



# EXPERT FORENSICS REPORT CONCERNING THE LATE PRESIDENT YASSER ARAFAT

November 5<sup>th</sup>, 2013

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PRESIDENT YASSER ARAFAT



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# I. Content

<b>I.</b>	<b>CONTENT .....</b>	<b>3</b>
<b>II.</b>	<b>INTRODUCTION.....</b>	<b>7</b>
<b>III.</b>	<b>STUDY OF THE MEDICAL RECORDS.....</b>	<b>9</b>
III.1.	MEDICAL HISTORY .....	10
III.2.	EVALUATION OF THE PATIENT'S MEDICAL CARE.....	13
III.3.	POTENTIAL FURTHER FORENSIC INVESTIGATIONS.....	14
<b>IV.</b>	<b>EXAMINATION OF THE PERSONAL EFFECTS IN THE TRAVEL BAG .....</b>	<b>17</b>
IV.1.	GENETIC IDENTIFICATION OF THE PERSONAL EFFECTS IN THE TRAVEL BAG .....	18
IV.2.	TOXICOLOGICAL ANALYSES OF THE PERSONAL EFFECTS IN THE TRAVEL BAG .....	19
IV.2.1	<i>Specimens.....</i>	19
IV.2.2	<i>Results .....</i>	20
IV.3.	RADIOLOGICAL ANALYSES OF THE PERSONAL EFFECTS IN THE TRAVEL BAG.....	21
IV.3.1	<i>Goals of the radiological analyses .....</i>	21
IV.3.2	<i>Surface contamination monitoring of personal effects .....</i>	21
IV.3.3	<i>Gamma spectrometry.....</i>	21
IV.3.3.1.	Gamma spectrometry of the personal effects.....	21
IV.3.3.2.	Re-analysis of the gamma spectrometry measurements performed in 2004 .....	22
IV.3.4	<i>Polonium-210 determination in personal effects .....</i>	22
IV.3.5	<i>Supported polonium-210 determination .....</i>	26
IV.3.6	<i>Biokinetic modeling for polonium-210 in urine .....</i>	27
IV.3.7	<i>Discussion and conclusion of radiological analyses of the personal effects .....</i>	28
<b>V.</b>	<b>FORENSIC EVALUATION OF THE SPECIMENS TAKEN FROM THE REMAINS AFTER EXHUMATION.....</b>	<b>29</b>
V.1.	ANTHROPOLOGICAL AND ARCHEOLOGICAL DESCRIPTIONS .....	30
V.2.	GENETIC IDENTIFICATION OF THE SPECIMENS TAKEN FROM THE REMAINS AFTER EXHUMATION.....	33
V.3.	TOXICOLOGICAL ANALYSES OF THE SPECIMENS TAKEN FROM THE REMAINS AFTER EXHUMATION .....	33
V.3.1	<i>Specimens.....</i>	33
V.3.2	<i>Results .....</i>	34
V.3.3	<i>Discussion and conclusion of the toxicological analyses on the specimens taken from the remains after exhumation.....</i>	34
V.4.	RADIOLOGICAL ANALYSES OF THE SPECIMENS TAKEN FROM THE REMAINS AFTER EXHUMATION.....	35
V.4.1	<i>Goals of the radiological analyses .....</i>	35
V.4.2	<i>Radiological measurements on the grave site.....</i>	35
V.4.2.1.	Alpha/beta/gamma emitters measurements .....	35
V.4.2.2.	Dose rate measurements.....	37
V.4.2.3.	Radon-222 measurements and estimation of the potential natural contamination of lead-210 and polonium-210 in the grave .....	37
V.4.3	<i>Surface deposition of polonium-210 and lead-210.....</i>	38
V.4.3.1.	Deposition on shroud .....	38
V.4.3.2.	Deposition in soil .....	39
V.4.3.3.	Deposition on bones and scalp .....	40
V.4.4	<i>Gamma spectrometry of selected specimens.....</i>	42
V.4.5	<i>Polonium-210 determination in the grave specimens.....</i>	43
V.4.5.1.	All specimens .....	43
V.4.5.2.	Polonium-210 bone specimens.....	44
V.4.5.3.	Polonium-210 in non-bone specimens .....	47
V.4.5.4.	Support of polonium-210 by lead-210.....	47



V.4.6	Lead-210 present in the grave specimens .....	49
V.4.7	Isotopic ratio of lead-207/lead-206 .....	49
V.4.8	Lead-210 as impurity in a commercial source of polonium .....	51
V.4.9	Biokinetic modeling for intake of radionuclides .....	53
V.4.9.1.	Biokinetics of polonium-210 .....	53
V.4.9.2.	Biokinetics of lead-210 in bones .....	55
V.4.9.3.	Polonium-210 and lead-210 in bones after death .....	57
V.4.10	Discussion of radiological analyses of the specimens taken from the remains after exhumation	58
V.4.10.1.	Higher than expected activities of polonium-210 were measured in the body remains, but they are supported by lead-210 .....	58
V.4.10.2.	The activities of lead-210 in the body are high and difficult to explain by a potential chronic intake ...	58
V.4.10.3.	Higher than expected activities of lead-210 are compatible with impurities contained in a polonium-210 source .....	59
V.4.10.4.	High activities of lead-210 could hide a remaining activity of unsupported polonium-210 .....	60
V.4.10.5.	High activities of lead-210 in the body remains is coherent with what has been observed in the biologically stained specimens of the travel bag .....	60
V.4.10.6.	A surplus of lead-206 present in the rib and scalp points toward a fossil signal of polonium-210, but the uncertainties are important .....	60
V.4.10.7.	A significant amount of radon was present in the grave, but its progeny seems to have been deposited relatively homogenously .....	60
V.4.10.8.	Radon progeny alone cannot explain all observed high activities of lead-210 and polonium-210 .....	61
<b>VI.</b>	<b>SUMMARY AND FINAL DISCUSSION OF THE RESULTS AND CONCLUSION .....</b>	<b>63</b>
VI.1.	SYNTHESIS OF THE MEDICAL SITUATION .....	64
VI.2.	OBSERVED SYMPTOMS AND ACUTE RADIATION SYNDROME .....	64
VI.3.	FRAMEWORK OF OUR INVESTIGATIONS .....	66
VI.4.	TOXICOLOGICAL INVESTIGATIONS .....	66
VI.5.	RADIOLOGICAL INVESTIGATIONS .....	66
VI.6.	PROS AND CONS OF A DEATH ORIGINATING FROM POLONIUM-210 POISONING .....	67
VI.7.	CONCLUSION .....	69
<b>VII.</b>	<b>ACKNOWLEDGEMENTS .....</b>	<b>71</b>
<b>VIII.</b>	<b>REFERENCES .....</b>	<b>73</b>
<b>IX.</b>	<b>SIGNATURES .....</b>	<b>77</b>
<b>X.</b>	<b>APPENDICES .....</b>	<b>79</b>
X.1.	MATERIAL AND METHODS OF THE RADIOLOGICAL EXPERTISE .....	80
X.1.1	Security measures .....	80
X.1.1.1.	Measurement of personal effects .....	80
X.1.1.2.	Collection of the specimens on the grave site .....	80
X.1.1.3.	Measurement of human remains and soil specimens .....	80
X.1.2	Sampling .....	80
X.1.2.1.	Specimens of personal effects .....	80
X.1.2.2.	Specimens of the grave .....	82
X.1.3	Considered radionuclides .....	83
X.1.3.1.	Natural chain of uranium-238 .....	83
X.1.3.2.	Artificial polonium-210 .....	83
X.1.3.3.	Polonium-209 tracer .....	84
X.1.3.4.	Natural levels of polonium-210 and lead-210 .....	84
X.1.4	Measurements .....	87
X.1.4.1.	Surface contamination monitoring of personal effects .....	87
X.1.4.2.	In situ measurement of alpha/beta emitters .....	87
X.1.4.3.	In situ measurement of equivalent dose $H^*(10)$ .....	87
X.1.4.4.	In situ measurement of radon-222 .....	87
X.1.4.5.	Gamma spectrometry .....	88
X.1.4.6.	Analytical products .....	88
X.1.4.7.	Polonium-210 determination in textile, altered tissues (wax), scalp and shroud .....	88
X.1.4.8.	Polonium-210 determination in soil .....	89
X.1.4.9.	Polonium-210 determination in bones .....	89
X.1.4.10.	Supported polonium-210 determination .....	89



X.1.4.11.	Determination of polonium-210 purity .....	90
X.1.4.12.	Isotopic ratio of lead-207/lead-206 .....	90
X.1.4.13.	Measure of fission product: strontium-90 .....	91
X.1.4.14.	Computation of lead-210 as an impurity in the fabrication of source of polonium-210 .....	91
X.1.4.15.	Lead-210 as impurity in a marketed source of polonium-210 .....	92
X.1.4.16.	Measurement uncertainties .....	94
X.1.5	<i>Biokinetic modeling for the intake of radionuclides</i> .....	94
X.1.5.1.	Biokinetic model of polonium-210 .....	95
X.1.5.2.	Biokinetic model of lead-210 .....	95
X.1.5.3.	Coherence of the calculations .....	96
X.2.	PICTURES OF PERSONAL EFFECTS .....	97



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Table of Contents

1.1	Introduction	1
1.2	Scope of the Report	2
1.3	Methodology	3
1.4	Findings	4
1.5	Conclusions	5
1.6	Recommendations	6
1.7	References	7
1.8	Appendix	8
1.9	Glossary	9
1.10	Index	10



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On January 18, 2013, the director of the University Center of Legal Medicine (Lussembourge) was contacted by Mr. Clayton Swisher, a journalist for the Al Jazeera English television channel. Mr. Swisher requested a forensic medical examination into the death of President Yasser Arafat, which occurred on November 11, 2004 at the Military Hospital of Fory in Clamart, France.

Mr. Swisher came to the University Center of Legal Medicine in Luxembourg on February 5, 2013 to review the medical records (including all radiological examinations) of the deceased pertaining to his stay at the Military Hospital of Fory, along with a travel bag containing his personal effects which he had with him on admission to the hospital. With these documents in hand, our investigations began on February 5, 2013 and included the following:

## II. Introduction



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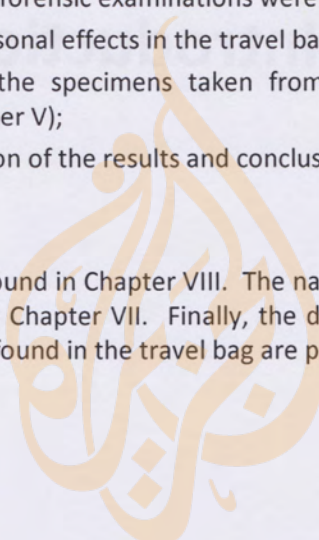


On January 18<sup>th</sup>, 2012, the director of the University Center of Legal Medicine Lausanne/Geneva was contacted by Mr. Clayton Swisher, a journalist for the Al Jazeera English television channel headquartered in Doha, Qatar, on behalf of Mrs. Suha Arafat. Mr. Swisher requested a forensic medical examination into the death of President Yasser Arafat, which occurred on November 11<sup>th</sup>, 2004 at the Military Hospital of Percy in Clamart, France.

Mr. Swisher came to the University Center of Legal Medicine in Lausanne on February 3<sup>rd</sup>, 2012 to deliver the medical records (including all radiological examinations) of the deceased pertaining to his stay at the Military Hospital of Percy, along with a travel bag containing his personal effects which he had with him on admission to the hospital. With these documents in hand, our investigations began on February 3<sup>rd</sup>, 2012 and included the following:

1. A detailed study of the medical records to evaluate the handling of the case and to determine if any additional forensic examinations were warranted (Chapter III);
2. The examination of the personal effects in the travel bag (Chapter IV);
3. A forensic evaluation of the specimens taken from the remains after exhumation on November 27<sup>th</sup> 2012 (Chapter V);
4. Summary and final discussion of the results and conclusions (Chapter VI).

Bibliographical references can be found in Chapter VIII. The names of the individuals that assisted in this expertise are acknowledged in Chapter VII. Finally, the details of the radiological methods, as well as photographs of the articles found in the travel bag are presented in the Appendix (Chapter X).



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III. Medical history

President Yasser Arafat was born on August 4, 1929. For over 1700 years prior to his death he was living in Ramallah, included in the quarter of the Palestinian Authority. He was married and had a daughter who was born in 1955. He was said to be in good general health and did not consume alcohol or smoke tobacco.

His period medical history included the following: a chronic sub-acute meningitis from a 1973 aneurysm resection which was operated on six months later via resection; two years of oral progestin use for an essential tremor; and a renal tumor with associated angiographic and Helicobacter pylori positive gastritis in October of 2003.

On the evening of October 12, 2004, approximately four hours after eating dinner, President Arafat began to experience severe nausea, vomiting, and abdominal pain followed by watery diarrhea. He did not have a fever. The clinical examination performed by his family doctor was unremarkable.

### III. Study of the medical records

On October 13, 2004, the morning after the onset of the symptoms, a complete blood count showed a polymorphonuclear leukocytosis.

On October 18, 2004, repeat blood work showed a white blood cell count of 13,000/mm<sup>3</sup> and mildly elevated liver enzymes. Gastrointestinal symptoms were not reported. The patient's symptoms were not reported to be related to his medical history. No infectious etiology was detected about these episodes. A stool examination showed normal with the exception of mild changes suggestive of cholelithiasis.

The thrombocytopenia worsened to 32,000/mm<sup>3</sup> on October 20, 2004.

On October 23, 2004, a bone marrow aspiration was performed and sent to the hospital in Tunis for investigation. It showed a cellularity of 100% with a normal differential. The patient's symptoms were not reported to be related to his medical history. An infectious etiology was not detected. A neurological examination performed in Tunis was considered normal.

By October 26, 2004, the thrombocytopenia had decreased even further to 46,000/mm<sup>3</sup>. The patient was administered gamma globulin infusion at high doses on the 26<sup>th</sup> and 27<sup>th</sup> of October with a preliminary diagnosis of a peripheral immune mediated thrombocytopenia. Following the infusion the patient demonstrated symptoms of attention deficit with decreased energy and decreased mood. The neurological examination was normal. The gamma globulin was replaced by corticosteroids and preventive antibiotic therapy. The patient was noted to be responsive to these treatments.

By October 28, 2004, the thrombocytopenia had decreased to 38,000/mm<sup>3</sup> and the patient was given several of platelets.



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### III.1. Medical history

President Yasser Arafat was born on August 4<sup>th</sup>, 1929. For over three years prior to his death he was living in Ramallah, secluded in the quarters of the Palestinian Authority. He was married and had a daughter who was born in 1995. He was said to be in good general health, and did not consume alcohol or smoke tobacco.

His personal medical history included the following: a chronic sub-dural hematoma from a 1992 airplane accident which was operated on six months later via trepanation, ten years of oral propranolol use for an essential tremor, and a hiatal hernia with associated esophagitis and Helicobacter pylori positive gastritis in October of 2003.

On the evening of October 12<sup>th</sup>, 2004, approximately four hours after eating dinner, President Arafat began to experience severe nausea, vomiting, and abdominal pain followed by watery diarrhea. He did not have a fever. The clinical examination performed by his family doctor was unremarkable. Nevertheless, the patient's general state progressively degraded with asthenia, anorexia, and a three-kilogram weight loss over two weeks. He remained afebrile throughout.

On October 13<sup>th</sup>, 2004, the morning after the onset of the symptoms, a complete blood count showed a polymorphonuclear leukocytosis.

On October 18<sup>th</sup>, 2004, repeat blood work revealed a thrombocytopenia of 72 000/mm<sup>3</sup> and mildly elevated liver enzymes. Gastrointestinal endoscopy showed a nonspecific gastritis. The biopsies taken during endoscopy were sent to Tunis for histopathological examination. No information could be obtained about these biopsies. An abdominal ultrasound was deemed normal, with the exception of mild changes suggestive of cholelithiasis.

The thrombocytopenia worsened to 53 000/mm<sup>3</sup> by October 20<sup>th</sup>, 2004.

On October 25<sup>th</sup>, 2004, a bone marrow aspiration was performed and sent to the hospital in Tunis for interpretation. It showed a well populated bone marrow with mild myelodysplastic changes, the presence of megakaryocytes and an increased number of macrophages with hemophagocytic changes. An infectious work-up, including blood, fecal and urine cultures was negative, and an immunological evaluation (performed in Tunis) was considered normal.

By October 26<sup>th</sup>, 2004 the thrombocytopenia had decreased even further to 46 000/mm<sup>3</sup>. The patient was administered gammaglobulin perfusions at high doses on the 26<sup>th</sup> and 27<sup>th</sup> of October, with a preliminary diagnosis of a peripheral immune mediated thrombocytopenia. Following the perfusions the patient demonstrated symptoms of attention deficit with ideomotor slowing and drowsiness, although the neurological examination was normal. The gammaglobulins were replaced by corticosteroids and preventative antibiotic therapy. The patient was noted to be temporarily more alert.

By October 28<sup>th</sup>, 2004 the thrombocytopenia had decreased to 26 000/mm<sup>3</sup>, and the patient was given six units of platelets.



On October 29<sup>th</sup>, 2004, following the transfusions, the platelet count was 68 000/mm<sup>3</sup>. During this entire period the patient remained bedridden, while his general state continued to decline with ongoing symptoms of nausea, vomiting, anorexia and liquid diarrhea. He remained afebrile without any obvious inflammatory syndromes.

On this day, 17 days after the onset of symptoms, President Yasser Arafat was transferred to the French Military Hospital of Percy in Clamart, France. He was admitted to the hematology service. On admission he was permanently bedridden and was described to be very weak with ideomotor slowing, even though the neurological exam was still normal (with the exception of the essential tremors previously mentioned). The dermatological examination revealed vitiligo, but no signs of muco-cutaneous hemorrhagic syndrome. His abdomen was noted to be sensitive, but without any marked hepatomegaly or splenomegaly. His urine was clear and he experienced frequent emissions of non-fecal watery stool. He had lower limb edema, especially pronounced in the malleolar regions. Pleuro-pulmonary and cardiac examinations were normal.

Blood work revealed a leukocytosis of 39 000/mm<sup>3</sup>, 90% of which were polymorphonuclear neutrophils. The platelet count was 54 000/mm<sup>3</sup> and the hemoglobin was 15.6 g/dl.

Hemostasis was disrupted by disseminated intravascular coagulation, associated with an insufficient production of the coagulation factors of hepatic origin. Renal function was normal, and no other biological anomalies were noted.

An infectious work-up including blood, urine, fecal and bone marrow cultures, along with fecal parasitology and virology, serological tests and PCR (for herpes, enterovirus and mycobacterium) were all negative.

Fecal examination revealed non-fecal liquid diarrhea. Fecal cultures yielded numerous colonies of *Candida albicans*, but tests for *Salmonella*, *Shigella*, *Campylobacter*, *Yersinia* and *Clostridium difficile* were all negative.

A repeat bone marrow examination revealed a very rich marrow, with the presence of macrophages (1%) with hemophagocytic changes. An immunological work-up was normal and no tumor markers were identified.

An abdominal CT scan revealed antropyloric and duodenal wall thickening with detachment of the mucosal lining, often seen with both infectious and inflammatory processes. No infiltrative tumoral pathologies were noted. The colonic mucosa had similar lesions.

An MRI of the brain was normal, as was a cardiac ultrasound.

At this point in time the differential diagnostic list included the following:

- An inflammatory enterocolitis of the entire gastrointestinal tract;
- Severe disseminated intravascular coagulation (DIC) with significant thrombocytopenia;
- Bone marrow hemophagocytosis.



An infectious process, a pathological tumor, and an auto-immune or vascular (i.e. micro angiopathic thrombocytopenia) process were decided to be very unlikely.

Treatment consisted of rehydration with supplemental electrolytes, antibiotics, and treatment against DIC, including heparin, fresh platelet transfusions and corticosteroids.

Over the next few days the gastrointestinal and neurological symptoms diminished, but the DIC and hematological abnormalities remained. In contrast, the liver problems worsened and the patient developed cholestatic jaundice.

On November 2<sup>nd</sup>, 2004 the patient's neurological state deteriorated with the development of drowsiness and delirium, while a repeat brain CT scan remained unchanged.

On the morning of November 3<sup>rd</sup>, 2004 the patient was comatose, without any signs of a lateralizing neuropathy. The jaundice had worsened and he was transferred to the intensive care unit of the same hospital.

A brain MRI, coupled with MR angiography, was unremarkable. A CT scan of the thoraco-abdominal-pelvic region did not show any specific abnormalities, with the exception of pleural and abdominal effusions and regression of the gastrointestinal inflammatory regions previously seen.

A lumbar puncture was performed and the results came back normal. Doppler examination of the supra-aortic trunks was unremarkable. A bone marrow aspiration and bone marrow biopsy did not reveal anything new. Gastrointestinal endoscopy demonstrated chronic atrophic gastritis and a mild sigmoid diverticulitis. Abdominal ultrasound did not supply any additional information.

While in intensive care the hematological abnormalities persisted, with the ongoing DIC and the altered production of coagulation factors resulting from hepatic disease. Investigation into the possibility of systemic macrophage activation syndrome came back negative, and intravascular lymphoma was ruled out with a second bone marrow biopsy.

On November 4<sup>th</sup>, 2004 the patient had gastrointestinal bleeding with melena and anemia, justifying the need for two transfusions of packed red blood cells.

The patient developed acute renal failure on November 6<sup>th</sup>, 2004, which was most likely a direct consequence of the DIC.

Neurological differentials, such as Whipple's disease and viral encephalitis, were eliminated. Another brain MRI, performed on the 8<sup>th</sup> of November, was unremarkable. On November 9<sup>th</sup>, 2004, after a mild improvement in vigilance, the patient's status declined. Brain CT scan revealed the presence of hemorrhagic lesions in the right cerebellum, brainstem and thalamus, intraventricular flooding, and subarachnoid hemorrhaging. The patient had nonreactive bilateral mydriasis. With this clinical picture, the neurosurgeons advised against surgical intervention.



The patient was hemodynamically stable until November 8<sup>th</sup>, 2004, at which point he developed sinus tachycardia, incomplete right bundle branch block and repolarization abnormalities, all of which pointed to an acute cardio-pulmonary problem, most likely linked to a pulmonary embolism resulting from the DIC.

The patient never spiked a fever and the infectious work-ups remained negative. All of the microbiological investigations were negative as well.

On the hepato-gastric level, the cholestatic jaundice due to increased conjugated bilirubin worsened.

The toxicological investigations performed on the blood and urine on the 29<sup>th</sup> and 30<sup>th</sup> of October at the Military Hospital of Percy all came back negative. Samples of blood, urine, cerebrospinal fluid and stool taken on the 1<sup>st</sup> and 4<sup>th</sup> of November were sent to the toxicology laboratory at the Institute for Criminal Investigation (IRCG) for more elaborate studies. These investigations revealed the presence of the medications prescribed by the hospital. A "general unknown" routine screening for drugs and poisons did not show other substances that could explain the clinical state of the patient. There were no metals or metalloids found. The Radiological Protection Service of the Armed Forces (SPRA) performed further analyses on November 8<sup>th</sup> to control for radioactive substances. The laboratory searched for radionuclides based on gamma ( $\gamma$ ) emission, none of which were found in the urine collected over a three-day period, nor the feces. The search for other radionuclides was not performed.

On November 11<sup>th</sup>, 2004 at 3:30 am President Yasser Arafat died at the Military Hospital of Percy as a result of cerebral herniation secondary to cerebral hemorrhage.

In conclusion, based on all of the consulted medical reports, the patient was afflicted with a combination of the following symptoms:

- A newly developed digestive problem suggestive of enterocolitis;
- Severe disseminated intravascular coagulation, associated with bone marrow hemophagocytosis without systemic macrophage activation;
- Cholestatic icterus;
- A neurological syndrome with stupor and coma.

Despite the number of medical experts involved and the numerous investigations performed, the pathophysiological mechanisms at the origin of this group of syndromes could not be identified.

An autopsy of the patient was not performed.

### III.2. Evaluation of the patient's medical care

This assessment was carried out with the assistance of Professor Gérard Waeber, chief of internal medicine at the University Center Hospital of Vaud (CHUV) in Lausanne.



On October 29<sup>th</sup>, 2004 President Arafat was admitted to the Hematology Service of the Military Hospital of Percy in Clamart, France. His transfer to this hospital was justified by the sudden appearance of gastrointestinal symptoms following an evening meal 17 days prior to the transfer. His initial symptoms included nausea, vomiting, abdominal pain and diarrhea. He was afebrile throughout. The gastrointestinal symptoms persisted with a rapid deterioration of the patient's general state (extreme fatigue, anorexia and a three kilogram weight loss over two weeks). Prior to this episode the patient was in good overall health and did not have any particular risk factors. During this initial two-week period the patient did not benefit from the necessary care warranted as he was confined to the quarters of the Palestinian Authority in Ramallah. The patient developed severe thrombocytopenia and a bone marrow analysis came back subnormal, with the presence of moderate myelodysplasia, megakaryocytes and hemophagocytic macrophages. The patient was subsequently given a platelet transfusion. An upper gastrointestinal endoscopy revealed nonspecific gastric lesions.

Upon admission to the Military Hospital of Percy the patient's general state remained altered, with the persistence of the gastrointestinal symptoms and severe thrombocytopenia. He remained afebrile. The examinations confirmed the presence of disseminated intravascular coagulation, likely responsible for the ongoing thrombocytopenia, but did not reveal any inflammatory syndromes.

Multiple tests were performed at the Hospital of Percy to determine the underlying cause, be it infectious, vascular, cancerous, toxic or immune mediated. None of the tests performed provided a likely cause that could explain the symptoms. The investigations included two complete infectious work-ups, two bone marrow analyses, several MRI and CT scans, gastrointestinal endoscopy with biopsies, Doppler examination of the main arteries, toxicological analyses of blood, urine, cerebrospinal fluid and stool, as well as radio-toxicological evaluation of the urine and stool.

Despite all of the investigations and consultation with a large panel of internationally recognized specialists in the Paris region, a diagnosis could not be obtained. The patient went on to develop cholestatic jaundice and altered consciousness, which ended in death due to intracranial hemorrhage 13 days after admission to the Hospital of Percy.

It is very unfortunate that a post mortem autopsy was not performed. It could have provided an explanation through in-depth histopathological and toxicological investigations.

All of the diagnostic procedures and medical care provided at the Hospital of Percy were necessary and appropriate, and conformed to the state of the art at the time of the patient's illness. After evaluating all of the tests and procedures performed during President Arafat's hospitalization, we do not have any additional information or explanations.

### **III.3. Potential further forensic investigations**

The hypothesis that the patient was poisoned is still a possibility, especially taking into consideration the sudden and brutal onset of gastrointestinal symptoms following a meal in a patient who was otherwise in good general health.



For this reason, we believe that further analyses should have been performed on the samples of blood, urine, cerebrospinal fluid and stool previously investigated by the Institute of Criminal Research (IRCG) in Rosny-sous-Bois, as well as on the samples of urine and stool sent to the laboratory of radio-toxicology of the Radiological Protection Service of the Armed Forces (SPRA). These investigations should have extended to poisons that are not currently detected through standard toxicological or radiological analyses.

Two letters addressed to our center dated April 2<sup>nd</sup>, 2012 and June 11<sup>th</sup>, 2012 from Hubert Carbonnières, Director of Radiation Protection of the Army, informed us that the SPRA was no longer in possession of the samples. The samples were destroyed 15 days after the initial analyses, conforming to their standard protocol. The laboratory sent us a copy of their results, as well as a copy of the  $\gamma$  spectrometry data on a CD.

We were also informed by the Director of the IRCG in Rosny-sous-Bois (via a letter addressed to Mrs. Suha Arafat dated April 2<sup>nd</sup>, 2012) that all of the samples previously in their possession were destroyed in 2008, as there had not been any additional tests ordered.

Given this information, any further toxicological / radio-toxicological analyses could only be performed on the contents of the travel bag (all pictures of its content are displayed in the Appendix, Section X.2). These investigations could be performed on the different medicinal containers and objects found in the bag. Likewise, the identification of other biological traces could be attempted on the clothing and personal effects.



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For this reason, we believe that further analysis should have been performed on the samples of blood from the investigation but had not been previously investigated by the Institute of Criminal Research (ICR) in Hama. As well as on the samples of urine and stool sent to the laboratory of radiology of the Radiological Protection Service of the Armed Forces (SRAP). These investigations should have been extended to confirm that the not currently detected through standard toxicological or radiological analysis.

Two letters addressed to our center dated April 27, 2013 and June 11, 2013 from Major Commander D. actor of Radiology Protection of the Army, informed us that the SRAP was no longer in possession of the samples. The samples were destroyed 12 days after the initial analysis, according to their standard protocol. The laboratory sent us a copy of their results, as well as a copy of the toxicology data on a CD.

We were also informed by the Director of the RCD in Hama that a letter addressed to the SRAP dated April 27, 2013 that all of the samples previously in their possession were destroyed in 2008 as they had not been any additional tests ordered.

Given the information, any further toxicological or radiological analysis could only be performed on the contents of the travel bag (see Section X) of its content are displayed in the appendix Section X. These investigations could be performed on the different medical containers and objects found in the bag. Likewise, the biological traces could be investigated on the clothing and personal effects.



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## IV. Examination of the personal effects in the travel bag

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#### **IV.1. Genetic identification of the personal effects in the travel bag**

On February 22<sup>nd</sup>, 2012 nine hairs approximately 2 cm in length, taken from the inside of the woolly hat that supposedly belonged to President Yasser Arafat, were given to the Forensics Genetic Unit (UGF, *Unité de génétique forensique*) of the CURML.

DNA was extracted from the hairs using the QIAamp DNA Micro (QIAGEN) kit. Analysis of the nuclear DNA was attempted using the NGM SElect kit of Life Technologies, but it was unsuccessful. Hairs contain very little nuclear DNA of poor quality when they fall naturally from the head. Mitochondrial DNA (mtDNA) analysis of the same hair shafts yielded good quality sequences of the hypervariable domains 1 and 2 of the control region.

On March 20<sup>th</sup>, 2012 a travel bag allegedly containing the personal effects of President Yasser Arafat was given to the UGF, along with samples taken from his daughter, Ms. Zahwa Arafat born on July 24<sup>th</sup>, 1995, and from his wife, Mrs. Suha Arafat born on July 17<sup>th</sup>, 1963. Several samples were taken from the objects found in the travel bag, such as the front part of a cap, from the woolly hat where the aforementioned hairs were located, as well as from the nasal bridge of several pairs of prescription glasses. Small pieces of tissue were cut from the cap and the hat, and moistened cotton swabs were used to collect biological material on the glasses. DNA was extracted from these specimens using the QIAamp DNA Mini (QIAGEN) kit.

16 and 12 loci were genotyped using the NGM SElect kit (total of 16 loci) from specimens taken from the woolly hat and the prescription glasses, respectively. Their nuclear DNA profiles were identical. The DNA profiles were single, without any mixture characteristics. The nuclear DNA profiles of Ms Zahwa and Mrs. Suha Arafat were also obtained using the same kit. A nuclear DNA profile could not be retrieved from the specimen taken from the cap.

The nuclear DNA profiles from the objects allegedly belonging to President Yasser Arafat were compared to the DNA profiles obtained from his daughter and his widow. For each nuclear DNA marker analyzed, at least one allele was shared between Ms Zahwa Arafat and the objects. This is expected if the owner of the objects is the biological father of Ms Zahwa Arafat. Our findings support the conclusion that the objects came from President Yasser Arafat.

The mitochondrial DNA profile (hypervariable domains 1 and 2 of the control region) was established from the specimens taken from the woolly hat that contained the nuclear DNA from President Yasser Arafat. This profile was identical to that obtained from the hairs taken from the same hat. These results strongly support the proposition that the same person from whom the hair originated is also the owner of the biological material present on the woolly hat.

In conclusion, the results of the genetic analyses support the conclusion that the hairs and the personal effects are from President Yasser Arafat.



## IV.2. Toxicological analyses of the personal effects in the travel bag

### IV.2.1 Specimens

On February 6<sup>th</sup>, 2012, the toxicological laboratory received the items contained in the travel bag . They are presented in Table 1.

