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Evaluating the Polonium Activity Concentration in a well-known Cuban cigar

Daniel Marcos Bonotto*, Fábio de Oliveira Thomazini**
**(Geology Department, IGCE-Geosciences and Exact Sciences Institute, UNESP-São Paulo State University, Rio Claro, São Paulo State, Brazil)*
*** (Geology Department, IGCE-Geosciences and Exact Sciences Institute, UNESP-São Paulo State University, Rio Claro, São Paulo State, Brazil)*

ABSTRACT
^{210}Po is a natural radionuclide produced continuously in rocks and soils through the decay of ^{226}Ra in the ^{238}U radioactive decay series. Although the atmosphere contains ^{210}Po arising from ^{226}Ra that occurs naturally in the earth's crust, the majority of the ^{210}Po in tobacco plants generally comes from high-phosphate fertilizers applied to the tobacco crop. This radionuclide persists even after the plant leaves are dried and processed, constituting a major health threat in cigarettes and cigars due to its high volatility at the burning temperature of 600-800°C and subsequent inhalation in the vapour phase or adsorption on smoke particles, leading to increasing concentration in the human body, particularly in the lungs. This paper describes the characterization of the ^{210}Po activity concentration in a Cuban cigar of a very traditional brand and comparison with other reported data in the literature for various countries, discussing the results obtained for different types of smokers, and taking into account calculations realized for the CED-Committed Effective Dose.

***Keywords* -** natural radioactivity, polonium, Cuban cigar, alpha spectrometry, Committed Effective Dose

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I. Introduction

Polonium is a rare and highly radioactive metalloid. It belongs to the *chalcogens* family and is represented by p-block elements in the Periodic Table. Marie Curie and collaborators discovered it in 1898 in the bismuth fraction obtained by dissolution and analytical treatment of pitchblende ore. Polonium occurs sparsely as a natural product of the radioactive decay series. Its abundance in the Earth's crust is about 2×10^{-15} % [1].

Polonium isotopes are generated in the actinium, uranium, and thorium natural decay series, which are alpha emitters (Table 1). Table 1 also includes some data for ^{209}Po, which is an artificial radionuclide used as a spike in the experiments described in this paper, permitting the measurement of the ^{210}Po activity concentration after its spontaneous deposition in selected metallic surfaces. Among the polonium isotopes reported in Table 1, ^{210}Po from the uranium series exhibits a longer half-life (138 days). It is a natural radionuclide produced continuously in rocks and soils through the decay of ^{226}Ra in the ^{238}U radioactive decay series (4n+2), since uranium is among the main elements contributing to natural terrestrial radioactivity (Fig. 1). ^{210}Po is generated after the ^{222}Rn-production in the ^{238}U decay series. Because some of the ^{222}Rn escapes from the rocks, minerals, and soils to the surrounding fluid phase, such as air, ^{222}Rn emanating from land surfaces is responsible for ^{210}Po present in the atmosphere.

Table 1. Summary of polonium isotopes data, according to [2].

Series	Nuclide	Half-life[1]	Decay type	Energy (MeV)	TP[2] (%)
Thorium	^{216}Po	0.15 s	α	5.984	0.002
				6.777	100
	^{212}Po	0.3 µs	α	8.785	100
Uranium	^{218}Po	3.05 m	α	6.000	100
			β^-	0.277	0.02
	^{214}Po	0.16ms	α	7.688	100
	^{210}Po	138 d	α	5.305	100
Actinium	^{215}Po	1.78ms	α	7.384	100
Neptunium	^{213}Po	4.2 µs	α	8.336	100
	^{209}Po[3]	102 y	α	4.979	99.52

[1] y = years, m = minutes, s = seconds; [2] TP = transition probability; [3] ^{209}Po is an artificial radionuclide used as a spike in the experiments described in this paper.

Polonium exists as a solid of very complex structure, which has the following valences: -2 (in polonium hydride, H_2Po), +2 (in polonium dichloride, $PoCl_2$), +4 (in polonium dioxide, PoO_2) and +6 (in polonium trioxide, PoO_3) [4]. Some other general properties of polonium are: Atomic number- 84; Atomic weight- 207.21; Atomic volume (mL)- 18.2; Configuration- $6s^2 6p^4$; Ionization energy (kcal)- 194; Atomic radius (Å)- 1.64; Melting point (K)- 527; Boiling point (K)- 1235; ΔH_{sub} (kcal)- 34.5; Ionic radius (Å)- 1.32 [4].

