Concentration levels of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in dry tobacco leaves in Greece

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Abstract

Tobacco leaves are large and have sticky exudates that retain the radon decay products once they deposit on the leaves. The study of $^{210}\text{Po}$ in tobacco is required, because of the cumulative alpha-radiation dose delivered to humans from inhaled $^{210}\text{Po}$ in cigarette smoke. $^{210}\text{Pb}$ is the other element of interest since it is the $^{210}\text{Po}$ precursor in the radioactive decay chain. In the present study, the concentrations of these two radionuclides were determined in tobacco samples from seven regions in Greece. $^{210}\text{Po}$ was determined by alpha spectrometry using a surface barrier detector after radiochemical separation and spontaneous deposition of polonium on a nickel disk. The $^{210}\text{Pb}$ activity in the samples was determined via the $^{210}\text{Po}$ resulting from the decay of $^{210}\text{Pb}$. The results of the present study indicate that $^{210}\text{Po}$ concentrations ranged from 3.6 to 17.0 mBq g$^{-1}$ (average 13.1 mBq g$^{-1}$) of dry tobacco, while $^{210}\text{Pb}$ concentrations ranged from 7.3 to 18.0 mBq g$^{-1}$ (average 13.4 mBq g$^{-1}$). The mean value of the annual committed effective dose for smokers (20 cigarettes per day) of Greek tobacco was estimated to be 287 μSv (124 μSv from $^{210}\text{Po}$ and 163 μSv from $^{210}\text{Pb}$). The inhalation dose for smokers is on average about 12 times higher than for non-smokers living in the mid-latitudes of the northern hemisphere.

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1. Introduction

Among the several hazardous chemicals found in cigarettes, $^{210}$Po that is an $\alpha$-decay radionuclide, is the major radioactive element of interest because it can be inhaled with cigarette smoke, due to its relatively high volatility at the temperature of a burning cigarette. Polonium is volatilised at the temperature of a burning cigarette between 600 and 800 °C and it is inhaled in the vapour phase (Singh and Nikelani, 1976), or adsorbed on smoke particles (Martell, 1974). Lead is also volatile at temperatures over 500 °C (Parfenov, 1974). Cigarette smoking increases the intake of $^{210}$Pb and $^{210}$Po (Martell, 1983) leading to increased concentrations in the human body, particularly in the lungs. $^{210}$Po is deposited on the bronchial tissue and may play a role in the initiation of bronchial cancer in smokers (Martell, 1975; Singh and Nikelani, 1976). $^{210}$Pb decays by $\beta$-emission and it is the $^{210}$Po precursor in the radioactive decay chain of $^{238}$U. In the mid-latitudes of the northern hemisphere, the average concentration of $^{210}$Pb in surface air is 0.5 mBq m$^{-3}$, while that of $^{210}$Po is about 0.05 Bq m$^{-3}$ (UNSCEAR, 2000). The annual intakes of non-smokers through inhalation would be 4 Bq of $^{210}$Pb and 0.37 Bq of $^{210}$Po.

About 10% of the $^{210}$Pb and 20% of the $^{210}$Po contained in the cigarette will enter the lungs with the main smoke stream (Parfenov, 1974). Other authors have reported different percentage values, based on different tobacco-burning temperatures during smoking, between 500 and 700 °C (Martell, 1975). On average 75% of the $^{210}$Po contained in cigarettes was found in the cigarette smoke (Khater, 2004; Skwarzec et al., 2001). In these studies the polonium content in cigarette smoke was estimated on the basis of its activity in tobacco leaves, ash, fresh filters and post-smoking filters. The reported percentage values are ranging from 33 to 94%. Both studies assumed that 50% of the smoke aerosol generated is inhaled into a smoker’s lungs. Therefore on average we can assume that 37% of the $^{210}$Po contained in cigarettes is inhaled via smoking. Since both radionuclides have similar burning behaviour over 500 °C we can assume the same percentage value of $^{210}$Pb inhaled via smoking.

