# THE LANCET

## Supplementary appendix

This appendix formed part of the original submission and has been peer reviewed. We post it as supplied by the authors.

Supplement to: Froidevaux P, Baechler S, Bailat CJ, at al. Improving forensic investigation for polonium poisoning. *Lancet* 2013; **382:** 1308.



#### Figure: <sup>210</sup>Po activities for each measured sample.

Stained samples were visually used or worn (#1 and #2 underwear with urine stain; #3 chapka; #4 used toothbrush; #5 and #6 hospital cap with blood stain; #7chapka; #8 sportswear.) The horizontal dotted line at 3-5 mBq defines the upper range of the measured reference activities. The activity of the highest contaminated sample (#1) is about 50 times the value of the reference samples. Uncertainties (coverage factor k=2) are about 5% for most active, about 10% at 5 mBq and about 40% for least active samples.

### Methods

**DNA analyses.** DNA analyses were performed in order to establish that the hairs tested belonged to Mr. Arafat. As no reference sample from his DNA was directly available, Mrs. Souha Arafat and Ms Zahwa Arafat, respectively Mr. Arafat's wife and daughter, kindly accepted to provide us with buccal swabs in order to perform DNA tests. From the personal items found in the bag, we decided to analyse a sport cap, a wool cap and 5 pairs of medical glasses. About 10 cm<sup>2</sup> of subsamples were cut from the 2 caps, and moistened swabs were used to sample cells from the glasses. DNA was extracted and purified from these samples using standard techniques.<sup>1</sup> Recovered DNA was amplified with the NGM SElect kit (Life technologies) following the manufacturer's instructions. An ABI3500 (Life technologies) was used for the analysis of the amplified DNA in a standard way. The hypervariable regions one and two (HVI and HVII) of the control region of the mitochondrial DNA (mtDNA) were analyzed following the methodology described in Castella et al.<sup>2</sup>, using an ABI 3130 (Life technologies) for the electrophoresis.

Unfortunately, no nuclear DNA (nDNA) profile could be obtained from the hairs. An nDNA profile was nevertheless obtained from the caps and glasses. This precluded a direct genetic identification of the hairs using Souha and Zahwa Arafat's buccal swabs. Nonetheless, we succeeded in establishing the mtDNA profile of the hairs. Since mitochondria are maternally inherited, Souha and Zahwa Arafat's mtDNA are the same but differ from Yasser Arafat's mtDNA. We therefore had to perform an indirect genetic identification using the following procedure: first, we established an nDNA profile from the glasses and caps found in the bag. Then we used a paternity test to assess the compatibility of this DNA with the daughter's nDNA. The probability that the DNA from items found in the bag is DNA belonging to Zahwa Arafat's father exceeds 99.999%. We then established an mtDNA profile from these items and compared it with the one obtained from the hairs. The two mtDNA profiles were identical. Since this mtDNA profile was not observed within a database containing the mitochondria DNA profiles of 10841 individuals (http://empop.org/), our analytical results strongly support the hypotheses that the hairs were effectively those of Mr. Arafat.

**Toxicological analyses.** Analyses of elements on 20 mg of hair samples found in a woollen cap were performed by ICP-MS (Agilent 7700 series) after acidic mineralisation (1 hour at 80°C in 0.5 ml of nitric acid 65% Suprapur<sup>®</sup>). Analyses included a full screening for the following elements: Be, Al, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Mo, Ag, Cd, Sn, Sb, Te, I, Ba, Pt, Hg, Tl, Pb, Bi, Th and U. Screening for organic and drug molecules was carried out on several items found in the bag by GC-MS (Agilent 5973N) and by LC-MS/MS (AB Sciex 5500) after dilution of 1 mg of powder or liquid into 1 ml of methanol. Systematic toxicological analysis was performed according to procedures described by Maurer.<sup>3</sup>

 $\gamma$ -radioactivity. For gamma spectrometry measurements, personal belongings were pooled into 11 samples. Each sample was then measured using a high purity germanium spectrometer p-type coupled with the software Winner 6.0 (measuring time > 24h). Artificial radionuclides, such as  ${}^{60}$ Co,  ${}^{134}$ Cs and  ${}^{137}$ Cs were investigated as well as  ${}^{210}$ Pb.

