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NOTE

^{210}Po IN SOIL AND TOBACCO LEAVES IN QUANG XUONG, VIETNAM AND ESTIMATION OF ANNUAL EFFECTIVE DOSE TO SMOKERS

Hao Van Duong[1], Duong Thanh Nguyen[1], Anita Peka[2], Edit Tóth-Bodrogi[2] and Tibor Kovács[2,*]
[1]Hanoi University of Mining and Geology, 18 Vien Street, Bac Tu Liem District, Hanoi 100000, Vietnam
[2]Institute of Radiochemistry and Radioecology, University of Pannonia, Veszprem 8200, Hungary

*Corresponding author: kt@almos.uni-pannon.hu

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^{210}Po is the main radiotoxic chemical in tobacco smoke and one of the primary causes of lung cancer. Investigating ^{210}Po concentration in tobacco is important in estimating the annual effective dose (AED) due to smoking. In this study, the ^{210}Po concentrations in tobacco leaves and soil in Quang Xuong, Vietnam were measured using a high-resolution passivated and implanted planar silicon detector. Based on these data the AEDs to smokers were estimated. The ^{210}Po activity concentration in tobacco varied significantly from 28.7 to 254.0 mBq g^{-1}, whereas its variation in soil was insignificant. The AED due to smoking fresh tobacco leaves in Vietnam (average 565 µSv y^{-1}) was significantly higher than the values reported for other countries (36–361 µSv y^{-1}).

INTRODUCTION

Tobacco leaves are commonly used to manufacture tobacco products (e.g. cigarettes, cigars, bidis) for human consumption in many countries worldwide. Tobacco products are most commonly consumed by smoking and passive smoking[(1)]. Tobacco smoke is predominantly a gas containing over 8000 toxic chemicals that are harmful to human health[(2)]. In addition, over the last century, naturally occurring radionuclides such as ^{234}U, ^{238}U, ^{232}Th and their decay products, e.g. ^{226}Ra, ^{210}Pb and ^{210}Po, have been detected in tobacco leaves (both wet and dry)[(3,4)]. ^{210}Po is counted as one of the causes of lung cancer[(5)] and is one of the most toxic substances on earth (^{210}Po is an alpha-emitting isotope, 5.4 MeV, half-life of 138 d). When a cigarette is burned, ^{210}Po is volatilised and smokers inhale as well as absorb the toxicant ^{210}Po. Therefore, the analysis of the ^{210}Po activity concentration in tobacco leaves and tobacco cigarettes should be evaluated to estimate the annual effective dose (AED) absorbed by smokers.

The ^{210}Po activity concentrations in tobacco leaves as well as tobacco cigarettes and the AED absorbed by smokers have been extensively measured and estimated by many authors in the world[(1,6–22)]. In general, these investigations showed that the activity concentration of ^{210}Po in tobacco leaves and tobacco cigarettes ranged from 12.6 to 47.6 mBq g^{-1} dry wt. or from 8.0 to 26.4 mBq cigarette^{-1}. The AED to smokers in these investigations varied from 36.0 to 361.0 µSv y^{-1}. A previous paper stated that the high concentration of ^{210}Po in tobacco leaves is due to its absorption from soil via the roots of tobacco plants and the atmospheric deposition of ^{210}Po on the surface of tobacco leaves[(8)]. In addition, phosphate fertilisers used for growing tobacco are also a major source of ^{210}Po in tobacco leaves[(23)]

Besides the activity concentration of ^{210}Po, the uptake of ^{210}Po in tobacco leaves is also crucial in terms of assessing its pathways into cigarettes. The transfer of ^{210}Po from soil to plants has been extensively studied. Based on the transfer of ^{210}Po from soil to some plants such as potatoes, vegetables, cereals, grass and alfalfa[(24)] showed that the transfer of ^{210}Po via the root system is rather small. The research also indicated that the main source of ^{210}Po in the parts of plants above ground is atmospheric deposition. Skwarzec *et al.*[(8)] also confirmed that the transfer of ^{210}Po from soil to tobacco leaves in Poland via the root system of tobacco plants is insignificant. The activity concentration of ^{210}Po in the roots was five times less than that in leaves. On the other hand, other authors indicated that the uptake through the root system is not negligible[(25)]. The pathway of polonium uptake is somewhat disputed, but it is extremely important for NORM (Naturally Occurring Radioactive Material) biomonitoring applications, since the tobacco plants have favourable characteristics as biomonitor and bioindicator species[(26,27)] Furthermore, the transfer of ^{210}Po from soil to plants can vary from region to region because they depend on several properties, e.g. the organic matter and pH of soil, plant species and other environmental conditions[28–30].