The main source of $^{210}$Pb and $^{210}$Po in the environment is the gaseous $^{222}$Rn, which escapes from the earth’s crust into the atmosphere. The daughter products of $^{222}$Rn are removed from the atmosphere with aerosol particles wet and dry deposition on the land surface and oceans (Skwarzec et al., 2001; Ladinskaya et al., 1973). Direct deposition of $^{210}$Pb and $^{210}$Po on the leaf surfaces of tobacco is the most important route for their accumulation by the plant (Karunakara et al., 2000). The large surface area of tobacco leaves and their hairy and sticky nature facilitates enhanced deposition of $^{210}$Pb on the leaves during the plant’s life. In Greece, tobacco leaves are dried after harvest on air and continues to be exposed to radon progeny in ambient air. Another source of $^{210}$Pb and $^{210}$Po are the phosphate fertilizers. A higher polonium concentration can be expected in tobacco leaves from fields rich in phosphate fertilizers (Tso et al., 1966).

The objective of this study was to determine the activity concentrations of $^{210}$Po and $^{210}$Pb in Greek tobacco just before packaging for consumption. The variation of concentration levels with respect to geographical origin of the tobacco is also
investigated. Although the activity concentrations of $^{210}\text{Pb}$ in Greek tobacco were determined by a previous study (Papastefanou, 2001), it was the first time that $^{210}\text{Po}$ concentrations in Greek tobacco were determined.

### 2. Materials and methods

Tobacco from seven regions in Greece was collected. From each region about eight samples from different tobacco producers were collected and mixed to produce a composite sample. An amount of 10 g of each composite sample was used for the analysis of $^{210}\text{Po}$ and $^{210}\text{Pb}$. For each region, at least two composite samples were analysed.

For the determination of both radionuclides, $^{209}\text{Po}$ tracer and lead carrier solution were added to 10 g of dried and homogenized tobacco. The sample was leached with nitric acid for 1 h. The pH was then adjusted to 3.5–4.0, followed by successive precipitation of thioacetamide solution, according to the procedure described by Fisenne (1990). The polonium isotope was spontaneously deposited on a nickel disc and the activity was measured by an alpha spectrometry system consisting of a 24 mm ruggedized surface barrier detector connected to a 7401 VR Canberra spectrometer. The spectra were obtained by a TRUMP 8 K multichannel buffer card on PC. The efficiency of the detector was determined by means of an $^{241}\text{Am}$ surface source with geometry identical to the nickel sample disks and was found to be 15%. The $^{210}\text{Pb}$ activity in the samples was measured from the solutions prepared earlier for the extraction of $^{210}\text{Po}$. They were stored for a period of 5–7 months to allow production of $^{210}\text{Po}$ resulting from the decay of $^{210}\text{Pb}$ at a known time interval. Saturated L-ascorbic acid was added and the $^{210}\text{Po}$ was again deposited as described above. The activity concentration of $^{210}\text{Pb}$ was calculated by measuring the activity of $^{210}\text{Po}$, as described below:

The rate of decrease of $^{210}\text{Pb}$ is given by:

$$\frac{dN_0}{dt} = -\lambda_0 N_0$$  \hspace{1cm} (1)

where $N_0$ is the number of $^{210}\text{Pb}$ nuclei and $\lambda_0$ the decay constant of $^{210}\text{Pb}$.

The rate of change of $^{210}\text{Po}$ is given by:

$$\frac{dN_1}{dt} = \lambda_0 N_0 - \lambda_1 N_1$$  \hspace{1cm} (2)

where $N_1$ is the number of $^{210}\text{Po}$ nuclei and $\lambda_1$ the decay constant of $^{210}\text{Po}$.