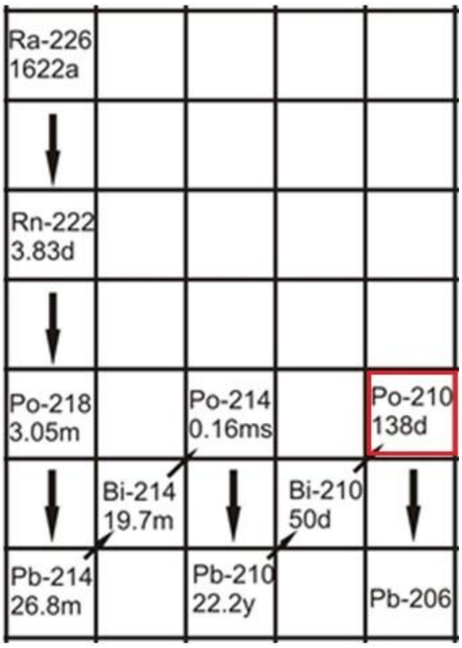

Fig. 1. The natural uranium (4n+2) decay series from ^{226}Ra. Modified from [3].

Abe and Abe [5], when studying the polonium volatility in the air dust, found that 40% volatilizes between 200-700°C, whilst 100% volatilizes at 900°C. These authors concluded that polonium was also present as PoO_2 in other unidentified compounds. Mabuchi [6] studied the volatility of a few polonium organic compounds and found that all 11 chelates examined were volatile below 200°C at atmospheric pressure. Among the most common polonium compounds, the dioxide sublimates at 885°C, whereas the tetrachloride boils at 390°C. In fact, significant losses have been recorded when heating this radioactive element at temperatures above 150°C.

^{210}Po in water is a well-documented radionuclide for health risk, as the World Health Organization (WHO) proposed a guidance level of 0.1 Bq/L for the activity concentration in drinking water to not exceed the reference dose level of the committed effective dose equal to 0.1 mSv from 1 year's consumption [7].

Tobacco is a plant of the genus *Nicotiana* whose leaves are mostly used for smoking [8]. It can be smoked in different forms and shapes (e.g., cigarettes, cigars, pipes, and rolling tobacco), with cigarettes accounting for more than 90% of the market share among all tobacco products [9]. In 1964, ^{210}Po was reported to be a tobacco smoke constituent due to its emission of alpha radiation, which is carcinogenic [10-12]. An excellent review of such a situation was done by [13], whose major highlights are now summarized. Inhalation experiments have shown that ^{210}Po is the cause of lung cancers in animals [14, 15]. ^{210}Po is thought to deposit in the bronchial segmental bifurcations, resulting in substantial doses of high-energy alpha radiation in the pulmonary sites where bronchogenic carcinomas frequently arise [16, 17]. Alpha radiation has also been shown to induce signaling pathways in cells that are not directly exposed (the so-called bystander effect) [18]. Mean tissue concentrations of ^{210}Po in cigarette smokers have been observed to be more than double those of nonsmokers [19]. It is estimated that smokers of 1.5 packs of cigarettes a day are exposed to as much radiation as they would receive from 300 chest X-rays a year [11]. ^{210}Po has been estimated to be responsible for 1% of all US lung cancers [20]. Because the American Cancer Society [21] estimates that each year, 162,460 deaths in the United States and 1.3 million deaths worldwide [22] are attributable to lung cancer, ^{210}Po may be responsible for more than 1,600 deaths in the United States and 11,700 deaths in the world every year [13].

Therefore, this short review highlighted the relevance of monitoring the polonium presence in tobacco and by-products consisting of cigarettes, cigars, tobacco pipes, snuff, etc., due to the concerns associated with human health. This paper describes the results of the analysis of ^{210}Po occurring in a selected sample of well-known cigars, revealing a novel, useful dataset for scientists interested in this research topic elsewhere.

II. Selected sample for analysis

The experiment described in this paper was realized by selecting one cigar sample from the Cuban brand Cohiba (Fig. 2), much appreciated by smokers [23]. The tobacco cultivation occurs in Pinar del Río Province, western Cuba (Fig. 3), subjected to a tropical climate with an annual mean temperature of 25.1°C, evaporation of 1,860 mm, precipitation of 1,766 mm, and a defined rainy season between May and October [24, 25]. Agricultural production is the main economic activity in the region, which includes forestry, tobacco, rice, etc., with the tobacco production (Fig. 4) constituting the major source for the national industry [25].

