Radiation dose from cigarette tobacco

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Abstract. The radioactivity in tobacco leaves collected from 15 different regions of Greece before cigarette production was studied in order to estimate the effective dose from cigarette tobacco due to the naturally occurring primordial radionuclides, such as $^{226}$Ra and $^{210}$Pb of the uranium series and $^{228}$Ra of the thorium series and or man-made produced radionuclides, such as $^{137}$Cs of Chernobyl origin. Gamma-ray spectrometry was applied using Ge planar and coaxial type detectors of high resolution and high efficiency. It was concluded that the annual effective dose due to inhalation for adults (smokers) for $^{226}$Ra varied from 42.5 to 178.6 μSv y$^{-1}$ (average 79.7 μSv y$^{-1}$), while for $^{228}$Ra from 19.3 to 116.0 μSv y$^{-1}$ (average 67.1 μSv y$^{-1}$) and for $^{210}$Pb from 47.0 to 134.9 μSv y$^{-1}$ (average 104.7 μSv y$^{-1}$), that is the same order of magnitude for each radionuclide. The sum of the effective dose of the three natural radionuclides varied from 151.9 to 401.3 μSv y$^{-1}$ (average 251.5 μSv y$^{-1}$). The annual effective dose from $^{137}$Cs of Chernobyl origin was three orders of magnitude lower as it varied from 70.4 to 410.4 nSv y$^{-1}$ (average 199.3 nSv y$^{-1}$).

KEYWORDS: Radiation dose, cigarette tobacco, tobacco leaves, $^{226}$Ra, $^{228}$Ra, $^{210}$Pb, $^{137}$Cs.

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1. Introduction

Radioactive nuclides of the uranium series, such as $^{210}\text{Pb}$ and $^{210}\text{Po}$, have long been associated with tobacco plants [1-2]. The issue of radioactivity in tobacco smoke has received much attention in the scientific press and, increasingly, in the medical press [3]. Beside this, Martell and Sweder (1982) [4] reported that radon decay products naturally found in the environment are altered when they pass through burning cigarettes into mainstream smoke. Martell (1975) [5] stated that the cumulative α-, β- and γ-radiation dose, particularly that from α-radiation, from inhaled radionuclides deposited in small volumes of bronchial tissue may be an important factor in the initiation of bronchial (lung) cancer in smokers. The smokers are subjected to α-radiation in the bronchial epithelium from three sources: 1) from indoor radon and thoron decay products inhaled between cigarettes, 2) from $^{214}\text{Po}$, $^{212}\text{Po}$ and $^{212}\text{Bi}$ in large mainstream smoke particles, and 3) from $^{210}\text{Po}$ which grows into $^{210}\text{Pb}$-enriched particles which persist at bronchial bifurcations [6].

Radford Jr. and Hunt (1964) [7] at Harvard School of Public Health (Boston, MA) reported that for an individual smoking two packages of cigarettes a day, the radiation dose to bronchial epithelium from $^{210}\text{Po}$ inhaled in cigarette smoke probably is at least seven times that from background sources, and in localized areas may be up to 10 Sv (1000 rem) or more in 25 years. Beside, Winters and DiFranza (1982) [8] at the University of Massachusetts (Boston, MA) much later reported that in a person smoking 1 ½ packs of cigarettes (i.e. 30 cigarettes) per day, the radiation dose to the bronchial epithelium in areas of bifurcation is 80 mSv (8000 mrem) per year – the equivalent of the dose to the skin from 300 X-ray films of the chest per year. This figure was comparable to total-body exposure to natural background radiation containing 0.8 mSv (80 mrem) per year in someone living in the Boston area.

Takizawa et al., (1994) [9] reported that the range of $^{210}\text{Po}$ contained in the tobacco grinds in Japan varied from 13.0 to 20.1 Bq kg$^{-1}$ (mean 15.4 Bq kg$^{-1}$), about 50 % of $^{210}\text{Po}$ present in tobaccos was transferred into the smoke and the other 50 % remained in the ash and butt. One pack-a-day smoker inhaled 24 mBq of $^{210}\text{Po}$ per day through smoking and the annual inhalation was 8.8 Bq. Peres and Hiromoto (2002) [10] reported that $^{210}\text{Po}$ in (dry) tobacco in Brazil ranged from 10.9 to 27.4 Bq kg$^{-1}$ and $^{210}\text{Pb}$ from 11.9 to 30.2 Bq kg$^{-1}$. The collective committed effective dose resulting from the use of cigarettes produced in Brazil per year was estimated to be 1.5x10$^7$ man-Sv, considering an annual production of 5x10$^8$ kg of cigarettes in Brazil and the committed effective dose of 0.16 mSv per year of cigarette smoking.

