# **Levels of 210Po in blood, urine and hair of some Saudi smokers**

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The activity concentration of <sup>210</sup>Po was investigated in blood, urine and hair samples of some non-smokers, cigarette-smokers (tobacco-smokers) and shisha smokers (jurak- and mehassel-smokers). The results indicated that <sup>210</sup>Po concentration was variable within each group of volunteers and fluctuated within certain range. The activity concentration in the blood of the non-smokers, the cigarette-smokers and the shisha-smokers was found to be ranged from 7–77, 17–86 and 22–92 mBq/l, respectively. These values were ranged from 1.5–10, 3.3–15.9 and 2.2–19.6 mBq/l in the urine samples of the same volunteers, respectively. The <sup>210</sup>Po activity concentration in their hair was found to be ranged from 1.9–4.8, 1.9–6.4 and 2–6.5 Bq/kg, respectively. The obtained results are discussed and some conclusions, based upon the average values, were drawn.

# **Introduction**

Several studies<sup>1–4</sup> indicated that <sup>210</sup>Pb and <sup>210</sup>Po, the members of the uranium decay series, have long been associated with tobacco plants and induce radiation dose to smokers. In a previous work,<sup>5</sup> the activity concentration of 210Po was measured in different types of tobacco and other smoking paste-mixtures (jurak and mehassel) available in the Saudi market. Jurak is a paste-mixture composed of 30% tobacco leaves, 50% molasses and 20% spices and minced fruits, whereas mehassel is a local name of a paste-mixture of unknown ratios of tobacco leaves to minced fruits and spices only. Both jurak and mehassel are smoked by shisha (water pipe used for smoking and has a water filter). 210Po was also measured in the post-smoking ashes, butts and water filters. It was found that cigarette tobacco (almost 100% tobacco leaves) contains high concentrations of  $^{210}P_0$ , which ranged from 14.7–19.2 Bq/kg and a mean value of 16.4 Bq/kg. The major part (about 68%) of this amount of activity was released with the main smoke stream to the smoker's respiratory system, whereas about 7% was retained by the butt and about 25% remained in the ash. The study also indicated that 210Po activity concentration in jurak and mehassel ranged from 3.1–4.8 and 6.4–8.6 Bq/kg and mean values of 3.8 and 7.2 Bq/kg, respectively. It was concluded that  $^{210}Po$ content in these paste-mixtures were related to the tobacco ingredient in the mixture. As in the case of cigarette smoking, the major part of the 210Po content was released with the main smoke stream to the smoker's respiratory system. These results recommended and encouraged the extension of the study to include the investigation of 210Po concentration in blood, urine and hair of a number of volunteers in Saudi Arabia. Although previous studies  $6.7$  indicated that 210Pb and 210Po are usually in state of radioactive

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equilibrium in tobacco, the present study focuses on 210Po because it is more volatile, and emits a particularly energetic  $\alpha$ -radiation which is more hazardous when received internally. Although the main route of 210Po intake by the human body is the ingestion with foodstuffs, $8-10$  other studies reported that smoking also represents a significant route.<sup>11,12</sup> <sup>210</sup>P<sub>0</sub> from tobacco smoke is carried by the blood and accumulates in the liver, kidneys, bone marrow and blood vessel walls. Ingestion with drinking water, especially of underground origin represents another route of 210Po intakes.13 Inhalation of 222Rn released from the soil also contributes in 210Po body burden. The radiation exposure, delivered to sensitive tissues, as that of air pathways in the respiratory system, may induce cancer both alone and synergistically with non-radioactive carcinogens.14 The absorption coefficient of 210Po into blood from digestive tract was estimated to be 35%, and the excretion of this radionuclide from the body in the urine was 14–15 times less than in feces.8 However, the body burden of 210Po in normal human body may differ from one person to another depending upon the mode of life as diet habits, origin of drinking water, residence place which affects radon exposure rate and also smoking habits as smoking rate, smoking period and the type of the smoking material. However, many factors may affect the <sup>210</sup>Po intake and lead to the variation in the body burden in different individuals. Due to the large variance in the estimates of  $210P<sub>P</sub>$  content among different populations, the prospective of this article is limited to estimating the contribution of smoking to the levels of 210Po in the blood, urine and hair of some Saudi smoker-volunteers. Some volunteers were cigarette-smokers (tobacco-smokers) and others were shiha-smokers (jurak- and mehassel-smokers). The study had included some number of non-smokers for comparison.