Just after the deposition of $^{210}\text{Po}$, $t_0 = 0$

$$N_0 = N_{0,0}$$  \hspace{1cm} (3)

$$N_1 = 0$$  \hspace{1cm} (4)
From Eq. (1) and initial condition (3) the number of $^{210}$Pb nuclei is given as follows:

$$N_0 = N_{0,0} e^{-\lambda_0 t}$$  \hspace{1cm} (5)

Combining Eqs. (2), (5) and initial condition (4) the number of $^{210}$Po nuclei is given as follows:

$$N_1 = \frac{\lambda_0 N_{0,0}}{\lambda_1 - \lambda_0} \left[ e^{-\lambda_0 t} - e^{-\lambda_1 t} \right]$$  \hspace{1cm} (6)

By measuring the decay rate of $^{210}$Po, \( (dN_a/dt) = \lambda_1 N_1 \) at a certain time \( t \) and using Eq. (1) at \( t = 0 \), Eq. (6) can be written as:

$$\frac{dN_{0,0}}{dt} = \frac{\lambda_1 - \lambda_0}{\lambda_1} \frac{dN_a}{dt} \left[ e^{-\lambda_0 t} - e^{-\lambda_1 t} \right]^{-1}$$  \hspace{1cm} (7)

yielding the decay rate or activity of $^{210}$Pb.

The chemical yield of the $^{210}$Po process was 23—62% and the minimum detection activity of this method (95% confidence level) was 0.3 mBq g$^{-1}$ with a counting time of 1000 min (Fig. 1).

Fig. 1. Typical spectrum for $^{210}$Po.
3. Results and discussion

$^{210}$Po concentrations ranged from 3.6 to 17.0 mBq g$^{-1}$ (average 13.1 mBq g$^{-1}$) of dry tobacco, while $^{210}$Pb concentrations ranged from 7.3 to 18.0 mBq g$^{-1}$ (average 13.4 mBq g$^{-1}$) of dry tobacco (Table 1).

The activity concentrations of $^{210}$Pb determined by the present study are in accordance with the results of a previous study on Greek tobacco. Papastefanou (2001) reported $^{210}$Pb concentrations in dried tobacco leaves collected from 17 different regions in Greece, ranging from 6.34 to 18.2 mBq g$^{-1}$ (average 14.12 mBq g$^{-1}$).

The results of this study are in the range of values measured elsewhere in the world. A review of the radioactivity levels in cigarettes consumed worldwide (Watson, 1983; Khater, 2004; Peres and Hiromoto, 2002) showed that $^{210}$Po concentration ranged from 3.3 to 31.1 mBq g$^{-1}$ of dry sample. For comparison two countries are mentioned here, Brazil that produced tobacco with relatively high concentrations in these two radionuclides and Syria that produced tobacco with relatively low concentrations. The concentrations of $^{210}$Pb in Brazilian cigarette tobacco (Peres and Hiromoto, 2002) ranged from 11.9 to 30.2 mBq g$^{-1}$ of dry tobacco while $^{210}$Po concentrations ranged from 10.9 to 27.4 mBq g$^{-1}$. For Syrian tobacco (Batarsekh and Teherani, 1987), $^{210}$Po concentrations are reported from 0.74 to 2.96 mBq g$^{-1}$. The concentrations in Greek tobacco lie between the concentrations determined in these two countries.

The ratio of $^{210}$Po/$^{210}$Pb in the examined samples is close to 1, except the samples from the region of Messinia. When the time interval between the harvesting of tobacco leaves and cigarettes production is more than 2 y, the $^{210}$Po and $^{210}$Pb in the smoked tobacco are practically in radioactive equilibrium and their activities are equal. In the case of shorter tobacco storage their activities are not generally equal. Then $^{210}$Po/$^{210}$Pb activity ratio depends on their ratio at the time of harvest and the time interval of storage.