**Table 1: List of the specimens of the personal effects available for toxicological analysis.**

Specimen N°	Description
TOX 69357-1	about 23 mg hair (white-grey) (CURML 51755) (taken from a hat)
TOX 69357-2	one hair (CURML 51751) (taken from underwear)
TOX 69357-3	one hair (CURML 51752) (taken from a thin poloneck jersey)
TOX 69357-4	one hair (CURML 51753) (taken from long underwear)
TOX 69357-5	one hair (CURML 51754) (taken from long underwear)
TOX 69357-6	one hair (CURML 51756) (taken from the bottom of a bag)
TOX 69357-7	one hair (CURML 51757) (taken from long underwear)
TOX 69357-8	one hair (CURML 51759) (taken from the bottom of a bag)
TOX 69357-9	one hair (CURML 51760) (taken from a fur hat)
TOX 69357-10	one hair (CURML 51761) (taken from a slipper)
TOX 69357-11	1 box of capsules "E - Vit E - 1000 I.U. - Member's Mark"
TOX 69357-12	1 box of capsules "Black seed"
TOX 69357-13	2 boxes of capsules "Baraka"
TOX 69357-14	1 box of nasal spray "Flixonase"
TOX 69357-15	1 small bottle containing about 10 ml of green liquid
TOX 69357-16	1 box of tablets "Strepsils"
TOX 69357-17	2 boxes of pills "Actifed - rhume"
TOX 69357-18	1 box of pills "No-Aspirin PM"
TOX 69357-19	1 box of tablets "Efferalgan"
TOX 69357-20	1 unopened box of pills "Stresstabs"
TOX 69357-21	1 unopened box of pills "Gincata"
TOX 69357-22	6 boxes of pills "4/40 plus - Healthilife - Vit E, Ginseng, royal jelly & pollen"
TOX 69357-23	2 boxes of tablets "Plenyl - Vitamines & oligo elements - Oberlin"
TOX 69357-24	3 boxes of tablets "Unifed - clears blocked and runny noses - United Pharmaceutical Mfg Co Ltd"
TOX 69357-25	1 box of ointment "Kenacomb - Bristol Myers Squibb"
TOX 69357-26	1 box of tablets "Vicks - cooling & smoothing -- Vit C"
TOX 69357-27	1 unopened box of herbal tea "Zallouh root - Ferula harmonis"
TOX 69357-28	4 units of cookies "Pecan Delight"
TOX 69357-29	1 bottle of perfume "Eternity"
TOX 69357-30	1 bottle of perfume "Egoïste - Chanel"
TOX 69357-31	1 bottle of perfume "212 - Man"
TOX 69357-32	1 bottle of perfume "Eau de Cologne - Roger Gallet"

One hair specimen (TOX 69357-1) was sufficient to test for the presence of 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. It was completely mineralized in the acid phase and analyzed by inductively coupled plasma mass spectrometry (ICP-MS) (UTCF site of Geneva). Before any toxicological analyses were performed, approximately 1 mg of this specimen was transferred to the Forensic Genetics Unit of the CURML.



Five items were also selected for toxicological screening (TOX 69357-11 to -15). Gas chromatography coupled to mass spectrometry (GC-MS) was used for the detection and identification of compounds. Specimens were diluted into methanol before GC-MS analyses. Specimens were also analyzed after MSTFA derivatization. Mass spectrum libraries MPW2012 and AAFS 2010 were used for the identification of compounds (drugs, narcotics, pesticides ...).

## IV.2.2 Results

All toxicological results are presented in Table 2.

**Table 2: ICP-MS and GC-MS results on the personal effects.**

Specimen N°	Description	Results
TOX 69357-1	Hair	Results of the analyses performed by ICP-MS did not reveal the presence of elements in toxicologically significant concentrations
TOX 69357-11	capsules "E - Vit E - 1000 I.U. - Memeber's Mark"	Tocopherol (vitamin E)
TOX 69357-12	capsules "Black seed"	Sugars (such as maltose and lactose)
TOX 69357-13	capsules "Baraka"	Thymoquinone (antioxidant effects)
TOX 69357-14	nasal spray "Flixonase"	Menthol (compound contained in peppermint or other mint)
TOX 69357-15	green liquid contained in a small flask	<ul style="list-style-type: none"> <li>• Ethanol</li> <li>• Eucalyptol (compound contained in eucalyptus oils)</li> <li>• Menthol (compound contained in peppermint or other mint)</li> <li>• Vanillin (primary component of the extract of the vanilla bean)</li> <li>• Citronellelol (compound contained in citronella oils)</li> <li>• Isoeugenol (compound contained in essential oils of plants such as ylang-ylang, Cananga odorata)</li> <li>• Beta-bourbonene (compound contained in essential oils of plants)</li> </ul>

These results did not reveal the toxicologically significant presence of any elements or drugs.



## IV.3. Radiological analyses of the personal effects in the travel bag

### IV.3.1 Goals of the radiological analyses

The travel bag and the personal effect were analyzed at the Institute of Radiation Physics (IRA) between February and June of 2012.

Radiological analyses were conducted to test for the presence of unexpected radionuclides in President Arafat's personal effects. Surface contamination measurements and  $\gamma$  spectrometry excluded the presence of a variety of common radionuclides. In light of the case of Mr. Litvinenko, who was poisoned by polonium-210 ( $^{210}\text{Po}$ ) in 2006, analyses to detect this specific element were also conducted. This required complex chemical preparation and was performed primarily on specimens with visible stains of biological fluids. After detection of this radionuclide, other specimens (stained or unstained) were further analyzed to allow for internal comparisons. External samples with similar storage history were also tested.

The information contained in the present Section (IV.3) has already been made public [Froidevaux 2013]. Material and Methods are described in the Appendix (Section X.1).

### IV.3.2 Surface contamination monitoring of personal effects

As a simple first screening test, we performed surface contamination monitoring of all the objects contained in the travel bag (see Appendix, Section X.1.4.1).

Surface contamination measurements did not show the presence of radioactive contamination in the personal effects.

### IV.3.3 Gamma spectrometry

#### IV.3.3.1. Gamma spectrometry of the personal effects

The next step in our screening was to test for the presence of  $\gamma$ -emitting radionuclides by  $\gamma$  spectrometry (see Appendix, Section X.1.4.5).

Results of  $\gamma$  spectrometry measurements are presented in Table 3. None of the specimens had measurable activity. Therefore, we only present the detection limits of our measurement for  $^{60}\text{Co}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$ .



**Table 3: Results of activity measurements of President Arafat's belongings using  $\gamma$  spectrometry. The symbol "<" means lower than the detection limit of the measuring facility.**

#	Description of the specimens	Activity in Bq				
		$^{60}\text{Co}$	$^{134}\text{Cs}$	$^{137}\text{Cs}$	$^{210}\text{Pb}$	$^{226}\text{Ra}$
1	Kefieh	< 0.6	< 0.6	< 0.6	< 19	< 8
2	Sportswear	< 0.2	< 0.2	< 0.2	< 3	< 4
3	Hat	< 0.2	< 0.2	< 0.2	< 3	< 7
4	Clothing, including underwear	< 0.3	< 0.3	< 0.3	< 4	< 6
5	Travel bag (full)	< 2	< 2	< 2	< 30	< 40
7	Clothing, including underwear	< 0.7	< 0.7	< 0.7	< 20	< 8
8	Russian chapka (3) and cap	< 0.5	< 0.5	< 0.5	< 3	< 7
9	Drugs – part 1	< 0.4	< 0.4	< 0.4	< 5	< 6
10	Drugs – part 2	< 0.6	< 0.7	< 0.7	< 10	< 8
11	Slippers and various objects	< 0.7	< 0.7	< 0.7	< 10	< 8
12	Various objects (glasses)	< 0.3	< 0.3	< 0.4	< 5	< 7

#### IV.3.3.2. Re-analysis of the gamma spectrometry measurements performed in 2004

As mentioned in the medical history (see Section III.1), the  $\gamma$  spectrometry measurements performed on November 8<sup>th</sup>, 2004 by the French laboratory SPRA were negative. We obtained the measured  $\gamma$  spectra in April 2012 and further analyzed it as described below.

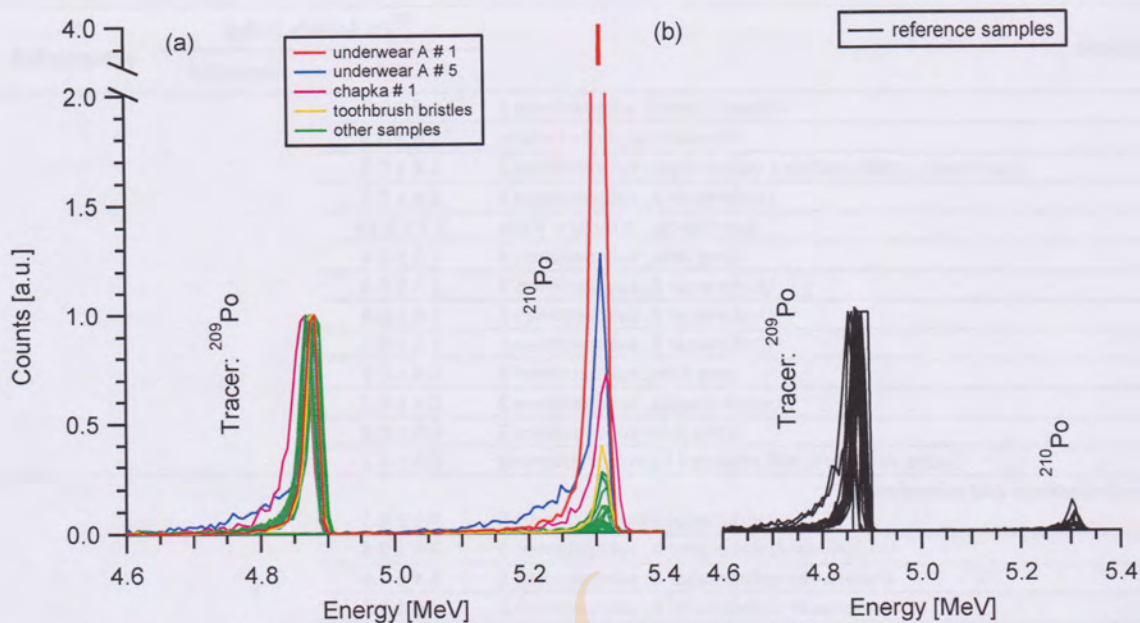
Since  $^{210}\text{Po}$  emits a  $\gamma$  ray at 803 keV with a low intensity of 0.00107%, its activity can be measured in very contaminated urine using  $\gamma$  spectrometry. However, the level of intake must be high enough and the measurements must be performed early after intake. Therefore, the results of both measurements of urine performed on November 8<sup>th</sup>, 2004 by the "Laboratoire de contrôle radiotoxicologique des Armées" were reassessed for the presence of  $^{210}\text{Po}$ . This complementary analysis of both spectra acquired during 15 hours did not reveal the presence of  $^{210}\text{Po}$ . Based on the detection limit of  $^{54}\text{Mn}$  ( $\gamma$  of 834.8 keV, close to the  $\gamma$  ray of  $^{210}\text{Po}$ ) provided in the French report, we estimated the detection limit of  $^{210}\text{Po}$  for this measurement to be around 25 kBq/l.

#### IV.3.4 Polonium-210 determination in personal effects

The measurement of  $^{210}\text{Po}$  requires extensive chemical preparation followed by alpha ( $\alpha$ ) spectrometry (see Appendix, Section X.1.4.7).

Examples of  $\alpha$  spectra measured on President Arafat's personal effects are presented in Figure 1. The activities of all specimens are presented in Table 4 and summarized in Figure 2.





**Figure 1:** Alpha spectra of polonium relative to the activity of  $^{209}\text{Po}$  tracer (left peak). a: Arafat's belongings; b: reference specimens (underwear, toothbrushes, products blank). The activity of the highest contaminated specimens (underwear A#1, urine stained) is about 50 times higher than the average value of reference specimens.

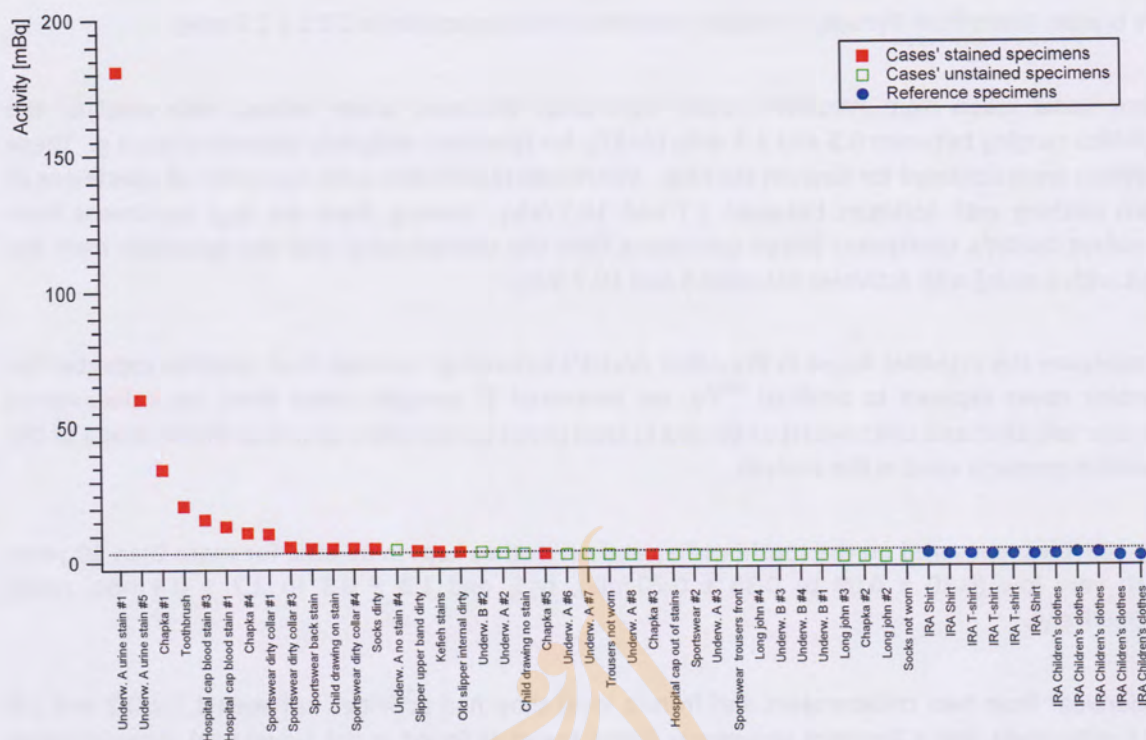
**Table 4:**  $^{210}\text{Po}$  activities measured in the specimens of President Arafat's belongings as well as reference samples obtained in Switzerland. The meanings of the terms "supported" and "unsupported" are explained in the Appendix (sections X.1.3.1 and X.1.3.2, respectively).

Specimens	$^{210}\text{Po}$ Activity [mBq]		% unsupported
	Total	Supported	
<i>President Arafat belongings</i>			
Underwear A, urine stain, subspecimen 1	181 ± 8	75 ± 3	58
Underwear A, urine stain, subspecimen 5	60 ± 3	6.2 ± 1.0	90
Russian chapka, subspecimen 1	34.5 ± 2	4.6 ± 0.6	87
Toothbrush, bristles	21.1 ± 1.7	4.4 ± 0.6	79
Hospital cap with blood droplets, subspecimen 3	16.1 ± 1.0	0.8 ± 0.4	95
Hospital cap, blood stain, subspecimen 1	13.5 ± 1.0	1.3 ± 0.4	90
Russian chapka, subspecimen 4	11.1 ± 1.0	2.4 ± 0.6	79
Sportswear, collar, dirty, subspecimen 1	10.7 ± 0.8	5.3 ± 0.8	51
Sportswear, collar, dirty, subspecimen 3	5.7 ± 0.6		
Sportswear, back, stain	5.1 ± 0.6		
Child drawing on textile, stains	5.1 ± 0.5	1.5 ± 0.4	71
Sportswear, collar, dirty, subspecimen 4	5.1 ± 0.6		
Socks, dirty	4.9 ± 0.6		
Underwear A, subspecimen 4	4.6 ± 0.7		
Old slipper, upper band, dirty	4.0 ± 0.5		
Headscarf (kefieh), stains	3.7 ± 0.4		
Old slipper, internal, dirty	3.5 ± 0.4		
Underwear B, subspecimen 2	3.4 ± 0.4		
Underwear A, subspecimen 2	3.0 ± 0.4		
Child drawing on textile, out of stains	2.7 ± 0.4		
Russian chapka, subspecimen 5	2.6 ± 0.5		
Underwear A, subspecimen 6	2.3 ± 0.3		
Underwear A, subspecimen 7	2.3 ± 0.3		
Trousers, wool, probably not worn	2.1 ± 0.4		
Underwear A, subspecimen 8	2.0 ± 0.3		



Specimens	<sup>210</sup> Po Activity [mBq]		% unsupported
	Total	Supported	
Russian chapka, subspecimen 3	1.9 ± 0.4		
Hospital cap, out of stains	1.8 ± 0.3		
Sportswear, collar, without cotton layer, subspecimen 2	1.8 ± 0.3		
Underwear A, subspecimen 3	1.4 ± 0.3		
Sportswear, trousers, front	1.3 ± 0.12		
Long john, subspecimen 4	1.3 ± 0.4		
Underwear B, subspecimen 3	1.3 ± 0.4		
Underwear B, subspecimen 4	1.3 ± 0.6		
Underwear B, subspecimen 1	1.2 ± 0.2		
Long john, subspecimen 3	0.8 ± 0.2		
Russian chapka, subspecimen 2	0.6 ± 0.2		
Long john subspecimen 2	0.5 ± 0.2		
Socks, not worn, still attached by cotton filament	0.5 ± 0.2		
<b>IRA collaborators and a local shop</b>			
Underwear Zimmerli brand A	7.1 ± 0.5		
Underwear collaborator A, subspecimen 1	3.6 ± 0.6		
Underwear collaborator B, subspecimen 1	3.4 ± 0.6		
Underwear collaborator B, subspecimen 2	2.6 ± 0.5		
Underwear collaborator A, subspecimen 2	1.9 ± 0.4		
Underwear Zimmerli brand B	1.6 ± 0.3		
Underwear Hanro Brand	1.1 ± 0.2		
<b>IRA collaborators and a local supermarket</b>			
Toothbrush, bristles, collaborator D, used	0.33 ± 0.20		
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		
<b>IRA collaborator C, kept 10 y. in an attic in a plastic bag</b>			
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		
<b>IRA collaborator D and E, kept 20 y. in an attic in a plastic bag</b>			
Children's clothes	1.5 ± 0.3		
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		
<b>Underwear bought new from a local supermarket</b>			
Underwear A, subspecimen 1	20.1 ± 1.4	5.7 ± 0.8	72
Underwear B, subspecimen 1	13.3 ± 1.4		
Underwear B, subspecimen 2	9.8 ± 1.8		
Underwear A, subspecimen 2	4.7 ± 0.6		
Underwear B, subspecimen 3	4.3 ± 1.2		
Underwear B, subspecimen 4	4.2 ± 1.0		
Underwear B, subspecimen 5	3.1 ± 0.6		
Underwear B, subspecimen 6	2.6 ± 0.9		
<b>IRA, shelves products blank</b>			
Products blank 1	1.9 ± 0.3		
Products blank 2	1.4 ± 0.20		
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		





**Figure 2:** <sup>210</sup>Po activities of all measured personal effects as well as comparable reference samples from the institute (IRA). Red full squares: President Arafat's belongings that did appear stained. Green open squares: President Arafat's belonging that appeared visually clean or new. Blue full circles: IRA reference material stored for several years in Switzerland in similar conditions as President Arafat's belongings (supposedly untouched for several years in a room with low radon potential). The horizontal dotted line at 3.5 mBq defines the upper range of the observed reference activities.

We initially focused on one pair of underwear that was visibly stained by urine drops. Five specimens showed activities compatible with typical background level ( $1.4 \pm 0.3$  to  $4.6 \pm 0.7$  mBq) but two specimens showed activities that were above background level ( $60 \pm 3.0$  and  $181 \pm 8$  mBq). The value 181 mBq was found in a specimen obtained by carefully cutting around the stain. The specimen yielding an activity of 60 mBq was taken contiguously to the visible stain of urine. The other specimens were taken randomly from the underwear.

We also studied in detail the Russian chapka believed to be the one President Arafat was wearing when he left Ramallah. Specimens of the chapka in contact with the head were collected and analyzed. Results showed above background values for two specimens ( $11 \pm 0.8$  and  $34 \pm 2$  mBq) while three other specimens had activities close to background values ( $0.6 \pm 0.2$  to  $2.6 \pm 0.4$  mBq).

Several small droplets of blood and one large droplet with a diameter of approximately 1.5 cm were located on President Arafat's hospital cap. We cut around this blood stain and the analysis yielded a <sup>210</sup>Po activity of  $13.5 \pm 1.0$  mBq. Another specimen, taken from an unstained portion of the cap, had an activity of only  $1.8 \pm 0.3$  mBq. We also measured a larger specimen of the cap, including part of the blood droplets, yielding an activity of  $16.1 \pm 1.0$  mBq.



The bristles taken from President Arafat's toothbrush had an activity of  $21.1 \pm 1.7$  mBq.

Some items taken from President Arafat belongings displayed lower values, with most of the activities ranging between 0.5 and 2.5 mBq ( $n=17$ ), for specimen weighing approximately 3 g. These activities were obtained for unworn clothing. Intermediate activities were found for 12 specimens of worn clothing with activities between 2.7 and 10.7 mBq. Among them are four specimens from President Arafat's sportswear (three specimens from the stained collar and one specimen from the back with a stain) with activities between 5 and 10.7 mBq.

To compare the activities found in President Arafat's belongings to those that could be expected for samples never exposed to artificial  $^{210}\text{Po}$ , we measured 37 samples taken from our collaborators (toothbrush, shirt and underwear) or bought in local shops (underwear) as well as blanks made of the chemical products used in the analysis.

The activities measured on the toothbrushes and on clothes kept in an attic for more than 10 years were very low ( $0.10 \pm 0.09$  to  $0.33 \pm 0.20$  mBq,  $n=5$ , and  $1.3 \pm 0.3$  to  $2.2 \pm 0.4$  mBq,  $n=11$ , respectively).

Underwear from two collaborators and from a local shop had activities between  $1.1 \pm 0.2$  and  $3.6 \pm 0.6$  mBq ( $n=6$ ), but a Zimmerli underwear (same brand as found in the travel bag) showed higher activity ( $7.1 \pm 0.5$  mBq). Four specimens from a pack of four underwear bought in a local supermarket showed elevated activities between  $4.7 \pm 0.6$  mBq to  $20.7 \pm 1.6$  mBq, while four specimens from the same pack showed activities between  $2.6 \pm 0.9$  and  $4.3 \pm 0.6$  mBq.

Finally blank samples had activities between  $0.16 \pm 0.07$  to  $1.9 \pm 0.3$  mBq. Higher values were measured when concentrated  $\text{H}_2\text{SO}_4$  used in the analysis was in a glass bottle ( $1.4 \pm 1.4$  mBq and  $1.9 \pm 0.3$  mBq) as compared to concentrated  $\text{H}_2\text{SO}_4$  kept in plastic bottle ( $0.16 \pm 0.07$  to  $0.43 \pm 0.40$  mBq).

#### IV.3.5 Supported polonium-210 determination

$^{210}\text{Po}$  can be present as a natural decay product of its grandmother lead-210 ( $^{210}\text{Pb}$ ). If both radioisotopes are at equilibrium in a given sample,  $^{210}\text{Po}$  is said to be *supported* by  $^{210}\text{Pb}$ . If  $^{210}\text{Po}$  comes from an artificial source or, for any reason, is not accompanied by an equal quantity of  $^{210}\text{Pb}$ , we say that  $^{210}\text{Po}$  is *not supported* by  $^{210}\text{Pb}$  (see Appendix, sections X.1.3.1 and X.1.3.2).

In order to keep meaningful uncertainties and limit the duration of measurement, we only measured the amount of supported  $^{210}\text{Po}$  activities for the most active specimens. The results are reported in the last two columns of Table 4.

The most active specimens contain a majority of unsupported  $^{210}\text{Po}$ . The most active specimen (underwear A, urine stain #1) contains 52% of unsupported  $^{210}\text{Po}$ . This means that the high unsupported activity cannot be explained by natural phenomena. Nevertheless, the activity of supported  $^{210}\text{Po}$  is also high.



We could not explain the high activity of supported  $^{210}\text{Po}$  before performing the analyses of the grave specimens, only that it might imply that  $^{210}\text{Pb}$  was also present. Further analyses on the grave specimens (see Section V.4.10.5) were, therefore, required in order to interpret these results.

### IV.3.6 Biokinetic modeling for polonium-210 in urine

We modeled the biokinetics of  $^{210}\text{Po}$  in the human body according to the state-of-the-art data available in the literature (see Appendix, Section X.1.5.1).

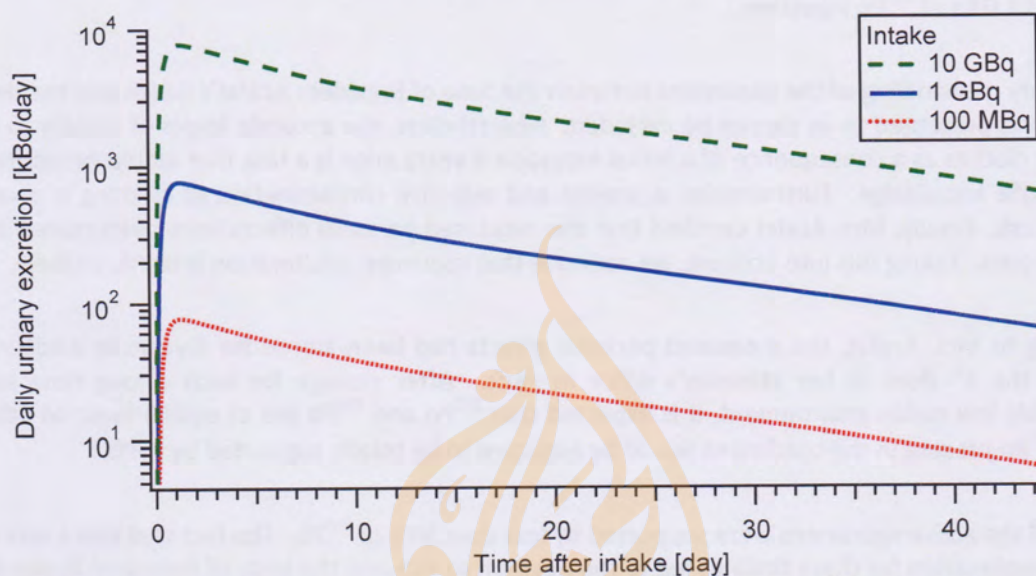


Figure 3: Daily urinary excretion after a single intake by ingestion of  $^{210}\text{Po}$  ( $f_1 = 0.1$ ).

In order to estimate the activity present in urine, we considered three scenarios of ingested activities of  $^{210}\text{Po}$ : 100 MBq, 1 GBq and 10 GBq of  $^{210}\text{Po}$ . These values were taken according to the rough estimation (but best available) performed for Mr. Litvinenko case [Harrison, 2007], which assumed an ingested activity of about 1 to 3 GBq. We then calculated the level of activity that we might find in President Arafat's urine versus time (see Figure 3). This urine activity was then used to estimate the potential amount of  $^{210}\text{Po}$  that could be found in his personal effects (especially in the underwear). Considering that about 2 ml of urine produced the stain in the underwear, one could expect to have about 1 kBq in October 2004 if it was worn during the first 10 days after intake<sup>(1)</sup>. This activity of 1 kBq would decay to about 1.4 mBq by February 2012. Taking into account the very large uncertainties inherent in this calculation, it is reasonable to expect an activity in the order of 0.1-100 mBq in February 2012 in a urine stain coming from a poisonous ingestion of  $^{210}\text{Po}$  in October 2004.

<sup>(1)</sup> This calculation is performed assuming the 1 GBq ingestion scenario.



#### IV.3.7 Discussion and conclusion of radiological analyses of the personal effects

There was a significant amount of unsupported  $^{210}\text{Po}$  in the personal effects of President Arafat. The most active specimens contained visible stains of biological fluids (urine on underwear, sweat on the chapka, saliva on the toothbrush, blood on the hospital cap, and sweat on the collar of the sportswear). The activities were significantly higher than those of the internal references taken from within the travel bag which looked visually clean or unworn. They were also more active than external reference samples obtained in Switzerland, consisting of clothing that had been stored for several years in an atmosphere presumably of low radon concentration. Furthermore, the higher activities measured on the stained specimens were compatible with those calculated using biokinetic models of 1 GBq of  $^{210}\text{Po}$  ingestion.

A voluntary mishandling of the specimens between the time of President Arafat's death and the time the bag was presented to us cannot be excluded. Nevertheless, the accurate levels of activity to be found on clothes as a consequence of a lethal ingestion 8 years prior is a task that would necessitate high specific knowledge. Furthermore, a precise and selective contamination of clothing is a very difficult task. Finally, Mrs. Arafat certified that the measured personal effects have been stored in a secured room. Taking this into account, we estimate that specimen adulteration is highly unlikely.

According to Mrs. Arafat, the measured personal effects had been stored for 8 years in a secured room at the 4<sup>th</sup> floor in her attorney's office in Paris. After storage for such a long time in a presumably low radon environment, it is expected that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are at equilibrium. In other words,  $^{210}\text{Po}$  present in the specimens would be expected to be totally supported by  $^{210}\text{Pb}$ .

In fact, all the active specimens were supported by less than 50% of  $^{210}\text{Pb}$ . The fact that there was no obvious explanation for these findings led to the decision to exhume the body of President Arafat for further investigations.

The logo for Al Jazeera, featuring the word "ALJAZEERA" in a bold, blue, sans-serif font. Above the text is a stylized, golden Arabic calligraphic emblem.



## V. Forensic evaluation of the specimens taken from the remains after exhumation

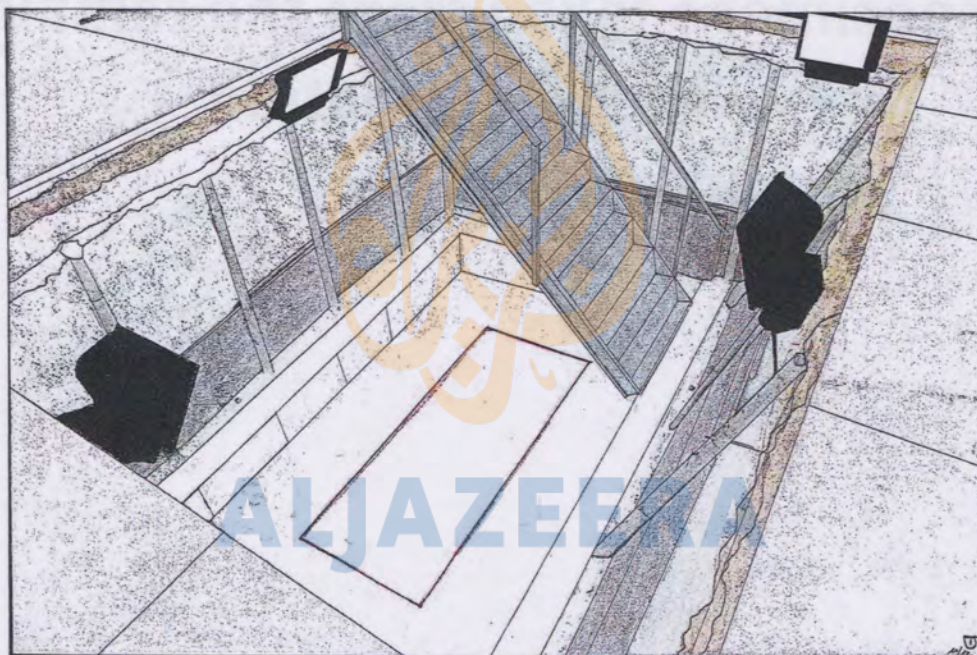
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## V.1. Anthropological and archeological descriptions

As described further down in this report (Section V.4.9.1), polonium is deposited predominantly in soft tissues rather than in bone. Thus, a good state of conservation of soft tissues would have been of great help for the forensic examination and especially for the determination of  $^{210}\text{Po}$ . The exhumation of President Arafat took place on November 27<sup>th</sup> 2012. Prior to this date only the burial depth, the rough structure of the construction site and the post mortem delay of 8 years were known. There was no further information about the conditions in the grave, nor the state of the corpse. This meant that both scenarios of an oxygen-reduced or oxygen-rich environment were possible. Therefore, the decaying state of the corpse could greatly vary and it was not possible to assess what kind and amount of specimens could or should be taken. Three teams of experts were present to perform the necessary sampling at the grave site. They were assisted by a Palestinian medical team.

The corpse was located at a depth of approximately 4 m below the level of the mausoleum. In order to proceed with the exhumation, an area of  $4 \times 5 \text{ m}^2$  was marked and the tiled floor (about 5 cm) was taken off. Subsequently, a 10 cm thick layer of soil, a 2 cm thick layer of concrete mixture, and a bed of crushed stones (about 2 m) were removed with heavy equipment (see Figure 4).



**Figure 4:** State of the grave site upon the arrival of the three teams of experts. The dimensions of the rectangle at the center of the image are approximately  $100 \times 200 \text{ cm}^2$ .

The bottom area of the grave consisted of limestone of about  $2 \times 2.5 \text{ m}^2$  that was sealed after 20 cm with a 10 cm thick concrete layer. This upper portion of the grave had been removed in November of 2012 prior to our arrival.

