Estimation of the Amount of $^{210}$Po Released with the Smoke Stream into Smoker's Lungs from Cigarette Tobacco and Some Smoking-pastes in Saudi Arabia

Mohammed Nasir Al-Arifi

To estimate the amount of $^{210}$Po released with the smoke stream to smoker's lungs, $^{210}$Po was measured in some types of tobacco, jurak and mehassal samples and in the post-smoking ashes, butts and water filters. Jurak is a paste mixture composed of 30% tobacco, 50% molasses and 20% spices and minced fruits. Mehassal is a local name of a paste mixture of unknown ratios of tobacco to spices and minced fruits. Both jurak and mehassal are used for smoking by Shisha (pipe used for smoking and has a water filter). $^{210}$Po activity concentration in cigarette tobacco, jurak and mehassal ranged from about 1.47-19.2, 3.1-4.8 and 6.4-8.6 Bq kg$^{-1}$ dry weight, respectively with mean values of about 16.4, 3.8 and 7.2 Bq kg$^{-1}$, respectively. The results indicated that about 68% of the $^{210}$Po content released with the smoke stream in case of cigarette and mehassal samples. This value was about 40% in jurak samples.

Key words: Cigarette tobacco, Polonium-210, radiation hazards, tobacco, smoking pastes
INTRODUCTION

$^{210}$Pb and $^{210}$Po, the members of the uranium decay series, have long been associated with tobacco plants\textsuperscript{[1-4]}. It has been reported that the principal mechanism of incorporation involves uptake into roots from the soil and phosphate fertilizers\textsuperscript{[5,6]}. Other studies suggested that the deposition of $^{210}$Pb as a radon daughter product by rainfall is the principal mechanism of $^{210}$Pb and $^{211}$Po entry to the plant\textsuperscript{[7,8]}. Lighted cigarettes produce $^{211}$Po and insoluble $^{210}$Pb in the gaseous and particulate phases of tobacco smoke\textsuperscript{[9]}, which inhaled by the smokers into their lungs delivering radiation doses. $^{210}$Po being soluble gets removed from the inner linings of the lung to the blood. Blood circulating in the lung absorbs it partly and carries it to every tissue and cell in the body. Polonium isotopes are amongst the most radio nuclides to human being\textsuperscript{[10]}. Special attention has been given to the pure $\alpha$-emitter $^{210}$Po as the major radio nuclide of interest due to its volatility (B.P. = 962$^\circ$C) and relatively higher radio toxicity.

Accordingly, a previous preliminary study\textsuperscript{[11]} was carried out just to evaluate the $^{210}$Po levels in the blood of smokers and comparing it with that in the blood of nonsmokers. Based upon analyzing 18 blood samples of smokers and 12 samples of nonsmokers, it was concluded that about 30% of blood-$^{210}$Po is related to smoking. The results recommend and encourage a more comprehensive study to understand all possible local factors that may affect the $^{210}$Po intake in the Saudi population and lead to variation in $^{210}$Po concentration in the blood of the different individuals.

Therefore, the present study was dedicated to determine the $^{210}$Po content in tobacco of some cigarette brands and some paste mixtures used for smoking and available in the Saudi market and commonly used. In addition, the post smoking ashes, butts and water filters were also analyzed to estimate the amount of $^{210}$Po released with the smoke stream to the smoker's lungs.

MATERIALS AND METHODS

Standard reference material: $^{209}$Po standard reference material, purchased from the National Bureau of Standards, USA, under the code numbers STM 4326, was diluted and used as spiking tracer for analyzing $^{209}$Po.

The most frequently used cigarette brands and smoking paste mixtures were investigated. Cigarettes are packed with 100% tobacco leaves, whereas jurak is a paste mixture composed of 30% tobacco, 50% molasses and 20% spices and minced fruits. All available jurak samples were imported from India. Mehassel is local name of a paste mixture of unknown ratios of tobacco to minced fruits and spices. Mehassel is prepared by individuals and locally packed and distributed under no trade mark. Both jurak and mehassel are smoked by Shisha (pipe used for smoking and have a water filter).