Colangelo et al., (1992) [11] reported $^{210}\text{Po}$ in tobacco in Argentina ranged from 10 to 80 Bq kg$^{-1}$ and the lung dose due to the use of tobacco varied from 75 to 600 µSv y$^{-1}$. Khater (2004) [12] that the range of $^{210}\text{Po}$ in cigarette tobacco in Egypt ranged from 9.7 to 22.5 mBq/cigarette (average 16.6 mBq/cigarette). The average percentages of $^{210}\text{Po}$ content in fresh tobacco plus wrapping paper that were recovered by post-smoking filters, ash and smoke were 4.6, 20.7 and 74.7, respectively. Cigarette smokers who are smoking one pack (20 cigarettes) per day, are inhaling on average 123 mBq/day of $^{210}\text{Po}$ and $^{210}\text{Pb}$ each. The mean values of the annual effective dose for smokers (one pack per day) were estimated to be 193 and 251 µSv y$^{-1}$ from $^{210}\text{Po}$ and $^{210}\text{Pb}$, respectively.

Approximately 10 litre/day is inhaled through 40 cigarettes – this is about 1/2000 of the amount of air usually breathed per day (20 m$^3$/day [13]) [4]. Published radiochemical data for inhaled mainstream smoke showed an average $^{210}\text{Po}$ content of about 1.33 mBq (0.036 pCi) per cigarette or 96.2 Bq per kg smoke (2.6 pCi $^{210}\text{Po}$ per g smoke) [14] with a $^{210}\text{Pb}$:$^{210}\text{Po}$ ratio of 0.66±0.23 [15].

This paper reports data on the radioactivity of tobacco leaves after the collection from tobacco fields and before cigarette production in order to estimate the effective dose from cigarette tobacco due to the naturally occurring radioactive nuclides and the Cesium-137 of Chernobyl origin.
2. Materials and Methods

Seventeen different samples of tobacco leaves produced in the year 1990 at different locations in Greece (Fig. 1) were examined for radioactivity using γ-ray spectrometry. The spectrometric system consisted of two different high-purity Germanium low-background detectors. One planar Ge detector, of active area 2000 mm², thickness 20 mm, Be window 0.5 mm and energy resolution (FWHM) 400 eV for 5.9 keV γ-rays (\(^{55}\text{Fe}\)) and 700 eV for 122 keV γ-rays (\(^{57}\text{Co}\)), was appropriate for γ-rays ranging from 5 to 186 keV for determination of \(^{210}\text{Pb}\) (47 keV) and \(^{226}\text{Ra}\) (186 keV). A second coaxial Ge detector (p-type), crystal size 155 cc, resolution (FWHM) 1.9 keV at 1.33 MeV (\(^{60}\text{Co}\)) and 900 eV at 122 keV (\(^{57}\text{Co}\)), peak-to-Compton ratio 55:1 and efficiency 42%, was appropriate for γ-rays ranging from 240 to 2614 keV for determination of \(^{226}\text{Ra}\) via its decay products \(^{214}\text{Pb}\) and \(^{214}\text{Bi}\) of uranium series, \(^{226}\text{Ra}\) of thorium series and \(^{137}\text{Cs}\) (662 keV) of Chernobyl origin.

The samples were dried before radioactivity measurement for 3-4 days at a temperature of 30°C to avoid any moisture adsorption. After that, the samples were cut into very small pieces using a blender and were mixed with active charcoal and then sealed appropriately for about 1 month to reduce radon leaching and to attain radioactive equilibrium between radon and thoron decay products as eight-half-lives of \(^{222}\text{Rn}\) \((T_{1/2} = 3.82 \text{ d})\), the decay product of \(^{222}\text{Ra}\) and precursor of \(^{214}\text{Pb}\) and \(^{214}\text{Bi}\), correspond to 1 month. About eight half-lives of \(^{224}\text{Ra}\) \((T_{1/2} = 3.66 \text{ d})\), the precursor of \(^{228}\text{Ra}\), are also 1 month.

The samples were measured in two geometries, that is, in a standard geometry 40 g plastic can of 6 cm diameter and a Marinelli beaker of 1 l (volume). The overall efficiency of the counting system was known to an accuracy of better than 5% for the plastic can geometry and about 12% for the Marinelli beaker. The collecting time was 120,000 s.

Gamma-ray spectra obtained with both HPGe detectors are presented in Fig. 2a for the planar detector and Fig. 2b for the coaxial detector.

3. Results and Discussion

3.1. Radionuclide concentrations

Table 1 presents the concentrations in Bq kg\(^{-1}\) of \(^{226}\text{Ra}\), \(^{210}\text{Pb}\), \(^{228}\text{Ra}\) and \(^{137}\text{Cs}\) in Greek tobacco leaves produced in 1990, i.e. 4 years after the Chernobyl reactor accident (26 April 1986).

