



# The Radiological Accident in Tammiku



THE RADIOLOGICAL ACCIDENT  
IN TAMMIKU

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# THE RADIOLOGICAL ACCIDENT IN TAMMIKU

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## FOREWORD

On 21 October 1994, three brothers entered the radioactive waste repository at Tammiku, Estonia, without authorization and removed a metal container enclosing a radiation source. This action initiated the sequence of events in a radiological accident.

The accident resulted in the death of one person and injury to a number of others. The death was not originally attributed to radiation exposure. However, a physician who examined the injuries of the stepson of the dead person realized the radiological nature of the accident and initiated the rescue actions that limited the extent of the consequences.

The Estonian authorities requested international assistance to analyse the accident and to advise on remedial actions. Because of this request, it was possible to learn lessons from the accident's consequences and to advise measures for future accident prevention.

The IAEA wishes to express its gratitude to the Estonian authorities for their help in enabling others to benefit from the lessons to be drawn from the accident.

The IAEA officers responsible for this publication are P. Ortiz-López and I. Turai.

#### *EDITORIAL NOTE*

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# 1. INTRODUCTION

## 1.1. BACKGROUND

The use of radiation sources in industrial, medical and other applications is widespread in both developed and developing countries. An important function of the IAEA is the provision of support and assistance to Member States in establishing the infrastructure necessary to ensure the safety of the sources, i.e. a system for regulatory control and staff training. In recent years there have been several serious accidents associated with industrial uses of radiation, and the study and analysis of these accidents by the IAEA have provided useful lessons on the consequences of deficiencies of control, and on methods of safety improvement. The IAEA has already prepared reports of the fatal accidents in El Salvador [1], Israel [2], Viet Nam [3] and Belarus [4], where failures of safety interlocking systems in complex applications and/or inadequate training were the main causes of injury to operators. In the accident in Goiânia [5], the cause was more complex and members of the general public were affected. That accident also gave rise to extensive environmental contamination.

With the dissolution of the USSR, the governmental infrastructure, including the regulatory structure for ensuring radiation protection and waste management, had to be adapted to the new States, a process that will take many years to complete. The change to a market economy has also had a very large impact on some aspects of everyday life, and the reduction in State support has led some persons to seek to collect and sell scrap materials from obsolete industries. Many of these industries will have used radioactive sources for various applications and thus there is a risk that radiation sources, perhaps in metal housings, are inadvertently recovered and unwittingly sold with scrap metal. The likelihood of this happening is greatly increased if the source accountancy controls in the country are inadequate. Such an incident can cause significant doses to the persons involved, and may result in the source ending up in the public domain, outside the established system of control for radiation protection. The accident described in this Safety Report illustrates the consequences of such a sequence of events.

On 21 October 1994, three brothers entered a waste repository at Tammiku, Estonia, without authorization and removed a metal container enclosing a caesium-137 source. During the removal the source was dislodged and fell to the ground. One of the men picked up the source, placed it in his pocket and took it to his home in the nearby village of Kiisa. Very soon after entry into the repository he began to feel ill, and a few hours later he began to vomit. The man was subsequently admitted to hospital with severe injuries to his leg and hip, and died on 2 November 1994.

The injury and subsequent death were not attributed to radiation exposure, and the source remained in the man's house with his wife and stepson and the boy's great-grandmother. The boy was hospitalized on 17 November with severe burns on his hands, and these were identified by a doctor as radiation induced. The authorities were alerted, and the Estonian Rescue Board recovered the source from the house. The source was returned to the Tammiku repository on 18 November. The occupants of the house and one of the two surviving brothers were hospitalized and diagnosed as suffering from radiation induced injuries of varying severity. All were subsequently released from hospital, but, at the time of writing this report, the treatment to the most exposed individuals is still continuing.