In Vietnam, tobacco has been consumed for centuries and is now one of the 15 top consumers of tobacco worldwide. In Vietnam, traditionally tobacco leaves are chewed with betel and smoked using water pipes and cigarettes. The most common form of tobacco consumption is smoking that kills ~40 000 people annually[31]. As reported, tobacco smoke contains many toxic components including radiotoxic chemicals. The activity concentration of ^{210}Po plays an important role in estimating the AED to smokers[1]. Therefore, the investigation of radiotoxic chemicals such as ^{210}Po in tobacco is very important in terms of estimating the AED and proposing solutions to reduce the health effects of tobacco consumption in Quang Xuong, Vietnam. The ^{210}Po activity concentrations in Vietnamese tobacco products, including commercial cigarettes and pipe tobacco, and the AED to Vietnamese smokers have been investigated and estimated by previous works[18,19]. However, no studies have been published concerning the activity concentrations of ^{210}Po in soil and tobacco leaves in Vietnam to date. The main objective of this study is to determine the activity concentration of ^{210}Po in tobacco leaves as well as soils. The tobacco leaves were collected from Quang Xuong, a district in Thanh Hoa Province, which is one of the most significant areas for the cultivation of tobacco plants in Vietnam. The results of this research may expand the global database and prove valuable in terms of estimating the AED due to smoking tobacco in Vietnam.

SAMPLES AND METHODS

Study area

The study area was Quang Linh, the provincial capital of Quang Xuong District, Thanh Hóa Province. The area of the Quang Linh commune is about 50 000 m^2 and is one of the biggest regions for growing tobacco plants in Quang Xuong District. Tobacco leaves from here are mainly used to produce tobacco for cigarettes and pipes.

Sampling and preprocessing

Sampling

Six samples of old leaves ~10 months old (lower part of tobacco plant) and young leaves ~6 months old (upper part of tobacco plant) were picked at the commune of Quang Linh. The samples of old leaves were denoted as 1, 2 and 3, whereas the samples of young leaves were labelled as 4, 5 and 6. Sufficient amounts of tobacco leaves were picked to ensure that the dry weight of each sample was ~5 g. The soil samples consisted of ~800 g soil taken from a depth of 25–40 cm around the roots of the tobacco plants from which leaves were picked.

Preprocessing samples and method

Samples of tobacco leaves were washed to remove any traces of soil, dust and surface contamination. The samples of leaves were then chopped into small pieces and dried in an oven at a temperature of 60°C to a constant dry weight. The dried samples were then milled into a powder and sieved through a 0.02-mm mesh sieve. One gram of each from the dry sample of leaves in powdered form was extracted for analysis.

In the laboratory, the powdered samples were wetted by distilled water before 100 mBq of ^{209}Po tracer solution (Polonium nitrate in 1 M HNO_3) (4.88 MeV alpha emission) was added. Subsequently, the samples were wet digested using a mixed solution of HNO_3:HCl (1:3) before H_2O_2 was added to complete the digestion (H_2O_2 acted as an oxidising agent). For the separation of the Po solution co-precipitation with MnO_2 was performed by adding a solution of $MnCl_2$ with $KMnO_4$ in a solution of NH_4OH at pH > 9[32], in combination with the use of Dowex 1 × 8 (100–200 mesh) anion-exchange resins[33] as used previously for other organic samples[34]. The obtained solution was evaporated to almost dryness, then dissolved in a solution of 0.5 M HCl acid before catalytic chemicals were added. ^{210}Po was spontaneously deposited on one side of a polished silver disc from the solution over ~4 h at a temperature of ~80°C. The obtained sample was left to dry at room temperature and measured using a high-resolution passivated and implanted planar silicon (PIPS) detectors ORTEC Alpha-Ensemble-4 spectrometer with AlphaVision software and a minimum detection limit of 0.5 mBq. Quality control was performed using 1 g of each sample of reference material IAEA-446 (Baltic Sea Seaweed) and the recovery rate of ^{209}Po tracer was up to 90%.

