CONTAMINATION WITH POLONIUM-226, URANIUM AND RADIUM-226 DUE TO SMOKING

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Abstract—The content of ^{210}Po, ^{226}Ra and uranium has been examined in well-known Yugoslavian cigarettes and in tobaccos from regions of Yugoslavia where the best tobacco is produced. Simultaneous measurements were made of the content of these nuclides in the urine of smokers and non-smokers.

The distribution of ^{210}Po in smoke, ash and cigarette stubs as a function of combustion temperature was studied. Values of ^{210}Po in cigarettes ranged from 0.4–0.6 pCi/g, ^{226}Ra below 10^{-12} Ci/g and uranium was less than 5γ/g of cigarette.

Analysis of the distribution of ^{210}Po has shown that about 40% is found in smoke. The urinary ^{210}Po values in smokers and non-smokers are different, but there is no significant statistical difference between them.

So far more than 270 organic substances and 15 elements have been identified in cigarette smoke. These could be divided into four main groups from the toxicological point of view:

(1) nicotine and similar alkaloids,
(2) irritants like aldehydes,
(3) substances binding on blood pigments like carbon monoxide (1), cyanides (2), etc.,
(4) carcinogenic substances like radionuclides, arsenic, chromium, nickel, etc.

Recently more and more attention has been paid to the radionuclide content of tobacco and cigarettes. Turner and Radley[3] and Mulvaney[4] observed high radioactivity in cigarette ash. Mulvaney[4] ascribed this radioactivity to the presence of ^{40}K, and later to ^{87}Rb. He discussed the hazard to the respiratory tract due to smoking. Spiers and Passey[5] consider that the specific radioactivity of ^{40}K is very low and could not represent a real hazard. On the other hand Ash[6] calculated that the radioactivity of ^{40}K in the smoke could cause carcinogenic effects. Runeckless[7] studied the radioactivity of the whole cigarette, the stub and smoke. This author established that 90% of the total radioactivity was in the ash and only 1% is inhaled.

Turner and Radley[3] paid attention to the alpha-radioactivity of the smoke. They showed low alpha-activity due to radon and thoron, the relative quantity being 1% of the total activity. Radford and Hunt[8] studied the ^{210}Po content in the cigarette, stub and smoke. The quantity of ^{210}Po in one cigarette is within the limits 0·39–0·48 pCi and about half of this quantity is inhaled by smoking. These authors calculated that by smoking two packs of cigarettes daily a dose of about 5 rem could be reached in 25 years of smoking. If the distribution is not uniform, local doses could reach 165 rem in 25 years. These doses could cause carcinogenic effects. Skrable *et al.*[9] pointed out that the lung model used by Radford and Hunt is not identical with the lung model proposed by ICRP and concluded that the real doses are lower.

Gregory[10] determined ^{210}Po in tobacco leaves from four countries. Tobacco from New Zealand had a mean value of 0·15 pCi of ^{210}Po per gram; from the U.S.A. and South Africa about 0·49 pCi/g and in Rhodesian tobacco even more. Russian authors[11] established that

tobacco contains 0·43 pCi ^{210}Po per gram and that filter tips could contain 10% of ^{210}Po from the smoke. Brown and Jarvis[12] analyzed the ^{90}Sr content of Canadian tobacco, concluding that this radionuclide does not represent a carcinogenic hazard. Kelly[13] studied ^{210}Po in the inhaled cigarette smoke and established values between 0·029 to 0·044 pCi per type of cigarette. Yavir *et al.*[14] performed spectroscopic analysis of the ^{210}Po content of cigarettes and found quantities similar to Radford and Hunt. Chaterjee *et al.*[15] analyzed the radium content in tobacco ash and found about 0·5 pCi per cigarette.

Some time ago we started to study various sources of radioactive contamination of uranium miners[16-19] and therefore we wished to establish the content of ^{210}Po uranium and radium in Yugoslavian tobacco and cigarettes.

METHOD

Tobacco was digested in the following way: to one gram of tobacco (content of one cigarette) 1 ml of distilled water and 2 ml of fuming HNO_3 was added and slowly digested for 2 hr. Another 5 ml of distilled water and 1 ml of conc. H_2O_2 was added and heated to dryness. Some drops of perchloric acid were then added to digest organic substances and the standard procedure was followed for the determination of ^{210}Po by deposition on silver foils.[20] Determination of polonium in the urine of smokers and non-smokers was performed by the same method.

Uranium was determined using a fluorimetric method[21] and radium indirectly by the determination of radon.[22]

RESULTS AND DISCUSSION

We studied the content of ^{210}Po, uranium and radium in the four most popular kinds of cigarettes and tobaccos from the most important region in Yugoslavia. We paid special attention to tobaccos from the region around uranium mines.

We also studied the distribution of ^{210}Po in the cigarette, smoke and butt and the smoking habits of 25 smokers.

We also analyzed the uranium and radium content of cigarettes and tobaccos. The results obtained were very low and so unreliable that statistical evaluation was impossible.

In Table 1 the results of ^{210}Po determinations of four popular kinds of Yugoslavian cigarettes are presented. It is obvious that some differences exist in the ^{210}Po content. The mean value of ^{210}Po in cigarettes is 0·38 pCi/g. These results are in accordance with those of Radford and Hunt,[8] and very similar to American[10] and Russian[11] results.