Fig. 2. General view of the cigar sample analyzed in this study.

Fig. 3. Location of the Pinar del Río Province in Cuba. Adapted from [25].

In Cuba, more than 40% of agricultural lands are affected by severe erosion problems, with some studies developed to identify the major soil characteristics and properties as reported by [24, 25]. The results of the analysis of 19 soils by [25] indicated the following textural classes: clay, sandy loam, loam, clay loam, sandy clay, and loamy sand The soil textures range were [25]: clay = 6.8-75.7%; silt = 10.5-38.1%; modified silt + fine sand content = 14.8-62.8%; fine sand = 2.1-31.7%; 0.1-0.2 mm = 2.7-30.9%; 0.2-0.5 mm = 2.7-13.3%; >0.5 mm = 0-23.9%; coarse sand = 9.5-55.5%; sand = 13.7-74.1%.

Fig. 4. Example of tobacco cultivation at Pinar del Río Province in Cuba. Modified from [26].

Major soil types in Pinar del Río Province are Yellowish Ferralitic Leached, Alitic Low Activity Yellowish Red Clay, and Arenosol or Acrisol Chromic Ferric, Nitisol Rhodic, and Arenosol, depending on the classification system [27]. Some of these soil types are related to the occurrence in the area of lateritic deposits, associated with nickel and iron-bearing weathering crusts possessing the following average concentration [28]: Ni=0.96-1.26%; Fe=8.6-43.1%; Co=0.018-0.136%; Si=3.6-20.0%; Mg=1.0-16.5%; Al=1.6-7.9%.

Red clayey soils (Ferralsols/Fersialitic) are ideal for tobacco, but also suffer severe erosion, with key soil types often acidic with low nutrient capacity, requiring careful management for crops like tobacco and rice, with microbial studies showing potential for improving phosphorus uptake [27]. Heavy metal levels are generally naturally high, but often bioavailable at low risk, as indicated by [27] on the evaluation of the total concentrations and fractions of heavy metals Cd, Pb, Zn, Fe, Mn, Ni, Cu, Cr, and Co and their relationship with the physicochemical properties of soil.

The agricultural stages (on the farm) for the cigar production are cultivation, tending, harvesting,

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curing, fermentation, sorting/stripping, and aging, while the manufacturing stages (in the factory) involve blending, rolling, quality control, conditioning, and packaging [29].

III. Experimental

In this study, 1.85 g of dry weight of the cigar shown in Fig. 2 was inserted into a Teflon beaker. Then, for its dissolution, 5 mL of concentrated HNO_3 was added, with the beaker being covered with a Teflon lid, and put over a hot plate at 45°C for two days. The dissolution was complete after this period, with a known amount (0.2 mL; activity = 40 dpm/mL) of ^{209}Po spike being also added to assess ^{210}Po recovery. Afterwards, the solution was evaporated to dryness, and the dry residue was dissolved with 8M HCl to a volume of 15 mL. which was complemented with more 5 mL of ferric chloride (concentration = 60 mg/mL). ^{210}Po was co-precipitated with $Fe(OH)_3$ by increasing the pH to 7-8 through the addition of concentrated NH_4OH solution. The precipitate was recovered, dissolved in 8M HCl and Fe^{3+} was extracted into an equal volume of isopropyl ether.

Subsequently, 5 mL of 20% hydroxylamine hydrochloride and 2 mL of 25% sodium citrate solution were added, and the pH was adjusted to 2 with concentrated ammonia solution [30]. Polonium was plated onto a copper disc (2.5-cm diameter) suspended in the solution placed on a hot plate magnetic stirrer, which was heated to 85- 90 °C, and stirred for 75-90 min with a Teflon stirrer. Then, the disc was removed, washed with demineralized water, dried in a heating lamp, and counted by conventional alpha spectroscopy (Fig. 5).