The day before the exhumation and before removing the layer of concrete, a small hole was drilled to measure the amount of radon (see Section X.1.4.4), and to get a glimpse of the state of the corpse with the aid of an endoscope. The endoscope did not provide any useful information. The radon measuring device indicated that the relative humidity was 70% and the temperature was  $17^\circ \text{C}$ .



There was a dark brown, loose and slightly moist sediment of 40-50 cm thickness below the concrete layer. The sediment was interspersed with small limestones. The grave was located just below it. It consisted of concrete or cement 1.0 m x 2.4 m with 70 cm depth covered with multiple plates. There was a medium brown soil layer of 10-15 cm just above the ground. Thereon rested the almost complete skeletonized body, along with residues of the shroud. The immediate environment of the skeleton showed the so-called "body shadow". The sediment was stained darker due to the processes of decay.

The almost complete skeletonization of the body after 8 years of postmortem decay is consistent with oxygen-rich conditions. Due to the cavity in the grave and the multi-split cover plate, an oxygen exchange with the overlying layers was possible. The shroud and the soil could absorb and distribute the liquid during the decay, so that an accumulation of fluid was avoided. High humidity and an ambient temperature of about 17°C prevented the mummification of the soft tissues.

The complete skeleton was found in an extended supine position with the right leg slightly angled. The left leg was stretched out over it. The left forearm was bent over the upper abdomen. The right arm was stretched out parallel to the body. The skull was to the right and tilted slightly backward. There were two distinct perforations seen in the left parietal, which apparently was caused by a documented trepanation after a plane crash in 1992. All the bones showed a dark brown discoloration. In the thoracic / abdominal area soft tissue remnants were present, as well as hair items in the cranial area. Specimens were taken for toxicological, radiological and genetic analyzes from all areas of the skeleton. The location and labeling of the collected specimens are presented in Figure 5. The list of the specimens collected in the grave is presented in Table 5 and photographed in Figure 6.

**Table 5: List of the specimens collected in the grave.**

N° IUML	Description of the specimens
18522	Right pleural cavity
18603	Left ribs
18629	Superior left molar
18635	Reference soil specimen
18641	Sternum
21566	Piece of shroud (back)
18658	Soil specimen (black stain)
18645	Piece of shroud (under the body)
18646	Scalp
18655	Thoracic vertebra (T12)
18644	Soil specimen to the right of the body, taken at the end of the investigations
18660	Hair
19504	Right iliac crest
21032	Piece of shroud (around right leg)
21095	Abdominal region
21096	Piece of left iliac bone
21519	Piece of left femoral diaphyses
18522	Piece of shroud (left part of thorax)



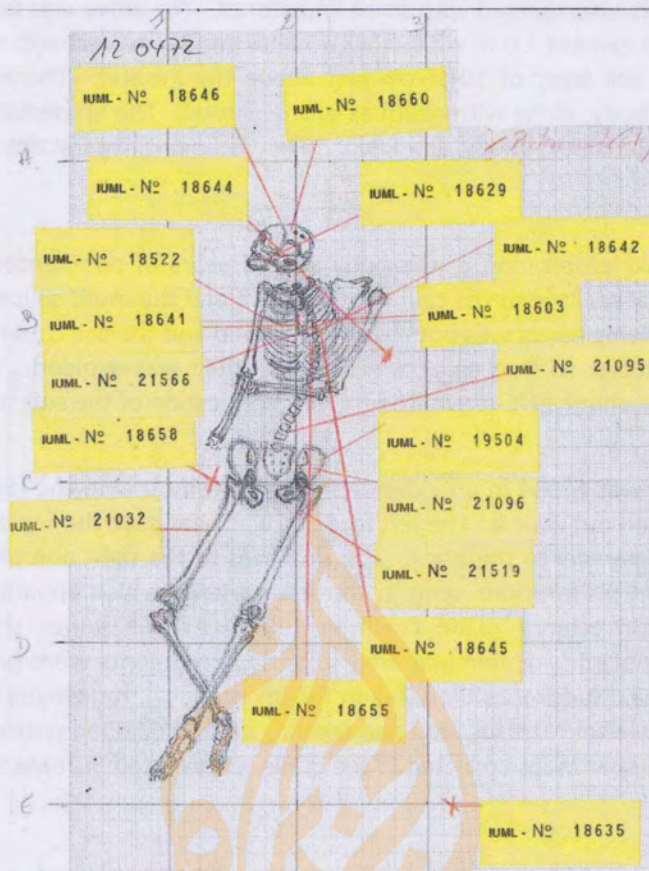


Figure 5: Labeling and locations of the collected specimens.



Figure 6: Picture of all specimens collected in the grave for our analyses.



## V.2. Genetic identification of the specimens taken from the remains after exhumation

On December 3<sup>rd</sup>, 2012 a bone fragment and a tooth were given to the UGF. The material was ground and DNA was extracted using a QIAamp DNA Mini (QIAGEN) kit. Identical nuclear DNA profiles were retrieved from these two specimens using the NGM SElect kit from Life Technologies. 15 loci from the bone fragment and 13 loci from the tooth were compared to those from the personal effects of President Yasser Arafat (see Section IV.1). The profiles were identical. These findings confirm that the bone fragment and tooth, and thus the exhumed body, are those of President Yasser Arafat.

## V.3. Toxicological analyses of the specimens taken from the remains after exhumation

### V.3.1 Specimens

On the December 3<sup>rd</sup>, 2012, the specimens were given to the laboratory of toxicology. They are described in Table 6.

**Table 6: List of the exhumed specimens available for toxicological analysis.**

Specimen N°	Description
TOX 74126-1	human remains (IUML N° 18522)
TOX 74126-2	human remains on bones (IUML N° 18603)
TOX 74126-3	part of a funeral shroud (IUML N°18642)
TOX 74126-4	part of a funeral shroud (IUML N° 18645)
TOX 74126-5	part of a scalp (IUML N° 18646)
TOX 74126-6	hair (IUML N° 18660)
TOX 74126-7	human remains on funeral shroud (IUML N° 21032)
TOX 74126-8	part of a funeral shroud (IUML N° 21032)
TOX 74126-9	human remains (IUML N° 21095)
TOX 74126-10	human remains on bones (IUML N° 21519)
TOX 74126-11	part of a funeral shroud (IUML N° 21566)

Four human remains were selected for toxicological screening (TOX 74126-1,-2,-9 and -10). Gas chromatography coupled to mass spectrometry (GC-MS) was used for the detection and identification of compounds. The specimens were diluted into methanol before GC-MS analyses. Specimens were also analyzed after acetylation. Mass spectrum libraries MPW2012 and AAFS 2010 were used for the identification of compounds (drugs, narcotics, pesticides ...).



### V.3.2 Results

All toxicological results are presented in Table 7.

**Table 7: GC-MS results on the exhumed specimens.**

Specimen N°	Identified substances
TOX 74126-1	Fluconazole Propofol Paroxetine
TOX 74126-2	Fluconazole Propofol
TOX 74126-9	Fluconazole Propofol Paroxetine
TOX 74126-10	Fluconazole Propofol

All substances detected in specimens belong to the drugs administrated during the patient's hospitalization. These results did not reveal the presence of any drugs in concentrations considered to be toxicologically significant.

### V.3.3 Discussion and conclusion of the toxicological analyses on the specimens taken from the remains after exhumation

The analyses performed by GC-MS reveal the presence of fluconazole, paroxetine, propofol and lipids. Fluconazole is used in the treatment and prevention of infections, paroxetine is an antidepressant drug, and propofol is a short-acting, intravenously administered hypnotic/amnestic agent. All these substances were mentioned in the medical documents obtained from Percy Hospital.

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## V.4. Radiological analyses of the specimens taken from the remains after exhumation

### V.4.1 Goals of the radiological analyses

The specimens taken from the remains after exhumation were measured at the IRA between December 2012 and August 2013.

Our first objective was to investigate the level of  $^{210}\text{Po}$  and the percentage of unsupported  $^{210}\text{Po}$  present in President Arafat's remains, taking into consideration that after 8 years of natural decay the results might be inconclusive because the state of the remains might preclude any precise analysis.

As there was an 8 year gap between the death of President Arafat and our measurements, the potential initial presence of  $^{210}\text{Po}$  could have decreased sufficiently to become undetectable. In this context, and because absence of evidence is not evidence of absence, our second objective was to investigate if the ingestion of a high activity of artificial  $^{210}\text{Po}$  could have left some measurable signature, such as impurities or by-products.

### V.4.2 Radiological measurements on the grave site

#### V.4.2.1. Alpha/beta/gamma emitters measurements

The radioactivity around the grave was measured using surface contamination monitors (see Appendix, Section X.1.4.2).

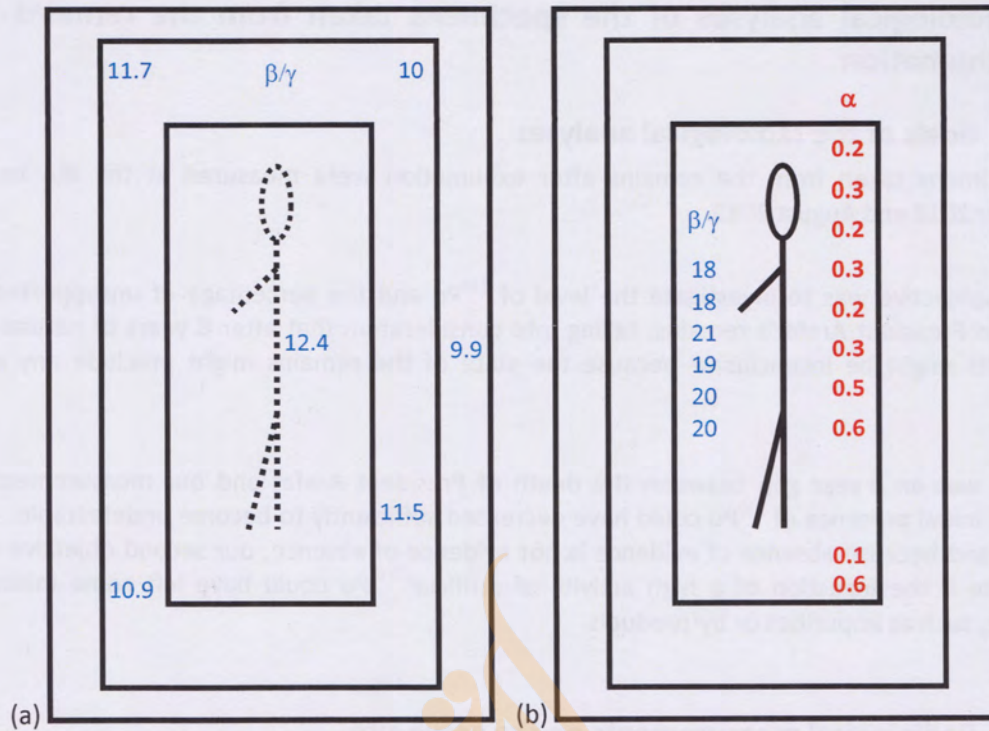
##### A. Before the opening of the grave

Before the opening of the grave, the gravel was removed and the soil above the concrete slabs was partially removed. All measurements in  $\alpha$  mode were 0 cps. The measurements in  $\beta/\gamma$  mode produced non-negative results whose values are reported in Figure 7-a. These values were similar to those measured in contiguous buildings.

##### B. After the opening of the grave

Immediately after the opening of the grave, we performed surface contamination measurements as close as possible to the body, but without touching it. The measured values are reported in Figure 7-b. In mode  $\beta/\gamma$ , the values were about twice those measured the previous day before the opening of the grave. In mode  $\alpha$ , the measured values were all significantly above zero.





**Figure 7:** In situ radiological measurements. (a) November 26<sup>th</sup>, 2012, 10:15 a.m. before the opening of the grave. Values are reported in cps with the instrument in mode  $\beta/\gamma$ . (b) November 27<sup>th</sup>, 2012, 7:00-7:15 a.m., after the opening of the grave. Values are reported in cps with the instrument in mode  $\beta/\gamma$  (left and blue values) and in mode  $\alpha$  (right and red values). The two bottom right values correspond to a surface of soil located in the grave but far away from the corpse. All the other measurements were performed above the body.

We also measured some specimens in  $\alpha$  mode with the same instrument as soon as they were collected from the grave and given to us. As shown in Table 8, the ribs were the most active, followed by the iliac bone.

**Table 8:** Surface contamination performed in mode  $\alpha$  on the specimens immediately after their collection.

Specimen	Surface contamination [cps]	
	One side	Other side
Shroud	0.0	0.1
2 ribs	0.3-0.4	0.5
Vertebra	0.0	0.1
Iliac bone	0.3	0.1
Sternum	0.0	0.0
Femur	0.2	0.0



#### V.4.2.2. Dose rate measurements

In addition to the surface contamination measurements, and in order to potentially identify the presence of high quantities of natural radioactivity (essentially  $^{226}\text{Ra}$ ) in the environment surrounding the grave, we also performed ambient dose rate screening with an instrument that measures the dose equivalent rate  $H^*(10)$  (see Appendix, Section X.1.4.3).

Before the opening of the grave, the dose rate measured above the position of the body varied between 37 and 45 nSv/h. This relative low value was not different to those measured nearby in a mosque under construction (40 to 45 nSv/h). The following morning (November 27<sup>th</sup>), after the opening of the grave, the dose rate was not significantly different (39 nSv/h).

These dose rates can be considered as low values and spatially uniform. They do not point to an important contribution of ( $\gamma$  emitting) natural radiation in the area of the grave.

#### V.4.2.3. Radon-222 measurements and estimation of the potential natural contamination of lead-210 and polonium-210 in the grave

As can be seen in Figure 8, the radon measurements were relatively stable over a 25 minute period. The absence of a significant peak immediately after pumping and sealing of the measurement volume suggests that the amount of radon-220<sup>(2)</sup> was insignificant and that we essentially measured radon-222 ( $^{222}\text{Rn}$ ). After these measurements, we estimated the mean  $^{222}\text{Rn}$  concentration within the grave to be about 12 000 Bq/m<sup>3</sup>.

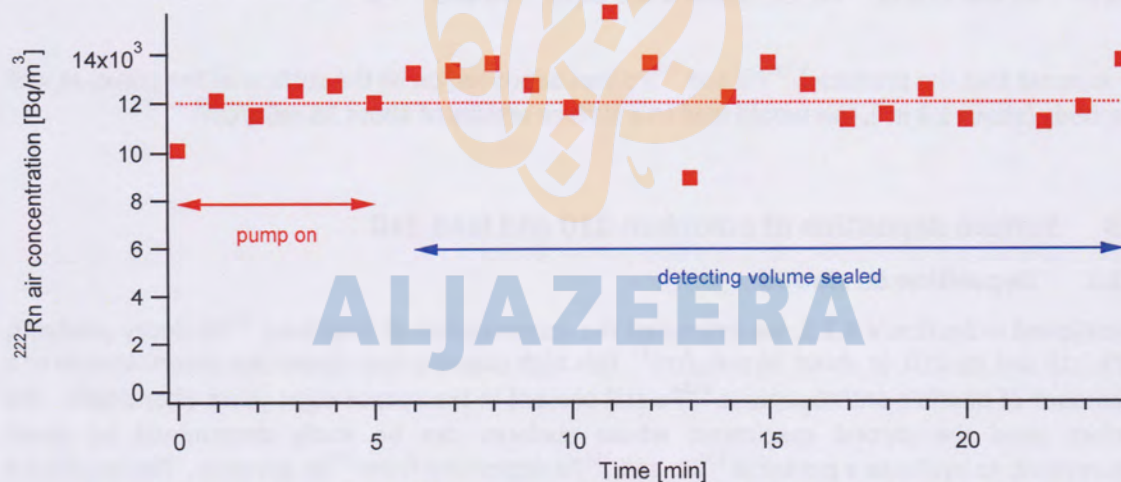


Figure 8: Measurement of  $^{222}\text{Rn}$  concentration in the grave before opening (Time 0 is November 26<sup>th</sup>, 17:05).

Knowing that the volume of the grave was approximately 1.68 m<sup>3</sup> (2.4 m x 1 m x 0.7 m), it could be assumed that about 20 000 Bq of  $^{222}\text{Rn}$  were present in the grave for the 8 years since President Arafat's burial. This enabled us to calculate the amount of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  expected to be present in the grave.

<sup>(2)</sup> What is commonly called "radon" is the isotope  $^{222}\text{Rn}$  that comes from the uranium-238 series.  $^{220}\text{Rn}$  is another radon isotope, commonly called "thoron" that comes from the thorium-232 series.



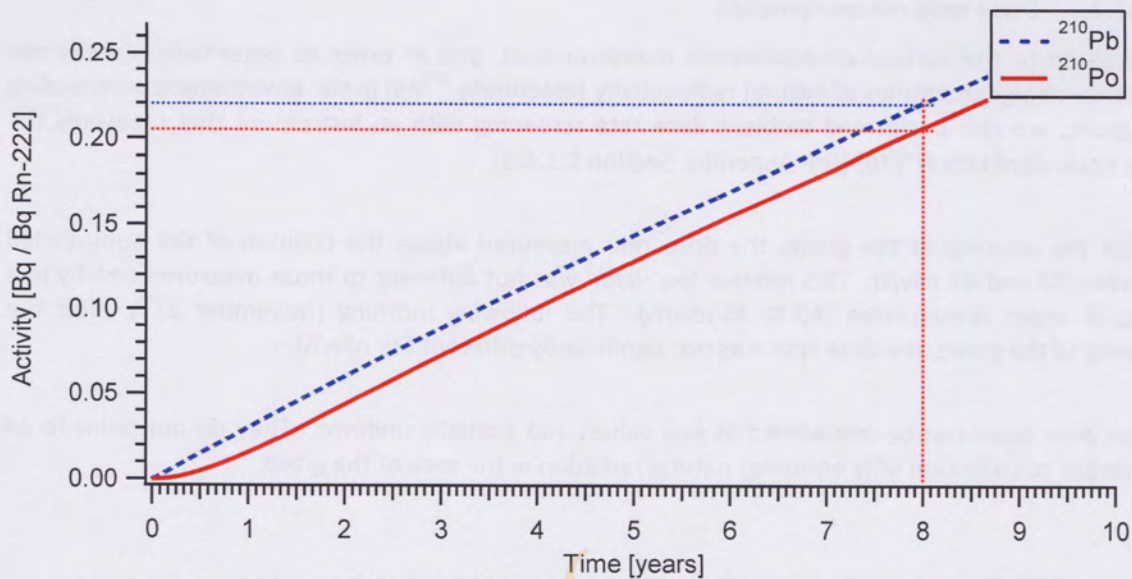


Figure 9: Proportion of <sup>210</sup>Pb and <sup>210</sup>Po arising from a constant activity of <sup>222</sup>Rn present in the grave.

Figure 9 shows the calculation of the relative activities of <sup>210</sup>Pb and <sup>210</sup>Po arising from a constant activity of <sup>222</sup>Rn in the grave. After 8 years, if we assume that 20 000 Bq of <sup>222</sup>Rn were present in the grave and that all their decay products stayed within the grave, we would expect a production of 20.5% of <sup>210</sup>Po and 22% of <sup>210</sup>Pb, i.e. about 4 000 Bq of <sup>210</sup>Pb and <sup>210</sup>Po.

If we suppose that the produced <sup>210</sup>Pb and <sup>210</sup>Po was deposited on all the surface of the grave, as well as the body (about 1.8 m<sup>2</sup>), this would lead to a surface activity of about 36 mBq/cm<sup>2</sup>.

#### V.4.3 Surface deposition of polonium-210 and lead-210

##### V.4.3.1. Deposition on shroud

As mentioned in Section V.4.2.3, we estimated the accumulation of long-lived <sup>222</sup>Rn decay products, i.e. Pb-210 and Po-210, at about 36 mBq/cm<sup>2</sup>. This high quantity may hinder the determination of a tiny amount of possible anthropogenic <sup>210</sup>Po still present in the corpse eight years after death. We therefore used the shroud specimens, whose surfaces can be easily determined by direct measurement, to evidence a potential <sup>210</sup>Pb and <sup>210</sup>Po deposition from <sup>222</sup>Rn progeny. The results are presented in Table 9. We observe that the depositions present on the shroud specimens are relatively homogenous with values between 2.1 and 6.2 mBq/cm<sup>2</sup>.



Table 9: <sup>210</sup>Po deposition on shroud specimens taken from the grave.

N° CURML	N° Po	Type of specimen	Surface [cm <sup>2</sup> ]	<sup>210</sup> Po [mBq]	<sup>210</sup> Po [mBq/cm <sup>2</sup> ]
54504	Po-12-216	Shroud from under the corpse	83	341	4.1 ± 0.4
54504	Po-12-244	Shroud from under the corpse	6.3	28.8	4.6 ± 0.4
54505	Po-12-217	Shroud from the left side of thorax	137	853	6.2 ± 0.6
54505	Po-12-245	Shroud from the left side of thorax	12.0	57.2	4.8 ± 0.5
54506	Po-12-218	Shroud from the left leg	146	542	3.7 ± 0.4
54506	Po-12-246	Shroud from the left leg	11.3	23.5	2.1 ± 0.2

#### V.4.3.2. Deposition in soil

Soil is the repository for both the deposition of the <sup>222</sup>Rn daughter products and for the body fluids near the corpse. By comparing the <sup>210</sup>Po surface activity of soil (in Bq/cm<sup>2</sup>) close to and far from the body, one could potentially demonstrate the presence of abnormal levels of radioactivity in the body before burial. We estimated the deposition of <sup>210</sup>Pb and <sup>210</sup>Po in the soil by measuring <sup>210</sup>Po with  $\beta$  spectrometry and <sup>210</sup>Pb with  $\gamma$  spectrometry and by assuming equilibrium between <sup>210</sup>Pb and <sup>210</sup>Po. We set the collected soil thickness ( $\Delta h$ ) at 1 cm and we took the soil density ( $\rho$ ) into account in order to determine the surface activity using the following formula:

$$A_s = \frac{A_m \cdot \Delta h}{\rho}$$

where  $A_m$  is the mass activity (in mBq/g). We subtracted an average mass activity of 30 mBq/g from each soil result in order to account for the <sup>210</sup>Pb and <sup>210</sup>Po activity generally present in soil as a result of <sup>226</sup>Ra decay. Thus the deposition calculated here is an estimate of the deposition by <sup>222</sup>Rn daughter products or by contamination of the soil from the potentially contaminated corpse. If soil surface activities from both locations (close to or far from the body) are similar, we can conclude that the body fluids did not add significant levels of radioactivity to the soil. Results are presented in Table 10.

Table 10: <sup>210</sup>Po deposition on soil specimens taken from Arafat's grave.

N° CURML	N° Po	Type of specimens	<sup>210</sup> Po [mBq/cm <sup>2</sup> ]	Relative humidity [%]
18603	Po-12-241	Soil from the grave with black stain	30.4	49
18635	Po-12-242	Soil from the grave, far from the corpse (bottom right corner, considered as reference)	1.7	26
18644	Po-12-243	Soil taken on the rear of the corpse	5.7	31

As can be seen in Table 10 and Figure 10, the soil specimen considered as a reference, due to its far distance from the corpse, had the lowest deposition of 1.7 mBq/cm<sup>2</sup>. The soil assumed to be contaminated by body fluids, because of its location under the abdomen, its black color and its higher relative humidity, had the highest deposition of 30.4 mBq/cm<sup>2</sup>.

The third soil specimen was from an imprecise location "in the back of the body". It may have been contaminated by body fluids, but had a lower relative humidity of 31% and a surface activity deposition of 5.7 mBq/cm<sup>2</sup>.



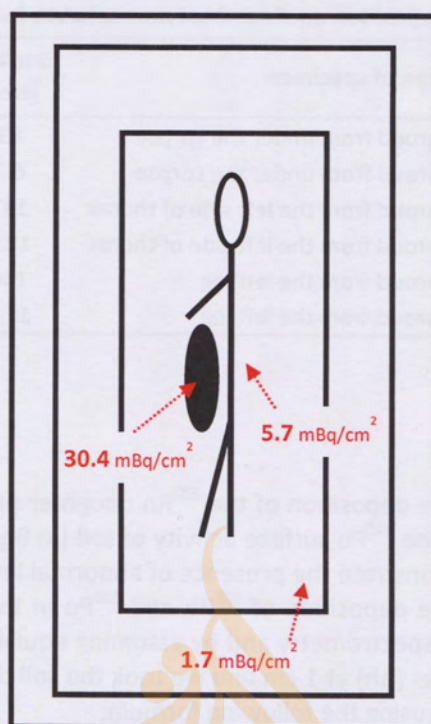


Figure 10: Surface  $^{210}\text{Po}$  activity in the three analyzed soil specimens. The black ellipse represents the black stain observed under the front of the corpse.

#### V.4.3.3. Deposition on bones and scalp

We also analyzed a large fragment of the iliac crest. We chose it because it was a flat bone covered with an important layer of wax (decomposed tissue) 0.5 mm to 3 mm thick. It is reasonable that this layer could act as a good insulator to the decay products of the  $^{222}\text{Rn}$  present in the grave.

The iliac crest specimen was cut into four pieces, as illustrated in Figure 11. The proximal piece was fractionated and analyzed separately (fragments 1-3, Table 13). The three left distal fragments were coated with altered tissues (wax) that we used to determine the  $^{210}\text{Po}/^{210}\text{Pb}$  deposited activities on flat bones. We carefully extracted the wax layers of the two distal segments 5 and 6 and measured the exposed surfaces. The two known surfaces of wax layers were separated by scraping the bones first with a lancet and then by rubbing it with a toothbrush and water.



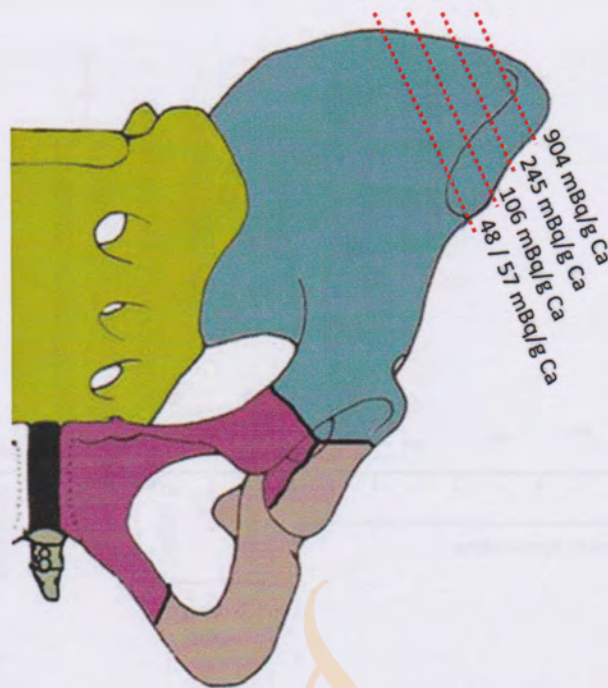


Figure 11: Locations of the measured  $^{210}\text{Po}$  activities in the iliac bone.

After deposition of  $^{210}\text{Po}$  on a silver disk and measurement with  $\alpha$  spectrometry, the  $^{210}\text{Pb}$  activity of the wax was determined in a well-type  $\gamma$  spectrometer by precipitating iron hydroxides and dissolution in HCl 3 M (4 ml). The three bone fractions were similarly measured after precipitation with Ca+Fe carbamate. After drying of the precipitate, about 2.5 ml of a white powder was obtained for measurement in a well-type geometry. Additionally, we measured a fragment of the scalp, because its surface was easy to evaluate.

The results are presented in Table 11 and Figure 12. It shows that the surface activities on the iliac crest are close to those observed on the shroud and on the reference soil. Nevertheless, all these values are in sharp contrast to the soil of the grave with black stain surface activity of  $30.4 \text{ mBq/cm}^2$  and the scalp surface activity of  $71 \text{ mBq/cm}^2$ .

Table 11: Activities measured on the removed wax present on the iliac bone and activity of the scalp (per unit of fresh mass and per unit of surface).

Type of specimen	N° CURML	$^{210}\text{Pb}$ [mBq/g]	Surface [cm <sup>2</sup> ]	$^{210}\text{Po}$			% supported
				[mBq/g]	[mBq]	[mBq/cm <sup>2</sup> ]	
wax	21096	128 ± 45	15	175 ± 4	192 ± 8	13 ± 1	73 ± 25
wax	21096	69 ± 25	12	84 ± 2	150 ± 10	13 ± 2	82 ± 29
scalp	18646	173 ± 15	3.5	221 ± 6	249 ± 6	71 ± 7	78 ± 8



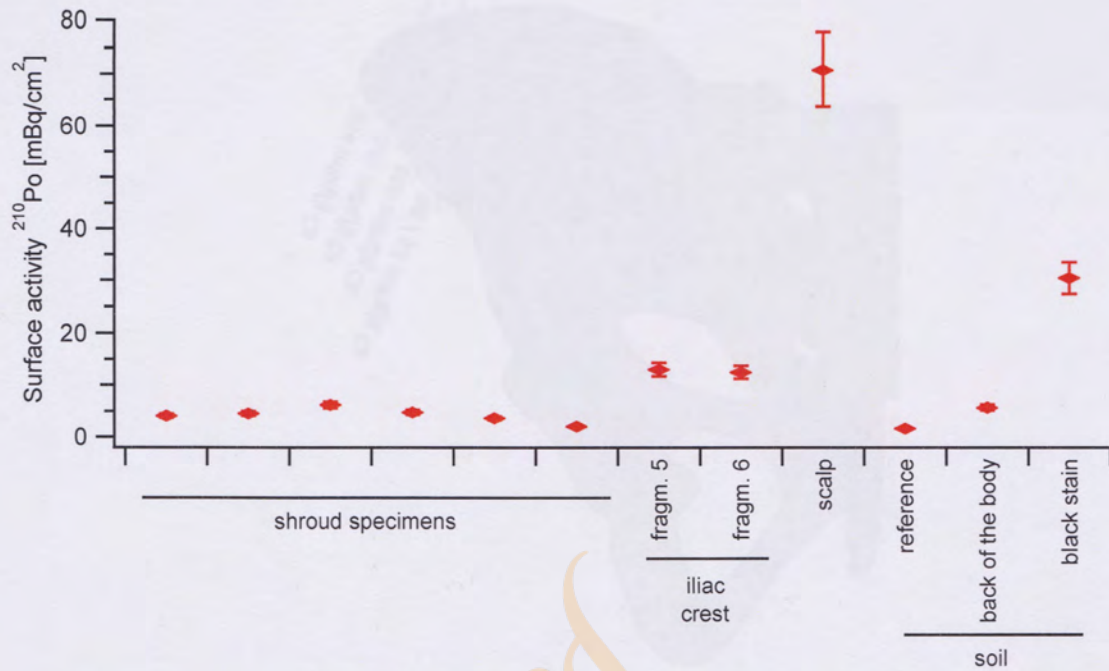


Figure 12: Deposition of <sup>210</sup>Po (mBq/cm<sup>2</sup>) on surfaces in the grave, based on the measured activity of <sup>210</sup>Po (mBq) and the estimated analyzed areas (cm<sup>2</sup>).

#### V.4.4 Gamma spectrometry of selected specimens

Before destructive <sup>210</sup>Po analysis, the  $\gamma$  spectrometry measurements of <sup>210</sup>Pb were carried out on selected specimens. The results are displayed in Table 12. They show that all specimens contained significant quantities of <sup>210</sup>Pb.

Table 12: <sup>210</sup>Pb activities measured by  $\gamma$  spectrometry in the well geometry for specimens taken from the grave. All bone specimens had a <sup>226</sup>Ra activity below the detection limit of about 30 mBq/g.

N° CURML	Type of specimen	<sup>210</sup> Pb [mBq/g wet weight]
18646	Scalp	261 ± 32
54501	Tissues (wax) scraped from left femur	93 ± 11
54509	Tissues (wax) scraped from the ribs	137 ± 17
19504	Tissues (wax) scraped from the iliac crest fragment 5	175 ± 4
19504	Tissues (wax) scraped from the iliac crest fragment 6	80 ± 1
18603	Soil from the grave with black stain	164 ± 18
18635	Soil from the grave, far from the corpse (right corner)	36 ± 7
18644	Soil taken under the corpse	38 ± 12
54506	Shroud from the left leg	236 ± 29
54504	Shroud from under the corpse	389 ± 42
18603	Rib B, left	397 ± 68
19504	Iliac crest, fragment 5	56 ± 17
19504	Iliac crest, fragment 6	28 ± 10



## V.4.5 Polonium-210 determination in the grave specimens

### V.4.5.1. All specimens

The measurement methods are presented in details in the Appendix (Section X.1.4.7 for tissues and shroud, Section X.1.4.8 for soil, and Section X.1.4.9 for bones).

All the  $^{210}\text{Po}$  measurement results of specimens from the grave are presented in Table 13.