<sup>210</sup>**Po** determination: a subsample of 2 to 3 g of textile (mostly cotton wool) was traced with  $50\pm0.3$  mBq of <sup>209</sup>Po then charred with 10 ml of conc. H<sub>2</sub>SO<sub>4</sub> at about 70°C. The sample was oxidised cautiously by adding 5 x 3 ml portions of conc. HNO<sub>3</sub>. When NO<sub>x</sub> production almost stopped, the solution was left covered with a watch glass for 2 hours at 80°C, afterwards the complete removal of organic matter was performed in a microwave digester under pressure (Ultraclave IV, Milestone, Germany). The condition for microwave mineralisation was: initial pressure 50 bars; maximal temperature: 180°C, mineralisation time: 30 minutes. If synthetic fibres are present, the solution must be filtered because they are not soluble. <sup>210</sup>Po was co-precipitated from the acidic solution along with iron hydroxide. After centrifugation, the precipitate was dissolved in 80 ml 1 M HCl , 500 mg of ascorbic acid was added and the polonium spontaneously electrodeposited on a silver disc during 4 hours at 70°C.<sup>4</sup> The sources were counted (170'000 to 400'000 s) on a PIPS detector (silicium detector of 450 mm<sup>2</sup>) in an alpha spectrometer (Canberra Alpha Analyst, France). Quality control was carried out through participation to the PROCORAD interlaboratories comparison annual exercise for the determination of <sup>210</sup>Po in urine (2008-2011, average bias: 4.2%).<sup>5</sup>

**Supported** <sup>210</sup>Po determination: after depositing <sup>210</sup>Po on a Ag° disc, the solution was evaporated to dryness and conc. HNO<sub>3</sub> portions were added to destroy all remaining ascorbic acid. The residue was dissolved in 10 ml 9 M HCl and this solution was passed through an anionic chromatography column (2 ml of Dowex AG1x8) to extract [PoCl<sub>6</sub>]<sup>2-</sup>. <sup>210</sup>Pb will pass into the elution solution which was evaporated to dryness. The residue was dissolved in 80 ml 1M HCl and left in the refrigerator for at least 3 months (35 % re-growth of <sup>210</sup>Po from <sup>210</sup>Pb) afterwards, 50 mBq of <sup>209</sup>Po was added and polonium electrodeposited on a silver disc as before. The method was checked for consistency by measuring the co-precipitation yield of Pb (stable) iron hydroxides (98 ± 2%) by ICP-OES (Perkin-Elmer Optima 3300 DV). We also checked that the column (Dowex AG 1x8, 2 ml, Bio-Rad, Switzerland) used to separate <sup>210</sup>Pb from <sup>210</sup>Po was able to produce a pure <sup>210</sup>Pb source without <sup>210</sup>Po. To do this, we spiked three aliquots with 168 mBq of <sup>210</sup>Pb/<sup>210</sup>Po at equilibrium and we separated Pb from Po. No <sup>210</sup>Po was found in the <sup>210</sup>Pb fraction, which means that the method quantitatively separated <sup>210</sup>Pb from <sup>210</sup>Po. Using stable Pb and ICP-OES measurements, we also confirmed that Pb was not extracted on the Dowex AG 1x8 column (extraction < 1%). Eventually, we checked that the <sup>210</sup>Po deposited on the Ag° disk did not contain co-deposited <sup>210</sup>Pb; for that we re-measured, three months after the first measurement, the 5 more active <sup>210</sup>Po sources (underwear, 2 subsamples; hospital cap, 2 subsamples and Russian chapka, one subsample). Based on the activity measured at t<sub>0</sub> and three months later, we determined the half-life of the <sup>210</sup>Po deposited on the source. We obtained T<sub>1/2</sub> between 134 and 164 days, which meant that the <sup>210</sup>Po sources are very pure (target value: 138 days).