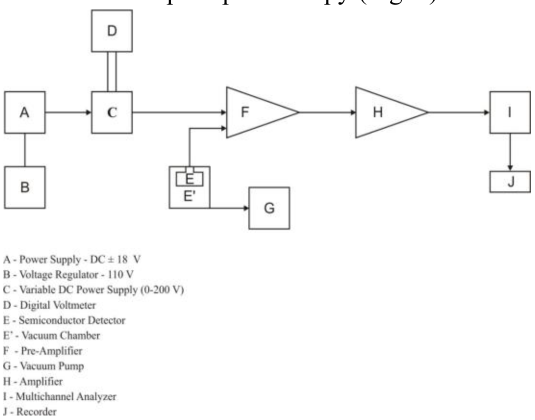

Fig. 5. A typical spectrometric system for detecting alpha particles as used in the experiment described in this study.

The alpha counting was realized with a 450 mm^2 area, 300 μm depletion depth, 26 keV resolution Si(Au) surface barrier detector (Fig. 5). The detector was coupled to EG&G ORTEC multichannel buffer, and the MAESTRO simulator software provided 1024 channels to plot the alpha spectrum containing the ^{209}Po and ^{210}Po peaks. The system was calibrated in the 4.8-5.5 MeV region of energy through a ^{232}U-^{228}Th standard radioactive source prepared at the *Centre de Faibles Radioactivités*, CNRS-CEA, Gif-sur-Yvette, France, and containing 0.91 Bq of ^{238}U. In addition, the same source included ^{234}U (α-particles energy = 4.77 MeV) and 0.17 Bq of ^{232}U, that is an artificial uranium isotope produced in nuclear reactor by neutron activation of ^{232}Th, has a half-life of 72 years, and originates ^{228}Th by the emission of α-particles of 5.3 MeV energy [2]. The ^{228}Th-daughters correspond to those occurring in the ^{232}Th decay series [2].

The channels identified in the alpha spectrum and the corresponding particle energy for the identified radionuclides allowed tracing the energy calibration curve of the alpha spectrometer (Fig. 6), which may be written according to the equation $E = 0.01075\ Ch$, where E is the energy (in MeV), and Ch is the channel number in the multichannel analyzer.

^{210}Po emits α particles of 5.3 MeV energy, practically the same as the alpha emissions from ^{232}U+^{228}Th, whereas ^{209}Po emits α particles of ~4.9 MeV energy, very close to that of ^{234}U. The isotope dilution technique was used to generate the ^{210}Po activity concentration in the sample [31].

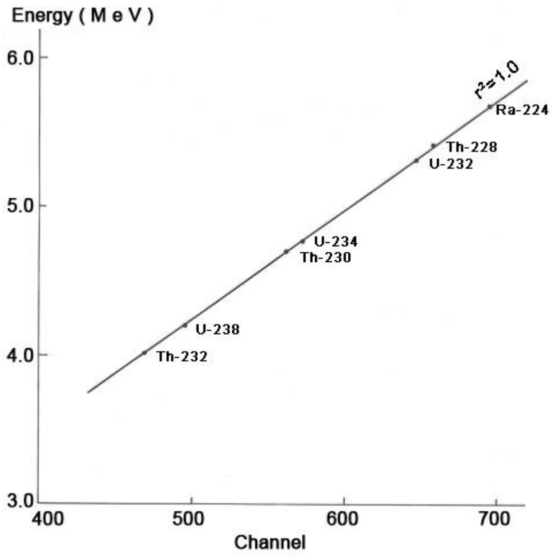

Fig. 6. The calibration curve of the Si(Au) spectrometer for α-particles energy readings as used in the experiment described in this study. It is also illustrated the position of the radionuclide ^{230}Th that is a ^{234}U-descendant in the ^{238}U decay series.

IV. Results and Discussion

Fig. 7 shows the alpha spectrum obtained during data acquisition, which highlights the peaks of both polonium isotopes (^{209}Po and ^{210}Po) and indicates their successful recovery. The results obtained in the alpha counting are summarized in Table 2, indicating a ^{210}Po activity concentration of 0.052 Bq/g (=52 mBq/g). The cigar analyzed is of type Robustos (ring gauge = 50; length = 124 mm; weight = 11.66 g), thus, the total ^{210}Po activity is 606 mBq/cigar.