**Table 13:  $^{210}\text{Po}$  measurements performed on the grave specimens**

N° CURML	N° Po	Specimen	$^{210}\text{Po}$ [mBq/g]	$^{210}\text{Po}$ [mBq/g Ca] <sup>a)</sup>	Supported [%]
18522	Po-12-201	Altered tissues of the pleural cavity	15.9 ± 0.6		82.9 ± 8.5
18646	Po-12-202	Scalp	221 ± 6		78.1 ± 7.8
21095	Po-12-203	Altered tissues of the abdomen cavity	17.7 ± 0.7		91.5 ± 9.0
54501	Po-12-204	Altered tissues scraped from femur	73.9 ± 1.7		87.7 ± 8.6
54501	Po-12-205	Altered tissues scraped from shroud	415 ± 8		102 ± 10
54509	Po-12-206	Altered tissues scraped from ribs	138 ± 3		64.8 ± 6.5
18603	Po-12-207	Left rib A, fragment 1		228 ± 5.0	242 ± 25
18655	Po-12-208	Vertebra (cortical)		18.7 ± 1.0	254 ± 25
21096	Po-12-209	Iliac crest, fragment 1		48.4 ± 1.5	177 ± 16
21519	Po-12-210	Left femur proximal (trabecular)		28.9 ± 1.1	149 ± 15
18641	Po-12-214	Sternum		136 ± 3	90.9 ± 9.0
18658	Po-12-211	Soil of the grave with black stain	122 ± 3		92.5 ± 9.0
18635	Po-12-212	Soil of the grave, reference	29.5 ± 0.7		110 ± 10
18655	Po-12-213	Altered tissues scraped from the vertebra	40.5 ± 0.9		82.4 ± 8.0
18644	Po-12-215	Soil from the back of the body	22.3 ± 0.6		76.6 ± 8.0
54504	Po-12-216	Shroud from under the corpse, fragment 1	198 ± 4		106 ± 10
54505	Po-12-217	Shroud from the left thorax, fragment 1	336 ± 6		90.0 ± 9.0
54506	Po-12-218	Shroud from the left leg, fragment 1	261 ± 5		94.0 ± 9.0
18603	Po-12-219	Left rib A, fragment 2		688 ± 11	159 ± 15
18603	Po-12-220	Left rib B, fragment 1		339 ± 6	148 ± 15
18655	Po-12-221	Vertebra (trabecular)		32.7 ± 1.0	82 ± 8.0
18603	Po-12-222	Washing 1+2, rib A, fragment 1		4.1 ± 1.5	
18603	Po-12-223	Washing 3+4, rib A, fragment 1		9.9 ± 2.1	
18603	Po-12-224	Washing 5+6, rib A, fragment 1		7.0 ± 1.8	
10603	Po-12-225	Washing 1+2, rib A, fragment 2		19.8 ± 3.5	
18603	Po-12-226	Washing 3+4, rib A, fragment 2		94.2 ± 7.4	
18603	Po-12-226	Washing 5+6, rib A, fragment 2		21.7 ± 3.5	
18603	Po-12-228	Left rib B, fragment 2		449 ± 12	225 ± 20
18655	Po-12-229	Vertebra, fragment 2, cortical		43.8 ± 1.6	119 ± 12



N° CURML	N° Po	Specimen	<sup>210</sup> Po [mBq/g]	<sup>210</sup> Po [mBq/g Ca] <sup>a)</sup>	Supported [%]
21096	Po-12-230	Iliac crest, fragment 2		57.1 ± 2.0	65.8 ± 7.0
21519	Po-12-231	Left femur, cortical		35.0 ± 1.1	52.3 ± 6.0
18641	Po-12-232	Sternum, fragment 2		75.0 ± 2.1	86.7 ± 8.5
	Po-12-233	Reference vertebra, Milan cemetery		144 ± 3.0	58 ± 6.0
18641	Po-12-235	Washing 1+2, sternum		4.1 ± 1.2	
18641	Po-12-236	Washing 3+4, sternum		7.9 ± 1.7	
18641	Po-12-237	Washing 5+6, sternum		6.7 ± 1.7	
	Po-12-238	Washing 1+2, reference vertebra		14.4 ± 2.4	
	Po-12-239	Washing 3+4, reference vertebra		21.5 ± 2.7	
	Po-12-240	Washing 5+6, reference vertebra		11.2 ± 2.1	
18658	Po-12-241	Soil of the grave with black stain	131 ± 3		88.6 ± 9.0
18635	Po-12-242	Soil of the grave, reference	35.4 ± 0.7		98.3 ± 10
18644	Po-12-243	Soil from the back of the body	26.0 ± 0.6		98.5 ± 10
54504	Po-12-244	Shroud from under the corpse, fragment 2	221 ± 4.2		44.5 ± 5.0
54505	Po-12-245	Shroud from the left thorax, fragment 2	257 ± 4		105 ± 10
54506	Po-12-246	Shroud from the left leg, fragment 2	146 ± 3		96.1 ± 10
18655	Po-13-040	Vertebra (cortical)		32.6 ± 0.4 <sup>b)</sup>	
21096	Po-13-041	Iliac crest, fragment 3		76.7 ± 0.6 <sup>b)</sup>	
18603	Po-13-042	Left rib B, fragment 2		879 ± 4.6 <sup>b)</sup>	
18658	Po-13-043	Soil from the grave with black stain	123 ± 2.4		
18635	Po-13-044	Soil from the grave, reference	28.4 ± 0.9		
21519	Po-13-045	Left femur, cortical		60.1 ± 0.5 <sup>b)</sup>	
19504	Po-12-047	Iliac crest, fragment 4		904 ± 12	91 ± 37
19504	Po-13-048	Altered tissues scraped from the iliac crest, fragment 5	175 ± 3.5		
19504	Po-13-049	Iliac crest, fragment 5		245 ± 5.0	126 ± 40
19504	Po-13-050	Altered tissues scraped from the iliac crest, fragment 6	79.5 ± 0.9		
19504	Po-13-051	Iliac crest, fragment 6		106.0 ± 3.0	137 ± 74

<sup>a)</sup> the unit mBq/g Ca is only applicable to bone specimens

<sup>b)</sup> these values are only indicative because <sup>210</sup>Po was isolated from the elution solution from the <sup>206</sup>Pb/<sup>207</sup>Pb experiment and the <sup>210</sup>Po was traced only after the chemical isolation of lead to avoid <sup>209</sup>Po interference in lead isotopes measurement.

#### V.4.5.2. Polonium-210 bone specimens

We took great care to decontaminate the bone specimens from the potential diagenetic <sup>210</sup>Pb and <sup>210</sup>Po coming from <sup>222</sup>Rn by performing a solubility profile [Schrag, 2012]. Our hypothesis was that diagenetic <sup>210</sup>Pb and <sup>210</sup>Po was deposited on the surface of the bone, while biogenic <sup>210</sup>Pb and <sup>210</sup>Po was introduced inside the bone structure (apatite) by metabolic reactions. Because bone apatite is very resistant to solubilization by weak buffer, it is possible to decontaminate the surface of the bone (see Appendix, Section X.1.4.8).



The activities of  $^{210}\text{Po}$  are presented in Table 14 for the bone specimens. 5 of the 13 specimens have activities above 100 mBq/g Ca, which defines the maximum of our reference values for  $^{210}\text{Po}$  in bone.

**Table 14:**  $^{210}\text{Po}$  activities for bone specimen taken from the body as well as a reference sample (last line).

N° CURML	N° Po	Type of specimen / sample	$^{210}\text{Po}$ [mBq/g Ca]	$^{210}\text{Pb}$ [mBq/g Ca]	% supported
18603	Po-12-207	Left rib A, fragment 1	228 ± 5	552 ± 11	242
18603	Po-12-219	Left rib A, fragment 2	688 ± 11	1094 ± 22	159
18603	Po-12-220	Left rib A, fragment 3	339 ± 6	502 ± 10	148
18603	Po-12-228	Left rib B, fragment 1	449 ± 12	1010 ± 20	225
18655	Po-12-208	Vertebra, fragment 1 (cortical + trabecular)	18.7 ± 1.0	47.5 ± 0.9	254
18655	Po-12-221	Vertebra, fragment 2 (trabecular)	32.7 ± 1.0	26.8 ± 0.5	82
18655	Po-12-229	Vertebra, fragment 3 (cortical + trabecular)	43.8 ± 1.6	52.1 ± 1.0	119
21096	Po-12-209	Iliac crest, left, fragment 1	48.4 ± 1.5	85.7 ± 1.7	177
21096	Po-12-230	Iliac crest, left, fragment 2	57.1 ± 2.0	37.0 ± 0.7	66
21519	Po-12-210	Left femur, trabecular (inside)	28.9 ± 1.1	43.1 ± 0.9	149
21519	Po-12-231	Left femur, fragment 2, mostly cortical	35.0 ± 1.1	18.2 ± 0.4	52
18641	Po-12-214	Sternum, fragment A	136 ± 3	124 ± 2.5	91
18641	Po-12-232	Sternum, fragment B	75.0 ± 2.1	65.3 ± 1.3	87
	Po-12-233	Reference vertebrae, sampled from a grave near Milan (I), death 1990, sampling 2008	144 ± 3	83.5 ± 1.7	58

As explained below (Section V.4.5-V.4.5.4), the decontamination procedure is efficient and the measured  $^{210}\text{Po}$  activity is likely to be underestimated. For this reason we also performed another series of  $^{210}\text{Po}/^{210}\text{Pb}$  measurements without a full decontamination method in a remaining iliac bone specimen in order to complete the present expertise. Figure 11 shows the locations of the three fragments that were measured on the iliac bone specimen and Table 15 shows the activities.

We measured  $^{210}\text{Po}$  in 35 specimens of vertebrae or ribs collected during autopsy. This work was intended to yield a  $^{210}\text{Po}$  ( $^{210}\text{Pb}$ ) reference value at the date of death.  $^{210}\text{Po}$  was also measured on 12 bone specimens of forensic interest [Schrag, 2013]. Bones taken during these autopsies were not submitted to diagenesis, thus were not submitted to solubility profiling for decontamination. On the other hand, bones of forensic interest were often found buried in soil or in aquatic system (glacier, river, marsh, lake). These specimens could have undertaken a strong diagenesis and submitted to decontamination, mostly through solubility profiling. The results of the analysis of  $^{210}\text{Po}$  in these specimens are compared to the results of this case for comparison, in Figure 13 and Figure 14.

**Table 15:** Activities measured on three fragments collected on the iliac bone (not decontaminated, scraped and brushed, n° CURML 19504).  $^{210}\text{Po}$  activities were obtained by  $\alpha$  spectrometry.  $^{210}\text{Pb}$  activities were obtained by  $\gamma$  spectrometry.

N° GRE	N° Po	$^{210}\text{Po}$ [mBq/g Ca]	$^{210}\text{Pb}$ [mBq/g Ca]	% supported
13-208	Po-13-047	904 ± 12	819 ± 330	91 ± 37
13-210	Po-13-049	245 ± 5	310 ± 99	126 ± 40
13-221	Po-13-051	106 ± 3	145 ± 78	137 ± 74



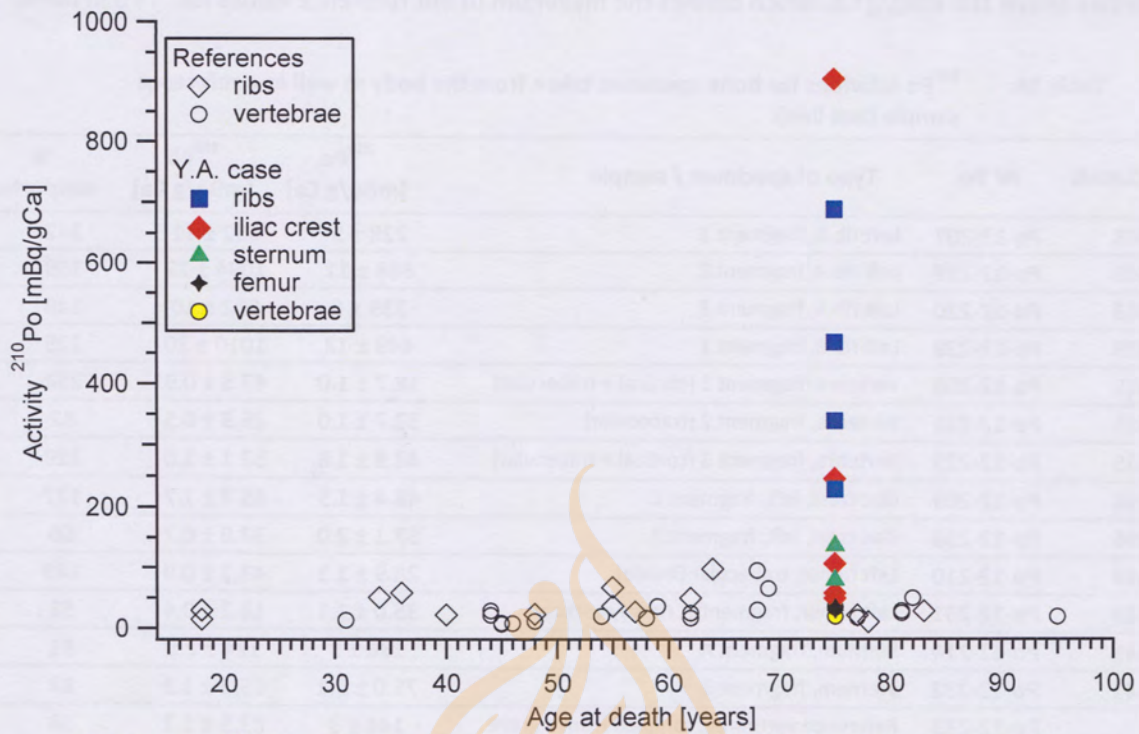


Figure 13:  $^{210}\text{Po}$  activities of all bone specimens compared with our references reported versus the age at death.

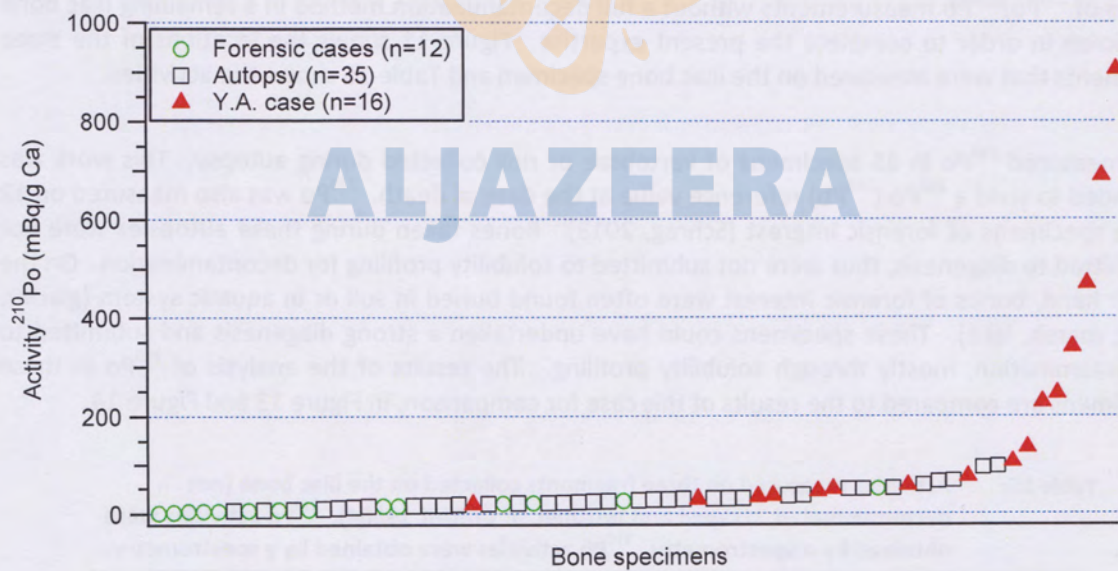


Figure 14:  $^{210}\text{Po}$  activities of all measured bones from President Arafat's grave, as well as comparable reference bones from our own study [Schrag 2013]. Full triangles: President Arafat's bones. Open circles: bones of forensic interest (CURML). Open squares: IRA reference bones collected at autopsy.



#### V.4.5.3. Polonium-210 in non-bone specimens

Table 16 presents the  $^{210}\text{Po}$  activities of the non-bone specimens. Most of them contain significant quantities of  $^{210}\text{Po}$ .

Table 16:  $^{210}\text{Po}$  activities for specimens other than bones taken from the body and the grave.

N° CURML	N° Po	Type of specimen	$^{210}\text{Po}$ [mBq/g]	$^{210}\text{Pb}$ [mBq/g]	% supported
18522	Po-12-201	Pleural cavity	15.9 ± 0.6	13.2 ± 0.3	83
18646	Po-12-202	Scalp	221 ± 6	172 ± 3.4	78
21095	Po-12-203	Tissues of the abdominal cavity	17.7 ± 0.7	16.3 ± 0.3	92
54501	Po-12-204	Tissues (wax) scraped from left femur	73.9 ± 1.7	65.0 ± 1.3	88
54505	Po-12-205	Tissues (wax) scraped from the shroud	415 ± 8	423 ± 8.5	102
54509	Po-12-206	Tissues (wax) scraped from the ribs	138 ± 4	89.7 ± 1.8	65
18603	Po-12-211	Soil from the grave with black stain	122 ± 3	114 ± 2.3	93
18658	Po-12-241	Soil from the grave with black stain	131 ± 3	117 ± 2.3	89
18635	Po-12-212	Soil from the grave, far from the corpse (bottom right corner)	29.5 ± 0.7	32.5 ± 0.6	110
18635	Po-12-242	Soil from the grave, far from the corpse (bottom right corner)	35.4 ± 0.7	34.7 ± 0.7	98
18644	Po-12-215	Soil taken under the corpse	22.3 ± 0.6	17.2 ± 0.3	77
18644	Po-12-243	Soil taken under the corpse	26 ± 0.6	25.7 ± 0.5	99
18655	Po-12-213	Tissues (wax) scraped from the vertebrae	40.5 ± 0.9	33.2 ± 0.7	82
54504	Po-12-216	Shroud from under the corpse	198 ± 4	201 ± 4.2	106
54504	Po-12-244	Shroud from under the corpse	221 ± 4	99.5 ± 2.0	45
54505	Po-12-217	Shroud from the left side of thorax	336 ± 6	302 ± 6.0	90
54505	Po-12-245	Shroud from the left side of thorax	257 ± 4	270 ± 5.4	105
54506	Po-12-218	Shroud from the left leg	261 ± 5	245 ± 4.9	94
54506	Po-12-246	Shroud from the left leg	146 ± 3	140 ± 2.8	96

#### V.4.5.4. Support of polonium-210 by lead-210

##### A. Bone specimens

As shown in the last column of Table 14, most of the bone specimens (n=7) have supported  $^{210}\text{Po}$  activity significantly higher than 100% (up to 254%). While it is well known that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are not in equilibrium in environmental samples and human samples just after death [Blanchard, 1970; Tukizawa, 1990], there is no reason why biogenic  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  could not be at equilibrium 8 years after death.

We attributed this disequilibrium to the decontamination method, which may extract  $^{210}\text{Po}$  more efficiently than  $^{210}\text{Pb}$  from bone. We explain this by the fact that biogenic  $^{210}\text{Pb}$  belongs to the crystalline structure of the apatite, while  $^{210}\text{Po}$  appears after two successive decays of nuclei  $^{210}\text{Pb}$  and  $^{210}\text{Bi}$  (see Section X.1.3.1), whose recoil energy can destroy the local crystalline structure. Thus,



$^{210}\text{Po}$  is more available for extraction with a weak complexing agent (NaOH 1M, acetate buffer). Nevertheless,  $^{210}\text{Po}$  was not present in significant quantities in washings 1 to 6, indicating that some  $^{210}\text{Po}$  was probably already lost during the previous NaOH 1 M washing or during the mineralization step using 10%  $\text{H}_2\text{O}_2$  and a pressurized microwave oven (see Appendix, Section X.1.4.8).

Performing further measurements on specimen of President Arafat's vertebra tested this hypothesis. In this case, we collected the washing fraction of NaOH 1 M in order to measure  $^{210}\text{Po}$ . Then  $^{210}\text{Pb}$  was isolated from this fraction and kept in the refrigerator for two months.  $^{210}\text{Po}$  was isolated again and measured a second time. Taking into account the regrowth factor expected after two months, we observed that the percentage of supported  $^{210}\text{Po}$  was only 45%. This means that the unsupported percentage was 55%. This demonstrates that NaOH 1 M preferentially extracts polonium as compared to lead. This most likely explains why there are percentages of supported  $^{210}\text{Po}$  higher than 100%.

#### B. Non-bone specimens

15 of the 19 non-bone specimens had a supported percentage of  $^{210}\text{Pb}$  between 80% and 120%, which can be considered to be within the uncertainty of 100% supported. Figure 15 shows percentages of supported  $^{210}\text{Po}$  in the non-bone specimens. There is a tendency towards unsupported  $^{210}\text{Po}$ .

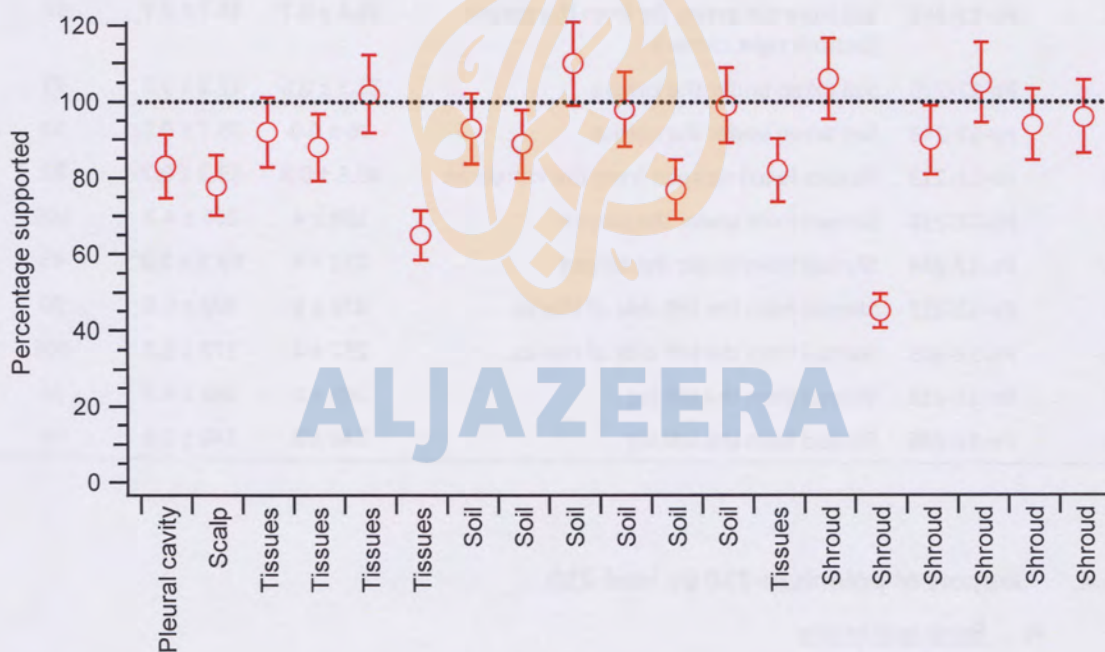


Figure 15: Percentage of  $^{210}\text{Po}$  supported by  $^{210}\text{Pb}$  for all non-bone specimens collected in the grave (same order as in Table 16).

The measurements of the wax specimens collected on the iliac bone (see Table 11) were performed with  $^{210}\text{Pb}$  measured directly by  $\gamma$  spectrometry instead of indirectly by  $\alpha$  spectrometry (see Appendix, Section X.1.4.5-B). The supported percentages also tended to be lower than 100%.



#### V.4.6 Lead-210 present in the grave specimens

$^{210}\text{Pb}$  can be directly measured by  $\gamma$  spectrometry if its activity is sufficiently high. It can also be measured indirectly by  $\alpha$  spectrometry through its granddaughter  $^{210}\text{Po}$ , either with only one measurement that assumes equilibrium between  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , or with two successive measurements typically performed 2-3 months apart.

Figure 16 synthetically presents all of the  $^{210}\text{Pb}$  measurements performed on the bones and compares them with the values of our reference [Schrag, 2013].

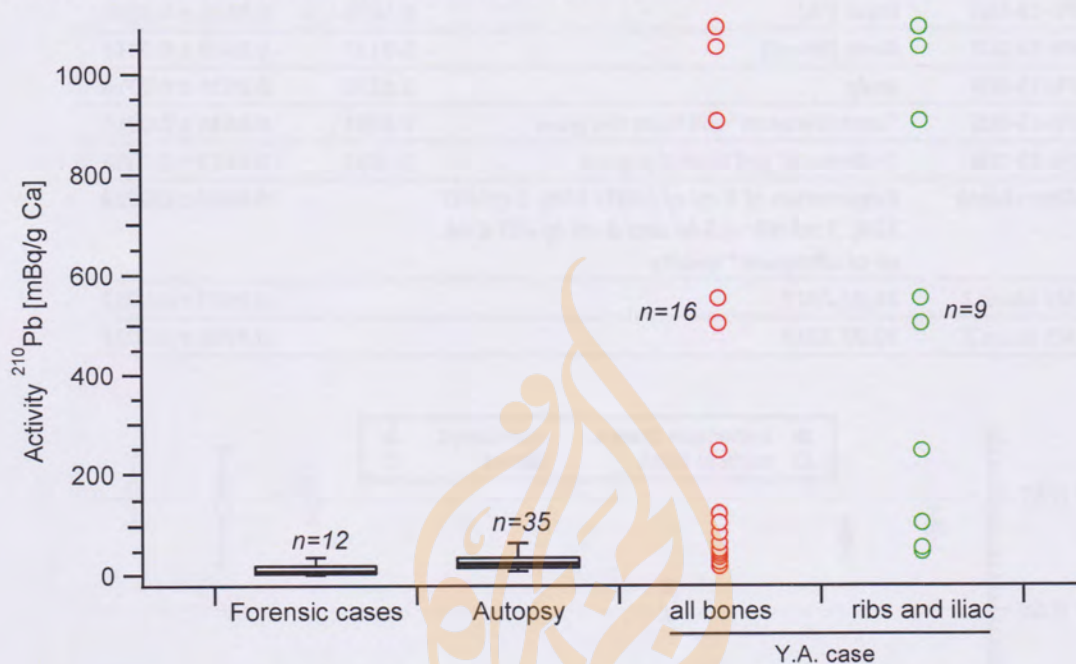


Figure 16:  $^{210}\text{Pb}$  activities measured in bone specimens. The two first columns (Forensic cases and Autopsy) come from our reference (Schrag, 2013). The two last columns correspond to the present study. Box-plots display quartiles.

#### V.4.7 Isotopic ratio of lead-207/lead-206

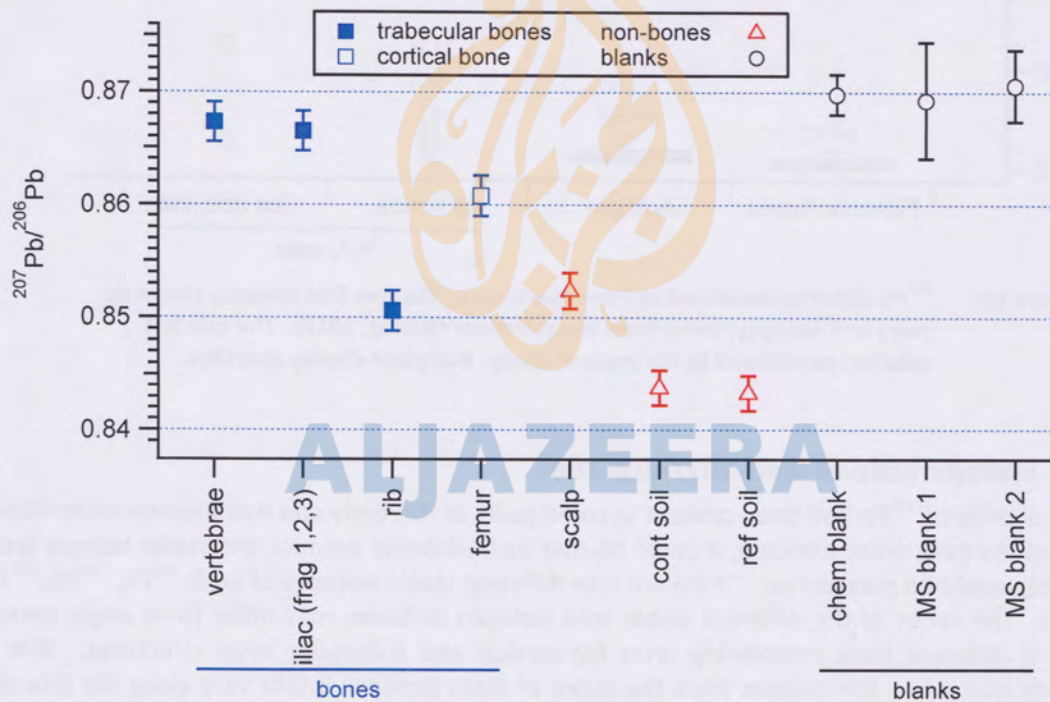
If a large activity of  $^{210}\text{Po}$  had been present in some parts of the body and had become insignificant because of the long decay duration, it could be that its final decay product, the stable isotope lead-206 ( $^{206}\text{Pb}$ ), would be measurable. There are four different stable isotopes of lead:  $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$ . The ratios of the different stable lead isotopes in bones may differ for a single person because of different bone remodeling rates for cortical and trabecular bone structures. This is particularly true when the sources (thus the ratio) of leads isotopes intake vary along life [Martin, 2013; Yoshinaga, 1996]. Nevertheless, for the trabecular bone structure of iliac crest, vertebrae and ribs, the lead isotopes ratios should be similar, if not differently altered by an acute intake. On the other hand, the lead isotopes ratio of the cortical fraction may represent a past intake because of a much longer bone remodeling rate [Martin, 2013; Zoeger, 2005].

The  $^{207}\text{Pb}/^{206}\text{Pb}$  isotopic ratio of 4 bones, 1 scalp and 2 soil specimens, as well as blank products, were determined by the Paul Scherrer Institute (PSI) in July 2013 (see Appendix, Section X.1.4.12). The results are presented in Table 17 and in Figure 17, as given by the mass spectrometry analysis. No corrections were made to account for the lead content of the *chem blank* and the specimens.



**Table 17:** Specimens measured for  $^{207}\text{Pb}/^{206}\text{Pb}$  isotopic ratio. "Chem blank" is a sample made of all the reactants used in the preparation of the samples for MS determination. "MS blanks" are 0.2 M  $\text{HNO}_3$  from Fisher Chemical Optima suprapure from the PSI shelves.

N°	Type of specimen / sample	m [g]	$^{207}\text{Pb}/^{206}\text{Pb}$
Pb-13-001	Bone (vertebra)	0.8733	$0.8673 \pm 0.0018$
Pb-13-002	Bone (iliac crest)	1.0693	$0.8665 \pm 0.0018$
Pb-13-003	Bone (rib)	0.5070	$0.8506 \pm 0.0018$
Pb-13-007	Bone (femur)	0.9127	$0.8608 \pm 0.0018$
Pb-13-004	Scalp	1.1275	$0.8524 \pm 0.0016$
Pb-13-005	"contaminated" soil from the grave	0.6091	$0.8438 \pm 0.0016$
Pb-13-006	"reference" soil from the grave	0.5883	$0.8433 \pm 0.0016$
Chem blank	Evaporation of 6 ml of $\text{HNO}_3$ 65%, 3 ml $\text{HCl}$ 32%, 3 ml $\text{HBr}$ 0.5 M and 8 ml of $\text{HCl}$ 6 M, all of Ultrapure® quality		$0.8698 \pm 0.0018$
MS blank 1	26.07.2013		$0.8693 \pm 0.0052$
MS blank 2	30.07.2013		$0.8706 \pm 0.0032$



**Figure 17:** Lead-206/lead-207 ratio measured on a selection of specimens.

Two trabecular bone specimens have a ratio close to 0.8670. A cortical bone specimen (femur) has a ratio slightly below this value (0.8609), which may be explained by a contribution from the typical environmental lead intake [Keenleyside, 1995; Martin, 2013]. Finally, the third trabecular bone specimen (rib) has a significantly lower ratio of 0.8507. This is noteworthy because the rib is the most radioactive specimen in terms of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  and a lower  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio means that there is more  $^{206}\text{Pb}$  in this specimen than in the others.



The scalp is also a specimen with high  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  activity as compared to other exposed surfaces, and its  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio is very similar to that of the rib. This higher  $^{206}\text{Pb}$  content than expected may be due to significant  $^{210}\text{Po}$  decay in these two specimens. However, due to analytical problems related to the high content of Ca in the bone specimens, the Pb content was significantly lower than expected and these measurements must be interpreted cautiously. In particular, when a blank correction is carried out, the uncertainties of the results are significantly larger than those of the direct measurements.