Table 1
Composite tobacco samples activity concentrations ± 1σ of $^{210}$Po and $^{210}$Pb (dry weight) (mBq g$^{-1}$ and mBq cigarette$^{-1}$)

<table>
<thead>
<tr>
<th>Tobacco samples</th>
<th>Prefecture</th>
<th>$^{210}$Po (mBq g$^{-1}$)</th>
<th>$^{210}$Pb (mBq g$^{-1}$)</th>
<th>$^{210}$Po (mBq cig.$^{-1}$)$^a$</th>
<th>$^{210}$Pb (mBq cig.$^{-1}$)$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS1 Seres$^b$</td>
<td>10.8 ± 1.2</td>
<td>12.3 ± 1.8</td>
<td>8.6</td>
<td>9.8</td>
<td></td>
</tr>
<tr>
<td>TS2 Imathia$^b$</td>
<td>16.7 ± 0.1</td>
<td>12.6 ± 1.3</td>
<td>13.4</td>
<td>10.1</td>
<td></td>
</tr>
<tr>
<td>TS3 Messinia$^c$</td>
<td>3.6 ± 0.2</td>
<td>7.3 ± 0.8</td>
<td>2.9</td>
<td>5.8</td>
<td></td>
</tr>
<tr>
<td>TS4 Thessaloniki$^b$</td>
<td>14.6 ± 0.4</td>
<td>13.5 ± 3.8</td>
<td>11.7</td>
<td>10.8</td>
<td></td>
</tr>
<tr>
<td>TS5 Xanthi$^d$</td>
<td>17.0 ± 1.1</td>
<td>13.6 ± 2.4</td>
<td>13.4</td>
<td>10.9</td>
<td></td>
</tr>
<tr>
<td>TS6 Kilkis$^b$</td>
<td>15.2 ± 1.3</td>
<td>18 ± 3.4</td>
<td>12.2</td>
<td>14.4</td>
<td></td>
</tr>
<tr>
<td>TS7 Kavala$^b$</td>
<td>14.0 ± 1.4</td>
<td>16.7 ± 2.7</td>
<td>11.2</td>
<td>13.4</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>13.1</td>
<td>13.4</td>
<td>10.5</td>
<td>10.7</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Assuming 0.8 g of tobacco per cigarette.
$^b$ Region of Macedonia.
$^c$ Region of Peloponissos.
$^d$ Region of Thrace.
Assuming that 75% of the $^{210}$Po contained in cigarettes was found in the cigarette smoke, 50% of the total smoke is inhaled into a smoker’s lungs and applying the dose coefficients for adults of $5.6 \times 10^{-6}$ Sv Bq$^{-1}$ for $^{210}$Pb and $4.3 \times 10^{-6}$ Sv Bq$^{-1}$ for $^{210}$Po (ICRP 72, 1996) the average committed effective dose for a smoker of 20 cigarettes per day, that have been produced from Greek tobacco is estimated to be 287 $\mu$Sv (Table 2). The annual committed effective dose through inhalation of non-smokers living in the mid-latitudes of the northern hemisphere is estimated to be 27 $\mu$Sv (UNSCEAR, 2000). The inhalation dose for smokers (20 cigarettes per day) is on average more than 12 times higher than for non-smokers.

The tobacco samples analysed in the present study cover most of the regions where tobacco is cultivated in Greece. These are mainly the north of the country (Macedonia and Thrace) and smaller regions in the south (Messinia and Aitolia) (Fig. 2). It is found that concentrations in Messinia are the lowest observed. Natural radioactivity in surface soils in Greece shows a similar variation with enhanced levels of natural radionuclides like $^{226}$Ra in the North compared to central and southern regions. The measured specific activity concentration of $^{226}$Ra for surface soils in the respective geographical areas is shown in Fig. 3 (Anagnostakis et al., 1996; Promponas, 1992). It is clear that the data available for comparison are not statistically significant. However, the results indicate that $^{210}$Po and $^{210}$Pb concentrations in tobacco grown in the same geographical area show less variation compared to the observed levels of their parent radionuclides in surface soil. Another observation is that although the $^{226}$Ra concentration values in surface soils of the areas where the tobacco samples TS3, TS4, TS6 (Fig. 3) were obtained are similar, the concentration values of $^{210}$Po and $^{210}$Pb in tobacco sample TS3 (Messinia) are