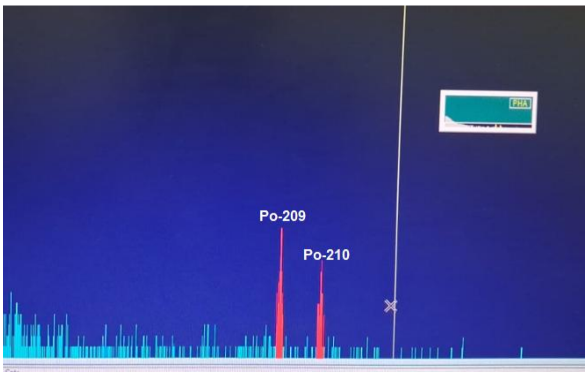

Fig. 7. Alpha spectrum obtained for the cigar sample analyzed in this study. Counting time = 150,000 s (41.7 h).

Table 2. Results obtained in the cigar sample analyzed in this study.

Parameter	Unit	^{209}Po	^{210}Po
Channels	-	597-606	649-658
Counts	-	64	48
Count rate	cps	0.00043	0.00032
Net count rate	cps	0.00039	0.00028
Activity	dpm	-	5.81
Activity concentration	dpm/g	-	3.14
Activity concentration	Bq/g	-	0.052

Table 3. ^{210}Po activity concentration data obtained in cigarette samples from various countries.

Country	Site	^{210}Po (mBq/g)	Ref.
India	Kalpakkan	13.0	[32]
India	Tiruchirappalli	13.2	[33]
Greece	Average	13.1	[34]
Vietnam	Average	26.4	[33]
Slovenia	Average	14.0	[9]
Italy	Average	15.7	[35]
China	Average	22.0	[36]
Hungary	Average	22.0	[37]
Brazil	Average	21.2	[38]
Egypt	Average	21.0	[39]
Malaysia	Average	13.8	[40]
Sudan	Average	36.5	[41]

Table 3 reports a compilation of the ^{210}Po activity concentration data obtained in cigarette samples from various countries, which range from 13.0 to 36.5 mBq/g, and are below the 52 mBq/g obtained in the cigar sample analyzed in this study. Although the atmosphere contains ^{210}Po arising from ^{226}Ra that occurs naturally in the earth's crust, the majority of the ^{210}Po in tobacco plants likely comes from high-phosphate fertilizers applied to the tobacco crop [11]. Such radionuclide persists even after the plant leaves are dried and processed, thus justifying its presence in the Cuban cigar analyzed because very low levels of natural and artificial radioactivity were reported in sediments occurring in that area, for instance, ^{226}Ra activity concentration between <3.5 and 27.1 mBq/g [42].

As an example of ^{226}Ra present in Brazilian fertilizers, Conceição and Bonotto [43] reported a mean value of 256 mBq/g in phosphate rocks of Tapira city, west of Minas Gerais State, which is almost 10 times higher than the maximum value of 27.1 mBq/g reported in Cuban sediments by [42]. Therefore, the industrial by-products of the Itapira phosphatic rocks also exhibit high ^{226}Ra activity concentration, for instance, phosphate concentrated of 140-189 mBq/g, simple superphosphate of 262-304 mBq/g, and phosphogypsum of 269-280 mBq/g [43].

The major health threat associated with polonium in cigarettes and cigars is due to its high volatility at the burning temperature (between 600 and 800°C) and subsequent inhalation in the vapour phase or adsorbed on smoke particles, leading to increasing concentration in the human body, particularly in the lungs [13].

DCF (Dose Conversion Factor) is a mandatory parameter to perform radiation dose calculation from the activity concentration value reported in Table 2. One value sometimes used for such a purpose is 4.3 μSv/Bq as reported by [34, 38]. Some CED (Committed Effective Dose) calculations by researchers consider individuals smoking 20 cigarettes per day for one year. There is no specific, legally established annual dose limit for ^{210}Po inhalation from tobacco smoke because smoking is a voluntary exposure to a known carcinogen, not a regulated occupational or public exposure scenario. Instead, the studies estimate the radiation dose a regular smoker receives, often comparing it to established public dose limits to highlight the risk.