**Biokinetic model**. The most up-to-date systemic biokinetic model of polonium<sup>6</sup> has been implemented in the simulation modelling tool Ecolego<sup>7</sup> in order to calculate the typical retention rate of <sup>210</sup>Po in organs and tissues after acute ingestion. In our case, ingestion is the most probable route of intake. Therefore, the systemic biokinetic model was coupled to the human alimentary tract model of ICRP 100<sup>6</sup>. Absorption of <sup>210</sup>Po is assumed to occur exclusively from the small intestine and is characterized by the fractional intestinal absorption  $f_1=0.1$  (inorganic form) or  $f_1=0.5$  (organic form). The  $f_1$  value is defined as the fraction of the activity leaving the stomach that is subsequently transferred to the blood by absorption from the small intestine. Using this model with  $f_1=0.1$ , the daily urinary excretion after acute ingestion of 1 Bq of <sup>210</sup>Po was determined. Based on activity estimates of ingested <sup>210</sup>Po in the case of Mr. Litvinenko's poisoning, we estimated the level of activity that one might find in urine and belongings, especially underwear, in the case of Mr. Arafat's poisoning.

#### Supplementary results and discussion

 $\gamma$ -radioactivity: None of the samples checked by surface contamination monitors and by  $\gamma$ -spectrometry measurements displayed any activity above the detection limit. This was coherent with the  $\gamma$ -spectrometry performed in France on 8 November 2004, by the "Laboratoire de contrôle radiotoxicologique des Armées" on two urine samples collected over 3 days. However, because the low intensity (0.00107%) gamma ray of 803 keV emitted by <sup>210</sup>Po had not been investigated at that time, we reassessed the raw data of the gamma spectra. This new analysis did not reveal the presence of <sup>210</sup>Po. A high activity of <sup>40</sup>K in urine was confirmed: about 400 Bq/l while the normal daily urinary excretion<sup>3</sup> is around 80 Bq/l. This is compatible with the medical report that mentions a treatment with furosemide from 6 to 10 November 2004 for acute kidney failure. Indeed, this treatment increases the rate of urine flow and the excretion of electrolytes such as potassium.

<sup>210</sup>**Po analyses:** We first focused on a pair of underwear that was stained with urine drops. Five subsamples showed activities compatible with typical background level  $(1.4\pm0.3 \text{ to } 4.6\pm0.7 \text{ mBq})$  but two subsamples showed activities that were above background level  $(60\pm3.0 \text{ and } 181\pm8 \text{ mBq})$ . The value 181 mBq was found in a subsample obtained by carefully cutting around a visible stain of urine. The subsample yielding an activity of 60 mBq was taken contiguously to the visible stain of urine. The other subsamples were taken randomly from the underwear. We also studied in detail the Russian chapka believed to be the one Mr. Arafat was wearing when he left Ramallah. Subsamples of the chapka in contact with the head were collected and analyzed. Results showed above background values for two subsamples ( $11\pm0.8$  and  $34\pm2$  mBq) while three other subsamples had activities close to background values ( $0.6\pm0.2$  to  $2.6\pm0.4$  mBq).

Mr. Arafat's hospital cap was stained with several droplets of blood and a larger one with a diameter of about 1.5 cm. We cut around this blood stain and the analysis yielded a <sup>210</sup>Po activity of 13.5±1.0 mBq. Another subsample, taken from blood stains, had an activity of only 1.8±0.3 mBq. Eventually, we measured a larger subsample, including part of the blood droplets. Its activity was 16±1 mBq. Additionally, the bristles taken from Mr. Arafat's toothbrush had an activity of 21.1±1.7 mBq.

Some items taken from Mr. Arafat's belongings displayed lower values, with most of the activities in a range between 0.5 and 2.5 mBq (n=17) per subsample weighting about 3 g. These activities were obtained for clothing that had not been worn. Intermediate activities were found for 12 samples with activities between 2.7 and 10.7 mBq. Among them were four subsamples from Mr. Arafat's sportswear (three subsamples from the dirty collar and one subsample from the back with a stain) with activities between 5 and 10.7 mBq.