<table>
<thead>
<tr>
<th>Tobacco samples</th>
<th>Prefecture</th>
<th>Inhaled via smoking per day (mBq d$^{-1}$)</th>
<th>Annual committed effective dose ($\mu$Sv y$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{210}$Po</td>
<td>$^{210}$Pb</td>
</tr>
<tr>
<td>TS1</td>
<td>Seres$^d$</td>
<td>64</td>
<td>74</td>
</tr>
<tr>
<td>TS2</td>
<td>Imathia$^d$</td>
<td>100</td>
<td>76</td>
</tr>
<tr>
<td>TS3</td>
<td>Messinia$^e$</td>
<td>22</td>
<td>44</td>
</tr>
<tr>
<td>TS4</td>
<td>Thessaloniki$^d$</td>
<td>89</td>
<td>77</td>
</tr>
<tr>
<td>TS5</td>
<td>Xanthi$^f$</td>
<td>100</td>
<td>82</td>
</tr>
<tr>
<td>TS6</td>
<td>Kilkis$^d$</td>
<td>92</td>
<td>108</td>
</tr>
<tr>
<td>TS7</td>
<td>Kavala$^d$</td>
<td>84</td>
<td>100</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td>79</td>
<td>80</td>
</tr>
</tbody>
</table>

$a$ 20 Cigarettes/day and 75% of the $^{210}$Po and $^{210}$Pb contained in cigarettes are in the cigarette smoke and 50% of $^{210}$Po and $^{210}$Pb in smoke is inhaled.

$b$ 4.3 $\mu$Sv Bq$^{-1}$, $^{210}$Po dose conversion factor.

$c$ 5.6 $\mu$Sv Bq$^{-1}$, $^{210}$Pb dose conversion factor.

$d$ Region of Macedonia.

$e$ Region of Peloponissos.

$f$ Region of Thrace.
clearly lower than that of TS4 and TS6. Atmospheric conditions may also play a role in the difference observed between Messinia and the North Regions as the former receives more regularly maritime air masses from the central Mediterranean, which may be depleted in radon and radon progeny (Fig. 2). The geographical distribution of tobacco cultivation does not exhibit the level of variation that would allow further statistical study.

It has been suggested that other sources for \(^{210}\text{Po}\) and \(^{210}\text{Pb}\) can include road traffic and industry, especially coal-fired power plants (Watson, 1983). Other studies in coastal areas (Carvalho, 1995) explained most of the variation in airborne \(^{210}\text{Po}\) and \(^{210}\text{Pb}\) concentrations by large or synoptic scale air mass transport of continental origin. The presence of \(^{210}\text{Po}\) and \(^{210}\text{Pb}\) in the coarse and fine aerosol fractions was studied extensively in the industrial areas of the U.S. (Pittsburgh) and were found to have similar apparent residence times, indicating that little excess \(^{210}\text{Po}\) could have come from soils or from coal-fired power plants (Gaffney et al., 2004). Considering that the samples examined here are collected from rural areas, where such sources could have an impact only via long or regional range transport of aerosol species contaminated by
$^{210}$Pb, their contribution can be regarded as not significant. The sources of the tobacco plant $^{210}$Po and $^{210}$Pb activities measured can be controlled by their sources in local soils, radon concentration in air and possibly level of fertilization.

The use of calcium phosphate fertilizers in tobacco-growing soils is also regarded as a source of $^{210}$Pb and $^{210}$Po. The raw material for the phosphate fertilizer production is the phosphoric acid. During the wet process manufacture of phosphoric acid, concentrated phosphate rock is reacted with sulphuric acid, which results in the production of by-product gypsum, the so-called “phosphogypsum” (Burnett et al., 1996). It is well known that more than 80% of Ra, Po and Pb originally present in the phosphate rock remains in phosphogypsum. About 80% of U and Th is present in the phosphoric acid (Perianez and Garcia-Leon, 1993; Bolivar et al., 2000). The most common fertilizers used in Greece are Nitrogen–Phosphorus–Potassium (NPK) fertilizers. Typical values measured for $^{226}$Ra in Greek fertilizers are between 7 and 35 Bq kg$^{-1}$. These levels suggest that phosphate fertilizers are not a major source for $^{210}$Po in Greek tobacco.

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