In the tobacco leaves, \(^{226}\text{Ra}\) activity concentrations ranged from 1.80 to 7.57 Bq kg\(^{-1}\) (average 3.38 Bq kg\(^{-1}\)), while \(^{228}\text{Ra}\) activity concentrations ranged from 1.10 to 6.62 Bq kg\(^{-1}\) (average 3.83 Bq kg\(^{-1}\)) and \(^{210}\text{Pb}\) activity concentrations ranged from 6.34 to 18.2 Bq kg\(^{-1}\) (average 14.12 Bq kg\(^{-1}\)). Cesium-137 activity concentrations in the tobacco leaves ranged from 1.20 to 7.00 Bq kg\(^{-1}\) (average 3.40 Bq kg\(^{-1}\)).

3.2. Effective dose estimate

Assuming that 0.82 g of tobacco per cigarette in Greek cigarettes and a smoker is smoking 30 cigarettes (1 ½ packs) per day or 24.6 g of tobacco per day, then the annual consumption of tobacco by cigarettes, \(M_T\) is estimated to be 9.0 kg per year. Taking into consideration the data of Table 1 for the radionuclide concentrations, \(C_i\) (Bq kg\(^{-1}\)) in tobacco dry leaves, the fraction of the radionuclide activity concentration that is recovered from cigarette tobacco to cigarette smoke which is 0.75 (75%) [12] and the most recent dose conversion coefficients of the radionuclides, \(F\) (Sv Bq\(^{-1}\)) for the case of inhalation.
for adults (smokers) [16-19] as presented in Table 2, then the data of Table 3 are derived for the annual effective dose, \( H_E \) (Sv y\(^{-1}\)) due to inhalation for adults (smokers), according to the equation

\[
H_E = 0.75 \times M_T \times C_i \times F
\]

\( M_T \) = Annual mass of tobacco consumed (kg y\(^{-1}\))

\( C_i \) = Radionuclide concentration (Bq kg\(^{-1}\))

\( F \) = Dose conversion factor (Sv Bq\(^{-1}\))

From the data of Table 3, it is resulted that the annual effective dose to \( ^{226}\)Ra varied from 42.7 to 178.6 \( \mu \)Sv y\(^{-1}\) (average 79.7 \( \mu \)Sv y\(^{-1}\)), while for \( ^{228}\)Ra from 19.3 to 116.0 \( \mu \)Sv y\(^{-1}\) (average 67.1 \( \mu \)Sv y\(^{-1}\)) and for \( ^{210}\)Pb from 47.0 to 134.9 \( \mu \)Sv y\(^{-1}\) (average 104.7 \( \mu \)Sv y\(^{-1}\)), that is of the same order of magnitude for each natural radionuclide. The dose from \( ^{210}\)Po (\( \alpha \)-emitter), a decay product of \( ^{210}\)Pb, should be of the same order of magnitude of the dose due to \( ^{210}\)Pb. Holtzman and Ilciewicz (1966) [15] reported that in smokers the \( ^{210}\)Po is nearly in radioactive equilibrium with its parent \( ^{210}\)Po: \( ^{210}\)Pb = 0.87±0.10. The sum of the effective doses of the three natural radionuclides \( ^{226}\)Ra, \( ^{228}\)Ra and \( ^{210}\)Pb varied from 151.9 to 401.3 \( \mu \)Sv y\(^{-1}\) (average 251.5 \( \mu \)Sv y\(^{-1}\)). Assuming an additional average annual effective dose due to \( ^{210}\)Po that of 104.7 \( \mu \)Sv y\(^{-1}\) of \( ^{210}\)Pb, then the sum of the effective doses of the four natural radionuclides \( ^{226}\)Ra, \( ^{228}\)Ra, \( ^{210}\)Pb and \( ^{210}\)Po should be varied from 256.6 to 506.0 \( \mu \)Sv y\(^{-1}\). This dose must be compared with the average world-wide exposure to natural radiation sources 2.4 mSv y\(^{-1}\) and the part due to inhalation which is 1.26 mSv [20] that is almost 50 % or a little higher.

The annual effective dose due to \( ^{137}\)Cs of Chernobyl origin was three orders of magnitude lower. It varied from 70.4 to 410.4 nSv y\(^{-1}\) (average 199.3 nSv y\(^{-1}\)) and so very little is contributing to the total dose due to inhalation to smokers. However, in the event of inhomogeneous irradiation, as is a well-defined case in smoking and deposition of radionuclides of the U- and Th-series in the lungs, irradiation effects must be compared in the absorbed rather than effective dose to the lungs, indicating that as homogeneously distributed \( ^{137}\)Cs irradiates the whole body rather uniformly.