To compare the activities found in Mr. Arafat's belongings to those that could be expected from samples never exposed to artificial <sup>210</sup>Po, we measured 37 samples taken from our collaborators (toothbrush, shirt and underwear) or bought in local shops (underwear) as well as blanks made from the products used in the analysis (see method). The activities of the toothbrushes were very low  $(0.10\pm0.09 \text{ to } 0.33\pm0.20 \text{ mBq}, n=5)$  as well as those from clothing items kept in an attic for more than 10 years, carefully packed in a plastic bag  $(1.3\pm0.3\pm0.3 \text{ to } 0.3\pm0.20 \text{ mBq})$ 

 $2.2\pm0.4$  mBq, n=11). Underwear from two collaborators and from a local shop had activities between  $1.1\pm0.2$  and  $3.6\pm0.6$  mBq (n=6) but a pair of Zimmerli brand underwear showed a higher activity ( $7.1\pm0.5$  mBq). Four subsamples from a four-pack of underwear bought in a local supermarket showed elevated activities between  $4.7\pm0.6$  mBq to  $20.7\pm1.6$  mBq, while four subsamples from the same pack showed activities between  $2.6\pm0.9$  and  $4.3\pm0.6$  mBq. Finally, blank samples had activities between  $0.16\pm0.07$  to  $1.9\pm0.3$  mBq, with an apparently higher value when the concentrated H<sub>2</sub>SO<sub>4</sub> used in the analysis was kept in a glass bottle ( $1.4\pm1.4$  mBq and  $1.9\pm0.3$  mBq) compared to the concentrated H<sub>2</sub>SO<sub>4</sub> kept in plastic bottle ( $0.16\pm0.07$  to  $0.43\pm0.40$  mBq).

Higher than background level activities of <sup>210</sup>Po were measured on some samples that had not been exposed to artificial <sup>210</sup>Po. Several subsamples from an underwear pack bought in a local supermarket and one sample from a pair of underwear bought in a local luxury shop, and which matched Mr. Arafat's brand of underwear, showed elevated levels of <sup>210</sup>Po. The highest activity measured on these samples was 20 mBq, 70% of it unsupported, raising questions about the background level of natural <sup>210</sup>Po that can be expected from new cotton fabrics. In addition, these results question the intensity of the radioactive disequilibrium between <sup>210</sup>Pb and its granddaughter <sup>210</sup>Po in environmental samples exposed to radionuclide plant uptake and <sup>222</sup>Rn progeny. In fact, cotton can contain <sup>210</sup>Po through the transfer of the element from soil to plant, due to the decay chain of the ubiquitous <sup>238</sup>U isotope. Furthermore, <sup>210</sup>Po can be found in cotton wool balls as a result of the atmospheric deposition of <sup>222</sup>Rn progeny.<sup>8</sup>, <sup>9</sup> Superphosphate fertilizers are known to increase <sup>210</sup>Po and <sup>210</sup>Pb concentration in plants.<sup>10</sup> When the cotton fabrics are new, they may present increased levels of unsupported <sup>210</sup>Po, as demonstrated by the results of analyses on the four-pack of underwear bought in a local supermarket. On the other hand, subsamples from items kept in an attic for 10 years protected from <sup>222</sup>Rn progeny show only background values (0.6±0.2 to 2.2±0.4 mBq), most probably supported by <sup>210</sup>Pb (not determined here because of the low activity). Thus, it is necessary to compare the increased activities found in the samples stained by body fluids to the activities of samples kept in similar conditions.

We determined the reference values of <sup>210</sup>Po activities internally with items from Mr. Arafat's bag and externally by measuring Swiss samples that had been stored in similar conditions for a comparable period of time. In particular, because Mr. Arafat's items had been stored for 8 years on the 4<sup>th</sup> floor of an office building, it is reasonable to assume that the radon contamination was very low. From the bag, we measured 9 subsamples from clothing that had not been worn. The activities were between  $0.5\pm0.2$  and  $3.4\pm0.4$  mBq and match the activities found in samples kept for 10 years or more in an attic ( $0.6\pm0.0.2$  to  $2.2\pm0.4$ , n=11). As a result, the reference value for cotton fabrics kept shielded from <sup>222</sup>Rn progeny for at least 8 years has an activity of <sup>210</sup>Po close to a maximum of 3 mBq. In this respect, the activities found in two subsamples of underwear stained by urine, two subsamples of a hospital cap stained with blood droplets, two subsamples of the Russian chapka and one subsample of a toothbrush are significantly above this value.