Soil samples were also dried in an oven at a temperature of 105°C to a constant weight. The dry soil samples were then milled into a powder and sieved through a 0.2-mm mesh sieve. The preparation of soil samples was carried out following the procedure outlined by Chandrashekara *et al.*[35]. Accordingly, 5 g of each powdered dry sample was transferred to a glass beaker, placed on a hotplate, digested with HNO_3 and then with a mixture of HNO_3 and H_2O_2 to eliminate the organic matter. Digestion was continued until a white residue was obtained. The residue in the form of a solution was evaporated to dryness, converted into its chloride salt by adding HCl (1:1) and dried by evaporation once more. The dried solution was dissolved in 0.5 M HCl and filtered through filter paper (Whatman: Grade 42) in order to remove any traces of residue present in the solution. The obtained solution was treated by following similar steps to those the samples of tobacco leaves were subjected to as outlined above. The obtained sample was left to dry at room temperature and measurements were also taken using a high-resolution PIPS detectors ORTEC Alpha-Ensemble-4 spectrometer with AlphaVision software and a minimum detection limit of 0.5 mBq. Quality control was performed using 5 g of each sample of reference material IAEA-384 (Fangataufa Sediment) and recovery rates of the ^{209}Po tracer were up to 85%.

Calculations

Calculation of activity concentration

The activity concentration of ^{210}Po was calculated based on the following formula:

$$c_{210Po} = A_{209Po} \frac{n_{210Po}}{n_{209Po} \times M} \quad (1)$$

c_{210Po}= activity concentration of ^{210}Po (Bq g^{-1});
A_{209Po}= activity of ^{209}Po (Bq);
n_{209Po}= count number of ^{209}Po (count);
n_{210Po}= count number of ^{210}Po (count);
M = mass of sample (g).

The system is calibrated using the standard material IAEA-384 (and the results are corrected for the difference in counting efficiency for different energies accordingly).

Annual effective dose

The AED is commonly used to evaluate the effect of smoke inhalation from smoking tobacco on human health. In this study, the AED to smokers was calculated based on the following formula:

$$\mathrm{AED} = \mathrm{A} \times \mathrm{F_1} \times \mathrm{F_2} \times \mathrm{D_c} \times \mathrm{N} \times \mathrm{T} \quad (2)$$

AED = the annual effective dose (μSv y^{-1})

A = activity concentration of ^{210}Po in tobacco leaves (Bq g^{-1})

F_1 = transfer factor of ^{210}Po from cigarettes to smoke

F_2 = the inhaled fraction of the total amount of smoke generated

D_c = the effective dose conversion factor (Sv Bq^{-1}) of ^{210}Po

N = daily consumption of tobacco (g) (N = 20 cigarettes, 14 g of dry tobacco leaves

T = the number of days in a year (T = 365).

In previous studies, the effective dose conversion factor (D_c), transfer factor (F_1) and inhaled fraction (F_2) used to estimate the dose of ^{210}Po were inconsistent. Two approaches are commonly used. Group A uses 100% for F_1 in Formula 2, whereas Group B uses some kind of F_1 transfer factor from cigarette to smoke. In general, the most common is the Group B method, 70% for F_1; 50% for F_2 and 3.3 μSv Bq^{-1} for the effective dose conversion factor. The method of Group B is also popular, but it is often customised; the value of F_2 varies significantly from 13 to 25%. The variation in the inhaled fractions (F_2) can be attributed to the smoking protocol, method of analysis and filter used. Variations in these parameters obtained from previous studies are shown in Table 1. In some studies, instead of the general 20 cigarettes per day, data from national statistics are used in the calculations[17].

The AED calculations were carried out according to Formula 2, using the factors presented in the UNSCEAR2000 Report, F_1 70%, F_2 50%, D_c 3.3 μSv Bq^{-1}[38].