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### **Experimental**

### *Volunteers and sample collection*

Samples were collected from a number of smoker and non-smoker volunteers whom, after the aims and risk of the study had been fully explained, had given their full consent to the procedure. Seventy-four male volunteers had been participated in this study, where 51 volunteers were smokers and 23 volunteers were nonsmokers. Among the smokers there were 27 cigarettesmokers and the rest (24 volunteers) were shishasmokers. Their ages dispersed along a wide range (23– 56 years old) and the smoking period ranged from 4–30 years. The non-smoker volunteers were the individuals whom had never smoked, and their samples were used as control samples. The continuous smoking individuals were identified as smokers whereas the irregularly smoking individuals were excluded from this study. After washing, drying and cutting his hair by a barber and keeping it in a labeled plastic bag, 50–60 ml of blood was collected from each volunteer and supplying him with a 3-l plastic container to collect 24-hour urine sample. The samples were collected in successive batches (3 volunteers each) so that each batch was collected after the analysis of the previous one to avoid storing the urine and the blood samples before analysis.

### *Standard reference material*

209Po standard reference material, purchased from the National Institute for Standards & Technology (NIST), USA, under the code number STM 4326, was diluted and used as spiking tracer for analyzing 210Po.

# *Analysis of 210Po in the different samples*

Owing to the volatility of polonium under dry ashing conditions, an aliquot of the collected sample was spiked with 209Po radiotracer, and wet-ashed with conc.  $HNO<sub>3</sub>/30\%$   $H<sub>2</sub>O<sub>2</sub>$  oxidizing mixture to destroy the organic material and release free polonium ions into the solution.15 For blood and urine samples 50 and 150 ml were used for analysis, respectively, whereas 10 grams were used in case of hair samples. The hair samples were wet ashed, exactly as that of the blood and the urine samples, where hair is easily attacked by the oxidizing mixture. Accordingly, and due to the high concentration of 210Po in the hair compared to that of blood and urine, results of blood and urine were reported in mBq/l whereas those of hair were reported in Bq/kg. The sample solution was gently evaporated to near dryness and 5 ml of conc. HCl was added with continuous gentle heating and again evaporated to near dryness. The last step was repeated twice to ensure complete nitrate removal. The medium was diluted with 0.5M HCl to about 100 ml. Ascorbic acid, 40 mg, was added as a reducing agent to prevent any inhibitory effect of ferric ions from the sample. The solution was placed in a modified plating cell and the polonium isotopes were allowed to deposit spontaneously on a silver disc for 4 hours at 90–98 °C with continuous stirring. According to RADFORD et al., <sup>16</sup> the modified spontaneous plating cell was made from commercial baby feeding bottle with the bottom removed. Silver disc (25 mm diameter), rested on a Teflon-base disc, and was held in the screw top of the bottle by a Neoprene gasket. A plastic cover with a center hole for the stirrer was used to reduce evaporation from the inverted bottle. The silver disc was removed carefully, rinsed with distilled water, dried and left for 2 hours after plating to allow decay of short-lived polonium isotopes and counted by α-ray spectrometry.

The samples were measured by using the 4.866 and 5.305 MeV  $\alpha$ -rays of <sup>209</sup>Po and <sup>210</sup>Po, respectively. The blood and urine samples were counted for 60 hours to get adequate counting statistics (counting error <15%), whereas the hair samples were counted for 18 hours which were enough to get a counting error <10%. The chemical yield of 210Po was determined by the material balance technique, using 209Po as a radiotracer for high accuracy and precision. The chemical yield of spontaneous polonium deposition under the conditions mentioned was 84±9%. Analytical quality control of the chemical procedure was regularly performed through IAEA-326 and IAEA-327 standard reference materials.

### *Apparatus*

An "Alpha Analyst" high-resolution α-ray spectrometry system (Canberra model S470) was used for sample measurement. The system consists of eight 450 mm2 silicon surface barrier detectors. Each two detectors are located in the same chamber with partition. The system is fully computer controlled and manage the spectra automatically, using OS/2 operating system, Genie-PC and Alpha Analyst software. The lower limit of detection (LLD) for the eight detectors of the system ranged from 1.1–1.5 mBq/l, for 24-hour counting time and 50 ml sample size. The efficiencies of the detectors in the chosen configuration ranged from 20.4 to 21.1% with a quite satisfactory resolution of about 35–50 keV for the 5.305 MeV alpha-particles of 210Po. A motordriven vacuum pump provided adequate evacuation (10–  $<sup>2</sup>$  mm Hg) of the vacuum chambers of the system.</sup>

The uncertainties given with the final results are one standard deviations resulting from the propagation of all random counting uncertainties occurred anywhere in the entire measurement process.