Because <sup>210</sup>Po can also be the decay product (grandchild) of <sup>210</sup>Pb, the next step of our analysis was to estimate the amount of <sup>210</sup>Po that was supported by <sup>210</sup>Pb. As it is not possible to directly measure such low activities of <sup>210</sup>Pb, we waited for three months in order to let <sup>210</sup>Po re-grow in the polonium-purified solution. Ten samples showing elevated levels of <sup>210</sup>Po were therefore measured again. This allowed us to determine the amount of <sup>210</sup>Po that is supported by <sup>210</sup>Pb. All the samples have supported <sup>210</sup>Po levels below 50%.

**Biokinetic model and calculations**: The ICRP alimentary tract model<sup>6</sup> and the systemic biokinetic model for polonium revised by Leggett and Eckerman<sup>11</sup> were used to calculate the daily urinary excretion after an acute ingestion of <sup>210</sup>Po, as shown in Fig. 2-SM. Regarding the poisoning of Mr Litvinenko by <sup>210</sup>Po, Harrison et al.<sup>12</sup> concluded that 0.1–0.3 GBq or more absorbed into the blood of an adult male is likely to be fatal within 1 month. This range would correspond to an intake of 1–3 GBq or more, assuming 10% absorption to blood. Considering a poisoning by ingestion of 1 GBq of <sup>210</sup>Po, Fig. 2-SM shows that we can expect to find about 500 kBq/day in urinary excretion the first 10 days after intake, about 250 kBq/day between 10 and 20 days and about 150 kBq/day between 20 and 30 days. Assuming that about 2 ml of urine produced the stain in the underwear we tested, one could expect to have 1 kBq in the underwear in October 2004 if it was worn during the first 10 days after intake. This activity of 1 kBq would decay (T<sub>1/2</sub> = 138.4 d) to about 1.4 mBq by February 2012. Therefore, taking into account the large uncertainties inherent to this kind of model calculation it is reasonable to expect an activity in the order of magnitude of 1-10 mBq in February 2012 in a urine stain coming from a person that incorporated 1 GBq in October 2004.



**Figure 2-SM**. Modelisation of a daily urinary excretion after a single ingestion of 1 GBq of <sup>210</sup>Po. The systemic biokinetic model was coupled to the human alimentary tract model of ICRP 100 and implemented in the ECOLEGO model software. The fractional intestinal absorption  $f_1$  has been set to 0.1.

**Consistency with Acute Radiation Syndrome**. It can be argued that the clinical features are not consistent with ARS. However, symptoms of ARS reported in the literature refer almost exclusively to situations of acute whole-body external exposure. In a case of protracted internal exposure, the clinical features would depend on many parameters, mainly the type of radionuclide, its chemical form, the route of exposure (inhalation or ingestion) and the amount of activity intake. The Litvinenko case is the only known case of a lethal ingestion of <sup>210</sup>Po in a human. Regrettably this case has not been reported in the scientific literature and so the clinical features of his illness had to be studied from media reports.

It is also true that aging cannot completely explain the absence of myelosuppression. Nevertheless, since aging is associated with decreased bone marrow cellularity (i.e. increased adipocyte concentration), one could expect that less alpha-particle energy is deposited within active bone marrow. This results in a lower absorbed dose to active bone marrow and thus a potentially reduced myelosuppression in the elderly.

Also, it cannot be ruled out that the gastrointestinal syndrome would not predominate in a case of ingested <sup>210</sup>Po. Indeed, Harrison et al., [12] reported results from a rat study suggesting damage to the gut mucosa as a possible cause of death in a case of <sup>210</sup>Po ingestion. Along with this statement, Scott reported that in the event of a <sup>210</sup>Po-210 ingestion: "Death occurs via one of the two modes (among several possible modes) with the two lowest thresholds: hematopoietic and gastrointestinal".