The  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios of the soils are the lowest of our series and they are not significantly different from one another. This is not surprising because soils contain much more lead than bone. Therefore, the relative amount of  $^{206}\text{Pb}$  potentially produced by a large activity of  $^{210}\text{Po}$  would be much smaller. However, the fact that both soil specimens have the same  $^{207}\text{Pb}/^{206}\text{Pb}$  ratios is an indicator of the coherence of the whole analytical process.

#### V.4.8 Lead-210 as impurity in a commercial source of polonium

The presence of an impurity of  $^{210}\text{Pb}$  in the production of a  $^{210}\text{Po}$  source is worth investigating, because it would not be separated from  $^{210}\text{Po}$  by distillation due to the formation of a volatile lead polonide,  $\text{PbPo}$  [Feueurstein, 1992; Loewen, 2005].

We, therefore, considered this possibility and bought a  $^{210}\text{Po}$  solution of 2.91 MBq (combined relative standard uncertainty of 0.9 %) in 4.931 g from the Czech Metrology Institute to determine the level of  $^{210}\text{Pb}$  impurity in a commercial source of  $^{210}\text{Po}$  (certificate number 9031-OL-501/13). The exact date of the neutron irradiation was not available.

The source was first measured by  $\gamma$  spectrometry, then  $^{210}\text{Pb}$  was separated from the  $^{210}\text{Po}$  solution in our hot lab. Then,  $^{210}\text{Pb}$  and  $^{210}\text{Bi}$  were precipitated from this eluate along with iron hydroxides and the micro-precipitate were measured by proportional counting (see Appendix, Section X.1.4.15). During the chemistry steps, the secular equilibrium between  $^{210}\text{Pb}$  and  $^{210}\text{Bi}$  was not preserved. Efficiency for  $^{210}\text{Pb}$  is below 5%, while it is about 33% for  $^{210}\text{Bi}$  in a gas flow proportional counter. Therefore, the increase of  $^{210}\text{Bi}$  in the source due to the decay of  $^{210}\text{Pb}$  can be observed. After counting, the source was dissolved in 1 M HCl and  $^{210}\text{Po}$  determined, as previously described. The radiochemical purification was carried out on two aliquots of 0.9 MBq each. The residual  $^{210}\text{Po}$  activity in the  $^{210}\text{Pb}$  fraction was 140 mBq and 108 mBq, yielding a decontamination factor of  $1.5 \cdot 10^{-7}$ .

Figure 18 displays the in-growing signal of  $^{210}\text{Bi}$  as measured by proportional counting on a source of  $^{210}\text{Pb}$  iron hydroxides precipitate. This signal clearly identifies the presence of  $^{210}\text{Pb}$  through  $^{210}\text{Bi}$  growth<sup>(3)</sup> in the  $^{210}\text{Po}$  source and confirms the results of the  $\gamma$  spectrometry.

<sup>(3)</sup> We fitted the data according to an adaptation of the standard Bateman formula:

$$A_{^{210}\text{Bi}}(t) = A_{0,^{210}\text{Pb}} \frac{T_{^{210}\text{Pb}}}{T_{^{210}\text{Pb}} - T_{^{210}\text{Bi}}} \left( e^{-\frac{\ln 2}{T_{^{210}\text{Pb}}} t} - e^{-\frac{\ln 2}{T_{^{210}\text{Bi}}} t} \right) + A_0(^{210}\text{Bi}) e^{-\frac{\ln 2}{T_{^{210}\text{Bi}}} t}$$



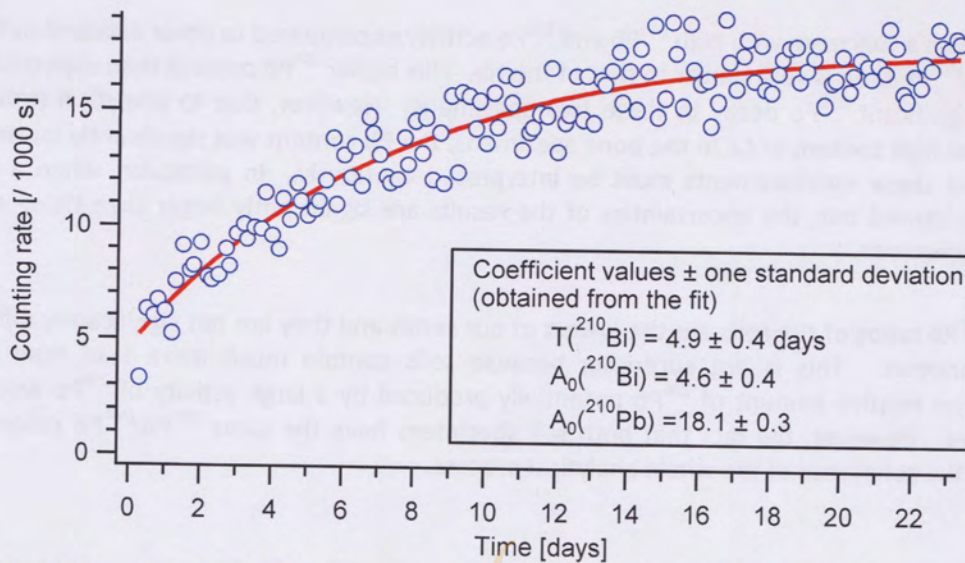


Figure 18: Proportional counting signal in the  $\beta$ -channel from an iron hydroxide source produced by purification of  $^{210}\text{Pb}$  from a  $^{210}\text{Po}$  0.9 MBq source. The fit according to the Bateman formulae leads to a  $^{210}\text{Bi}$  half-life of  $4.9 \pm 0.4$  days as compared to the reference value of 5.01 days.

Results of the  $\gamma$  spectrometry and proportional counting measurements are displayed in Table 18. The impurity factor ( $^{210}\text{Pb}/^{210}\text{Po}$ ) is approximately  $10^{-7}$ . Figure 19 shows the evolution of this factor with time. The impurity factor increases by a factor 10 after approximately 1.3 years. In other words, if a  $^{210}\text{Po}$  source has an impurity factor of  $10^{-7}$  at the time of production, 2.6 years after, the impurity factor would have increased to  $10^{-5}$ .

Table 18: Impurity factor ( $^{210}\text{Pb}/^{210}\text{Po}$ ) measured in a marketed source of polonium.

Counting time [s]	Measured radionuclide	Experimental conditions	$^{210}\text{Pb}$ activity [mBq/ 2.91 MBq $^{210}\text{Po}$ ]	Impurity factor ( $^{210}\text{Pb}/^{210}\text{Po}$ )
86 400	$^{210}\text{Pb}$	$\gamma$ spectrometry	$206 \pm 88$	$0.7 \cdot 10^{-7}$
450 767	$^{210}\text{Pb}$	$\gamma$ spectrometry	$238 \pm 30$	$0.8 \cdot 10^{-7}$
90 x 14 400	$^{210}\text{Bi}$	$\beta$ proportional counting	$183 \pm 25$	$0.6 - 0.7 \cdot 10^{-7}$



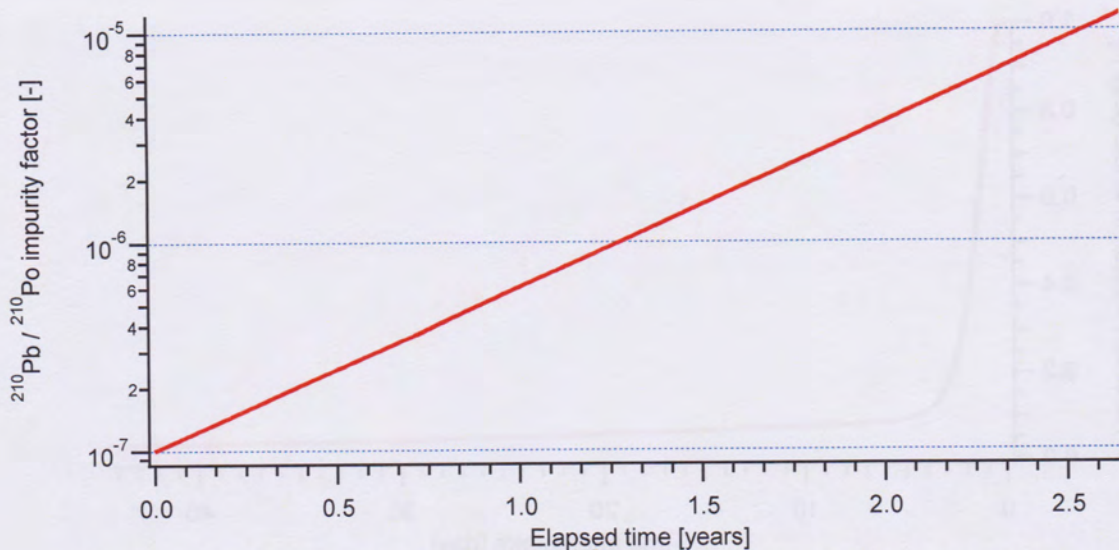


Figure 19: Temporal evolution of the  $^{210}\text{Pb}/^{210}\text{Po}$  impurity factor of a  $^{210}\text{Po}$  source starting with an initial value of  $10^{-7}$ , as found in this work. The impurity factor increases by a factor 10 after approximately 1.3 years.

#### V.4.9 Biokinetic modeling for intake of radionuclides

We modeled the biokinetics of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the human body according to the state-of-the-art data available in the literature (see Appendix, Section X.1.5.1, for polonium and Section X.1.5.2, for lead).

##### V.4.9.1. Biokinetics of polonium-210

###### A. Polonium-210 in the whole body

Whole-body retention is shown in Figure 20. A large portion of ingested  $^{210}\text{Po}$  is rapidly cleared through feces without entering the systemic circulation. Assuming a period of 30 days between intake and death, about 4 to 5% of ingested  $^{210}\text{Po}$  would remain in the body. For an ingestion of 1 GBq of  $^{210}\text{Po}$  in October 2004, one may expect to find a total activity of approximately 15 Bq in the corpse in November 2012 at the time of exhumation.



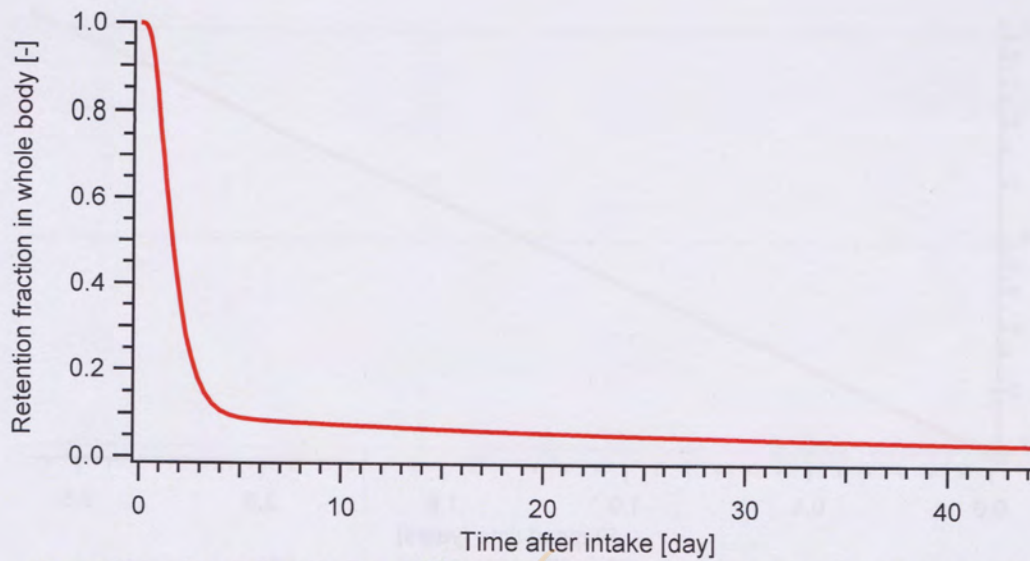


Figure 20: Retention fraction of  $^{210}\text{Po}$  in the whole body after a single intake by ingestion ( $f_1=0.1$ ).

$^{210}\text{Po}$  is deposited predominantly in soft tissues, mainly the liver, the skin, the kidneys and the red bone marrow (RBM). There is also a significant amount of  $^{210}\text{Po}$  in circulating blood (RBC and plasma) (see Figure 21).

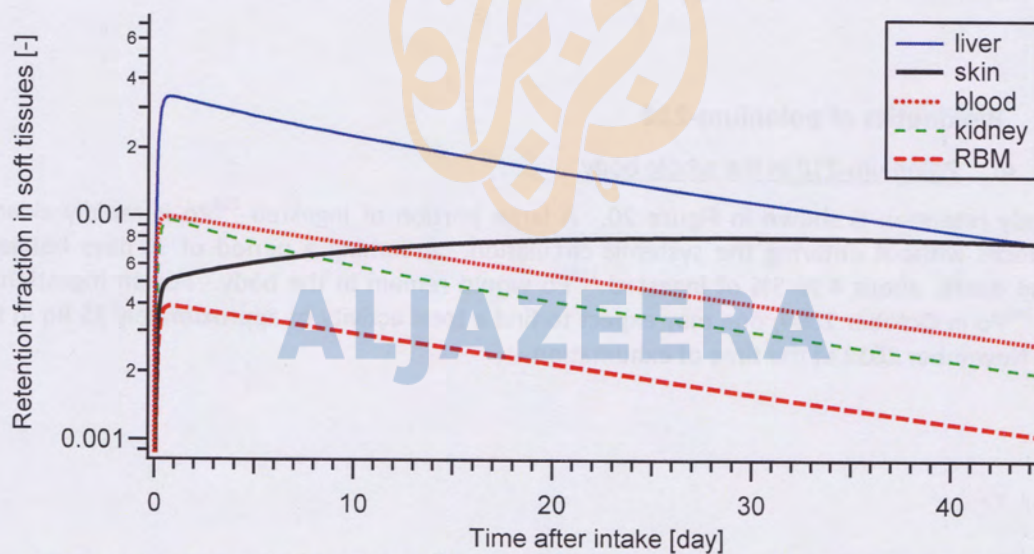


Figure 21: Retention fraction of  $^{210}\text{Po}$  in soft tissues and blood after a single intake by ingestion ( $f_1=0.1$ ).

#### B. Polonium-210 in bones

Figure 22 shows the uptake in the bone surface. The retention in bone surface is longer than the retention in other soft tissues. Immediately after intake, most of  $^{210}\text{Po}$  in the skeleton is located in the RBM, while 30 days after intake the retention fraction in the bone surface exceeds that in the RBM. About 0.2% of ingested  $^{210}\text{Po}$  is located in the bone surface 30 days after intake. For an ingestion of 1 GBq of  $^{210}\text{Po}$  in October 2004, one may expect to find a total activity in bones (without RBM) of around 1 Bq in November 2012 at the time of exhumation. According to the model, the



skeletal deposition of  $^{210}\text{Po}$  would occur exclusively in the bone surface in case of acute intake. This may provide another explanation for the partial loss of  $^{210}\text{Po}$  during the decontamination procedure of bone specimens from the grave, as reported in Section V.4.5.4.

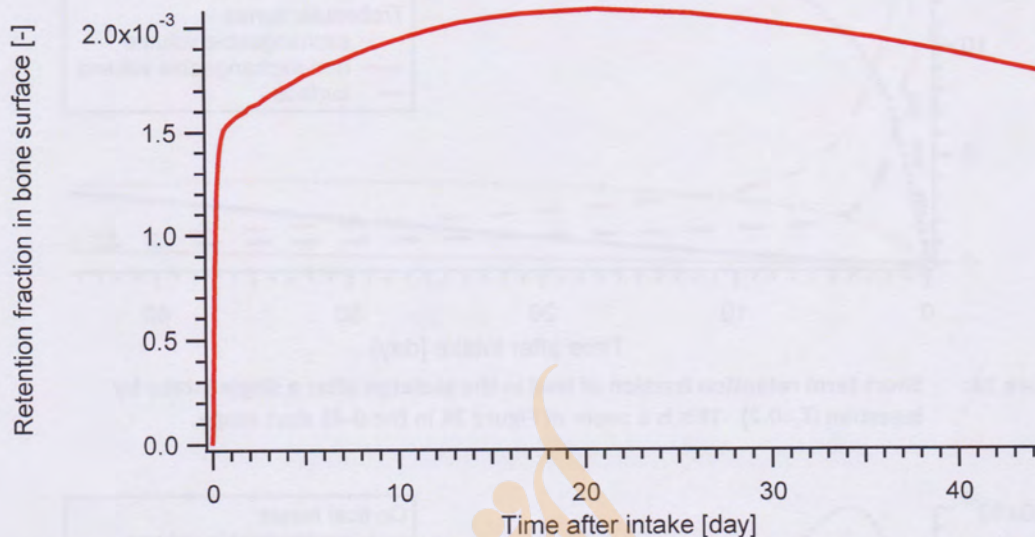


Figure 22: Retention fraction of  $^{210}\text{Po}$  in bone surface after a single intake by ingestion ( $f_1=0.1$ ).

#### V.4.9.2. Biokinetics of lead-210 in bones

Lead is known to be a bone-volume-seeking element (ICRP, 1992). 20% of ingested lead enters the blood pool, after which there is competition between the deposition in the skeleton and in other tissues, particularly red blood cells. Lead entering the skeleton deposits initially on bone surfaces and then migrates to exchangeable bone volumes within a few days. After 30 days, about 2% of ingested lead is located in the exchangeable bone volume (see Figure 23). A portion of lead leaves the exchangeable bone volume, and about one year after intake about 1.5% is assigned to the non-exchangeable bone volume and is stored for an extended period (biological half-life of 20-40 years for cortical bones), where it is gradually remobilized to blood by resorption (see Figure 24).



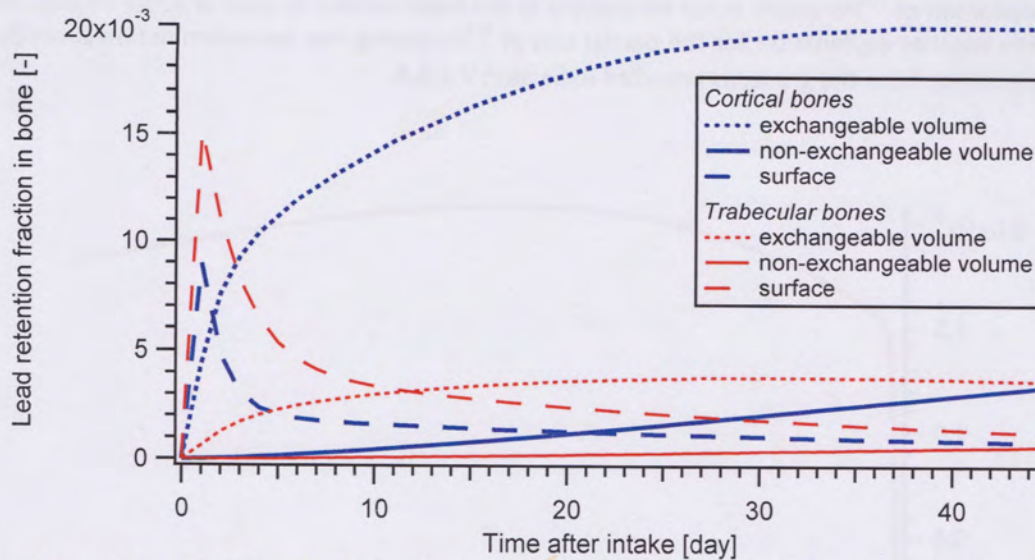


Figure 23: Short-term retention fraction of lead in the skeleton after a single intake by ingestion ( $f_1=0.2$ ). This is a zoom of Figure 24 in the 0-45 days range.

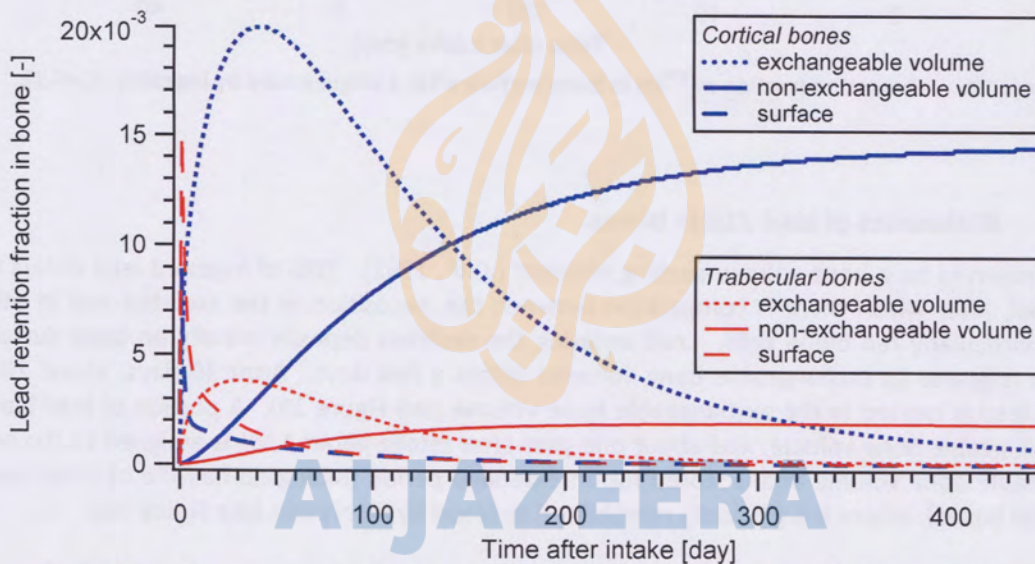


Figure 24: Long-term retention fraction of lead in the skeleton after a single intake by ingestion ( $f_1=0.2$ ).

The literature of chemical incorporation also reflects the typical one-month latency of lead fixation. For instance, by stating that it is well-known that “measurements of lead in bone reflect cumulative Pb exposure, whereas blood Pb levels are indices of absorption during the previous 21-30 days” [Rosen, 1993].

Our understanding of the biokinetics of lead in the skeleton is limited. The models only consider the broad categories of cortical and trabecular bones without differentiating between specific bones like vertebra, rib or iliac crest etc... This reduces the usefulness of the models because bones with different vascularization might incorporate different amounts of lead. After an acute ingestion, lead would initially deposit close to the bone surface and possibly in trabecular bones where exchange kinetics are rapid (Martin, 2013). One would, therefore, expect to observe large variations of lead between bones in the first few months following an acute ingestion. On a longer timescale, lead



would be uniformly distributed throughout the volume of the different bones. Such a uniform biodistribution might better reflect a chronic ingestion of lead [Martin, 2013].

#### V.4.9.3. Polonium-210 and lead-210 in bones after death

The two previous sections (V.4.9.1 and V.4.9.2) show that the total retention fraction of  $^{210}\text{Pb}$  in the bones 30 days after intake is about 10 times larger than the retention fraction of  $^{210}\text{Po}$ . This allows us to compute the activities of these two radionuclides in the skeleton after death by radioactive decay<sup>(4)</sup>. Figure 25 shows these activities for an intake of 1 GBq of  $^{210}\text{Po}$ . We considered three scenarios of  $^{210}\text{Pb}$  impurities in the source: 0.1 kBq, 1 kBq and 10 kBq. This corresponds to impurity factors of  $10^{-7}$ ,  $10^{-6}$  and  $10^{-5}$ , respectively. It can be observed that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  will reach equilibrium about 8.5 years, 7.1 years and 6 years after death, respectively. In other words, after those elapsed times, the activity of  $^{210}\text{Po}$  supported by  $^{210}\text{Pb}$  impurities significantly dominate the activity of the rapidly decaying unsupported  $^{210}\text{Po}$ .

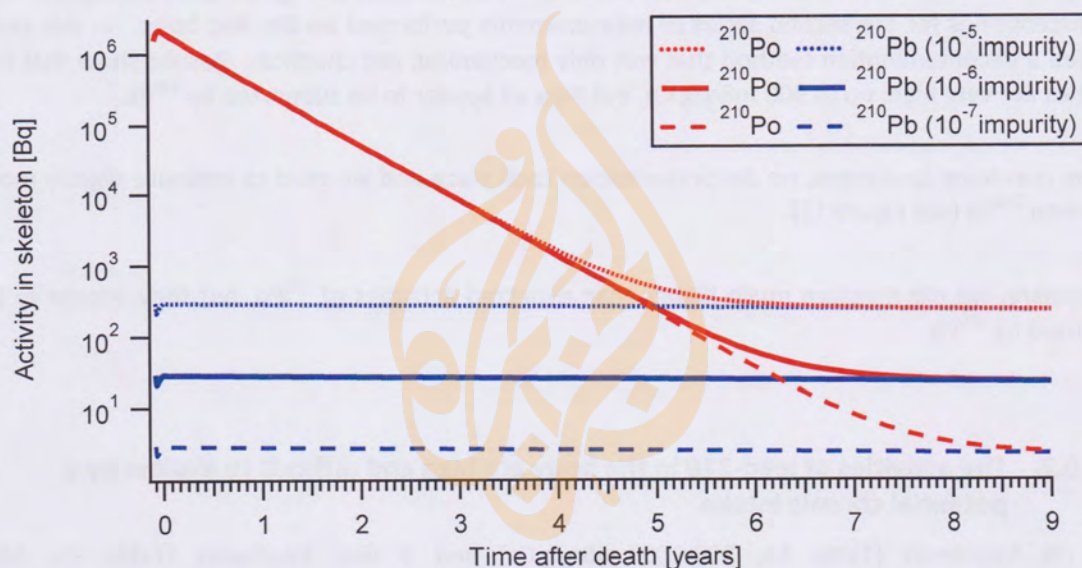


Figure 25: Total activity in the skeleton following a  $^{210}\text{Po}$  intake of 1 GBq. Three scenarios were computed with impurities of  $^{210}\text{Pb}$  in the source of 0.1 kBq (impurity factor  $10^{-7}$ ), 1 kBq ( $10^{-6}$ ) and 10 kBq ( $10^{-5}$ ). The activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  reach equilibrium after about 8.5 years, 7.1 years and 6 years respectively.

<sup>(4)</sup> Pure exponential decay for  $^{210}\text{Pb}$ . Exponential decay +  $^{210}\text{Pb}$  contribution for  $^{210}\text{Po}$ .



#### V.4.10 Discussion of radiological analyses of the specimens taken from the remains after exhumation

##### V.4.10.1. Higher than expected activities of polonium-210 were measured in the body remains, but they are supported by lead-210

We measured higher than expected activities of  $^{210}\text{Po}$ . This was particularly the case for 4 fragments of a rib (Table 14, 228-688 mBq/g Ca), as well as 3 fragments of the iliac bone (Table 15, 106-904 mBq/g Ca) that were all higher than the reference values from the literature (see Appendix, Table 22, around 30 mBq/g Ca).

Because we needed to fully decontaminate the bone specimens from any diagenetic  $^{210}\text{Po}$ , we used a relatively harsh decontamination method that removed more  $^{210}\text{Po}$  than  $^{210}\text{Pb}$  from the bone matrix. This is probably due to the destruction of the apatite crystalline structure when  $^{210}\text{Pb}$  disintegrates. This experimental fact prevented us from precisely measuring the amount of unsupported polonium in bone specimens. Nevertheless, the true value of  $^{210}\text{Po}$  is *at least* as high as what we report. The only exception is for the second series of measurements performed on the iliac bone. In this case, we used a decontamination method that was only mechanical, not chemical. Results show that the activities are very high, up to 900 mBq/g Ca, but they all appear to be supported by  $^{210}\text{Pb}$ .

For the non-bone specimens, no decontamination took place and we tend to measure slightly more  $^{210}\text{Po}$  than  $^{210}\text{Pb}$  (see Figure 15).

In summary, we did measure much higher than expected activities of  $^{210}\text{Po}$ , but they appear to be supported by  $^{210}\text{Pb}$ .

##### V.4.10.2. The activities of lead-210 in the body are high and difficult to explain by a potential chronic intake

Four rib fragments (Table 14, 502-1094 mBq/g Ca) and 3 iliac fragments (Table 15, 145-819 mBq/g Ca) had activities that were systematically higher than our reference values (see Appendix, Table 22, below 50 mBq/g Ca). The activity also tended to be higher in the scalp (Table 13, 172 mBq/g) than in reference tissues from the literature (see Appendix, Table 23, 1-10 mBq/g). Moreover, the stained soil below the body in the grave (Table 16, 122-131 mBq/g) had higher activity than the reference soil located away from the body (Table 16, 29-35 mBq/g).

If a  $^{210}\text{Pb}$  contamination had occurred a long time before death or had been chronic, one would have expected to observe relatively homogenous activities of  $^{210}\text{Pb}$  within the bones and potentially within what remained of the soft tissues (see Section V.4.9.2). This is not the case and we observed large differences in  $^{210}\text{Pb}$  concentrations across the different bones, with much more activity in the ribs and a large gradient of activity within the iliac bone in the distal direction.

In summary, we measured  $^{210}\text{Pb}$  activities in the bones and soft tissues that were up to 20 times larger than the references from the literature. The fact that they are not homogenous is compatible with an intake of  $^{210}\text{Pb}$  that occurred at the onset of the first symptoms (October 2004).



#### V.4.10.3. Higher than expected activities of lead-210 are compatible with impurities contained in a polonium-210 source

We tested different hypotheses about the possible origin of the observed high activities of  $^{210}\text{Pb}$ : smoking, radium and by-products of a high-activity source of polonium.

The high levels of  $^{210}\text{Po}$  observed in the personal effects of President Arafat have been explained many times in the media by a contamination of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  through (passive) smoking<sup>(5)</sup>. Daily urine (about 1.5 l) excretion of  $^{210}\text{Po}$  for a heavy smoker can be as high as 30 mBq/d, compared to a low value of 5 mBq/d for non smoker. Nevertheless the high excretion for heavy smoker cannot account for the 180 mBq activity found in President Arafat's underwear containing only 2 ml of urine. A heavy smoker would incorporate about twice as much  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  as a non-smoker [Holzmann, 1966]. This doubling would not be sufficient to explain the highest values that we measured. Furthermore, a chronic incorporation through (passive) smoking would not explain the large differences observed between the different bones.

$^{210}\text{Pb}$  and  $^{210}\text{Po}$  belong to the progeny of  $^{226}\text{Ra}$ , which has been used in the past in fluorescent painting for the watch and military industry. We measured specimens by  $\gamma$  spectrometry and found no  $^{226}\text{Ra}$  activities above the detection limit (see Table 12). Again, a chronic or an ancient acute contamination by  $^{226}\text{Ra}$  would, through time, have caused a homogenous deposition of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

We also tested for one potential fission product that specifically targets bones:  $^{90}\text{Sr}$  (see Appendix, Section X.1.4.13). None was found in the vertebra, nor in the femur that we measured (< 10 mBq/g Ca).

Finally, we tested the origin of  $^{210}\text{Pb}$  as being an impurity present in a potential  $^{210}\text{Po}$  source ingested by President Arafat. The Paul Scherrer Institute performed a computation (see Appendix, Section X.1.4.14) with limited knowledge about the raw material and the methodology actually used to produce a  $^{210}\text{Po}$  source. The calculation showed that  $^{210}\text{Pb}$  is indeed produced by neutron activation, but in a quantity too small to be detected in the bones of a person poisoned by polonium.

Simulation is useful, but real measurements provide more solid evidence. We measured a commercial  $^{210}\text{Po}$  source in order to directly estimate the impurity factor. The measured value ( $10^{-7}$ ) was significantly larger than the values obtained by simulation, and was sufficient to induce equilibrium between  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the bones at the time of their measurement at the beginning of 2013 (see Section V.4.8). Our calculation further showed that waiting more than a year before using such a source as a poison would greatly increase the activity of  $^{210}\text{Pb}$  in the bones.

In summary, the observed high activities of  $^{210}\text{Pb}$  present in the bones (and similarly also in the soil under the corpse) cannot be explained by any natural reason or by smoking. However, the amount of impurities of  $^{210}\text{Pb}$  that we measured in the commercial source of polonium can explain the high activity of  $^{210}\text{Pb}$  found in the bones of President Arafat.

<sup>(5)</sup> See for instance, the opinion of Deborah Blum on <http://blogs.smithsonianmag.com/smartnews/2012/12/was-yasser-arafat-poisoned-by-polonium/>



#### **V.4.10.4. High activities of lead-210 could hide a remaining activity of unsupported polonium-210**

After eight years of confinement in the grave, the high activity of  $^{210}\text{Pb}$  induces an equal activity of  $^{210}\text{Po}$ . This high activity of supported  $^{210}\text{Po}$  could prevent the detection of a smaller quantity of unsupported  $^{210}\text{Po}$  (see Figure 25).

We estimate that the detection limit of unsupported  $^{210}\text{Po}$  would be equal to about 20% of the activity of  $^{210}\text{Pb}$ . For the most radioactive bone ( $^{210}\text{Pb}$  activity of about 1 000 mBq/g Ca), the detection limit of unsupported  $^{210}\text{Po}$  is therefore equal to about 200 mBq/g Ca. According to the biokinetic models, this amount of activity is higher than what could be expected in bones after polonium poisoning 8 years prior to the measurement.