From the literature, it is seen that the dose estimated for smokers was considered only for \( ^{210}\)Po (\( \alpha \)-emitter) and its precursor \( ^{210}\)Pb (\( \beta \)-emitter). No consideration was taken on the radioisotopes of radium, i.e. \( ^{226}\)Ra of the uranium series and \( ^{228}\)Ra of the thorium series, although the dose for inhalation from each of them was shown to be of the same order of magnitude with that of \( ^{210}\)Pb and of course, of \( ^{210}\)Po [16-17].

For the dose due to \( ^{137}\)Cs of Chernobyl origin, Fletcher (1994) [21] was referred and concluded that the annual effective dose for inhalation of \( ^{137}\)Cs was estimated to be 7.44 nSv y\(^{-1}\) with the \( ^{137}\)Cs content in tobacco leaves averaging to 40 Bq kg\(^{-1}\), in Libya and annual intake 1 Bq y\(^{-1}\) for \( ^{137}\)Cs.

### 4. Conclusion

The radioactivity in tobacco leaves collected from 15 different regions of Greece before cigarette production has been studied in order to estimate the effective dose from cigarette tobacco due to the naturally occurring radionuclides and the Cesium-137 of Chernobyl origin. It was concluded that the annual effective dose due to inhalation for adults (smokers) for \( ^{226}\)Ra varied from 42.5 to 178.6 \( \mu \)Sv y\(^{-1}\) (average 79.7 \( \mu \)Sv y\(^{-1}\)), while for \( ^{228}\)Ra from 19.3 to 116.0 \( \mu \)Sv y\(^{-1}\) (average 67.1 \( \mu \)Sv y\(^{-1}\)) and for \( ^{210}\)Pb from 47.0 to 134.9 \( \mu \)Sv y\(^{-1}\) (average 104.7 \( \mu \)Sv y\(^{-1}\)), that is of the same order of magnitude for each radionuclide. The sum of the annual effective doses of the three natural radionuclides varied from 151.9 to 401.3 \( \mu \)Sv y\(^{-1}\) (average 251.5 \( \mu \)Sv y\(^{-1}\)) The annual effective dose from \( ^{137}\)Cs of Chernobyl origin was three orders of magnitude lower as it varied from 70.4 to 410.4 nSv y\(^{-1}\) (average 199.3 nSv y\(^{-1}\)).
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REFERENCES


[20] UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC

Figure captions

**Figure 1:** A map of Greece showing the sampling locations of tobacco leaves.

**Figure 2a:** Gamma-ray spectrum of tobacco leaves. Gamma photon energies ranged up to 200 keV obtained with a planar Ge detector.

**Figure 2b:** Gamma-ray spectrum of tobacco leaves. Gamma photon energies ranged up to 3000 keV obtained with a coaxial Ge detector.
Table 1: Activity concentrations in tobacco leaves

<table>
<thead>
<tr>
<th>No.</th>
<th>Lab. No.</th>
<th>$^{226}$Ra Bq kg$^{-1}$</th>
<th>$^{210}$Pb Bq kg$^{-1}$</th>
<th>$^{228}$Ra Bq kg$^{-1}$</th>
<th>$^{137}$Cs Bq kg$^{-1}$</th>
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<td>$6.21 \pm 1.01$</td>
<td>$16.40 \pm 0.87$</td>
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<td>$2.13 \pm 1.22$</td>
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<td>$5.62 \pm 0.89$</td>
<td>$4.02 \pm 1.00$</td>
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<td>$4.79 \pm 0.89$</td>
<td>$6.26 \pm 1.00$</td>
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<td>TAB-5</td>
<td>$3.66 \pm 1.12$</td>
<td>$18.20 \pm 4.10$</td>
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<td>$4.81 \pm 0.73$</td>
<td>$4.70 \pm 0.80$</td>
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<td>$1.70 \pm 1.00$</td>
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<td>$1.60 \pm 1.00$</td>
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Table 2: Dose conversion factors for inhalation for adults

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<tr>
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<td>$^{137}$Cs</td>
<td>$8.70 \times 10^{-9}$</td>
<td>[18-19]</td>
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Table 3: Annual effective dose of smokers in smoking 30 cigarettes per day

<table>
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<td>122.31</td>
<td>61.15</td>
<td>249.17</td>
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Figure 1: A map of Greece showing the sampling location of tobacco leaves
Figure 2a: Gamma-ray spectrum of tobacco leaves. Gamma photon energies ranged up to 200 keV obtained with a planar Ge detector.

Figure 2b: Gamma-ray spectrum of tobacco leaves. Gamma photon energies ranged up to 3000 keV obtained with a coaxial Ge detector.