## 1.2. OBJECTIVE

The objective of this report is to provide information to national authorities and regulatory organizations so that they can take steps to minimize the risks of similar accidents in the future, and also put in place arrangements to deal with such accidents if they do occur. It is hoped that this report will be of general interest in the radiation protection community, although it is aimed primarily at managers of waste disposal facilities, and legislators and regulators, both in developing countries and in all countries reviewing their radiation protection legislation.

## 1.3. SCOPE

This report describes the events leading up to the accident, the course of the accident, the remedial actions taken, and the lessons learnt from the sequence of events. It does not include discussions on theoretical aspects of the use and appropriateness of different methods for dose reconstruction.

## 1.4. STRUCTURE

This report presents brief information on the radiation protection infrastructure in Estonia (Section 2), the accident's chronology (Section 3), the recovery of the source (Section 4), the subsequent actions (Section 5), the physical and biological methods and results of dosimetry (Section 6), the medical description (Section 7) and the lessons to be learnt (Section 8), in the main body of the report. Detailed dosimetric information and medical findings are given in the Annexes.

## 2. RADIATION PROTECTION INFRASTRUCTURE IN ESTONIA

Estonia is the smallest country of the Baltic States, with an area of 45 125 km<sup>2</sup>, including some 800 islands. It is bounded by the Baltic Sea to the north and west, the Russian Federation to the east and Latvia to the south. The population of Estonia is 1 520 000, of which 62% are ethnically Estonians. There is a large minority of Russians (30%), and smaller minorities of Ukrainians (3%), Belorussians (2%) and Finns (1%). More than 70% of the population live in towns; almost 30% live in the capital, Tallinn.

After a short period of independence between the First and Second World Wars, Estonia was incorporated into the USSR. In September 1991 the USSR recognized the Republic of Estonia after independence was declared by the Estonian Parliament. The Parliament has approved a new constitution and the State is headed by a president.

### 2.1. RADIATION PROTECTION INFRASTRUCTURE AND RELEVANT ORGANIZATIONS

The legal framework of radiation protection in the former USSR existed in the form of a large set of government documents, instructions, norms and rules. However, there were no specific regulations covering nuclear energy, radiation protection or waste management. Basic documents in this area included the standards for radiation safety and basic requirements concerning the handling of radioactive materials and other sources of ionizing radiation. Soon after independence in 1991, the Estonian Government decided that the former Soviet guidelines for radiation protection and waste management would remain in force in Estonia until they are replaced by new national laws and regulations.

There was no single independent regulatory body for nuclear matters in the former State system until 1983, when the USSR State Committee for the Supervision of Safety in Industry and Nuclear Power (Gospromatomnadzor) was established. Before 1983, control functions in the field of nuclear applications were carried out by several organizations such as the National Public Health Centre of the Ministry of Health and the Meteorological and Hydrological Stations. These organizations were governed by a central authority in Moscow and its republican and regional branches.

Before the breakup of the USSR the central registers and inventories of radiation equipment and individual sources in Estonia were kept in the local Sanitary Epidemiological Centre, which reported to the ministers of health care. All radiation

equipment, radiation sources and radioactive compounds were sold through the regional branches of the USSR venture V/O IZOTOP. The types and models of radiation sources manufactured in the USSR were published in IZOTOP's catalogues and were available in regional offices. An enterprise could purchase radiation sources only after receiving written permission from the local Public Health and Epidemiological Centre (for approval of radiation safety in the workplace) and the local militia (for physical security requirements). All movements from the purchase to the disposal of the radioactive materials were supervised by the Public Health and Epidemiological Centre.

Some large defence related industrial establishments were outside local civil regulatory control and reported directly to their USSR ministerial authorities. These establishments used mainly sealed sources for a range of applications, and also assembled items such as thermoelectric generators containing strontium-90 sources. After independence, these companies came under Estonian regulatory control. Most have been reorganized and the larger radioactive sources have been transferred to the Russian Federation.