For each cigarette tobacco brand, four samples (one pack of 20 cigarettes each) were randomly collected at different places. Two cigarettes from each pack (total of 8 cigarettes) were dissected to isolate the tobacco, the filter and the wrapping paper and considered as one sample. Another 8 cigarettes of the same brand (selected by the same way) were smoked by volunteers, after which the ash and filters of each eight cigarettes were carefully collected and isolated as one sample of each. Eight cigarettes contain $4.72\pm0.24$ g of fresh dry tobacco, where each cigarette contains $0.59\pm0.03$ g. For jurak and mehassel, which were available in 900 g-packs, three packs were also randomly collected for each type at different places. Equal masses were taken up from each pack and mixed. The mixture was dried at 60$^\circ$C in the drying oven for constant weight carefully homogenized to represent one sample. Ten grams was taken for analysis. Another 10 g of the same sample was smoked by Shisha-smoker volunteers, where the ash and the water of the filter were carefully collected and analyzed. All the samples were analyzed\textsuperscript{[10]} in duplicates for $^{210}$Po and the mean value is reported in Bq kg$^{-1}$ dry weight, as a final result. The uncertainties given with the final results are one standard deviation resulting from the propagation of all random uncertainties incurred any where in the entire measurement process.

Owing to the volatility of polonium under dryashing conditions, the samples were spiked with $^{209}$Po radio tracer and wet ashed with conc. HNO$_3$/30% H$_2$O$_2$ oxidizing mixture to destroy the organic material and release free polonium ions into solutions. The solution was gently evaporated to near dryness and 5 mL of conc. HCl were 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 0.5 M HCl to about 100 mL. Ascorbic acid, 40 mg was added as a reducing agent to prevent any inhibitory effect of ferrous 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 h at 90-98$^\circ$C with continuous stirring\textsuperscript{[12,13]}. The modified spontaneous plating cell was made from commercial baby feeding bottle with the bottom removed. A 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 h after plating to allow decay of short-lived polonium isotopes and counted by α-spectrometry.

The samples were measured by using the 4.866 and 5.305 MeV α-peaks of 210Po and 214Po, respectively. The samples were counted for about 18 which was enough to get adequate counting statistics. The chemical yield of 214Po was determined by the material balance technique, using 210Po as a radio tracer for high precision. The chemical yield of spontaneous polonium deposition was 84±9%. Analytical quality control of the chemical procedure was regularly performed through IAEA-326 and IAEA-327 standard reference materials.

**Apparatus:** A Canberra model 8470 Alpha Analyst high-resolution α-spectrometry system was used to measure the α-emitters 210Po. 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 can automatically manage the spectra, 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 33-35 mBq kg⁻¹, for 48 h counting time and 5 g 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 55-50 keV for the 5.305 MeV alpha particles of 210Po.

A motor-driven vacuum pump provided adequate evacuation (10⁻² mm Hg) of the vacuum chambers of the system.

**RESULTS AND DISCUSSION**

Table 1 showed the results of the cigarette tobacco, where the activity concentration of 210Po ranged from 14.72-19.19 Bq kg⁻¹ and an average value of 16.39 Bq kg⁻¹. These values are in agreement with values reported by other investigators[15,16].

Due to these high concentrations and the volatility of polonium, it was also determined in the different post-smoking components (cigarette ash and butt) to estimate the amount of 210Po goes out with the smoke stream to the smoker lungs i.e. the smoke entering to the sucking machine only, excluding the side stream smoke in the surrounding environment. The estimation was based upon the assumption that the amount of 211Po goes out with the smoke stream equals the difference between the amount of 211Po in the fresh tobacco prior to smoking and the sum of the post-smoking amounts of 210Po in the cigarette ash and the butt (Table 1). 211Po content in the fresh filters and wrapping paper of the analyzed brands was measured and the levels were found to be below the detection limits of the counting system (<0.035 mBq kg⁻¹). Other investigator[17] reported that fresh filters of cigarettes contain 0.24-0.9 mBq/filter, where Khater[17] reported that the mean activity concentration of 210Po in fresh filters and wrapping paper were 7.5±3.6 and 7.2±5.8 mBq g⁻¹, respectively. According to the obtained results, the 210Po activity concentration remained in the ash ranged from 3.61-4.51 Bq kg⁻¹ of fresh tobacco after smoking, with an average value of 4.02 Bq kg⁻¹, whereas the amount trapped by the butt ranged from 0.95-1.4 Bq kg⁻¹ of fresh tobacco, with an average value of 1.22 Bq kg⁻¹. Based upon the average values, it can be concluded that during smoking 25% of the 210Po contained in tobacco remained in the tobacco ash and about 7% were trapped by the butt, whereas about 68% of the 211Po went out with the smoke stream to the smoker lungs. Other study[18] reported that the distribution of 210Po among ash, filter and smoke was 5.1-7.1, 0.2-16.5 and 28.9-70%, respectively, with a mean values of 25.7, 3.9 and 70%, respectively. These mean values are in agreement with the present findings. Such large differences in the 210Po content in ash in comparison with its content in tobacco are due to the different tobacco-burning temperatures of smoking (500-700°C), which cause 210Po to sublime into the smoke[19]. These differences may be related to the different burning temperatures and suction rate during smoking. 210Po concentration in fresh butts (prior to smoking) was investigated in the different brands and was found to be below the detection limit of the detectors (44-46 mBq kg⁻¹). Skwarec et al[20] reported that 210Po levels in fresh cigarette filter ranged from 0.03-0.76 mBq filter.