# **Results and discussion**

The results of <sup>210</sup>Po activity concentration in blood. urine and hair samples of some non-smoker, cigarettesmoker and shisha-smoker volunteers are illustrated in Figs 1 to 3 and summarized in Table 1. The activity concentration of 210Po in the blood was found to be ranged from 7–77, 17–86 and 22–92 mBq/l, respectively (Fig. 1). The average values and standard deviations were 29±13, 41±11 and 50±15 mBq/l, respectively. Based upon these average values, about 41% increase of blood  $2\overline{10}P_0$  was observed among the cigarette-smoker samples whereas about 71% increase was observed among the shisha-smoker samples. In other words, the levels of 210Po in blood samples could follow the sequence: shisha-smokers > cigarette-smokers > nonsmokers. Although a significant difference exists between 210Po concentration in blood of smokers and non-smokers, a three to five fold variation among individuals within the values of the same group was observed. This variation may be related to dietary factors and/or other environmental conditions that affect  $222$ Rn exposure rate.<sup>13,17</sup> Although the  $210$ Po content in cigarette tobacco (average value is 16.4 Bq/kg) and hence, in the main smoke stream is higher than that in jurak and mehassel (average values are 3.8 and 7.8 Bq/kg, respectively),<sup>5</sup> the average  $^{210}Po$ concentration in the blood of the later group (shishasmokers) was rather higher. This may be related to the nature of the smoke particulates of the paste-mixtures that introduced to the respiratory system, on burning the other paste-components that mixed with tobacco, and/or the nature of the puff which is deeper with the use of shisha as a tool of smoking the paste-mixtures causing more <sup>210</sup>Po retention in the lungs.

Figure 2 illustrates 210Po activity concentration in urine samples from the same individuals of the test groups. The results indicated that 210Po activity concentration ranged from 1.5–10, 3.3–15.9 and 2.2– 19.6 mBq/l in urine of non-smokers, cigarette-smokers and shisha-smokers, respectively, with average values and standard deviations of 5.9±1.8, 8.9±2.6 and 8.1 $\pm$ 1.8 mBq/l, respectively. The average <sup>210</sup>Po activity concentration data for urine samples follow the sequence: shisha-smokers  $\approx$  cigarette-smokers  $>$  nonsmokers. Based upon the average values these results reflect rather increase of  $210P<sub>Po</sub>$  in the smoker's urinary

excretion. It was about 50 and 38% among cigarettesmokers and shisha-smokers, respectively (Table 1). Forty-two percent-increase in  $^{210}P_0$  urinary excretion of some Filipino-smokers was reported which was 0.2673 pCi/24 h (9.90 mBq/24 h) compared to 0.1877 pCi/24 h  $(6.95 \text{ mBq}/24 \text{ h})$  of non-smokers.<sup>18</sup> Other studies<sup>19,20</sup> have shown a statistically significant increase in 210Po in smoker's urinary excretion relative to non-smokers among the uranium-miners who exposed to  $222$ Rn inhalation and have already increase in  $210$ Po level in the body burden due to this 222Rn exposure.

Accumulation of 210Po in the hair samples of the test groups was also investigated and the results are shown in Fig. 3. The  $^{210}P_0$  activity concentration in hair of non-smokers, cigarette smokers and shisha-smokers ranged from 1.9–4.8, 1.9–6.4 and 2.0–6.5 Bq/kg, respectively, with average values and standard deviations of  $2.8\pm0.4$ ,  $3.8\pm0.4$  and  $4.8\pm0.4$ , respectively. Based upon the average values, about 35% and 71% increase in 210Po activity concentration in hair is related to smoking among cigarette-smokers and shishasmokers, respectively (Table 1). These results showed the same trend as for blood (shisha-smokers > cigarettesmokers > non-smokers). It may be related to the same reasons mentioned above in case of blood, where more retention of 210Po in the blood is expected to increase 210Po accumulation in hair. The results also indicated that the accumulation of  $210P<sub>O</sub>$  in hair of smokers and even non-smokers was very high, compared to blood and urine. It is in agreement with other findings $8,18$ which reported that the highest concentration of <sup>210</sup>Po in the body was observed in the skeleton and hair.



*Fig. 1.* Activity concentration of <sup>210</sup>Po in blood of non-smokers (NS). cigarette-smokers (CS) and shisha-smokers (SS)

*Table 1.* Average <sup>210</sup>Po activity concentration (in Bq/l) and range in blood, urine and hair of some smokers and non-smoker volunteers

Sample	Non-smokers		Cigarette-smokers			Shisha-smokers		
	Range	Average	Range	Average	Increase, %	Range	Average	Increase, %
<b>Blood</b>	$7 - 77$	$29 \pm 13$	17–86	$41 + 11$	41	$22 - 92$	$50 \pm 15$	
Urine	$1.5 - 10$	$5.9 \pm 1.8$	$3.3 - 15.9$	$8.9 + 2.6$	50	$2.2 - 19.6$	$8.1 \pm 1.8$	38
Hair*	1.9–4.8	$2.8 \pm 0.4$	$-9 - 6.4$	$3.8 \pm 0.4$	35	$2.0 - 6.5$	$4.8 + 0.4$	

\* Activity concentration of  $^{210}$ Po in hair samples is reported in Bq/kg.