In Estonia there were many government organizations operating in the field of radiation protection despite the lack of comprehensive legislation. These organizations are summarized below.

- (1) National Health Board (reporting to the Ministry of Social Welfare)
  - occupational dosimetry
  - control of food irradiation
  - issuing of operating permits for the use of all radiation sources
  - issuing of permits for the acceptance and disposal of radioactive waste
  - maintaining inventory of sources in the country.
- (2) Radiation Centre (reporting to the Ministry of the Environment)
  - monitoring of environmental radioactivity.
- (3) Radiation Centre (reporting to the Ministry of the Interior)
  - monitoring of environmental radioactivity.
- (4) Standards Agency (reporting to the Ministry of the Economy)
  - approval and calibration of instruments for use in official radiological measurements.
- (5) AS ALARA Ltd: Radioactive Waste Management Organization (reporting to the Ministry of the Economy)
  - radioactive waste management and decommissioning of the Paldiski nuclear facility
  - responsibility for the Tammiku site since November 1995.
- (6) National Rescue Board: this organization was established from the former Civil Defence Forces and the Fire Department for the implementation of rescue operations in the event of major accidents or catastrophes.

However, the situation currently is as follows according to the Estonian Radiation Act adopted by the Riigikogu on 23 April 1997 and proclaimed by the President on 8 May 1997:

- (1) Estonian Radiation Protection Centre (reporting to the Ministry of Environment)
  - issuing of operating permits involving use of miscellaneous radiation activities
  - maintenance of an inventory of radiation sources in the country
  - occupational dosimetry
  - control of the the radioactivity of food
  - monitoring of environmental radioactivity
  - notification of radiological emergency situations.
- (2) National Health Board (reporting to the Ministry of Social Welfare)
  - dosimetry in radiotherapy, supervising the use of radionuclides and radiotherapy in hospitals.
- (3) National Rescue Board (reporting to the Ministry of the Interior)
  - monitoring of radioactivity level in emergency situations.
- (4) Radioactive Waste Management Organization AS ALARA Ltd (reporting to the Ministry of the Economy)
  - radioactive waste management and decommissioning of the Paldiski nuclear facility
  - responsibility for the Tammiku site since November 1995.
- (5) Standards Agency (reporting to the Ministry of the Economy)
  - approval and calibration of instruments for official radiological measurements.

## 2.2. RADIOACTIVE WASTE REPOSITORY, TAMMIKU

Estonia has neither a nuclear power programme nor nuclear research reactors, but there is still a need for a radioactive waste repository for waste arising from the use of radionuclides in medicine, research and industry.

The waste management facility for low and intermediate level waste was established at Tammiku, 12 km south of Tallinn, in 1963. It was the central storage/disposal facility for the waste generated from the use of radionuclides in Estonia.

In the mid-1980s reconstruction at Tammiku was started to upgrade the facility in accordance with the new revised criteria on the safe handling of radioactive waste, but this work was never completed because of a lack of resources.

At the time of the accident the repository was being managed by a municipal motor depot enterprise of Tallinn and supervised by the National Health Protection Board.

The facilities were established in accordance with criteria developed in Moscow in the late 1950s. The main disposal facility consists of a 15 m × 5 m × 3 m concrete vault providing 200 m<sup>3</sup> disposal capacity for solid radioactive waste. The vault is divided into nine compartments 1.6 m × 5 m × 3 m by concrete walls (Fig. 1). For storage of liquid waste, there is a 200 m<sup>3</sup> underground cylindrical concrete tank with a stainless steel lining. The internal diameter of the tank is 9 m and the height is 3.2 m. Both storage facilities are located below ground level.

The Tammiku facility is situated in a remote forested area. The site is surrounded by an outer barbed wire fence at 500 m radial distance with a gate and guard house, and an inner fence around the liquid waste storage tank and solid waste storage vaults. Both fences are only 1.5 m high and had been poorly maintained. The gate of the inner fence (Photo 1) as well as the doors of the temporary roof covering the vault compartment (Photo 2) currently used are locked and equipped with elementary alarm sensors.