For instance, Savidou et al. [34] estimated that the average CED due to ^{210}Po for a smoker of 20 cigarettes per day that has been produced from Greek tobacco corresponded to 124 μSv/yr. Such a value is about 5 times higher than the annual CED of 24 μSv through ^{210}Po inhalation of non-smokers living in the mid-latitudes of the northern hemisphere [34]. However, it is almost 23 times higher than the annual CED corresponding to 5.4 μSv for ^{210}Po due to air inhalation in Poland [44]. Other annual CED estimates for ^{210}Po, also considering 20 cigarettes smoked per day, are: 110 and 112 μSv/yr in India [32, 33], 223 μSv/yr in Vietnam [33], 61 μSv/yr in Slovenia [9], 55.2 μSv/yr in Italy [35], 123.2 μSv/yr in China [36], 185.6 μSv/yr in Hungary [37], 193 μSv/yr in Egypt [39], 100.2 and 111.9 μSv/yr in Malaysia [40], 251.3 61 μSv/yr in Sudan [41], and 160 μSv/yr in Brazil [38].

Despite the abundant data available in the literature for CED estimates involving ^{210}Po in cigarettes, there are only a few reports for cigars. One exception is the study of [33], in which the authors pointed out values between 50.5 and 103.7 μSv/yr (mean = 69.7 μSv/yr) for five Indian cigars, considering a cigar-smoking rate of 20 g/day. If such a rate is applied to the data reported in Table 2, then, 7300 g would be annually smoked, yielding a total ^{210}Po activity of 379.6 Bq for the Cuban cigar analyzed in this study. The DCF = 4.3 μSv/Bq would imply an annual CED = 1632 μSv, which is 23 times higher than the mean value reported by [33] for the Indian cigars.

Another relevant aspect should be considered involving the cigarettes and cigars smokers. In the case of cigars, the smoking frequency differs greatly, according to the following classification [45]: 1) the "special occasion" cigar smoker (low frequency), corresponding to an approximate cigar intake of 5-20 cigars per year; 2) the "I smoked when I feel like it but don´t need them" cigar smoker (low to mid frequency), corresponding to an approximate cigar intake of 20-50 cigars per year; 3) the "weekend warrior" cigar smoker (mid frequency), corresponding to an approximate cigar intake of 50-200 cigars per year; 4) the "daily" cigar smoker (high frequency), corresponding to an approximate cigar intake of 350-450 cigars per year; 5) the "daily multiple" cigar smoker (high frequency), corresponding to an approximate cigar intake of 720-2000 cigars per year. Well-known examples of this last type of smoker are Sir Winston Churchill, Mark Twain, and Sigmund Freud (typically smoked around 20 cigars a day) [45].

The annual CED estimate of 1632 μSv for the Cuban cigar analyzed in this study, considering a daily intake of 20 g, practically corresponds to smoking 2 of them per day, or 730 cigars per year, characterizing a "daily multiple" cigar smoker (high frequency). For a "special occasion" cigar smoker (low frequency) with an intake of 20 cigars per year, the annual CED would be 52 μSv, which is equivalent to the minimum value reported for Indian cigars [33]. Contrarily, for a smoker like Sigmund Freud with a daily intake of 20 cigars, the annual CED would reach the very high value of 19,032 μSv.

These possible scenarios illustrate the usefulness of the approach described in this study, increasing the database on the ^{210}Po activity concentration and annual CED related to this radionuclide in cigars, contributing to the development of novel insights by researchers interested in this subject.

V. Conclusion

^{210}Po is a radioactive metalloid, which was reported to be a tobacco smoke constituent due to its emission of carcinogenic alpha radiation. Estimates have pointed out that for an individual smoking two packs of cigarettes a day, the radiation dose to bronchial epithelium from ^{210}Po inhaled in cigarette smoke is at least seven times higher than that from background sources, implying that the radiation from this source could be significant in the genesis of bronchial cancer in smokers. Despite the large number of studies conducted worldwide to evaluate the ^{210}Po activity concentration and annual CED (Committed Effective Dose) associated with this radionuclide in cigarettes, only a few data have been published focusing on its presence in cigars. One sample of a Cuban cigar was digested with nitric acid and ^{210}Po was recovered and analyzed by alpha spectrometry, yielding an activity concentration of 52 mBq/g that is higher than values often reported in various countries for cigarettes. Additionally, the estimated annual CED value is 1632 µSv, which is 23 times higher than the mean value reported in the literature for five Indian cigars. The obtained dataset allowed to perform simulations of possible annual CED scenarios for cigar smokers characterized by different frequencies of cigar intake, i.e., low (5 to 20 cigars per year), mid (50 to 200 cigars per year), and high (350 to 450 or 720 to 2000 cigars per year). Such evaluation is useful for actions involving human protection against polonium inhalation in the vapour phase or adsorption on smoke particles in cigarettes and cigars.

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