . Although this is a rough estimation of collective dose, it is an indication that the cigarette smoking could be an important source of global dose resulting from avoidable natural radiation.

TABLE 1. Activity concentration of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in Brazilian cigarettes

BRAND	ACTIVITY CONCENTRATION (mBq.g <sup>-1</sup> )	
	<sup>210</sup> Pb	<sup>210</sup> Po
A	22.6 ± 3.7	20.9 ± 3.3
	23.6 ± 2.9	19.3 ± 0.8
B	27.8 ± 0.2	25.2 ± 3.1
	30.2 ± 2.2	27.4 ± 3.9
C	20.0 ± 1.9	22.9 ± 1.9
	23.3 ± 3.9	22.0 ± 2.7
D	11.9 ± 2.1	10.9 ± 2.2
	23.8 ± 1.6	20.7 ± 0.1
	(*)	23.5 ± 0.4
	22.8 ± 6.0	19.9 ± 3.4
E	20.1 ± 4.6	23.3 ± 0.3
	15.8 ± 1.2	(*)
F	20.7 ± 3.6	19.2 ± 3.4
	19.6 ± 3.0	26.2 ± 2.2
G	18.7 ± 2.2	20.5 ± 0.8
	20.4 ± 3.6	19.6 ± 2.8
H	20.4 ± 2.6	18.8 ± 5.3
	20.2 ± 3.7	21.6 ± 0.6

(\*) Not determined

The mean value of the mass of the cigarettes analyzed was (0.73 ± 0.03) g per cigarette.

## REFERENCES

1. G.F.Kilthau, *Cancer Risk in Relation to Radioactivity in Tobacco*. Radiologic Technology, vol. 67(3), p 217-222 (1996).
2. A.P.Watson, *Polonium-210 and Lead-210 in Food and Tobacco Products: A Review of Parameters and an Estimate of Potential Exposure and Dose*. Health and Safety Research Division. Flórida (1983).
3. S.R.D.Moreira, *Determinação de <sup>210</sup>Pb em águas minerais da região de Águas da Prata*. Dissertação de Mestrado. São Paulo, IPEN (1993).
4. A.E.Nevissi, *Measurement of <sup>210</sup>Lead, <sup>210</sup>Bismuth and <sup>210</sup>Polonium in environmental samples*. J. Radioanal. Nucl. Chem., vol. 148(2), p 121-131 (1991).
5. C.H.Colangelo, M.R.Huguet, M.A.Palacios, and A.A.Oliveira, *Levels of <sup>210</sup>Po in some beverages and in tobacco*. J.Radioanal. Nucl. Chem., vol. 166(3), p 195-202 (1992).
6. J.P.B.Raya, *Aplicaciones de la espectrometria gamma y alfa al estudio del impacto radiactivo producido por industrias no nucleares*. Tese de Doutorado. Sevilla, Universidad de Sevilla (1995).
7. R.T.Saito, *Determinação de <sup>210</sup>Pb e <sup>210</sup>Po em amostras marinhas e aerossóis*. Dissertação de Mestrado. São Paulo, IPEN (1996).
8. A.Nieri Neto, *Determinação de <sup>210</sup>Pb e <sup>210</sup>Po em Águas Minerais Radioativas*. Dissertação de Mestrado. São Paulo, IPEN (1996).