Regarding the dose level, Harrison et al. [12] reported that: "It is possible, therefore, that gut doses may have been substantially underestimated by not taking account of retained <sup>210</sup>Po". Moreover, using the average absorbed dose within the organ to characterize the dose-response relationship and the LD50 is questionable because of the potentially high level of heterogeneity of dose deposition within the organ in case of an alpha-emitting radionuclide, where devastating effects are localized in the direct vicinity of the decaying atom. For instance, Hobbs et al. [13] showed that cell level-based dosimetry, unlike usual red marrow dosimetry, was able to explain the unexpected lower marrow toxicities observed in clinical studies with <sup>223</sup>Ra, another alpha-emitting radionuclide (but a bone-seeker unlike <sup>210</sup>Po) used to treat bone metastases.

Finally, it should be mentioned that estimates of lethal activity and organ doses found in the literature addressing the Litvinenko case are based on standard models mainly developed for assessing cancer risk. In case of a lethal intake, high levels of a protracted dose delivered to target organs may significantly modify the metabolism over time (biokinetics).

Consequently, it is not possible to conclude that the clinical features of Mr. Arafat were incompatible with <sup>210</sup>Po poisoning.

**Limitations of the study**. We cut out subsamples of small sizes (from 0.3 to 4 g) in items found in the bag for toxicological analyses. The nature of the subsamples and the origin of the items could not be fully controlled. Particularly, we cannot reject a possible voluntary mishandling of the samples between the time of Mr. Arafat's death and the time the bag was presented to us. In particular, the addition of <sup>210</sup>Po to specific places on the clothes after Mr. Arafat's death cannot be excluded. Nevertheless, <sup>210</sup>Po is difficult to acquire and calculating the accurate levels of activity that would be found on clothes as a consequence of a lethal ingestion occurring 8 years before is a task that would necessitate specific expert knowledge.

Concerning the hair samples whose origin was confirmed, it was not possible to determine whether the hairs had been lost before or after the onset of the first symptoms.

According to his medical records, Mr. Arafat was a non-smoker and the toxicological analyses performed at Percy hospital were negative for cotinine. However, even if the medical files could not be trusted on this particular point, we could take into account that tobacco smokers excrete more <sup>210</sup>Po than non-smokers. From our own routine measurements, a heavy smoker excretes typically 0.015 mBq/ml of urine whereas a non-smoker excretes about a third of this quantity (0.005 mBq/ml of urine). This would only account for a negligible proportion of the <sup>210</sup>Po that were measured on Mr. Arafat's belonging.

**Table 1**. Activity (mBq) of <sup>210</sup>Po measured on subsamples of items taken from Mr. Arafat's belongings, IRA collaborators and local shops. Red colour indicates samples with a potential contamination with an unexplained level of <sup>210</sup>Po, green colour indicates samples with background levels of <sup>210</sup>Po and yellow colour shows an intermediate situation. The reported uncertainties are expanded with a coverage factor k=2.