*Fig. 2.* Activity concentration of  $^{210}$ Po in urine of non-smokers (NS), cigarette-smokers (CS) and shisha-smokers (SS)



*Fig. 3.* Activity concentration of <sup>210</sup>Po in hair of non-smokers (NS), cigarette-smokers (CS) and shisha-smokers (SS)

Generally, among the three test groups (nonsmokers, cigarette-smokers and shisha-smokers),  $^{210}Po$ activity concentration follows the same trend: hair  $\gg$  blood  $>$  urine in each group.

# **Conclusions**

Polonium-210 activity concentration, either in blood, urine or hair was variable within each group of volunteers, where a significant 3–5 fold variation was observed. This may be attributed to the fact that smoking is one significant route among other different routes of 210Po intake by human body. Based upon the

obtained average values, up to 71%, 50% and 71% increase in 210Po activity concentration was recorded in blood, urinary excretion and hair accumulation among shisha-smokers. Although the 210Po content in the smoking paste mixtures was considerably lower than that in cigarette tobacco, shisha smoking has rather higher effect on increasing the <sup>210</sup>Po body burden. The concentration of  $^{210}Po$  in hair was very high, compared with that of blood and urine. The concentration in the different components follows the sequence: hair >> blood > urine, among smokers and even non-smokers.

### **References**

- 1. E. A. MARTELL, Nature, 249 (1974) 215.
- 2. B. SKWARZEC, J. ULATOWSKI, D. I. STRUMINSKA, A. BORYLO, Environ. Radioact., 57 (2001) 221.
- 3. P. LOPES DOS SANTOS, E. M. WEINBERG, E. PENNA-FRANCA, Reviews Biol. Nuclides, 2 (1970) 73.
- 4. T. C. TSO, N. H. HARELY, I. T. ALEXANDER, Science, 153 (1966).
- 5. M. N. AL-ARIFI, J. Med. Sci., 5 (2005) 83.
- 6. M. S. SANTOS, A. M. G. F. AZEREDO, D. R. MELLO, L. M. O. C. JULIAO, J. Radioanal. Nucl. Chem., 182 (1994) 57.
- 7. J. M. GODY, V. A. GOUVEA, D. R. MELLO, A. M. G. AZEREDO, Radiat. Prot. Dosim., 45 (1992) 299.
- 8. L. A. LADINSKAYA, YU. D. PARFENOV, D. K. POPOV, A. V. FEDOROVA, Arch. Environ. Health, 27 (1973) 254.
- 9. F. P. CARVALHO, Health Phys., 69 (1995) 469.
- 10. P. MCDONALD, D. JACKSON, D. R. P. LEONARD, K. MCKAY, J. Environ. Radioact., 43 (1999) 15.
- 11. A. M. G. F. AZEREDO, J. L. LIPSZTEIN, Radiat. Prot. Dosim., 36 (1991) 51.
- 12. D. L. HENSHAW, K. J. HEYWARD, J. P. THOMAS, A. P. FEWS, Nucl. Track Radiat. Meas., 8 (1984) 453.
- 13. A. NIERI NETO, B. MAZZILLI, J. Environ. Radioact., 41 (1998) 11.
- 14. G. F. KILTHAU, Radiol-Technol., 67 (1996) 217.
- 15. K. SHAHEED, S. S. N. SOMASUNDARAM, P. S. HAMEED, M. A. R. IYENGAR, Environ. Pollut., 95 (1997) 371.
- 16. J. R. RADFORD, E. P. HUNT, V. R. SHERRY, Radiat. Res., 19 (1963) 298.
- 17. J. B. LITTLE, R. B. MCGANDY, Nature, 211 (1966) 842.
- 18. N. B. JUAN, E. BALLELOS, The Determination of Polonium in Urine of Filipino Non-smokers and Smokers, INIS Report, 1976, (PAECCD)-76004, 4 p. From: INIS Atomindex 1977, 8(6), Abstr. No. 296252.
- 19. P. L. SANTOS, R. C. GOUVEA, I. R. DUTRA, Sci. Total Environ., 148 (1994) 61.
- 20. A. M. G. F. AZEREDO, J. L. LIPSZTEIN, Radiat. Prot. Dosim., 36 (1991) 51.