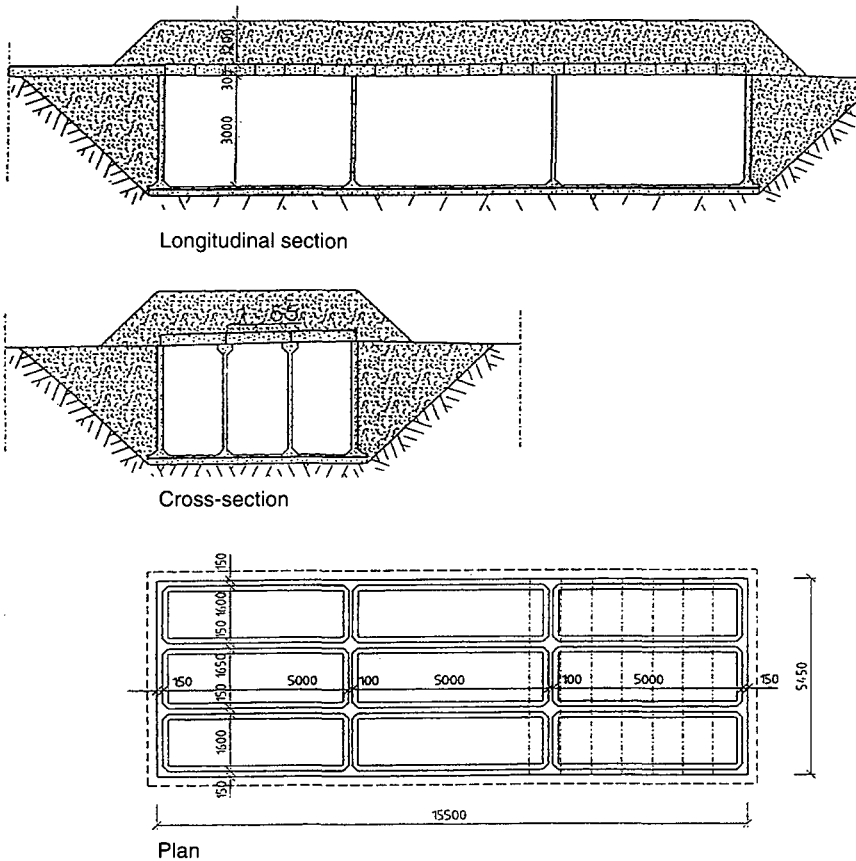
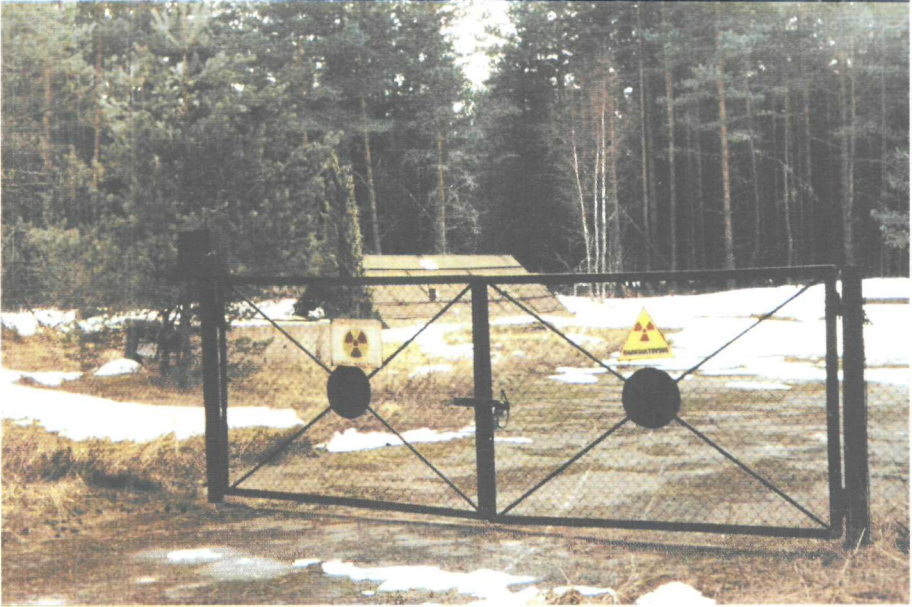