It is very interesting to correlate the absorption of ^{210}Po during smoking with excretion of ^{210}Po in the urine of smokers and non-smokers.

To study this correlation we had to establish the smoking habits of Yugoslavian smokers. We measured the temperature of the glowing part of Yugoslavian cigarettes and established that the temperature is 600–650°C. It is known[23] that ^{210}Po is volatilized at this temperature but we checked this fact by adding standard concentrations of ^{210}Po to cigarettes. The ratio of ^{210}Po in the smoke and butt was equal to the ratio between the mass of the whole cigarette and the butt (4: 1).

Table 1. *Activity of* ^{210}*Po in Yugoslavian Cigarettes*

Kind of cigarette	Number of samples	Mean value in pCi/g
"Morava"	5	0·36 ± 0·30
"Ibar"	10	0·20 ± 0·14
"Drava"	9	0·36 ± 0·10
"Zeta"	10	0·60 ± 0·08

Total mean value = 0·38 ± 0·13 pCi/g.
1 cigarette = 1 g.

Table 2. Activity of ^{210}Po in the Urine of Smokers and Non-smokers

	Number of samples	Mean value in pCi/litre	Standard error	*T*-test	
Smokers	20	0·60	±0·05	2·70	0·05
Non-smokers	9	0·37	±0·07		

We examined the smoking habits of 25 Yugoslavian smokers and established that in smoking a cigarette, the smoker:

inhales	30%
combusts	45%
leaves in the butt	25%

Because of ^{210}Po volatilization the same proportions are applicable for this substance. We conclude that smokers inhale about 30% of ^{210}Po present in the cigarette. Therefore, smokers are exposed to a higher quantity of ^{210}Po than non-smokers. In this case a higher excretion of ^{210}Po in the urine of smokers is to be expected.

Urine of 20 smokers and 9 non-smokers was analyzed for ^{210}Po and the results are presented in Table 2. After statistical evaluation of the results it was established that a significantly higher concentration exists in the urine of smokers. This increase is caused by the smoking of cigarettes.

We also wished to establish if differences exist in the ^{210}Po content of tobaccos from various regions of Yugoslavia. From the Institute of Tobacco Research in Beograd, we obtained representative samples of tobaccos from the most important producing areas of Yugoslavia. The results of these determinations are presented in Table 3. We established that statistically significant differences between these tobaccos are not evident. The mean value of these tobaccos was 0·405 pCi of ^{210}Po per gram and we presume that this value represents a mean value for Yugoslavian tobaccos.

We wished to compare this value with ^{210}Po content of tobaccos from some regions around uranium mines. We analyzed tobaccos grown around the uranium mine of Kalna and the uranium field at Pirot and these results are presented in Table 4. In only one case (Inovo at Kalna) we found a significant difference from the Yugoslavian mean value, presented in Table 5.

SUMMARY

We have established that the ^{210}Po content in Yugoslavian tobaccos is about 0·405 pCi/g and for cigarettes 0·38 pCi/g. If a man smokes 20 cigarettes per day containing 7·6 pCi of ^{210}Po he will inhale 30% which is 2·3 pCi per day.

In the urine of smokers we found an increase of 0·23 pCi of ^{210}Po compared with the

Table 3. Activity of ^{210}Po in Tobaccos from Most Important Yugoslavian Areas

	Area	Number of samples	Mean value in pCi/g	Standard error
1	Hercegovina, Stolac	4	0·36	±0·05
2	Vojvodina, Novi Sad	6	0·76	±0·12
3	Makedonija, Prilep	10	0·52	±0·08
4	Srbija, Knjaževac	11	0·36	±0·04
5	Srbija, Pirot	4	0·40	±0·04

Total mean value (excluding No. 2 sample): 0·405 ± 0·153 pCi/g.

Table 4.

	Area	Number of samples	Mean value in pCi/g	Standard error
1	Knjaževac, Trgoviste	11	0·36	0·09
2	Kalna, Inovo	6	0·84	0·09
3	Pirot, Cerova	4	0·40	0·04
4	Pirot, Sugrin	2	0·40	Small number
5	Pirot, Ragadeš	2	0·60	Small number
6	Pirot	2	0·52	Small number

Table 5. *Statistical Evaluation of Results obtained near Oil Field and Uranium Mine with Mean Value for Yugoslavian Tobaccos*

Number of samples	Area	Mean value	Standard error	*T*-test	*P*
35	Yugoslavian tobaccos	0·405	±0·153	4·73	0·01
6	Novi Sad (oil field)	0·76	±0·12		
6	Kalna, Inovo (uranium mines)	0·84	±0·09	6·12	0·01

excretion of non-smokers. Knowing that normal diuresis is 1–1·5 l. of urine per day, we would conclude that of the inhaled ^{210}Po about 10–15% is absorbed and excreted.

The polonium content in various tobaccos and cigarettes does not vary very much over various regions of the country. We obtained an increase in only one place in the vicinity of a uranium mine (Inovo at Kalna) and also in one case at Novi Sad (No. 2 in Table 3) where the oil fields are located.

It would be of interest to continue this study by analyzing the polonium content in plants, soils and waters in the vicinity of oil fields and uranium mines.

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