EXPERIMENTAL RESULTS AND DISCUSSION

Activity concentration of ^{210}Po

The activity concentrations of ^{210}Po obtained from six pairs of tobacco leaves and their respective soil samples are listed in Table 2. Due to the limited number of samples and the use of fresh tobacco leaves, our results are not representative for all smokers in Vietnam, but they can be used to estimate the relative risk of the fast cured pipe tobacco used locally. As shown in this table, the activity concentrations of ^{210}Po in tobacco leaves vary significantly from 28.7 to 254.4 mBq g^{-1} with an average value of 96.5 ± 84.2 mBq g^{-1}, whereas those in the samples of soil varied only slightly from 51.3 to 69.6 mBq g^{-1} with an average value of 62.6 ± 6.8 mBq g^{-1}. In general, the difference between the activity concentrations of ^{210}Po in the samples of soil is insignificant, possibly because the soil samples were taken from the same area. By contrast, the activity concentration of ^{210}Po in tobacco leaves varies greatly. The maximum activity concentration of ^{210}Po of 254.4 mBq g^{-1} was found in Sample 2 (old leaves), whereas the minimum of 28.7 mBq g^{-1} was found in Sample 5 (young leaves). The average activity concentrations of ^{210}Po in old and young leaves were 152.8 and 38.6 mBq g^{-1}, respectively. In general, the activity concentration of ^{210}Po in old leaves was approximately four times higher than that in young leaves. This finding is in good agreement with that of Skwarzec *et al.*[8] who indicated that the oldest and the youngest leaves contained up to 66% and only 1.8% of the ^{210}Po in the tobacco plants, respectively. Previous studies reported that the uptake of ^{210}Po via roots was insignificant[24]. Thus, the high activity concentration of ^{210}Po in old tobacco leaves can be attributed to the deposition

Table 1. Parameters for dose estimation from previous studies.

F_1	F_2	$F_1 \times F_2$	D_c (μSv Bq^{-1})	References
70%	50%	35%	4.3 (37)	10
75%	50%	37.5%	4.3 (37)	12
70%	50%	35%	3.3 (38)	13
70%	50%	35%	3.3 (38)	15
70%	50%	35%	3.3 (38)	16
70%	50%	35%	3.3 (38)	19
70%	50%	35%	3.3 (38)	14
70%	50%	35%	3.3 (38)	39
75%	50%	37.5%	3.3 (38)	20

Table 2. Activity concentration of ^{210}Po in tobacco leaves and soils.

No.	Age of leaves	Activity concentration of ^{210}Po (mBq g^{-1} dry wt)	
		Tobacco leaves	Soils
1	Old	91.7 ± 6.9	69.2 ± 5.8
2		254.4 ± 19.5	66.9 ± 4.9
3		112.4 ± 10.2	69.6 ± 5.2
4	Young	44.9 ± 4.3	62.9 ± 4.8
5		28.7 ± 2.6	51.3 ± 5.3
6		42.2 ± 5.1	62.2 ± 4.5
Minimum		28.7 ± 2.6	51.3 ± 5.3
Maximum		254.4 ± 19.5	69.6 ± 5.2
Average value (±SD)		95.7 ± 84.2	63.7 ± 6.8

of ^{210}Po from the atmosphere onto their surface[8]. Accordingly, the older leaves are subject to a longer time of exposure to the atmosphere resulting in a higher level of ^{210}Po deposition compared with that found in younger leaves. Thus, it is suggested that the use of young leaves is less harmful than that of old ones. Furthermore, the difference in activity concentration between old and young leaves has some implication for biomonitoring purposes for selecting the part of the plant to be investigated.

The ^{210}Po activity concentrations obtained in tobacco leaves in this study were compared with those from previous studies. Previous results of ^{210}Po activity concentrations were collected and presented in Table 3.

The mass of tobacco in one cigarette was estimated to be 0.7 g. It can be seen that the ^{210}Po activity concentration in the tobacco leaves in Vietnam, estimated to be 95.7 mBq g^{-1}, is the highest amongst the countries investigated; in fact, about 12 times higher than the lowest value of 11 mBq g^{-1} reported for cigarettes in Romania. The highest ^{210}Po activity concentration was found in tobacco leaves in Vietnam, followed by those in Argentina (47.6 mBq g^{-1}). Other countries from previous studies exhibit low ^{210}Po activity concentrations of <40 mBq g^{-1}. The high ^{210}Po activity concentration in tobacco leaves in Vietnam can be attributed to the use or overuse of phosphate fertilisers in growing tobacco plants. The use of phosphate fertilisers has been shown to have an increasing effect on ^{210}Po activity concentration in tobacco plants[41]. However, it should be noted that the ^{210}Po activity concentration in tobacco is strongly dependent on the duration and conditions of storage. The highest activity concentration of ^{210}Po in tobacco in Vietnam was found in pipe tobacco in Thanh Hoa Province[18], ~180 mBq g^{-1}, whereas in cigarettes the maximum reported value was 120 mBq g^{-1}[19]. ^{210}Po is an alpha-emitting isotope with a short half-life of 138 d. Thus, longer storage times will result in lower ^{210}Po activity concentrations. In this study, the ^{210}Po activity concentration in tobacco leaves was measured after having been picked from tobacco plants a few days beforehand. This may lead to a high activity concentration of ^{210}Po in samples of tobacco leaves. It is also suggested that the use of tobacco leaves that have been stored only for a short period of time is more harmful than those stored for longer periods.