To study the effect of suction rate on the 210Po concentration in the smoke stream, some cigarettes were installed in a suitable tube and connected to a suction pump and ignited. The cigarette was completely smoked within about 20 sec and the ash was carefully collected with the butts and analyzed for 210Po. The results showed that the remained 210Po in the ash was considerably less. It was found to be ranged from 0.74-1.50 Bq kg⁻¹ of tobacco with an average value of 1.08 Bq kg⁻¹ (Table 1). According to the average values, 6-7% of the tobacco's polonium remained in the ash and about 43% trapped by the butt and the rest (about 50%) supposed to be had gone out with the smoke stream. It is clear that suction rate has a considerable effect on the polonium distribution in the different cigarette components since the remained 210Po in the ash was considerably reduced from about 25%, in case of individual smoking, to about 7%, although the amount of 210Po in the smoke steam was
Table 1: Activity concentration of $^{210}$Po in tobacco of some cigarette brands and their post-smoking components (ash, butt and smoke stream)

<table>
<thead>
<tr>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>MERIT</td>
<td>15.52±0.94</td>
<td>4.06±0.39</td>
<td>26.16</td>
<td>1.18±0.24</td>
<td>7.60</td>
<td>10.28±1.05</td>
<td>66.24</td>
</tr>
<tr>
<td>KENT</td>
<td>16.10±0.93</td>
<td>3.61±0.34</td>
<td>22.42</td>
<td>1.08±0.21</td>
<td>6.71</td>
<td>11.41±1.01</td>
<td>70.87</td>
</tr>
<tr>
<td>Davidst</td>
<td>19.19±1.15</td>
<td>4.25±0.37</td>
<td>22.15</td>
<td>1.40±0.27</td>
<td>7.30</td>
<td>13.54±1.24</td>
<td>70.56</td>
</tr>
<tr>
<td>CARLTON</td>
<td>17.14±1.34</td>
<td>4.53±0.42</td>
<td>26.43</td>
<td>1.16±0.23</td>
<td>6.77</td>
<td>11.45±1.42</td>
<td>66.80</td>
</tr>
<tr>
<td>Marlboro</td>
<td>16.53±1.04</td>
<td>3.68±0.37</td>
<td>22.26</td>
<td>1.35±0.25</td>
<td>8.17</td>
<td>11.50±1.13</td>
<td>69.57</td>
</tr>
<tr>
<td>Marlboro*</td>
<td>14.72±1.35</td>
<td>3.09±0.39</td>
<td>27.11</td>
<td>1.24±0.22</td>
<td>8.42</td>
<td>9.49±1.42</td>
<td>64.47</td>
</tr>
<tr>
<td>L&amp;M *</td>
<td>15.39±1.03</td>
<td>3.75±0.35</td>
<td>24.37</td>
<td>1.41±0.27</td>
<td>9.16</td>
<td>10.23±1.12</td>
<td>66.47</td>
</tr>
<tr>
<td>L&amp;M **</td>
<td>16.51±1.02</td>
<td>4.30±0.40</td>
<td>26.05</td>
<td>0.95±0.20</td>
<td>5.75</td>
<td>11.26±1.11</td>
<td>68.20</td>
</tr>
<tr>
<td>Average</td>
<td>16.39±1.10</td>
<td>4.02±0.38</td>
<td>24.62</td>
<td>1.22±0.24</td>
<td>7.49</td>
<td>11.15±1.19</td>
<td>67.90</td>
</tr>
</tbody>
</table>

* = Lights  ** = Ultra lights

The increase in suction rate rises the ashing temperature of the tobacco during smoking, in addition to the intensity of the hot air wind carrying the smoke stream. Both may help in polonium evolution with the smoke February 17, 2005 stream before trapping by the butt. Higher suction rate produces bigger smoke particulates which are easily trapped by the butt resulted in reduction in $^{210}$Po releases with the smoke stream to the lungs. This explanation was supported by the observation that butts of the pump smoked cigarettes were dirtier than that of the individual smoked cigarettes.