Mr Arafat belongings181±8 $75\pm3$ 42Underwear A, urine stain, subsample 1 $60\pm3$ $6.2\pm1.0$ 10Russian chapka, subsample 1 $34.5\pm2$ $4.6\pm0.6$ 13Toothbrush bristles $211\pm1.7$ $4.4\pm0.6$ 21	
Underwear A, urine stain, subsample 1 $181\pm 8$ $75\pm 3$ $42$ Underwear A, urine stain, subsample 5 $60\pm 3$ $6.2\pm 1.0$ $10$ Russian chapka, subsample 1 $34.5\pm 2$ $4.6\pm 0.6$ $13$ Toothbrush bristles $211\pm 1.7$ $4.4\pm 0.6$ $21$	
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Russian chapka, subsample 1 $34.5\pm 2$ $4.6\pm 0.6$ $13$ Toothbrush bristles $21.1\pm 1.7$ $4.4\pm 0.6$ $21$	
Toothbrush bristles $21 + 17$ $44 + 0.6$ $21$	
Hospital cap with blood droplets, subsample 3 $16.1\pm1.0$ $0.8\pm0.4$ 5	
Hospital cap, blood stain, subsample 1 $13.5\pm1.0$ $1.3\pm0.4$ 10	
Russian chapka, subsample 4 $11.1\pm1.0$ $2.4\pm0.6$ 21	
Sportswear, collar, dirty, subsample 1 10.7±0.8 5.3±0.8 49	
Sportswear, collar, dirty, subsample 3 5.7±0.6	
Sportswear, back, stain $5.1\pm0.6$	
Child drawing on textile, stains $5.1\pm0.5$ $1.5\pm0.4$ 29	
Sportswear, collar, dirty, subsample 4 $5.1\pm0.6$	
Socks, dirty 4.9±0.6	
Underwear A, subsample 4 4.6±0.7	
Old slipper, upper band, dirty $4.0\pm0.5$	
Headscarf (kefieh), stains 3.7±0.4	
Old slipper, internal, dirty 3.5±0.4	
Underwear B, subsample 2 3.4±0.4	
Underwear A, subsample 2 $3.0\pm0.4$	
Child drawing on textile, out of stains $2.7\pm0.4$	
Russian chapka, subsample 5 2.6±0.5	
Underwear A, subsample 6 $2.3\pm0.3$	
Underwear A, subsample 7 $2.3\pm0.3$	
Trousers, wool, probably not worn 2.1±0.4	
Underwear A, subsample 8 $2.0\pm0.3$	
Russian chapka, subsample 3 $1.9\pm0.4$	
Hospital cap, out of stains $1.8\pm0.3$	
Sportswear, collar, without cotton layer, subsample 2 $1.8\pm0.3$	
Underwear A, subsample 3 $1.4\pm0.3$	
Sportswear, trousers, front 1.3±0.12	
Long john, subsample 4 $1.3\pm0.4$	
Underwear B, subsample 3 $1.3\pm0.4$	
Underwear B, subsample 4 $1.3\pm0.6$	
Underwear B, subsample 1 $1.2\pm0.2$	
Long john, subsample 3 $0.8\pm0.2$	
Russian chapka, subsample 2 0.6±0.2	
Long john subsample 2 $0.5\pm0.2$	
Socks, not worn, still attached by cotton filament 0.5±0.2	
IRA collaborators and a local shop	
Underwear Zimmerli brand A 7.1±0.5	
Underwear collaborator A, subsample 1 3.6±0.6	
Underwear collaborator B, subsample 1 3.4±0.6	

	Underwear collaborator B, subsample 2	2.6±0.5		
	Underwear collaborator A, subsample 2	1.9±0.4		
	Underwear Zimmerli brand B	1.6±0.3		
	Underwear Hanro Brand	1.1±0.2		
IRA collaborators and a local	Toothbrush, bristles, collaborator D, used	0.33±0.20		
supermarket				
	Toothbrush, bristles, collaborator C, used	0.32±0.11		
	Toothbrush, bristles, collaborator A, used and left untouched for 2 years	0.22±0.10		
	Toothbrush, bristles, collaborator B, used	0.19±0.10		
	Toothbrush, local supermarket, unused (new)	0.10±0.09		
IRA collaborator C, kept 10 years in an				
attic (3 <sup>rd</sup> floor) in a plastic bag				
	Shirt, back	2.2±0.4		
	Shirt, front	1.6±0.3		
	T-shirt, back	1.6±0.4		
	T-shirt, front	1.5±0.4		
	T-shirt, sleeve	1.4±0.3		
	Shirt, sleeve	1.3±0.3		
IRA collaborators D and E, kept 20	Children's clothes	1.5±0.3		
years in an attic in a plastic bag				
	Children's clothes	2.0±0.4		
	Children's clothes	2.0±0.4		
	Children's clothes	0.7±0.2		
	Children's clothes	0.6±0.2		
Specific bag of underwear bought new				
from a local supermarket				
-	Underwear A, subsample 1	20.1±1.4	5.7±0.8	28
	Underwear B, subsample 1	13.3±1.4		
	Underwear B, subsample 2	9.8±1.8		
	Underwear A, subsample 2	4.7±0.6		
	Underwear B, subsample 3	4.3±1.2		
	Underwear B, subsample 4	4.2±1.0		
	Underwear B, subsample 5	3.1±0.6		
	Underwear B, subsample 6	2.6±0.9		
IRA, shelves products blank	Products blank 1	1.9±0.3		
-	Products blank 2	$1.4{\pm}1.4$		
	Products blank 3	0.43±0.40		
	Products blank 4	0.16±0.07		
	Products blank 5	0.25±0.10		
	Products blank 6	0.20±0.10		