Table 3. Concentration of ^{210}Po in tobacco obtained from this study and previous studies.

Type of tobacco	Countries	^{210}Po activity concentration		References
		mBq cigarette^{-1}	mBq g^{-1}	
Tobacco leaves	Vietnam	—	95.7	This study
	Poland	—	24.0	8
	Greece	—	13.1	12
	Brazil	—	22.0	7
Tobacco cigarettes	Portugal	—	18.3	9
	Brazil	—	21.2	6
	Hungary	22.0	31.4	13
	Malaysia	—	12.6	15
	Slovenia	14.0	20.0	1
	India (Tiruchirap palli)	—	13.2	16
	Philippine	12.0	17.1	17
	Saudi and Egypt	—	13.0	11
	Argentina	—	47.6	22
	Egypt	21.0	30.0	10
	Italy	15.7	22.4	21
	China	—	21.6	14
	Japan	—	13.0	39
	Romania	8.4	11.9	20
	Vietnam	22.0	—	18
	Vietnam	26.4	37.7	19

Annual effective dose

The AED to Vietnamese smokers using fresh tobacco leaves was calculated based on Formula 2. The estimated AED varies over a wide range from 169 μSv y^{-1} (Sample 5) to 1501 μSv y^{-1} (Sample 2) with a mean value of 565 μSv y^{-1}. As reported, AED of ^{210}Po to smokers using fresh tobacco in Vietnam was lower in average than the reference level of 1000 and 20 000 μSv y^{-1} for existing situations in ICRP Publication 103[(40)]; however, Sample 2 exceeded 1,000 μSv y^{-1}. This indicates that the AED of ^{210}Po can potentially reach levels, where usually some mitigation measures are considered for planned public exposures. This reference level was used previously as a comparison for radiological exposure from smoking[(17)].

The AEDs calculated in previous studies worldwide were collected and are listed in Table 4. From the present study, it can be seen that the highest AED (565 μSv y^{-1} on average) was observed in Vietnamese tobacco leaves. It has to be noted that this does not represent the average dose from Vietnamese cigarettes, only the six samples from Quang Linh. The second highest value was calculated for Argentinian cigarettes (361 μSv y^{-1}) and the lowest value, of 36 μSv y^{-1}, was calculated for Portuguese cigarettes. In other countries, the range of AEDs was narrow with an average of $<$300 μSv y^{-1}. However, as previously mentioned, it should be noted that the parameters used to estimate the AED to smokers in different countries vary due to differences between types of cigarettes and smoking habits.

CONCLUSIONS

In this study, the activity concentrations of ^{210}Po in tobacco leaves and the soil in the vicinity of the tobacco plants were measured. In addition, the AED to Vietnamese smokers using fresh tobacco leaves was estimated based on the activity concentration of ^{210}Po in tobacco leaves. Based on the analysis of research results, the following conclusions were drawn:

The activity concentrations of ^{210}Po in tobacco leaves varied significantly between 28.7 and 254.4 mBq g^{-1}. The activity concentrations of ^{210}Po in old leaves were higher than those in young leaves. By contrast, the variation in the activity concentration of ^{210}Po in soil was insignificant.

The AED to Vietnamese smokers using fresh tobacco leaves was much higher than the global average. This indicates that smoking freshly harvested tobacco leaves in Vietnam is very harmful to human health, which might explain why the number of patients suffering from lung cancer due to smoking

Table 4. AED to smokers in Vietnam and other countries.

Countries	AED	References
Portugal	36	9
Brazil	160	6
Greece	124	12
Hungary	244	13
Malaysia	106	14
Slovenia	61	1
India (Tiruchirap palli)	112	16
Philippine	42	17
Argentina	361	22
Egypt	193	10
Italy	55.2	21
China	123	15
Japan	68	39
Romania	75.5	20
Vietnam (cigarettes)	74	18
Vietnam (cigarettes)	223	19
Vietnam (tobacco leaves)	565	This study

in Vietnam is rather high. The use of fresh (fast-cured) tobacco products in the area should be discouraged, not only because tobacco use is not beneficial to the population, but also because it can lead to radiological exposure exceeding 1 mSv, which is considered significant for other activities by the reference level in ICRP 103.

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