In other words, if poisoning with polonium actually occurred, the high level of  $^{210}\text{Pb}$  would probably not allow for the detection of its presence. Its detection would be masked.

#### **V.4.10.5. High activities of lead-210 in the body remains is coherent with what has been observed in the biologically stained specimens of the travel bag**

The high levels of  $^{210}\text{Pb}$  measured in the body remains were most likely also present before death, and potentially also in the biological fluids. If this were the case, it would explain why we also identified relatively high levels of supported  $^{210}\text{Po}$  in the most radioactive specimens of the personal effects (see Section IV.3.5).

#### **V.4.10.6. A surplus of lead-206 present in the rib and scalp points toward a fossil signal of polonium-210, but the uncertainties are important**

The trabecular bones of any given organism are expected to have the same proportions of  $^{206}\text{Pb}$  (i.e. the same  $^{207}\text{Pb}/^{206}\text{Pb}$  isotopic ratios). A measurable increase of  $^{206}\text{Pb}$  cannot be explained by the activity of  $^{210}\text{Pb}$  that we measured because the number of involved atoms is far too small. On the other hand, a very high activity of  $^{210}\text{Po}$  could produce a measurable increase of  $^{206}\text{Pb}$ .

The two specimens of trabecular bone with the lowest  $^{210}\text{Pb}$  activities (iliac crest and vertebra) had similar  $^{207}\text{Pb}/^{206}\text{Pb}$  isotopic ratios, which were significantly higher than the ratio of the other trabecular bone (rib) sampled that had the highest  $^{210}\text{Pb}$  activity (see Section V.4.7). In other words, the most radioactive trabecular bone, in term of  $^{210}\text{Pb}$ , is also the one that contains more  $^{206}\text{Pb}$ .

Examining the past presence of a radionuclide through the analysis of a stable remaining isotope is analytically difficult and subject to high uncertainty. Therefore, the interpretation of the results was, done cautiously.

#### **V.4.10.7. A significant amount of radon was present in the grave, but its progeny seems to have been deposited relatively homogenously**

The radon concentration measured the night before the opening of the grave was 12 000 Bq/m<sup>3</sup>. If one assumed that this exposure had been constant since burial and that the radon progeny was deposited homogenously on all surfaces, one would expect to have found a mean activity of  $^{210}\text{Po}$  or  $^{210}\text{Pb}$  equal to about 36 mBq/cm<sup>2</sup> (see Section V.4.3.1). The surface activities measured on the reference soil (Table 10, 1.7 mBq/cm<sup>2</sup>) and on the shroud (Table 9, 2.1 to 6.2 mBq/cm<sup>2</sup>) were all



significantly lower than 36 mBq/cm<sup>2</sup>. This may indicate that the radon concentration was lower in the past than during our measurement or that the deposition of the daughter products does not occur in totality on the grave surfaces. On the other hand, the removal of up to 3 meters of earth on top of the grave before the grave was opened might have increased <sup>222</sup>Rn flux into the grave by decreasing the pressure.

Nevertheless, the surface activities of the shroud were homogenous (Table 9, 2.1 to 6.2 mBq/cm<sup>2</sup>). Equally, the two measurements of the iliac bone wax were very similar (Table 11, 12.5 and 13 mBq/cm<sup>2</sup>). This indicates that radon progeny was deposited relatively uniformly on the corpses' exposed surfaces and cannot, therefore, explain the variability observed in the bone specimens.

#### **V.4.10.8. Radon progeny alone cannot explain all observed high activities of lead-210 and polonium-210**

The wax specimens had a surface activity that was approximately twice that of the reference soil and shroud specimens. This is compatible with an internal contribution of <sup>210</sup>Po and <sup>210</sup>Pb present in the now-disappeared soft tissues.

The surface activity of the soil visibly contaminated by the biological fluids was significantly higher than the reference soil in the grave (Table 10, 30.4 compared to 1.7 mBq/cm<sup>2</sup>). This is compatible with a contamination by <sup>210</sup>Po and <sup>210</sup>Pb from the body fluids. Furthermore, because the soil visibly contaminated by biological fluids contained the highest relative humidity, one would expect it to contain lower activities of radon progeny, e.g. <sup>210</sup>Pb and <sup>210</sup>Po<sup>(6)</sup>. Because we observe the opposite, this is also an argument in favor of a radiological contamination from the now disappeared body fluids and soft tissues, and not from the radon environment of the grave.

The surface activity of the scalp tissue was significantly higher than the shroud (Figure 12, 71 mBq/cm<sup>2</sup> and 5 mBq/cm<sup>2</sup> respectively). In fact, the shroud protected the scalp for a while, until the shroud decayed and exposed the scalp surface. Therefore, one would expect a lower surface activity for the scalp. This observation is therefore also compatible with a body contamination by <sup>210</sup>Po and <sup>210</sup>Pb.

Great attention was paid to measure the surrounding wax tissue separately from the iliac bone. While the surface activities of the wax tissue were almost constant, an increase of the bone <sup>210</sup>Po activity was observed in the distal direction (106, 245, 904 mBq/g Ca). This indicates that the variation of activity cannot be explained by external deposition and should, therefore, be interpreted by internal mechanisms. In other words, this is also compatible with a <sup>210</sup>Po and <sup>210</sup>Pb source present in the body and not deposited by radon within the grave.

In summary, radon progeny was present in the grave, but the detailed analysis of the activities in the different parts of the soil, the comparison of the surface activities of the scalp and the shroud, as well as the distribution of the activities on the surface and within the iliac bone clearly show that the radon progeny cannot explain the observed high activities of <sup>210</sup>Po and <sup>210</sup>Pb.

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<sup>(6)</sup> Raman et al. (2008) found a correlation with <sup>222</sup>Rn emanation from soil and soil humidity with a maximum emanation at 35% relative humidity (Hr). Sing et al (2011) found only a weak correlation between soil <sup>222</sup>Rn emanation and Hr, with higher emanation with increasing soil Hr. Thus, soils with high humidity usually contain less activity in radon progeny.



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## VI. Summary and final discussion of the results and conclusion

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## VI.1. Synthesis of the medical situation

On the evening of October 12<sup>th</sup>, 2004 President Yasser Arafat developed acute gastrointestinal symptoms of nausea, vomiting, abdominal pain and diarrhea, while remaining afebrile. These symptoms persisted with progressive deterioration of the general state of President Arafat, followed by the development of severe thrombocytopenia, disseminated intravascular coagulation (DIC), isolated hemophagocytic bone marrow changes, cholestatic jaundice and impaired vigilance. Despite the necessary and appropriate care administered, the patient died on November 11<sup>th</sup>, 2004 (30 days after the onset of symptoms) due to intracerebral hemorrhage.

Despite the multiple clinical investigations performed and the numerous renowned specialists consulted during the patient's stay at the Military Hospital of Percy, it was not possible to determine the nature and/or cause of these symptoms that began acutely in an otherwise healthy individual.

Post-mortem investigations (forensic autopsy, histology examination and toxicology) were not performed. Given that a diagnosis had not been reached and that the patient was a well known political figure, more information into the cause of his death should have been investigated. There were many rumors circulating at the time and the media was inventing hypotheses of their own.

Among the numerous hypotheses evoked, but not confirmed or excluded based on the negative results of the clinical investigations, was that the patient may have suffered from an acute intoxication. This hypothesis could not be ruled out for the following reasons:

- The presented clinical picture with the development of hematological abnormalities is highly suggestive of a pathology of toxic origin,
- No other cause to explain the onset of the symptoms could be identified,
- The toxicological investigations undertaken at the Military Hospital of Percy were performed on samples collected three weeks after the onset of the gastrointestinal symptoms. This three week delay could explain the negative results due to the metabolism / elimination of some toxic agent(s) possibly involved,
- Without calling into question the quality of the toxicological and radio-toxicological analyses performed, it should be recognized that one can not eliminate every possible toxic substance due to the non-exhaustive nature of the methods and analytical techniques employed. In addition, at that time, polonium-210 poisoning was not documented, except that it was known to be toxic.

## VI.2. Observed symptoms and acute radiation syndrome

The presented clinical symptoms of nausea, vomiting, fatigue, diarrhea and anorexia, followed by hepatic and renal failure are often seen in cases of acute radiation syndrome (ARS). ARS, however, usually also includes myelosuppression and hair loss, which were absent in this case. These latter two symptoms, however, are diversely observed and scantily documented in the case of internal exposure. The only well known case is that of Mr. Litvinenko, but his report has not been made public and we cannot access it.

Many patients have been treated with injected or ingested radionuclides at high activities in nuclear medicine therapy. Side-effects include nausea, vomiting, fatigue and abdominal pain, but hair loss is less common. Toxicity may occur in bone marrow and, to a lesser extent, kidneys and liver [Chatal, 1999]. Myelosuppression varies greatly among patients. In President Arafat's case, low marrow cellularity associated with aging and non-bone-seeking biodistribution of polonium-210 (<sup>210</sup>Po) might



have attenuated the myelosuppression [Watchman, 2007]. Since ingested  $^{210}\text{Po}$  is eliminated primarily through feces, the gastrointestinal syndrome, associated to multiple organ failure, could be a predominant cause of death [Casarett, 1964].

The symptoms of ARS reported in literature refer almost exclusively to situations of acute whole-body external exposure. In cases of protracted internal exposure, the clinical features would depend on many parameters, including 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 lethal ingestion of  $^{210}\text{Po}$  in humans, but it has not been reported in the scientific literature and the known clinical features were only those available from media reports.

Aging could also play a role in the absence of myelosuppression because it is associated with decreased bone marrow cellularity (i.e. increased adipocyte concentration). One would expect less  $\alpha$  particle energy deposited within active bone marrow. This would, therefore, result in a decreased absorption in active bone marrow and potentially reduced myelosuppression in the elderly.

It is also possible that the gastrointestinal syndrome would predominate in a case of ingested  $^{210}\text{Po}$ . Harrison et al., [2007] reported results from a rat study suggesting that damage to the gut mucosa was the possible cause of death in a case of ingestion of  $^{210}\text{Po}$ . Scott [2007] reported that, in case of ingested  $^{210}\text{Po}$ , "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. [2007] reported that "It is possible, therefore, that gut doses may have been substantially underestimated by not taking account of retained  $^{210}\text{Po}$ ". 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 a case of an  $\alpha$ -emitting radionuclide, where devastating effects are localized in the direct vicinity of the decaying atom. For instance, Hobbs et al. [2012] 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 radium-223 ( $^{223}\text{Ra}$ ), another  $\alpha$ -emitting radionuclide<sup>(7)</sup> 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 lethal intake, high levels of a protracted dose delivered to target organs may significantly modify the metabolism over time (biokinetics).

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<sup>(7)</sup>  $^{223}\text{Ra}$  is a bone-seeker radionuclide.



### VI.3. Framework of our investigations

In examining the toxicological and radio-toxicological investigations, four particular critical problems must be pointed out:

- There was a lack of adequate biological specimens, thus limiting the possibility to perform further analyses. This was partly due to the fact that the blood, urine, fecal and cerebrospinal fluid samples taken during the patient's hospitalization at Percy were subsequently destroyed. In addition, there were no samples taken at an early stage when the initial symptoms developed.
- As a result, our initial investigations were performed on very small specimens, such as a single hair shaft, or on atypical specimens, such as the sweat in the patient's clothing or traces of blood and urine found on his personal effects in the travel bag. The same investigations were performed on biological specimens (hair, bone, scalp) and on non-biological specimens (soil, shroud fragments) collected after exhumation of the deceased in Ramallah. In any event, all of these specimens proved problematic in terms of their analysis, as well as for the interpretation of the results. We have limited experience working with such specimens and very little has been published in the scientific literature.
- In addition, the fact that eight years passed between the death of the patient and the implementation of toxicological and radio-toxicological investigations contributes to the uncertainty of the analytical results and their interpretation. After such a long delay, especially under less than optimal conservation conditions (ambient temperature of the travel bag, burial), one cannot exclude the possibility of chemical degradation or redistribution with the surrounding environment. When considering radio-toxicological elements, one must keep in mind that they have a very short half-life (138 days for  $^{210}\text{Po}$ ), rendering their detection eight years after a possible administration very difficult and subject to large uncertainties. Furthermore, the elapsed 8 years prevented us from directly measuring the soft tissues (i.e. liver or kidneys) that would have been more suited to confirm the presence or absence of artificial polonium, as was found in the clothing of President Arafat.
- Finally, the "chain of custody" of the specimens contained in the bag cannot be documented between the death in November 2004 and their reception in Lausanne in February 2012. This was not the case for the specimens collected during the exhumation.

### VI.4. Toxicological investigations

The toxicological investigations of all of the biological specimens failed to highlight the presence of any exogenous substances other than those of therapeutic agents prescribed or administered to the patient before or during his hospitalization at the Military Hospital of Percy.

### VI.5. Radiological investigations

The radio-toxicological investigations demonstrated the presence of unsupported  $^{210}\text{Po}$  in the biological stains of President Arafat's personal effects and an unexpected high activity of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in several specimens from the remains after exhumation.



## VI.6. Pros and cons of a death originating from polonium-210 poisoning

In order to clarify our different findings and observations, Table 19 presents them in the context of two alternative propositions:

- $H_0$ : "The death was not caused by polonium-210 poisoning".
- $H_1$ : "The death was caused by polonium-210 poisoning".

**Table 19: Observed facts presented in the  $H_0$  /  $H_1$  propositions.**

Facts	Proposition $H_0$	Proposition $H_1$
The described symptoms have not been associated with a specific cause. (see Section III)	<ul style="list-style-type: none"> <li>• Not all the symptoms of acute radiation syndrome are present:               <ul style="list-style-type: none"> <li>○ No myelosuppression.</li> <li>○ No hair loss.</li> </ul> </li> </ul>	<ul style="list-style-type: none"> <li>• Myelosuppression and hair loss are associated with external radiation exposure, but not always with internal exposure.</li> <li>• Acute digestive symptoms remain unexplained.</li> <li>• Death occurred approximately one month after the onset of symptoms.</li> <li>• DIC and thrombocytopenia were documented.</li> <li>• No diagnosis had been obtained.</li> <li>• Sudden onset of symptoms after a meal in an apparent healthy individual</li> </ul>
Significant quantities of polonium-210 were found on the biological stains of the personal effects (see Section IV.3.7)		<ul style="list-style-type: none"> <li>• Unsupported polonium-210 was found on the stained personal effects.</li> <li>• A significant amount of supported polonium-210 was also present. It is compatible with the high activities of lead-210 found afterward in the body remains (see Section V.4.10.5).</li> <li>• Specimen adulteration is highly unlikely.</li> <li>• The other toxicological analyses were negative.</li> </ul>
Polonium-210 was measured in the body remains	<ul style="list-style-type: none"> <li>• All measured polonium-210 is supported by lead-210 (see Section V.4.10.1).</li> </ul>	<ul style="list-style-type: none"> <li>• The high activity of lead-210 could hide the presence of unsupported polonium-210. (see Section V.4.10.4)</li> </ul>
The observed activities of lead-210 in the remains are high and not homogenous. (see Section V.4.10.2)		<ul style="list-style-type: none"> <li>• The activities of lead-210 in the body are higher than all references analyzed.</li> <li>• The observed activities of lead-210 cannot be explained by chronic or long time intake.</li> <li>• This is coherent with an intake that occurred at the time of the first symptoms.</li> </ul>
The origin of the high lead-210 activity in the body remains has been investigated. (see Section V.4.10.3)	<ul style="list-style-type: none"> <li>• An unknown origin, totally unrelated to polonium poisoning cannot be ruled out.</li> <li>• A calculation performed with limited knowledge of the raw material and the methodology actually used to produce a polonium-210 estimated that the amount of lead-210 impurities would be too small to explain the values observed in the bones.</li> </ul>	<ul style="list-style-type: none"> <li>• Smoking and radium decay can be ruled out.</li> <li>• The amount of lead-210 impurity found in a marketed polonium-210 source was sufficient to explain the large activity of lead-210 observed in the bones.</li> </ul>
A surplus of lead-206 was present in the rib. (see Section V.4.10.6)	<ul style="list-style-type: none"> <li>• This measurement has a large uncertainty and may not be significant.</li> </ul>	<ul style="list-style-type: none"> <li>• This could be understood as a fossil signal of highly active polonium-210 source.</li> </ul>



Facts	Proposition H <sub>0</sub>	Proposition H <sub>1</sub>
A significant amount of radon was present in the grave.	<ul style="list-style-type: none"> <li>Lead-210 and polonium-210 are part of radon progeny.</li> </ul>	<ul style="list-style-type: none"> <li>Radon progeny appears to have been deposited relatively homogeneously. (see Section V.4.10.7)</li> <li>Radon alone is unable to explain the patterns of measured lead-210 and polonium-210.</li> <li>Most of the measured radioactivity is explainable by a source that was present within the body before death. (see Section V.4.10.8)</li> </ul>

In addition, the arguments in support of proposition H<sub>0</sub> are the following:

- The clinical description is not consistent with typical acute radiation syndrome, which includes bone marrow disruption (myelosuppression) and the loss of skin appendages (hair loss). However, myelosuppression and hair loss are essentially associated with external irradiation, but not necessarily with ingestion of radioactive substances. Furthermore, very little scientific literature exists on the acute exposure to polonium-210. For instance, no official documentation is available about the known case of Mr. Litvinenko at the time of this report.
- No significant amount of unsupported polonium-210 was measured in the body remains.

Alternatively, the arguments in support of proposition H<sub>1</sub> are the following:

- The acute onset of gastrointestinal symptoms with the progressive deterioration of the patient's general state is compatible with the ingestion of a large quantity of radioactivity.
- The development of thrombocytopenia and DIC has no known cause, but is compatible with an acute toxic pathology, possibly by the radioisotope polonium-210 (of which the acute toxicity in humans has been poorly described).
- The other toxicological investigations were negative.
- Significant quantities of unsupported polonium-210 were found on the biological stains of the personal effects. The smaller (but significant) quantity of supported polonium-210, also observed on these biological stains, is coherent with an impurity of lead-210 probably present in a poisonous polonium source.
- Despite the equilibrium between polonium-210 and lead-210, the activities are unexpectedly high in the specimens collected during exhumation of the corpse. Furthermore, this high activity could easily hide a remaining activity of unsupported polonium-210.
- The high activity of lead-210 measured in the body remains cannot be explained by smoking or radium-226 progeny.
- The high activity of lead-210 measured in the body remains is compatible with the amount of lead-210 impurities measured in a marketed source of polonium-210.
- The non-uniform distribution of lead-210 and polonium-210 is compatible with an acute intake occurring prior to the onset of the clinical symptoms.
- The lower lead-207/lead-206 ratios measured on two specimens with a high lead-210 and polonium-210 activity could result from the decay of past high polonium-210 activity.
- Radon-222 was present in the tomb, but does not explain the pattern and amount of lead-210 and polonium-210 measured in the body remains and the soils of the grave.



Taking into account the analytical limitations aforementioned, mostly time lapse since death and the nature and quality of specimens, the results moderately support the proposition  $H_1$ : that the death was the consequence of poisoning with polonium-210<sup>(8)</sup>.

## VI.7. Conclusion

On the evening of October 12<sup>th</sup>, 2004 President Yasser Arafat developed acute gastrointestinal symptoms eventually resulting in death one month later. His illness consisted of the persistence of the gastrointestinal symptoms, a rapid deterioration of his general state, development of severe thrombocytopenia with disseminated intravascular coagulation, cholestatic icterus and eventually intracerebral hemorrhage. The numerous clinical investigations performed prior to his death did not yield a diagnosis. The hypothesis that poisoning with polonium-210 was the cause of the clinical symptoms was proposed eight years after his death when new toxicological and radio-toxicological investigations were performed, demonstrating unexpectedly high levels of polonium-210 and lead-210 activity in many of the analyzed specimens.

Taking into account the analytical limitations aforementioned, mostly time lapse since death and the nature and quality of the specimens, the results moderately support the proposition that the death was the consequence of poisoning with polonium-210.



ALJAZEERA

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<sup>(8)</sup> This formulation has to be understood according to the *verbal probability scale* with the following 6 levels:

1. our results strongly support proposition  $H_0$ ;
2. our results moderately support proposition  $H_0$ ;
3. our results slightly support proposition  $H_0$ ;
4. our results slightly support proposition  $H_1$ ;
5. our results moderately support proposition  $H_1$ ;
6. our results strongly support proposition  $H_1$ .



...the results of the analytical findings demonstrated mostly that the level of lead and the ... the results of the analytical findings demonstrated mostly that the level of lead and the ... the results of the analytical findings demonstrated mostly that the level of lead and the ...

### VI.7. Conclusion

On the evening of October 12, 2004 President Yasser Arafat developed acute gastrointestinal symptoms resulting in death one month later. His illness consisted of the presence of the gastrointestinal symptoms, a rapid deterioration of his general state, development of severe hypotension with disseminated intravascular coagulation, chest pain and eventually neurological weakness. The numerous clinical investigations performed prior to his death did not yield a diagnosis. The hypothesis that poisoning with polonium-210 was the cause of the clinical symptoms was proposed eight years after the death when new toxicological and radio-toxicological investigations were performed demonstrating unexpectedly high levels of polonium-210 and lead-210 activity in some of the analyzed specimens.

Taking into account the analytical findings that were obtained, mostly three years after death and the nature and quality of the specimen, the results of the analytical findings support the proposition that the death was the consequence of poisoning with polonium-210.



# ALJAZEERA

The foundation was to be understood according to the actual probability scale with the following 5 levels:

- 1. our results strongly support proposition H1
- 2. our results moderately support proposition H1
- 3. our results slightly support proposition H1
- 4. our results do not support proposition H1
- 5. our results do not support proposition H1
- 6. our results do not support proposition H1





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## VII. Acknowledgements



# ALJAZEERA









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# X. Appendices

## X.1 Material and methods of the radiological expertise

## X.2 Pictures of personal effects

**ALJAZEERA**



## **X.1. Material and methods of the radiological expertise**

### **X.1.1 Security measures**

#### **X.1.1.1. Measurement of personal effects**

The fact that analyses were performed on President Arafat personal effects was kept secret to most of the IRA collaborators. A couple of hours before the release of the documentary from Al Jazeera (July 4<sup>th</sup>, 2012), the collaborators of IRA were informed via email.

#### **X.1.1.2. Collection of the specimens on the grave site**

The access to the grave was highly restricted and controlled by the Palestinian Authority. The grave itself was opened in the presence of representatives of the Palestinian Authority, the three involved laboratories (French, Russian and Swiss) and representatives of the French justice.

After the formal and registered reception of our specimens from the Palestinian Authority, they were sealed and transferred to Switzerland through diplomatic mailing by the Swiss mission in Ramallah. In Switzerland, the specimens were recovered in Bern at the Federal Department of Foreign Affairs and transported by a CHUV representative directly to Lausanne where they were stored in a restricted and secured area of the legal medicine department.

#### **X.1.1.3. Measurement of human remains and soil specimens**

For the radiological analyses, the specimens were transferred to IRA laboratories. A security guard was permanently on duty in front of the analytical laboratory door. The security exit door was permanently under alarm and movements' detectors were placed inside the analytical and the low levels counting laboratories. Alarm systems were connected to the Lausanne police department network. This insured that there was no manipulation of the specimens.

### **X.1.2 Sampling**

#### **X.1.2.1. Specimens of personal effects**

All the sampling was carried out with the aim to find contaminated specimens, thus explaining the choice of "stained" specimens. Cotton wool may contain a certain (unknown) amount of <sup>210</sup>Pb and <sup>210</sup>Po because of the large adsorption surface of cotton wool for atmospheric deposition during cultivation. Additionally, fertilizers contain a significant amount of <sup>210</sup>Pb and <sup>210</sup>Po.

Textiles specimens (2-3 g) were cut from underwear, Russian chapka, hospital hat, sportswear, keffiyeh, socks and others belongings present in President Arafat's travel bag. In some situations, careful cutting was performed around the stains. The specimens were chronologically analyzed in 14 batches. Descriptions and references are presented in Table 20.



**Table 20: Specimens of personal effects**

n° batch	n° CURML	n° IRA	Description
1	TOX 69357- 39	GRE-12-050	1 specimen of collar of a sportswear with dirt stains
	TOX 69357- 40	GRE-12-051	1 specimen from the front tigt of a pant (wool)
	TOX 69357-11-24	GRE-12-052	2 pills each of a mixture of medicines (about 38 g)
	TOX 69357- 44	GRE-12-053	2 specimens taken from underwear that appeared obviously worn (one with a urine stain)
2	TOX 69357- 44	GRE-12-053	6 other specimens from the stained underwear (3-4 g of cotton wool each)
3	TOX 69357- 49	GRE-12-054	1 specimen of hospital hat (cotton wool, around a blood stain, 0.7041 g))
	TOX 69357- 44	GRE-12-055	1 specimen from another underwear (visibly not worn)
	TOX 69357- 39	GRE-12-056	1 second specimen from the collar of the sportswear
	TOX 69357- 71	GRE-12-060	2 specimens made of the bristles of 2 toothbrushes (1 clearly used)
		GRE-12-061	1 reactants blank (H <sub>2</sub> SO <sub>4</sub> : 10 ml; HNO <sub>3</sub> : 25 ml; NH <sub>4</sub> OH: 40 ml, Fe: 20 mg)
4	TOX 69357- 44	GRE-12-062/063	2 specimens of a third underwear
	TOX 69357- 46	GRE-12-063	1 specimen from a kefieh taken around a stain (probably blood stain, 2.2540 g)
	TOX 69357- 35	GRE-12-065	1 specimen from a "Russian" chapka (internal band in contact with the head) worn by President Arafat on pictures taken shortly before his death
5		GRE-12-066/067/068	3 underwear (cotton) from 3 different IRA collaborators
6	TOX 69357-33		1 specimen from a sock possibly worn
	TOX 69357- 41	GRE-12-071	1 large piece from a pair of long johns (112.7529 g)
		GRE-12-073/074	2 new underwear bought directly from a shop (Bon Genie, Lausanne) of the brand Zimmerli
		GRE-12-075	1 underwear of the brand Hanro
7	TOX 69357- 49	GRE-12-076	1 specimen (far from the blood stain) of the hospital cap
	TOX 69357- 35	GRE-12-077	1 supplementary specimen of the Russian chapka
		GRE-12-078	1 specimen formed by the bristles of the toothbrush of an IRA collaborator
	TOX 69357- 41	GRE-12-079/080/081	3 specimens from a pair of long johns (taken along the leg)
8	TOX 69357- 50	GRE-12-083	1 specimen formed by 4 pieces of cotton wool taken from a child's drawing sampled around stains (possibly saliva, vomit and blood, total 1.4631 g)
	TOX 69357- 38	GRE-12-084/085	1 specimen taken from the interior and superior band (stained) of an old slipper
	TOX 69357-33	GRE-12-086	1 specimen from a new pair of socks (not worn, still attached by thread)
		GRE-12-087	1 reactants blank (H <sub>2</sub> SO <sub>4</sub> : 10 ml; HNO <sub>3</sub> : 25 ml; NH <sub>4</sub> OH: 40 ml, Fe: 20 mg)
9	TOX 69357- 39	GRE-12-101/102	2 more specimens from the collar of the sportswear
	TOX 69357- 39	GRE-12-103	1 specimen of the sportswear taken from the back, around a stain (possibly blood, 0.7565 g)
	TOX 69357- 35	GRE-12-104/105/106	3 more specimens from the interior band in contact with the skull of the Russian chapka
10	TOX 69357- 51	GRE-12-107/108	Smear of personal effects such as glasses, 1 blank smear
		GRE-12-109	1 specimen formed by the bristles of a toothbrush of an IRA collaborator
	TOX 69357- 49	GRE-12-110	1 specimen of the hospital cap with blood droplets
	TOX 69357- 50	GRE-12-111	1 specimen of the child drawing (out of the stains)
		GRE-12-112	1 reactants blank (H <sub>2</sub> SO <sub>4</sub> : 10 ml; HNO <sub>3</sub> : 25 ml; NH <sub>4</sub> OH: 40 ml, Fe: 20 mg)
11		GRE-12-315/316	Underwear IRA collaborator 3, 2 specimens
		GRE-12-317	Toothbrush, local supermarket, new
		GRE-12-318	Toothbrush, IRA collaborator 3, used
		GRE-12-319	Toothbrush, IRA collaborator 4, used



n° batch	n° CURML	n° IRA	Description
12		GRE-12-320/321/322	3 specimens from a T-shirt, IRA collaborator 3, left 10 years in an attic in plastic bag
		GRE-12-323/324/325	3 specimens from a shirt, IRA collaborator 3, left 10 years in an attic in plastic bag
13		GRE-12-333/334/335/336	4 specimens from an underwear pack (5 pieces), IRA collaborator 3, new from the market
		GRE-12-337	Toothbrush, IRA collaborator 2, used
14		GRE-12-340	6 specimens taken from a single underwear, IRA collaborator 3, new from the market

### X.1.2.2. Specimens of the grave

A Palestinian forensic medical investigator collected twenty specimens from President Arafat's corpse and grave during exhumation, in the presence of the three international teams. These specimens were: bones, shroud, tooth, scalp, abdominal and pleural altered tissues, and grave soil. Some specimens were shared between CURML and IRA. IRA specimens are described in Table 21.

**Table 21: Specimens of the grave analyzed at IRA.**

n° CURML	n° IRA	Description
18522	GRE-12-508	Altered tissues from the pleural cavity, mixed with soil
18646	GRE-12-509	Scalp
21095	GRE-12-510	Altered tissues from the abdominal cavity
54501	GRE-12-511	Altered tissues scraped from the femur (proximal)
54505	GRE-12-512	Altered tissues scraped from the shroud
54509	GRE-12-513	Altered tissues scraped from ribs
18603	GRE-12-514	Left ribs
18655	GRE-12-515	Vertebra, (thoracic)
19504 and 21096	GRE-12-516 and GRE-13-208/210/221	Iliac crest specimens
21519	GRE-12-517	Left femur (trabecular)
18641	GRE-12-518	Sternum
18658	GRE-12-519	Soil from the grave with black stain, close to the corpse
18635	GRE-12-520	Soil from the grave, reference, far from the corpse (right bottom corner)
18644	GRE-12-521	Soil from the grave, left of the body back
54504	GRE-12-522	Shroud from under the body
54505	GRE-12-523	Shroud from the left side of the thorax
54506	GRE-12-524	Shroud from the right leg

Unburied reference specimens consisted of vertebrae and ribs collected at autopsy. Vertebrae were obtained from individuals who died between 2006 and 2012. They were samples from the pathology institutes of Lausanne and Locarno, Switzerland. Ribs were obtained from the Institut Médico-Légal of Lyon, France. These samples were used to determine the  $^{210}\text{Po}$  reference value at the time of death for people aged 18 to 90 years (IRA data set, n=35). As they were not buried before sampling, they did not contain diagenetic  $^{210}\text{Po}$  [Schrag 2013]. We added 13 cases of forensic bones that were analyzed for  $^{90}\text{Sr}/^{210}\text{Po}$  to determine the post mortem interval [Schrag, 2013]. As these bones were recovered in nature and susceptible to diagenetic contamination, they were included as buried reference bones (CURML data set, n=12).



### X.1.3 Considered radionuclides

#### X.1.3.1. Natural chain of uranium-238

Among the natural radioactive chains present on earth, uranium-238 ( $^{238}\text{U}$ ) is of particular interest for this study. In its progeny is radium-226 ( $^{226}\text{Ra}$ ), which decays into radon-222 ( $^{222}\text{Rn}$ ). Because  $^{222}\text{Rn}$  is a noble gas, it can move rather easily through rock cracks or within the soil. It decays sequentially into a series of short-lived radionuclides until it reaches lead-210 ( $^{210}\text{Pb}$ ).

$^{210}\text{Pb}$  is radioactive with a relatively large half-life of  $22.23 \pm 0.12$  years [BIPM, 2008] (see Figure 26). It mainly decays by emitting a beta ( $\beta$ ) minus particle as well as a  $\gamma$  particle (46.5 keV), that is commonly used for its measurement by  $\gamma$  spectrometry. Its daughter product, bismuth-210 ( $^{210}\text{Bi}$ ), rapidly decays into polonium-210 ( $^{210}\text{Po}$ ).

$^{210}\text{Po}$  is also radioactive with a half-life of  $138.3763 \pm 0.0017$  days [BIPM, 2008]. It decays into the stable element of lead-206 ( $^{206}\text{Pb}$ ) by emitting an alpha ( $\alpha$ ) particle of 5.407 MeV as well as  $\gamma$ -particle of very low probability ( $p = 0.0000124$ ;  $E = 803$  keV).

When  $^{210}\text{Po}$  is obtained by the decay of  $^{210}\text{Pb}$ , one says that  $^{210}\text{Po}$  is supported by  $^{210}\text{Pb}$ .