A group of cigarettes (8 cigarettes, selected as mentioned before) of the same brand (CARLTON) was prepared so that 25% of the length of each cigarette was smoked by one volunteer and the same way. The remained part of the fresh tobacco (75% of the cigarette length of each cigarette) was carefully collected, homogenized, dried for constant weight and analyzed for $^{210}$Po, to study the effect of the flowing of the smoke stream on the $^{210}$Po level in the remained part. Other two groups of the same brand were prepared by the same way, except that 50% of the length of each cigarette was smoked in one group and 75% of the length of each cigarette was smoked in the other group. The remained tobacco of each group (50 and 25%, respectively) was analyzed and results indicated that $^{210}$Po concentration increases in the remained part during smoking. This means that the tobacco acts as an additional filter during smoking and hence, the smoke stream of the initial stage of smoking has a $^{210}$Po concentration lower than that of the final stage (Fig. 1). In other words, it can be concluded that the first part of the cigarette is comparatively have low amount of $^{210}$Po than the middle part and the middle have low amount than the final part. In the final part, the smoke stream is more saturated with $^{210}$Po because this part trapped an amount of $^{210}$Po from the smoke stream during the previous stages and smoked without additional filter.

Fig. 1: Change of $^{210}$Po level, in the remained part of cigarette tobacco, with smoking due to smoke stream flow

As mentioned above, jurak samples are composed of 30% tobacco and 70% other ingredients. $^{210}$Po levels ranged from 3.07-4.82 Bq kg$^{-1}$, with an average value of 3.79 Bq kg$^{-1}$. The results indicated that these samples, of 30% tobacco content, contain almost 30% of the $^{210}$Po activity concentration recorded in the cigarette tobacco (100% tobacco) samples. This means that $^{210}$Po levels in the samples are related to tobacco and the other ingredients may have a negligible $^{210}$Po concentration.

Table 2 presents also the $^{210}$Po activity concentration in some mehassel samples. It can be observed that the $^{210}$Po content in these samples is higher than that in jurak samples. It ranged from 6.39-8.59 Bq kg$^{-1}$ with an average value of 7.12 Bq kg$^{-1}$. Based upon the average $^{210}$Po concentration values and supposing that the other ingredients have a negligible levels of $^{210}$Po, it can be concluded that mehassel may be contain about 50% (by mass) tobacco and 50% other ingredients.

The distribution of $^{210}$Po in the different components (ash, water filter and smoke stream) of jurak and mehassel samples after smoking by Shisha was also studied. The results indicated that in the jurak samples the remained concentration of $^{210}$Po in the ash ranged from 1.72-2.38 Bq kg$^{-1}$ of fresh jurak, with an average value of 2.02 Bq kg$^{-1}$ (Table 2). In the water filter, the trapped
Table 2: Activity concentration of $^{210}$Po in some mixed pastes used for smoking by Shisha and its post-smoking distribution in the different components (ash, water filter and smoke stream)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Mixed paste</th>
<th>Conc. (Bq kg$^{-1}$)</th>
<th>%</th>
<th>Water filter</th>
<th>Conc. (Bq kg$^{-1}$)</th>
<th>%</th>
<th>Smoke stream</th>
<th>Conc. (Bq kg$^{-1}$)</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Indian Jurak</td>
<td>3.88±0.41</td>
<td>2.10±0.44</td>
<td>54.12</td>
<td>0.25±0.04</td>
<td>6.44</td>
<td>1.53±0.60</td>
<td>39.43</td>
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<tr>
<td>Indian Jurak</td>
<td>3.85±0.39</td>
<td>2.16±0.45</td>
<td>56.10</td>
<td>0.23±0.05</td>
<td>5.97</td>
<td>1.46±0.60</td>
<td>37.92</td>
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<tr>
<td>Indian Jurak</td>
<td>4.81±0.43</td>
<td>2.38±0.38</td>
<td>49.48</td>
<td>0.26±0.05</td>
<td>5.82</td>
<td>2.15±0.60</td>
<td>44.70</td>
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<tr>
<td>Indian Jurak</td>
<td>3.37±0.28</td>
<td>1.75±0.38</td>
<td>57.65</td>
<td>0.24±0.04</td>
<td>7.82</td>
<td>1.06±0.47</td>
<td>34.53</td>
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<tr>
<td>Indian Jurak</td>
<td>3.32±0.41</td>
<td>1.70±0.31</td>
<td>51.20</td>
<td>0.23±0.04</td>
<td>6.93</td>
<td>1.39±0.52</td>
<td>41.87</td>
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<tr>
<td>Average</td>
<td>3.79±0.38</td>
<td>2.02±0.39</td>
<td>53.71</td>
<td>0.25±0.04</td>
<td>6.60</td>
<td>1.52±0.56</td>
<td>39.69</td>
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<tr>
<td>Local mehassel</td>
<td>8.59±0.12</td>
<td>2.71±0.19</td>
<td>31.55</td>
<td>0.10±0.02</td>
<td>1.16</td>
<td>5.78±0.94</td>
<td>67.29</td>
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<td>Local mehassel</td>
<td>6.87±0.71</td>
<td>1.62±0.14</td>
<td>23.58</td>
<td>0.14±0.04</td>
<td>2.04</td>
<td>5.11±0.72</td>
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<td>Local mehassel</td>
<td>6.90±0.57</td>
<td>2.31±0.20</td>
<td>33.48</td>
<td>0.12±0.01</td>
<td>1.74</td>
<td>4.47±0.6</td>
<td>64.78</td>
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<tr>
<td>Sudanese mehassel</td>
<td>6.39±0.55</td>
<td>1.94±0.15</td>
<td>30.36</td>
<td>0.13±0.02</td>
<td>2.03</td>
<td>4.32±0.55</td>
<td>67.61</td>
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</tr>
<tr>
<td>Average</td>
<td>7.12±0.71</td>
<td>2.15±0.17</td>
<td>29.74</td>
<td>0.12±0.02</td>
<td>1.74</td>
<td>4.92±0.73</td>
<td>66.52</td>
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</tbody>
</table>