#### References

- 1. Castella V, Dimo-Simonin N, Brandt-Casadevall C, Mangin P. Forensic evaluation of the QIAshredder/QIAamp DNA extraction. *Forensic Sci. Int.* 2006; **156**: 70-73.
- 2. Castella V, Dimo-Simonin N, Brandt-Casadeval, C, Robinson N, Saugy M, Taroni, F, Mangin P. Forensic identification of urine samples: a comparison between nuclear and mitochondrial DNA markers. *Int. J. Legal Med.*, 2006; **120**: 67-72.
- 3. Maurer HH. Position of chromatographic techniques in screening for detection of drugs or poisons in clinical and forensic toxicology and/or doping control, *Clin. Chem. Lab. Med.* 2004; **42**: 1310-24.
- 4. Schrag B, Uldin T, Mangin P, Froidevaux P. Dating human skeletal remains using a radiometric method: biogenic versus diagenetic <sup>90</sup>Sr and <sup>210</sup>Pb in vertebrae. *Forensic Sci. Int.* 2012; **220**: 271-8.
- PROCORAD. Association pour la Promotion du Contrôle de Qualité des Analyses de Biologie Médicale en Radiotoxicologie. Radiotoxicological intercomparison exercices. <sup>210</sup>Po in urine. 2008-2011. www.procorad.org.
- ICRP. Human alimentary tract model for radiological protection: ICRP Publication 100. Ann. ICRP. 36 (1/2). Oxford: Elsevier, 2006. <u>http://dx.doi.org/10.1016/j.icrp.2006.03.001</u>
- ECOLEGO A toolbox for radio ecological risk assessment, Proceedings of the International Conference on the Protection from the Effects of Ionizing Radiation'', IAEA-CN-109/80. Stockholm: International Atomic Energy Agency. 229 – 232. <u>http://ecolego.facilia.se/ecolego/show/HomePage</u>.
- 8. Boryło A, Skwarzec B. Bioaccumulation of polonium (<sup>210</sup>Po) and uranium (<sup>234</sup>U, <sup>238</sup>U) in plants around phosphogypsum waste heap in Wiślinka (northern Poland). *Radiochim. Acta* 2011; **99**: 719–31.
- 9. Al-Marsi MS, Al-Hamwi A, Eadan, Z, Amin Y. Transfer factors of Polonium from soil to parsley and mint. *J. Environ. Radioact.* 2010; **101**: 1038-42.
- Vandenhove H, Olyslaegers G, Sanzharova N, Shubina O, Reed E, Shang Z, Velasco H. Proposal for new best estimates of the soil-to-plant transfer factor of U, Th, Ra, Pb and Po. *J. Environ. Radioact.* 2009; **100**: 721–32.
- 11. Leggett RW, Eckerman KF A. systemic biokinetic model for polonium. *Sci. Total Environ*.2001; **275:** 109-25.
- 12. Harrison J, Leggett R, Lloyd D, Phipps A, Scott B. Polonium-210 as a poison. J. Radiol Prot. 2007; 27: 17-40.
- Hobbs RF, Song H, Watchman CJ, Bolch WE, Aksnes AK, Ramdahl T, Flux GD, Sgouros G., A bone marrow toxicity model for 223-Ra alpha-emitter radiopharmaceutical therapy, *Phys. Med. Biol.* 2012; 57:3207-3222.