Po208 2.93 y	Po209 1.0E2 y	Po210 1.4E2 d	Po211 25.5 s   516 ms	Po212 45.1 s   298 ns
Bi207 31.78 y	Bi208 2.58 ms   3.7E5 y	Bi209 100 1.9E19 y	Bi210 3.0E6 y   5.01 d	Bi211 2.17 m
Pb206 stable 24.1	Pb207 806 ms   stable 22.1	Pb208 stable 52.4	Pb209 3.25 h	Pb210 22.17 y

Figure 26: Relevant radionuclides for this study.  $^{210}\text{Pb}$  is generally a decay product of  $^{222}\text{Rn}$ .  $^{210}\text{Po}$  is the "grand-daughter" of  $^{210}\text{Pb}$ . It decays into the stable isotope of  $^{206}\text{Pb}$  by emitting an  $\alpha$  particle.

#### X.1.3.2. Artificial polonium-210

$^{210}\text{Po}$  can also be artificially produced in a nuclear reactor. The (almost) stable radionuclide bismuth-209 ( $^{209}\text{Bi}$ ) is exposed in high neutron flux for several months depending on the necessary activity. By absorbing a neutron,  $^{209}\text{Bi}$  becomes bismuth-210 ( $^{210}\text{Bi}$ ). Because of its relative small half-life,  $^{210}\text{Bi}$  rapidly decays into  $^{210}\text{Po}$  by emitting a  $\beta$  minus particle.  $^{210}\text{Po}$  is usually extracted from the raw material by distillation [Endebrook, 1955]. In this case,  $^{210}\text{Po}$  is therefore not the decay product of



$^{210}\text{Pb}$ . A sample containing only one such type of  $^{210}\text{Po}$  can, therefore, be described as *unsupported* by  $^{210}\text{Pb}$ .

#### **X.1.3.3. Polonium-209 tracer**

We used polonium-209 ( $^{209}\text{Po}$ ) as a tracer for our measurement of  $^{210}\text{Po}$ . We chose this radionuclide because it has the same chemistry and because it is also an  $\alpha$  emitter, although with different kinetic energies: 4.978 MeV and 4.979 MeV. The tracer is used to account for all of the losses during the radiochemical process leading to a measurable  $\square$  polonium source.

#### **X.1.3.4. Natural levels of polonium-210 and lead-210**

##### **A. Polonium-210 in fabric**

We were not able to find any reference activity values of  $^{210}\text{Po}$  or  $^{210}\text{Pb}$  in garment or clothing. Nevertheless, we know that some quantities should be naturally present because of the presence of the radon decay products on raw materials, e.g. cotton. After about 2 years, however, it can be expected that both radionuclides are in equilibrium (they have the same activities).

In order to have reference activities of  $^{210}\text{Po}$ , we performed measurements on specimens found in President Arafat's travel bag that were obviously not worn or were visually clean. We also collected clothing from our institute's collaborators that had been stored for several years in similar conditions to those of President Arafat's belongings. As will be shown in the results (see section IV.3.4), a typical (and natural) value of 3.5 mBq of  $^{210}\text{Po}$  could be expected in a fabric sample weighing approximately 3 g.

##### **B. Polonium-210 and lead-210 in bones**

Apart from our own [Schrag, 2012, Schrag, 2013], few publications exist mentioning activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in bone. The ones we are aware of are summarized in Table 22. The values in the literature are often given in Bq/kg of wet weight or dry weight and it is not easy to compare such results with ours, given in Bq/g Ca. ICRP states that a wet bone has a Ca content of 4.5%. If the bone is dry and does not contain residual organic matter, a ratio of 0.3 g Ca per g dry bone is a good estimate. We think that the best unit to report results of activity (or mass) in bones is Bq (or mg) per g Ca because it is not dependent on the state (decomposition) of the organic matter in the bones.



**Table 22: Typical activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  observed in bones and reported in the literature.**

Study	Source	Type of specimens	Activity range [mBq/g Ca]	
			$^{210}\text{Po}$	$^{210}\text{Pb}$
[NRPB, 2003]	Autopsy	13 persons	6.29 – 12.95	
		Mixed bones		
	Autopsy*	Mixed bones	8.77-16.7	
	Autopsy*	All body ash	3.7-4.93	
	Autopsy*	Central US	8.87	
	Autopsy*	Mixed bones		
Autopsy*	Germany	13.6		
	Central US	18.0		
Autopsy*	Mixed bones			
[Swift, 2001]*	Burial	15 (old) adults, Portugal left femurs	0.2 – 12.9	0.2 – 12.9
[Johnston, 2005]*	Autopsy	3 adults, Belgium (age 68-92), many different types of bones for each case		7.0 - 9.6
[Ziad, 2012]* <sup>&amp;</sup>	Burial	2 children, Morocco	5.0 - 5.5	5.0 - 5.5
	Burial	2 adults, Morocco archeological (~ 800 y)	6.3 - 9.8	6.3 - 9.8
	Burial	3 adults, Morocco	5.3 - 15.3	5.3 - 15.3
[Blanchard and Moore, 1970]	Autopsy	Ribs, 5 adults from Alaska	13 - 20	16 - 30
[Schrag, 2012]	Burial, not decontaminated	20 adults, Northern Italy vertebrae	25 - 62	
	Burial, not decontaminated, strong diagenesis	11 adults, Northern Italy vertebrae	77 - 346	
[Schrag, 2013]	Burial	12 adults, Switzerland	2.0 - 50	
	Autopsy	20 adults, Switzerland vertebrae	6.4 - 65	(1 case at 95)
	Autopsy	15 adults, France ribs	10 - 67	(1 case at 98)

\* Values presented in Bq per unit of mass. We divided by 0.3 in order to estimate the activity per unit of g Ca. & The authors actually measured  $^{210}\text{Po}$  and reported  $^{210}\text{Pb}$  by supposing that both nuclides were at equilibrium.

All of the activities of  $^{210}\text{Po}$  of buried specimens reported in the literature were below 25 mBq/g Ca. Our measurements [Schrag, 2013] performed after autopsy showed a larger activity range than the rest of the literature, with the highest value being almost 100 mBq/g Ca and the majority of data well below 50 mBq/g Ca.

The specimens measured in the studies of Swift *et al.* [2001] and Ziad *et al.* [2012] were performed on exhumed corpses, which were potentially contaminated by diagenetic  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  present in the soil because of the decay of  $^{222}\text{Rn}$ . Because these two studies did not take extensive actions to remove this diagenetic component, and because the reported activities are similar to the values measured after autopsy, we can conclude that the diagenetic component was not dominant.



Nevertheless, Schrag *et al.* [2013] demonstrated that the occurrence of diagenetic  $^{210}\text{Po}$  can produce, in some specific cases, values as high as 350 mBq/g Ca when bones are buried for as long as 20 years and measured without decontamination. Schrag *et al.* [2013] also demonstrated that these high activities can be lowered below 50 mBq/g Ca, by using a decontamination method and solubility profiling.

### C. Polonium-210 and lead-210 in soft tissues

**Table 23: Typical activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  observed in soft tissues and biota reported in the literature.**

Study	Source	Type of specimens	Activity range [mBq/g wet weight]	
			$^{210}\text{Po}$	$^{210}\text{Pb}$
[Takizawa, 1990]	Autopsy	Human liver	1.7 (0.6-2.5)	0.56 (0.3-0.86)
		Human kidney	1.2 (0.8-2.0)	0.43 (0.18-0.75)
		Human lung	0.37 (0.18-0.56)	0.24 (0.10-0.44)
[Blanchard, 1970]	Autopsy	Human liver, > 20y	0.8-9.2	0.52-1.15
		Human kidney, >20y	1.8-7.9	0.33-1.26
		Human lung, >20y	0.10-0.93	0.13-0.38
[Khan, 2011]		Crab muscle	37-69	4.9-8.9
		Crab hepatopancreas	896-1877	38-59
		Crab intestine	131-189	4.9-29
[Aközcan, 2013]		Fish, sardine	54 (14-400)	4.5 (LD-15)
		Fish, Atlantic mackerel	25 (LD-42)	2.5 (LD-5)
		Fish, red mullet	30 (LD-60)	< LD
[Musthafa, 2012]		Seaweeds	13-32	2-4.7
		Bivalves	96-113	25-97
		Prawns	42-61	0.7-1.4
		Fish	41-81	0.7-2
[Calvahlo, 1995]		Mussels	132 ± 5	2.6 ± 0.1

Study	Source	Type of specimens	Activity range [mBq/g dry weight]	
			$^{210}\text{Po}$	$^{210}\text{Pb}$
Ugur, 2002		Mussels	52-1344	6-167
[Sert, 2011]		Lichens and mosses		
		<i>Cladonia convulata</i>	400 (151-593)	246 (111-360)
		<i>Tortilla torturosa</i>	500 (310-785)	384 (253-589)
		<i>Hypnum lacunosum</i>	506 (493-518)	387 (355-419)
		<i>Hypnum cupressiforme</i>	286 (230-342)	208 (159-257)
[Gwynn, 2013]		Wild edible berries	0.6-2.6	0.8-4.1
		Mushrooms	4.7-198	2.5-3.3

Human wet tissues have low  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities, well below 10 mBq/g wet weight, and most of the time only reach 1 mBq/g wet weight. One would not expect a significant contribution of this residual activity to the total body burden in a case of poisoning. On the other hand, tissues of seafood products can have 100 times this activity.



#### D. Polonium-210 and lead-210 in soil

**Table 24: Typical activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  observed in soil and reported in the literature.**

Study	Source	Type of specimens	Activity range [mBq/g]	
			$^{210}\text{Po}$	$^{210}\text{Pb}$
[Özden, 2013]]	Turkey	Cultivated soils, n=72	28 [12-86]	26 [17-36]
		Uncultivated soils, n=72	39 [10-134]	35 [23-78]
[Jia, 2006]	China	n=8	36 [8-82]	37 [7-81]
[Aslani, 2005]	Turkey	n=41	41 (13-135)	
[Saidou, 2007]	Northern Cameroon	Uranium enriched area, n=20	81 (19-222)	78 (14-250)

$^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities in soil are between a few mBq to 250 mBq/g, with an average around 40 mBq/g. It is very difficult to compare one specific soil activity to another. In this case, soils were compared from different locations in the grave (e.g. black stained under the corpse compared to reference soil far from the corpse), assuming that the thin (10 cm) layer of soil in the grave on which the body laid had similar  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity due to similar  $^{226}\text{Ra}$  occurrence all over the grave soil.

### X.1.4 Measurements

#### X.1.4.1. Surface contamination monitoring of personal effects

Surface contamination measurements were performed using two surface contamination detectors, a LB-122 monitor (Berthold, Germany) and a CoMo-170 (S.E.A. GmbH, Germany). The CoMo-170 surface contamination detector can discriminate  $\alpha$  and  $\beta/\gamma$ . All personal effects were scanned manually for radioactive contamination. Although the instruments have been verified according to the requirements of the Swiss Institute of Metrology (METAS) for a long list of specific radionuclides, we only used them semi-quantitatively in their native measurement units: counts per second (cps).

#### X.1.4.2. In situ measurement of alpha/beta emitters

During the exhumation, we carried out  $\alpha$  and  $\beta/\gamma$  measurements on the grave soil and corpse using the CoMo-170 surface contamination monitor (S.E.A. GmbH, Germany), as described in Section X.1.4.1.

#### X.1.4.3. In situ measurement of equivalent dose $H^*(10)$

During the exhumation, ambient dose rate measurement were taken ( $H^*(10)$ ) using an Automess 6150AD-b/H (Automess – Automation und Messtechnik GmbH, Germany), which had been previously verified according to the requirements of METAS. This low dose rate instrument uses a cylindrical organic scintillator detector (diameter 3", height 3", and density 1.032 g/cm<sup>3</sup>). According to the manufacturer, the instruments range is between 1 nSv/h and 99.9  $\mu\text{Sv/h}$ .

#### X.1.4.4. In situ measurement of radon-222

Because  $^{222}\text{Rn}$  is ubiquitous in soil and might have generated a significant burden of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , it was necessary to measure the  $^{222}\text{Rn}$  concentration (in Bq/m<sup>3</sup>) in the grave before opening. This was



done on the evening of November 25<sup>th</sup>, 2012 by perforating a 12 mm diameter hole through the grave slab. After insertion of a Teflon® tubing, an AlphaGuard (Genitron Instruments, Germany) was connected and pumped inside the apparatus for 5 minutes until equilibrium was reached. The detector volume was then tightly closed and performed a measurement every minute for 15 minutes in order to allow for potential thoron gas (<sup>220</sup>Rn) decay.

#### X.1.4.5. Gamma spectrometry

##### A. Personal effects

For  $\gamma$  spectrometry measurements, the personal effects were pooled into 11 specimens. Each specimen was then measured over 24 hours using a p-type high purity germanium (HPGe) spectrometer coupled with the software Winner 6.0. Artificial radionuclides, such as cobalt-60 (<sup>60</sup>Co), caesium-134 (<sup>134</sup>Cs) and caesium-137 (<sup>137</sup>Cs) were investigated as well as <sup>210</sup>Pb.

##### B. Grave specimens

Selected specimens from the grave (3 soils specimens, scalp, rib, shroud (2x) and altered tissues (4x scraped from the femur, iliac crest and rib) were measured in a well type HPGe p-type spectrometer coupled with the analysis software Genie 2000 (Canberra, France) in 5 ml vial. The particular low-energy <sup>210</sup>Pb calibration was done using a <sup>210</sup>Pb reference solution (Isotope Product Laboratories (IPL), USA).

#### X.1.4.6. Analytical products

If not otherwise stated, all products used in this work were of analytical grade and in some occasion of Suprapur® quality from Merck, Fluka or Carlo Erba suppliers. Water used for dilution was made by using an SG Ultra Clear UV system (< 0.05  $\mu$ S/cm) and in some occasions by dilution with Ultrapur® water (Merck).

Glassware and Teflon® bakers were decontaminated before and after use in an ultrasonic bath containing 10% RBS, then washed in a washing machine specific for analytical glassware (Renggli WA Combi, Switzerland).

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#### X.1.4.7. Polonium-210 determination in textile, altered tissues (wax), scalp and shroud

A specimen of 2 to 3 g of textile (mostly cotton wool) or 0.2 to 1.1 g of shroud, altered tissues or scalp, was traced with  $50 \pm 0.3$  mBq of <sup>209</sup>Po then charred with 10 ml of concentrated H<sub>2</sub>SO<sub>4</sub> at about 70°C. The specimen was cautiously oxidized by adding 5 x 3 ml portions of concentrate 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 conditions for microwave mineralization were: initial pressure 50 bars; maximal temperature: 180°C, mineralization time: 30 minutes. If synthetic fibers were present, the solution was 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 or overnight at room temperature. The sources were counted (170 000 to 400 000 s) on a PIPS detector (silicon detector of 450 mm<sup>2</sup>) in an alpha spectrometer (Canberra Alpha Analyst, France). Our quality control process was validated by the annual PROCORAD interlaboratory comparison for the determination of <sup>210</sup>Po in urine (2008-2011, average bias: 4.2%).



#### X.1.4.8. Polonium-210 determination in soil

A specimen (1-2 g) of soil was traced with  $50 \pm 0.3$  mBq of  $^{209}\text{Po}$  and 20 ml of  $\text{HNO}_3$  8 M was added. The final removal of organic matter, as well as polonium leaching, was performed in a microwave digester under high pressure (Ultraclave IV, Milestone, Germany). The conditions for microwave mineralization were: initial pressure 50 bars; maximal temperature:  $180^\circ\text{C}$ , mineralization time: 30 minutes. The residue was filtered and polonium was co-precipitated from the supernatant solution with iron-hydroxides. After centrifugation, the precipitate was dissolved in 80 ml 1 M HCl, 500 mg of ascorbic acid was then added and the polonium spontaneously electrodeposited on a silver disc overnight at room temperature. The sources were counted (170 000 to 400 000 s) on a PIPS detector (silicon detector of  $450\text{ mm}^2$ ) in an alpha spectrometer (Canberra Alpha Analyst, France).

#### X.1.4.9. Polonium-210 determination in bones

The determination of  $^{210}\text{Po}$  accumulated in bone specimens through metabolic mechanism (thereafter biogenic) required the removal of  $^{210}\text{Po}$  coming from the contamination by the  $^{222}\text{Rn}$  progeny (thereafter diagenetic  $^{210}\text{Po}$ ). This was done after removal of the organic matter (mostly residual altered tissues) and solubility profiling: a fragment of bone (rib, vertebrae, sternum, femur and iliac crest were tested) was weighed in a beaker and covered by NaOH 1 M. The solution was heated up to  $60^\circ\text{C}$  during one hour. The saponification of residual grease and wax helps the solubilization of the organic matter. After rinsing with ultrapure water the fragment was introduced in a Teflon beaker for total mineralization in 10%  $\text{H}_2\text{O}_2$  in a microwave digester under pressure (Ultraclave IV, Milestone, Germany). The conditions for microwave mineralization were: initial pressure 50 bars; maximal temperature:  $180^\circ\text{C}$ , mineralization time: 30 minutes. After this treatment, the bone was perfectly white and the trabecular structure preserved. After drying in an oven at  $45^\circ\text{C}$ , the bone was grinded to a fine powder in a mortar and 1.0 g was transferred to a conical plastic test tube. After adding 10 ml of an acetate buffer (0.2 M, pH 4.5), the tube was placed in an ultrasonic bath for exactly five minutes. Afterwards the solution was centrifuged. The supernatant was filtered on a syringe ( $0.45\text{ }\mu\text{m}$  glass fiber filter, Milipore) in a 50 ml flask. This constituted the first acetate washing. The washing was repeated a second time and filtered in the same 50 ml flask. This flask was labeled: washing 1+2. The washing operations were repeated until we obtained two more 50 ml flask labeled: washing 3+4 and washing 5+6. One ml of each flask was sampled for calcium determination with ICP-OES (Perkin Elmer Optima 3300 DV). After the last centrifugation, the bone residue was dissolved in 80 ml 1 M HCl (1 ml taken off for calcium determination), then 500 mg of ascorbic acid was added and the polonium spontaneously electrodeposited on a silver disc overnight at room temperature. The sources were measured between 170 000 and 400 000 s with a PIPS detector (silicon detector of  $450\text{ mm}^2$ ) in an alpha spectrometer (Canberra Alpha Analyst, France). Concentrated HCl was added to the washings to obtain a solution HCl 1 M, and they were analyzed for  $^{210}\text{Po}$  as described above.

#### X.1.4.10. Supported polonium-210 determination

To precisely determine the quantity of  $^{210}\text{Po}$  present in the sample, which is due to the presence of  $^{210}\text{Pb}$ , it is necessary to first remove the residual  $^{210}\text{Po}$  from the solution, and wait at least for 3 months of  $^{210}\text{Po}$ -ingrowing in the sample before being able to measure  $^{210}\text{Po}$  in growth. We did this because the sensitivity of the  $\alpha$  measurement of  $^{210}\text{Po}$  (detection limit: about 0.5 mBq) is much higher than the sensitivity of the direct  $\gamma$  measurement of  $^{210}\text{Pb}$  (detection limit about: 0.2 Bq).



After depositing  $^{210}\text{Po}$  on an  $\text{Ag}^\circ$  disc, the solution was evaporated to dryness and concentrated  $\text{HNO}_3$  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  $[\text{PoCl}_6]^{2-}$ .  $^{210}\text{Pb}$  will pass into the elution solution, which was evaporated to dryness. The residue was dissolved in 80 ml 1 M HCl and left in the refrigerator for at least 3 months (35% re-growth of  $^{210}\text{Po}$  from  $^{210}\text{Pb}$ ). Afterwards, 50 mBq of  $^{209}\text{Po}$  was added and polonium was electrodeposited on a silver disc as before. The method was checked for consistency by measuring the co-precipitation yield of lead (stable) iron hydroxides ( $98 \pm 2\%$ ) 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  $^{210}\text{Pb}$  from  $^{210}\text{Po}$  was able to produce a pure  $^{210}\text{Pb}$  source without  $^{210}\text{Po}$ . To do this, three aliquots were spiked with 168 mBq of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  at equilibrium and Pb was separated from Po. No  $^{210}\text{Po}$  was found in the  $^{210}\text{Pb}$  fraction, which means that the method quantitatively separated  $^{210}\text{Pb}$  from  $^{210}\text{Po}$ . Using stable Pb and ICP-OES measurements, it was also confirmed that Pb was not extracted on the Dowex AG 1x8 column (extraction  $< 1\%$ ).

#### X.1.4.11. Determination of polonium-210 purity

We checked that the  $^{210}\text{Po}$  deposited on the  $\text{Ag}^\circ$  disk did not contain co-deposited  $^{210}\text{Pb}$ . To accomplish this, 21 sources (3 from Arafat's belongings, 18 from items taken from the grave) were remeasured three months after the first measurement. From the measurement at  $t=0$  and  $t=3$  months, the apparent half-life of  $^{210}\text{Po}$  deposited on the source was calculated to check its purity.

The 21 apparent half-life values are within 125 and 152 days, with a mean value of 143 days (target value = 138 days). This shows that the  $^{210}\text{Po}$  sources produced by the analytical methods are very pure and do not contain co-deposited  $^{210}\text{Pb}$  or other interfering radionuclides.

#### X.1.4.12. Isotopic ratio of lead-207/lead-206

The Institute of Radio- and Environmental Chemistry of the Paul Scherrer Institute (PSI) in Switzerland was contacted in order to measure isotopic lead ratios. The  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio was measured using their mass spectrometer (Element 2, Thermo Scientific).

Isotopic and  $\alpha$ -spectrometry measurements are both destructive. We were therefore not able to measure both quantities on the exact same specimens.

##### A. Sources preparations

All the reactants used in this experimental section were of Ultrapur or Suprapur quality, from Merk (Darmstadt, Germany). Before experiment, all the vessels (Teflon) were cleaned by immersion in 1 M  $\text{HNO}_3$  during one week. Lead was isolated from the bone matrix following the method described by Yoshinaga (1996). Briefly, 0.5-1.0 g of mineralized bone (see X.1.4.X.1.4.9) was dissolved in 5 ml concentrated  $\text{HNO}_3$  and the solution is heated to dryness. The residue was dissolved in 5 ml 0.5 M HBr and loaded on an anion exchange column (AG 1-x8, 100-200 mesh, 1 ml) preconditioned with water, 6 M HCl, water and 0.5 M HBr. The resin bed was washed with 5 ml 0.5 M HBr and the extracted lead was eluted with 10 ml 6 M HCl. After evaporation, the residue was dissolved in 1%  $\text{HNO}_3$  prior to mass spectrometry.



Soils specimens (0.5 g) were leached by two fractions of 8 M HNO<sub>3</sub> (5 ml) at 80°C during one hour. After centrifugation and evaporation, the residue was treated as for bone specimens.

Scalp specimen was mineralized in a mixture of concentrated H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub>. After evaporation (not total for H<sub>2</sub>SO<sub>4</sub>), the residue was dissolved in 5 ml 0.5 M HBr and treated as for soils specimens.

#### B. Sources measurement

The specimens were analyzed in PSI (Paul Scherrer Institute, Villigen, Switzerland) using an Element 2 ICP-SFMS (Thermo Scientific). Measurements were referenced to the isotope scale using NIST certified reference material NBS-Pb-981 and another internal laboratory standard. Because the level of lead is very low, we focused the analysis on the <sup>206</sup>Pb/<sup>207</sup>Pb ratio only.

#### X.1.4.13. Measure of fission product: strontium-90

<sup>90</sup>Sr is a bone-seeking radionuclide produced by the fission of <sup>235</sup>U. Thus <sup>90</sup>Sr might be an impurity present in a <sup>210</sup>Po source made by irradiation of a <sup>209</sup>Bi source in a fission reactor. We thought it necessary to check for the presence of this radionuclide in the bones specimens in the grave. The measurements were carried out on an aliquot of the vertebra and femur specimens.

<sup>90</sup>Sr was analyzed through its daughter product, <sup>90</sup>Y, which is in secular equilibrium with <sup>90</sup>Sr in bones [Froidevaux, 2010]. After addition of 10 mg of Y carrier, the mineralization of 2 to 5 g of bone was performed in a microwave digester under pressure in 8M HNO<sub>3</sub> (Ultraclave IV, Milestone, Germany). The conditions for microwave mineralization were: initial pressure 50 bars; maximal temperature: 180°C, mineralization time: 30 minutes. The acidic solution was diluted to 300 ml and oxalic acid added. The precipitation of alkaline-earth cations was initiated by adding 25% NH<sub>4</sub>OH. The oxalate precipitate was mineralized in concentrated HNO<sub>3</sub> in a microwave digester under pressure. The acidic solution was diluted to 300 ml with water and the pH adjusted to 3. Yttrium was extracted on a 10 ml yttrium-specific ion-imprinted polymer [Chauvin, 2006]. After elution of yttrium from the extraction resin, yttrium oxalate was precipitated and filtered out. The source was counted in a low level proportional counter (Canberra Tennelec LB 4100w, France).

#### X.1.4.14. Computation of lead-210 as an impurity in the fabrication of source of polonium-210

Stable lead (isotopes <sup>206</sup>Pb, <sup>207</sup>Pb and <sup>208</sup>Pb in Figure 26) is probably always present in <sup>209</sup>Bi. For instance, high purity <sup>209</sup>Bi sold by the manufacturer Sigma-Aldrich is advertised with 99.99% purity and a maximal metal content of 100 ppm, including lead [Sigma-Aldrich].

In the present expertise, we wanted to know if <sup>210</sup>Pb could be produced during the neutron irradiation of <sup>209</sup>Bi performed during the fabrication of <sup>210</sup>Po. We, therefore, asked the team of Prof. Andreas Pautz from the Laboratory for Reactor Physics and System Behavior at EPFL, Lausanne, Switzerland to estimate the quantity of <sup>210</sup>Pb that could be present in the fabrication of <sup>210</sup>Po. The computation was performed at the Paul Scherrer Institut (PSI) and the final report was established on August 20<sup>th</sup>, 2013 [Pecchia, 2013]. They assumed several quantities of impurity of stable <sup>208</sup>Pb that could naturally be present within a source of <sup>209</sup>Bi and calculated the impurity factor of <sup>210</sup>Pb and <sup>210</sup>Po versus the duration of the neutron irradiation and for different types of neutron fluxes. As can be deduced from Figure 26, this involves the transmutation of <sup>208</sup>Pb into <sup>209</sup>Pb by absorption of one neutron, followed by another neutron absorption to transform <sup>209</sup>Pb into <sup>210</sup>Pb. The cross sections involved, as well as the half-life of <sup>209</sup>Pb, are relatively small. Orders of magnitudes of the impurity



factor ( $^{210}\text{Pb}/^{210}\text{Po}$ ) are displayed in Table 25. Although  $^{210}\text{Pb}$  is actually produced, the quantities appear to be small.

**Table 25:** Order of magnitude of the impurity factor ( $^{210}\text{Pb}/^{210}\text{Po}$ ) computed according to [Pecchia, 2013] after about 700 days of irradiation.

Proportion of $^{208}\text{Pb}$ present in the $^{209}\text{Bi}$ source	Impurity factor ( $^{210}\text{Pb}/^{210}\text{Po}$ )
0.001%	$10^{-15} - 10^{-14}$
0.01%	$10^{-14} - 10^{-13}$
0.1%	$10^{-13} - 10^{-12}$
1%	$10^{-12} - 10^{-11}$
5%	$10^{-11} - 10^{-10}$
10%	$10^{-11} - 10^{-10}$
25%	$10^{-11} - 10^{-9}$
50%	$10^{-10} - 10^{-9}$
80%	$10^{-10} - 10^{-8}$
95%	$10^{-9} - 10^{-7}$

#### X.1.4.15. Lead-210 as impurity in a marketed source of polonium-210

On September 10<sup>th</sup>, 2013, we received a  $^{210}\text{Po}$  source (Czech Metrology Institute, see Figure 27) of 2.91 MBq of activity in 2M HCl (4.9310 g), with no carrier added.

This source was measured as received during 86 400 s by  $\gamma$  spectrometry in a well geometry with a p-type HPGe detector (GCW4523, 45% relative efficiency) from Canberra (France) and the spectrum treated with the APEX GENIE 2000 software. Afterwards, the vial was opened in a hot laboratory (B-cell gloves box) to proceed with the radiochemical separation of  $^{210}\text{Pb}$  from  $^{210}\text{Po}$ : 1.5 ml of the solution was made 8 M HCl by adding 3.5 ml of concentrated HCl. This solution was passed through a 2 ml AG 1x 4 (100-200 mesh) anionic extraction resin conditioned by 10 ml of 9 M HCl (Bio-Rad econo-column). Polonium is extracted as  $[\text{PoCl}_6]^{2-}$  complex while lead is not retained. The column was washed with 10 ml 9 M HCl. All the eluates were conserved and slowly evaporated on a hot plate. The separation was carried out two more times, afterwards an aliquot of 0.5 ml was collected from the 15 ml elution solution to determine the level of residual  $^{210}\text{Po}$ . 0.5 mg of  $\text{Fe}^{3+}$  was added to the solution and iron hydroxides precipitated by adding 25%  $\text{NH}_4\text{OH}$ . The iron hydroxides precipitate was filtered off on 0.22  $\mu\text{m}$  filter (Millipore GSWPO2500 0.22  $\mu\text{m}$ ) and the filter counted during 90 times 4 hours in a low-level proportional counter (Tennelec LB 4100w, Canberra, France).

What remained of the initial source (3.5 ml) was sealed again in a glass vial and measured once again during 450 000 s by  $\gamma$  spectrometry in a well geometry. After counting, a second 1.5 ml aliquot of the source was treated as before to obtain a second filter that was also measured by  $\beta$  proportional counting.





CZECH METROLOGY INSTITUTE  
INSPECTORATE FOR IONIZING RADIATION

Radiová 1, 102 00 Praha 10



## CERTIFICATE

Cert. No.: 9031 - OL - 501/13

Type: ER X

Prod. No.: 280613-1430001

Radionuclide: Po-210

Half life: 138.4 days

Activity: 2910 kBq

Specific activity: 590,2 kBq /g

Mass: 4,9310 g

Combined standard uncertainty: 0,9 %

Radioactive impurities: -

Reference date: 12.9.2013

Measuring method:

The ER 2 and ER 25 standards are directly prepared from a standard solution activity of which was determined by absolute method, specific activity of ER 3, ER 1 and ER X is calculated from dilution ratio and specific activity of ER 2 solution. The activity of the standard marked with suffix K is determined by comparison with IIR standards on gamma ionization chamber.

Chemical composition:

PoCl<sub>4</sub> in 2 M HCl, carrier none added

Date of the certificate issue: 5.9.2013

Certificate validity: 2 years

Customer: CHUV

Rue du Grande-Pré 1  
1007 Lausanne  
Switzerland



Control: RNDr. R. Bludovský, CSc.

Ing. Jiří Surán, MBA  
director

Phone: +420 266020497 Fax: +420 266020466

Figure 27: Certificate of the marketed source of <sup>210</sup>Po measured to estimate its impurities.



#### X.1.4.16. Measurement uncertainties

Measurement uncertainties are fundamental for any comparison between a suspected value and a given reference level. We report our typical uncertainty evaluation in Table 26.

Table 26: Typical measurement uncertainties evaluated in the present study.

Type of specimen	Type of measurement	Range of value	Expanded uncertainty (k=2) in %
Fabric and other items found in the personal belonging	$^{210}\text{Po}$ activity by $\alpha$ spectrometry	Above 10 mBq	5%
		Around 5 mBq	10%
		Around 0.5 mBq	40%
Soil	$^{210}\text{Po}$ activity by $\alpha$ spectrometry	Above 10 mBq	5%
		$^{210}\text{Pb}$ activity by $\gamma$ spectrometry	Above 100 mBq/g
		Around 50 mBq/g	25%
Bones	$^{210}\text{Po}$ activity by $\alpha$ spectrometry	Above 200 mBq/g Ca	2%
		Around 50 mBq/g Ca	4%
	$^{210}\text{Pb}$ activity	100-200	20%
Soft tissues & shroud	$^{210}\text{Po}$ activity by $\alpha$ spectrometry	Above 10 mBq	5%
		$^{210}\text{Pb}$ activity	500-1000
Marketed $^{210}\text{Po}$ source	$^{210}\text{Pb}$ activity by $\gamma$ spectrometry	200-300 mBq	13%
	$^{210}\text{Pb}$ activity by $^{210}\text{Bi}$	200-300 mBq	12%

#### X.1.5 Biokinetic modeling for the intake of radionuclides

The uptake and retention of radionuclides in organs and tissues after intake is described by compartmentalized biokinetic models of the human body. Such biokinetic models have been proposed by ICRP for many radionuclides. They heavily depend on reliable biokinetic data that are hard to obtain and that are averaged across the human subjects, taking into consideration and/or extrapolated from laboratory animals. For  $^{210}\text{Po}$ , large amount of data are available from animal studies as well as from subjects exposed by inhalation at the workplace [Leggett, 2001]. Only limited data are available for ingestion by humans and come mainly from controlled studies on seriously sick volunteers whose conditions may have modified the biokinetics of polonium.