amount ranged from 0.23-0.28 Bq kg$^{-1}$ of fresh tobacco with an average value of 0.25 Bq kg$^{-1}$. According to the average values, it can be concluded that about 54% of the $^{210}$Po remained in the ash, 67% trapped by the water filter and the rest (about 40%) went out with the smoke stream.

In mehassel samples, the $^{210}$Po concentration remained in the ash ranged from 1.6-2.71 Bq kg$^{-1}$ of fresh mehassel, with an average value of 2.15 Bq kg$^{-1}$. In the water filter, the trapped amount of $^{210}$Po ranged from 0.1-0.14 Bq kg$^{-1}$ of fresh mehassel with an average value of 0.12 Bq kg$^{-1}$. According to the average values, it can be concluded that about 30% of the $^{210}$Po was remained in the ash, about 1.7% was trapped by the water filter and the rest (about 68%) had gone with the smoke stream.

Generally, it is clear that $^{210}$Po activity concentration in the analyzed samples follow the sequence: cigarette tobacco > mehassel > jurak. The concentration in the released smoke stream follows the same sequence. Based upon the percent release of $^{210}$Po in the smoke stream, it was about 68% in both cigarette tobacco and mehassel and about 40% in jurak. The less percent release of $^{210}$Po in jurak samples may be due to the nature of one or more of the other ingredients of this paste mixture. It may be due to molasses which represents 50% of the components of jurak and not present among the ingredients of mehassel. Jurak samples left a greater ash fraction which may be acted as an additional filter opposing release of $^{210}$Po with the smoke stream. This means that with regard to $^{210}$Po exposure, jurak smoking is safer to the smoker lungs than mehassel and cigarette tobacco.

CONCLUSIONS

Cigarette tobacco contains high concentration of $^{137}$Cs which ranged from 14.72-19.19 Bq kg$^{-1}$ and a mean value of 16.59 Bq kg$^{-1}$. On cigarette-smoking, about 68% of the $^{210}$Po content in the cigarette tobacco release with the smoke stream to the smoker lungs whereas about 25% remained in the ash and about 7% retained by the filter. Increasing the suction rate, resulted in reduction in the remained $^{210}$Po concentration in the ash from 25% to about 6.5% whereas about 43% is trapped by the butt and the rest (about 50%) supposed to be had gone out with the smoke stream. The filter trapped more amounts due to the comparatively larger smoke particulates which contain $^{210}$Po and are easily trapped by the filter.

$^{210}$Po activity concentration in jurak and mehassel ranged from 3.07-4.81 Bq kg$^{-1}$ and from 6.39-8.59 Bq kg$^{-1}$ and a mean values of 3.79 and 7.17 Bq kg$^{-1}$, respectively. These values indicate that the $^{210}$Po content in the paste mixtures is related to the tobacco ingredient in the paste mixture. On mehassel smoking, about 68% of the $^{210}$Po content release with the smoke stream to the smoker lungs, whereas about 30% remained in the ash and about 1.7% trapped by the water filter. These values were about 54%, about 7% and about 40%, respectively, on jurak smoking. This means that with regard to $^{210}$Po exposure, jurak smoking have more exposure to $^{210}$Po than cigarette tobacco and mehassel.

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