It should be mentioned that biokinetic models proposed by ICRP were developed for radiation protection purposes and apply to average healthy humans. In cases of acute lethal intakes of  $^{210}\text{Po}$ , the use of normal biokinetics may be inappropriate since severe tissue damage may have modified the behavior of polonium in the body. On the other hand, very few human data related to life-threatening intakes of  $^{210}\text{Po}$  are available: essentially one case of inhalation in which a Russian worker died [Harrison, 2007] and none of ingestion. It is unfortunate that the scientific data about the most famous case of Alexander Litvinenko have not been released by the British courts.



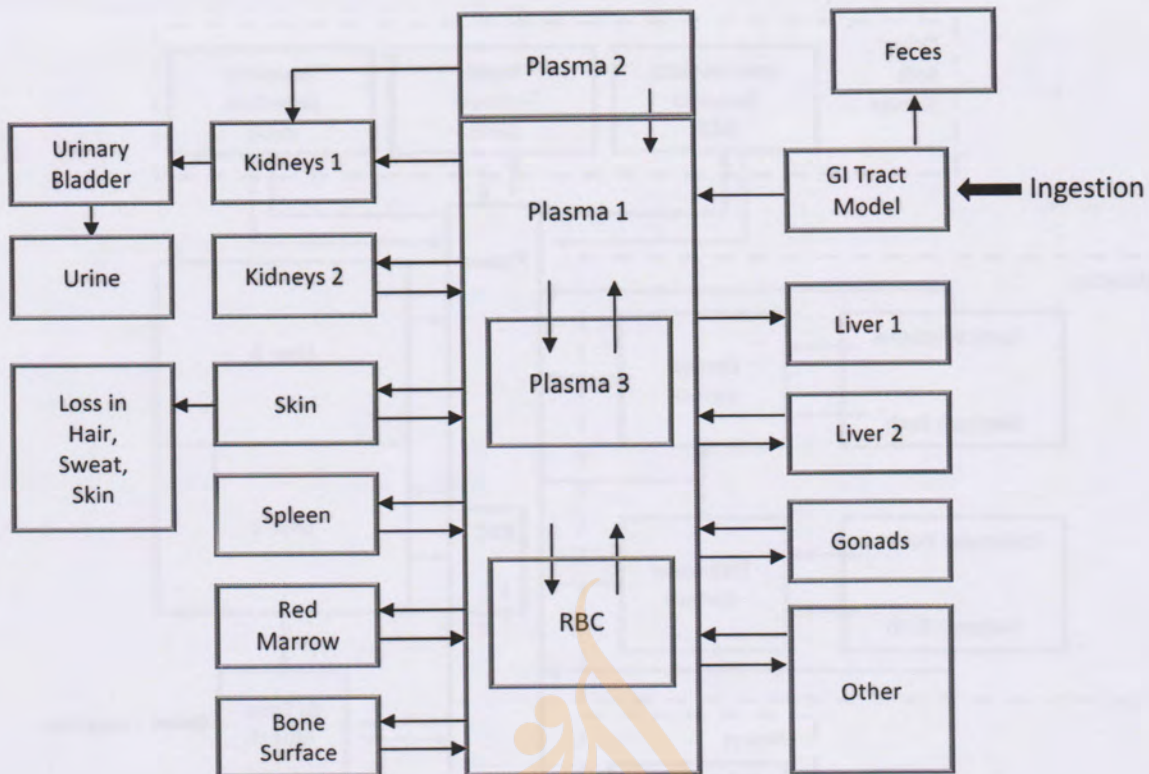


Figure 28: Compartments of the systemic model of polonium and connection with the model of the gastrointestinal tract.

#### X.1.5.1. Biokinetic model of polonium-210

We implemented the systemic biokinetic model of polonium proposed by Leggett and Eckerman [Leggett, 2001] (see Figure 28). This model is used here as an update of the model from the ICRP 67 Publication (ICRP, 1992). It has been implemented in the simulation modeling tool Ecolego (Ecolego, 2011), as well as SAAM II (Barrett, 1998), in order to calculate the typical retention of  $^{210}\text{Po}$  in organs and tissues in case of poisoning. In the latter situation, ingestion is the most probable route of intake, and we coupled the systemic biokinetic model to the human alimentary tract model described in ICRP Publication 30 (ICRP, 1979), as well as the revised version of ICRP Publication 100 (ICRP, 2006). Absorption of  $^{210}\text{Po}$  is assumed to occur exclusively from the small intestine and is characterized by the fractional intestinal absorption  $f_1=0.1$  for inorganic form. The  $f_1$  value is defined as the fraction of the activity leaving the stomach that is subsequently transferred to blood by absorption from the small intestine. Using this model, the daily urinary excretion, i.e. the fraction of the intake expected to be excreted in 24 hour urine after acute ingestion of  $^{210}\text{Po}$ , was determined at various times following intake.

#### X.1.5.2. Biokinetic model of lead-210

We also implemented the systemic biokinetic model of lead described in ICRP Publication 67 (ICRP, 1992) (see Figure 29) in the modeling software SAAM II (Barett, 1998) in order to calculate the typical retention of  $^{210}\text{Pb}$  in bones. Unlike polonium that is distributed mainly in soft tissues, lead is known to accumulate mainly in bone. The systemic biokinetic model of lead was coupled to the gastrointestinal tract model of ICRP 30 (ICRP, 1979). Absorption of lead from the small intestine to blood is characterized by  $f_1=0.2$ .



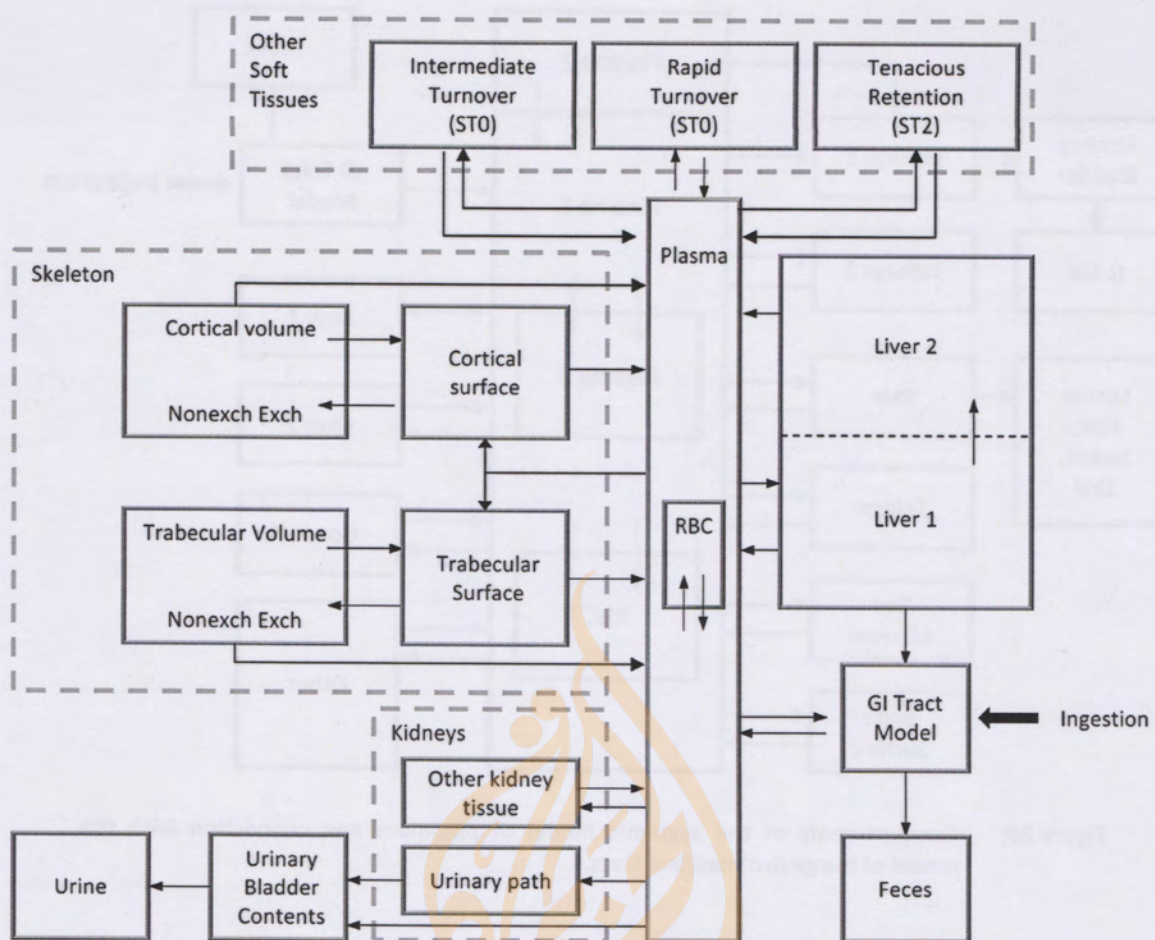


Figure 29: Compartments of the systemic model of lead according to ICRP Publication 67 (ICRP, 1992).

### X.1.5.3. Coherence of the calculations

The agreement of retention in organs computed with the softwares Ecolego and SAAM was excellent (compatible within 1%). In addition, results obtained either with the ICRP 30 or ICRP 100 models were very similar, with differences lower than 1% for all compartments of the models, except for the organs from the gastrointestinal tract model during the first days after intake.



## X.2. Pictures of personal effects

On the 6<sup>th</sup> February 2012, a travel bag was received at the University center of legal medicine, Lausanne-Geneva. Its content is pictured below in Figure 30 to Figure 94.



Figure 30: Travel bag.



Figure 31: 13 pairs of socks (TOX 69357-33).



Figure 32: 1 pair of gloves (TOX 69357- 34).





Figure 33: 3 fur hats  
(TOX 69357- 35).



Figure 36: 3 pairs of slippers  
(TOX 69357- 38).



Figure 34: 1 scar tissue  
(TOX 69357- 36).



Figure 37: Running suit  
(TOX 69357- 39).



Figure 35: 1 cap  
(TOX 69357- 37).



Figure 38: 1 underwear (polo  
and leggings)  
(TOX 69357- 40).





Figure 39: Underwear (1 polo and 3 long underwears) (TOX 69357- 41).



Figure 42: 9 men's underwear (TOX 69357- 44).



Figure 40: 1 underwear (polo) (TOX 69357- 42).



Figure 43: 2 plastrons (TOX 69357- 45).



Figure 41: 1 small towel (TOX 69357- 43).



Figure 44: 1 chech (TOX 69357- 46).



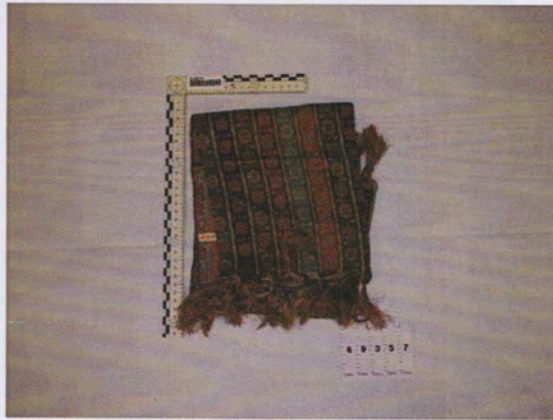


Figure 45: 1 scarf (TOX 69357- 47).



Figure 48: 1 scar tissue with child drawing (TOX 69357- 50).



Figure 46: 1 woolly hat (TOX 69357- 48).

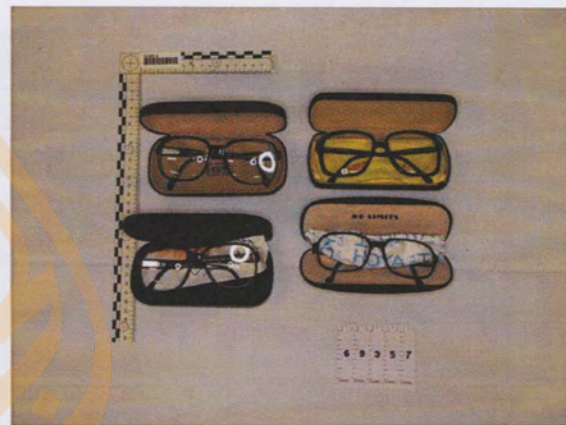


Figure 49: Pairs of glasses (TOX 69357- 51).

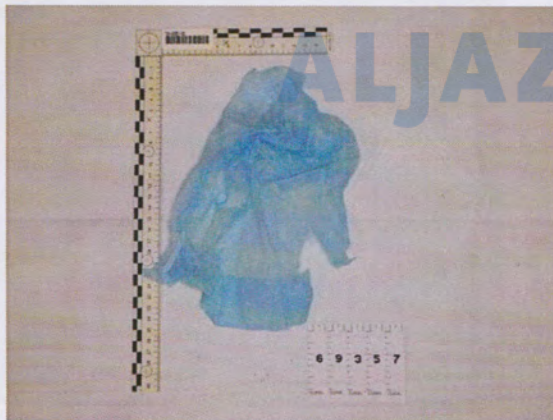


Figure 47: 1 hospital hat (TOX 69357- 49).



Figure 50: 3 boxes containing pins (TOX 69357- 42).



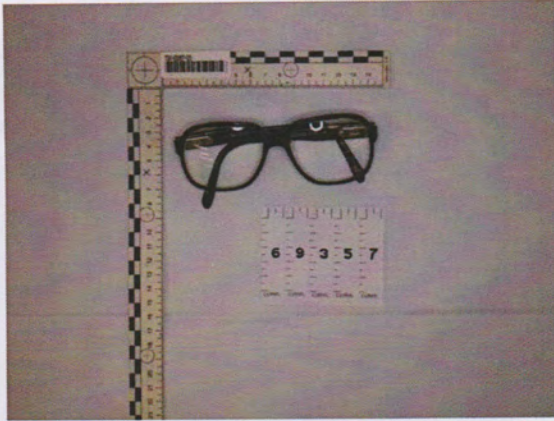


Figure 51: 1 pair of glasses  
(TOX 69357- 53).

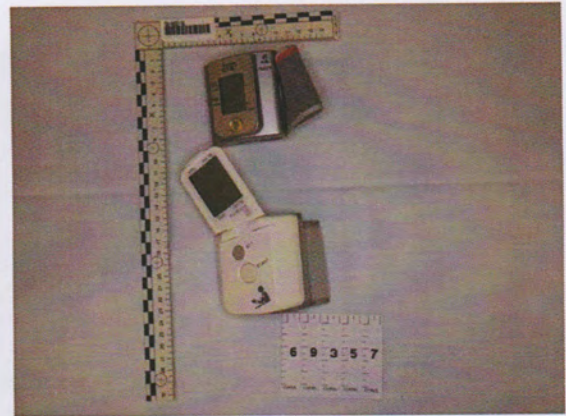


Figure 54: 2 medical devices  
(TOX 69357- 56).



Figure 52: 2 bracelets/rosary  
(TOX 69357- 54).

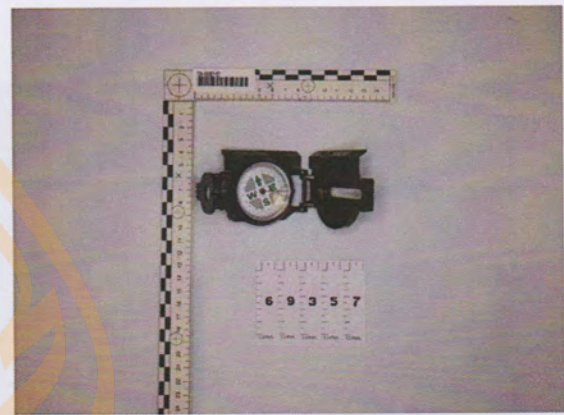


Figure 55: 1 compass  
(TOX 69357- 57).



Figure 53: 2 instruments for  
massage  
(TOX 69357- 55).



Figure 56: 2 digital doorbells  
(TOX 69357- 58).



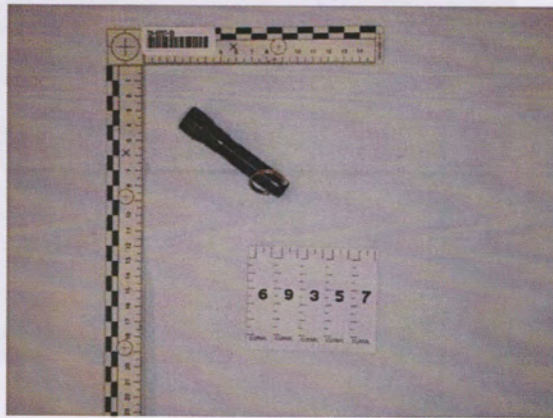


Figure 57: 1 small flashlight  
(TOX 69357- 59).



Figure 60: 1 toilet bag  
(TOX 69357- 62).



Figure 58: 1 pill-box  
(TOX 69357- 60).

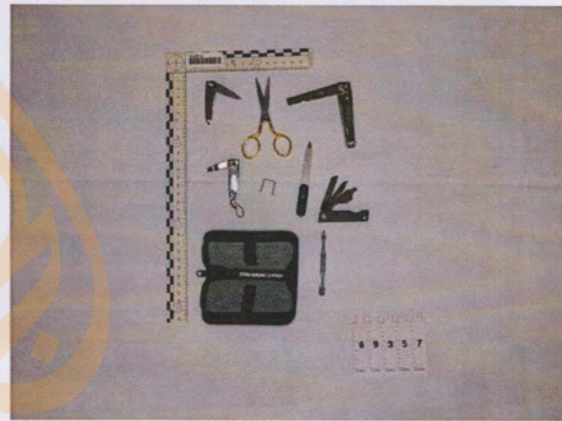


Figure 61: 1 manicure set  
(TOX 69357- 63).

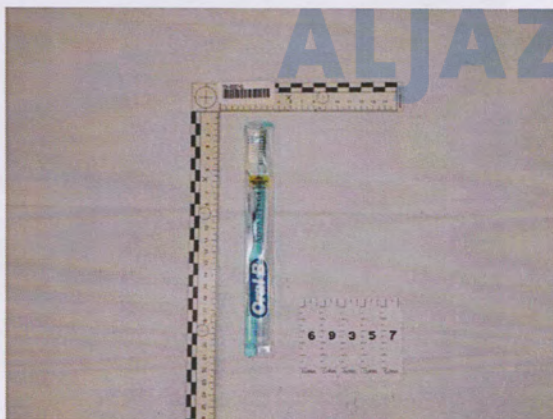


Figure 59: 1 toothbrush  
(TOX 69357- 61).



Figure 62: 2 bags (tissue)  
(TOX 69357- 64).





Figure 63: 1 small bag (tissue with ear taper fit (TOX 69357- 65)).

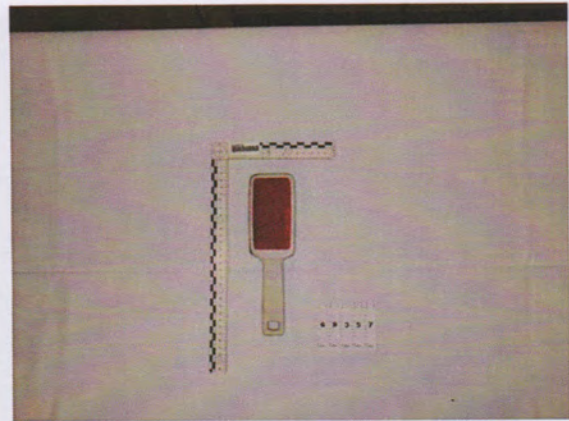


Figure 66: 1 small clothesbrush (TOX 69357- 68).



Figure 64: 1 box of matches (TOX 69357- 66).



Figure 67: 2 coins (TOX 69357- 69).



Figure 65: Travelling sewing case (TOX 69357- 67).



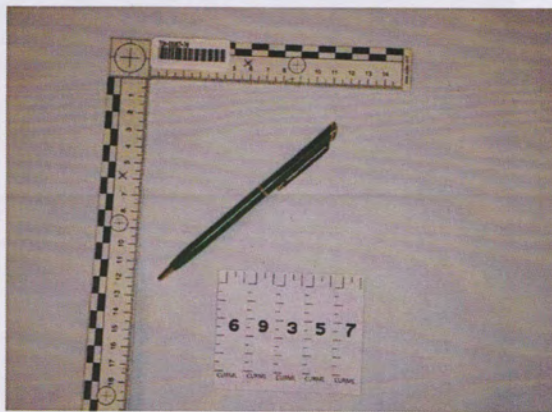


Figure 68: 1 pen  
(TOX 69357- 70).

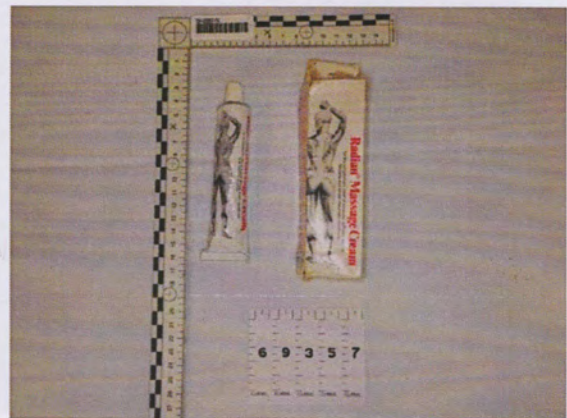


Figure 71 1 of tube massage cream  
with box  
(TOX 69357- 73).

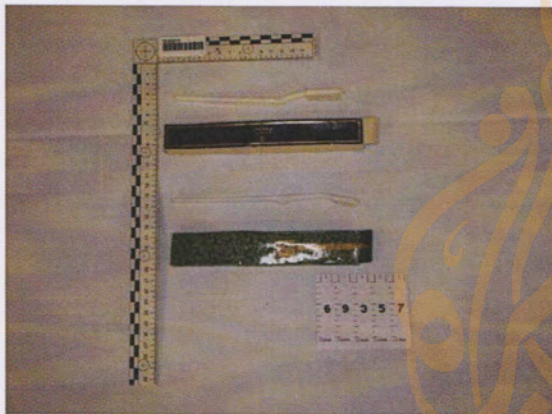


Figure 69: 2 toothbrushes  
(TOX 69357- 71).

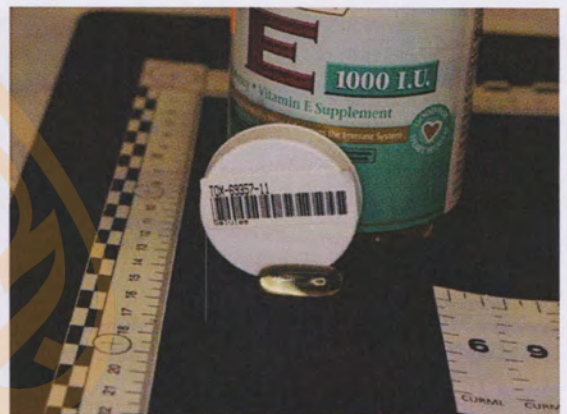


Figure 72: 1 box of capsules "E - Vit  
E - 1000 I.U. -  
Memeber's Mark"  
(TOX 69357-11).



Figure 70: 3 packs of padding ("Corn  
cushions")  
(TOX 69357- 72).



Figure 73: 1 box of capsules "Black  
seed"  
(TOX 69357-12).





Figure 74: 2 boxes of capsules "Baraka" (TOX 69357-13).



Figure 77: 1 box of tablets "Strepsils" (TOX 69357-16).



Figure 75: 1 box of nasal spray "Flixonase" (TOX 69357-14).

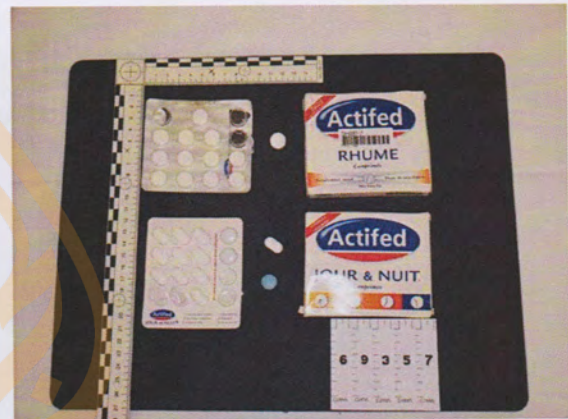


Figure 78: 2 boxes of pills "Actifed - rhume" / " Actifed - Jour&Nuit" (TOX 69357-17).

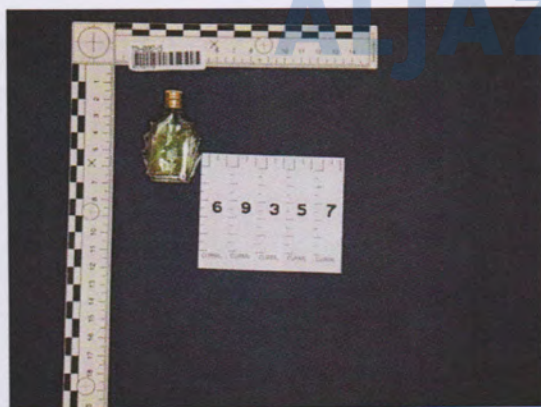


Figure 76: 1 small bottle containing a green liquid (TOX 69357-15).

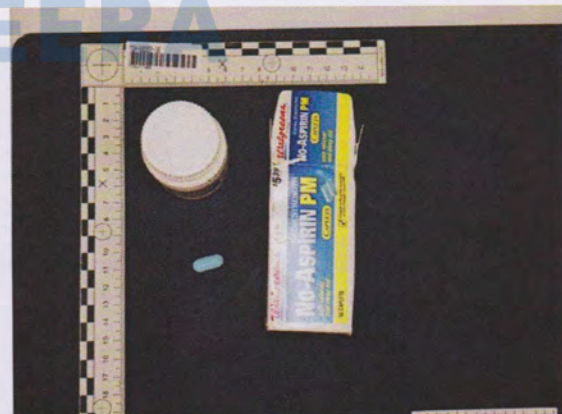


Figure 79: 1 box of pills "No-Aspirin PM" (TOX 69357-18).





Figure 80: Box of tablets "Efferalgan" (TOX 69357-19).

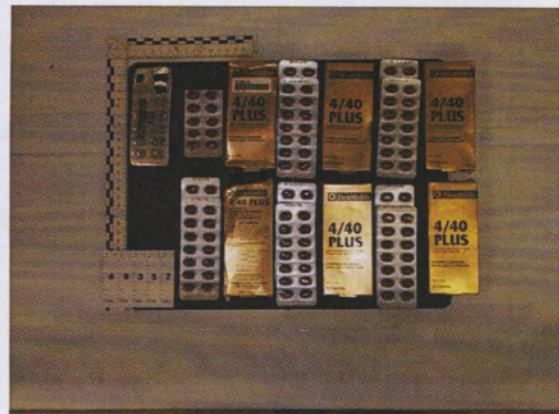


Figure 83: 6 boxes of pills "4/40 plus - Healthilife - Vit E, Ginseng, royal jelly & pollen" (TOX 69357-22).



Figure 81: 1 unopened box of pills "Stresstabs" (TOX 69357-20).



Figure 84: 2 boxes of tablets "Plenyl - Vitamines & oligo elements - Oberlin" (TOX 69357-23).



Figure 82: Unopened box of pills "Gincata" (TOX 69357-21).



Figure 85: 3 boxes of tablets "Unified - clears blocked and runny noses - United Pharmaceutical Mfg Co Ltd" (TOX 69357-24).



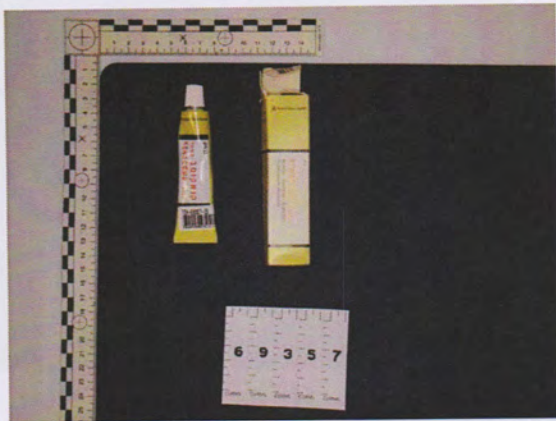


Figure 86: 1 box of ointment "Kenacomb - Bristol Myers Squibb" (TOX 69357-25).

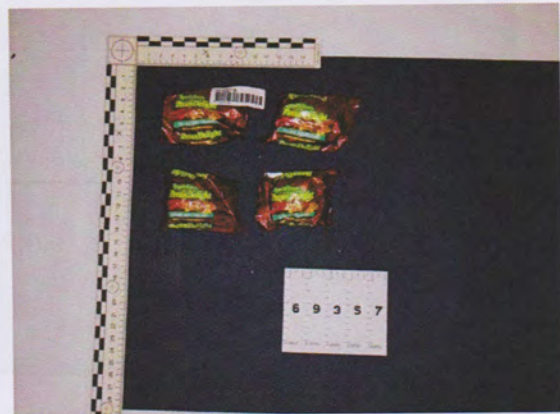


Figure 89: 4 units of cookies "Pecan Delight" (TOX 69357-28).



Figure 87: 1 box of tablets "Vicks - cooling & soothing - Vit C" (TOX 69357-26).

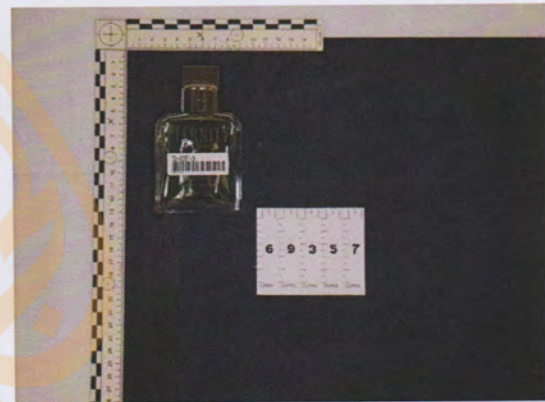


Figure 90: 1 bottle of perfume "Eternity" (TOX 69357-29).



Figure 88: 1 unopened box of herbal tea "Zallouh root - Ferula harmonis" (TOX 69357-27).

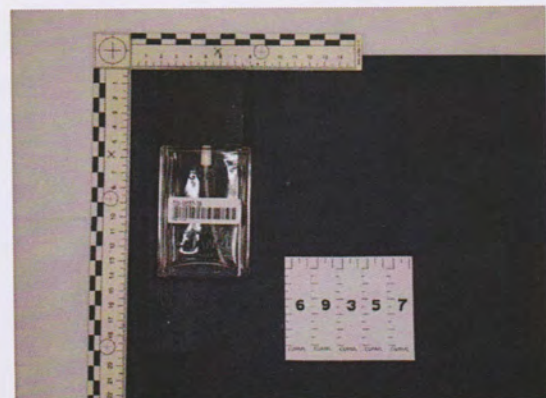


Figure 91: 1 bottle of perfume "Egoïste - Chanel" (TOX 69357-30).



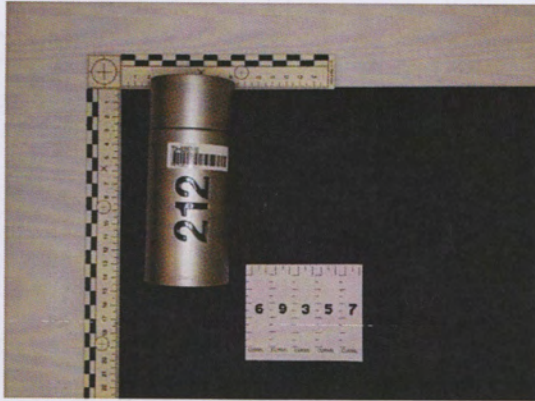


Figure 92: 1 bottle of perfume "212 - Man" (TOX 69357-31).

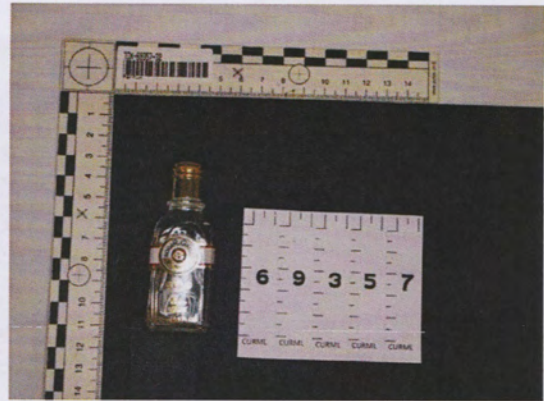


Figure 93: 1 bottle of perfume "Eau de Cologne - Roger Gallet" (TOX 69357-32).



Figure 94: 3 packs of plaster (TOX 69357- 74).

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ALJAZEERA