FIG. 1. Plan and sections of the vault in the radwaste repository at Tammiku, Estonia.



*Photo 1. Inner gate and fence of the Tammiku radwaste repository.*



*Photo 2. Locked roof cover of the vault.*



There is no treatment of radioactive waste in Estonia. All wastes generated from the use of radionuclides that cannot be treated as non-radioactive material are transported to Tammiku, where they are disposed of in the concrete vaults without any conditioning.

Over 30 years of operation, 1028 batches of waste have been disposed of at the facility, totalling approximately 97 t of waste with a total activity of 200 TBq. The remaining activity in January 1993 was 76 TBq. Sealed radiation sources, which were disposed of into a separate cell with additional shielding, comprised 99% of the known source inventory. The predominant radionuclides were strontium-90 and caesium-137, which represented approximately 75% and 24% respectively of the sealed source activity. The other radioactive waste, which included luminous articles containing radium-226, smoke detectors and antistatic devices containing plutonium-239, and smoke detectors containing americium-241, were disposed of without any segregation and packaging as mixed waste. The total activity of radium-226 and the long lived alpha emitting isotopes plutonium-239 and americium-241 is estimated to be less than 0.4 TBq.

### **3. THE ACCIDENT**

#### **3.1. BACKGROUND TO THE ACCIDENT**

The first indication of an anomaly was the discovery of a radioactive source in a shipment of scrap metal on 14 January 1994. The scrap metal had been delivered to the Estonian Metal Export Company (EMEX), Tallinn. As it was being routinely examined for radioactive contents with a hand held radiation dose rate monitor, a very high dose rate was detected. The dose rate measured during a general survey was in excess of 50 mGy/h and EMEX immediately informed the Estonian Rescue Board of the discovery. The Rescue Board subsequently visited the site and determined that the origin of the elevated dose rate was a metal container within the scrap. The dose rate close to the container was measured to be in excess of 2 Gy/h. The Rescue Board recovered the metal container and took it to the national waste disposal facility at Tammiku, where it was placed in the disposal repository.

The radionuclide and activity were not precisely determined prior to the disposal of the source, although the Estonian Health Board thought the source to be cobalt-60 with an estimated activity of about 7 TBq. The origin of the source assembly was not determined, but the shape of the metal container indicated that it may have been part of an irradiator. However, no industrial irradiators had ever been

located in Estonia. EMEX was also unable to trace the origin of the scrap shipment in which the container was found.

### 3.2. CHRONOLOGY OF THE ACCIDENT

The following description of the accident is based on information obtained during discussions with the Estonian authorities and interviews with the surviving brothers and their neighbours conducted during an IAEA mission to Estonia in December 1994. Account has also been taken of the findings of missions from Sweden, Finland and the Russian Federation, whose investigations were being completed as the IAEA team arrived. While there is no reason to doubt the essence of the description, a number of points differ slightly from the preliminary report prepared by the Estonian authorities in collaboration with the previous Russian team. These differences result from contradictions in the information provided by the surviving brothers, and in drawing up this chronology the most likely explanations have been used.

*21 October 1994*

The three brothers, RiH, RaH and IH, visited the waste disposal facility in the early hours of the morning. They approached the facility through the forest, climbed over the 1.5 m high fence, and broke into the repository and climbed down into the pit. In doing so, they overrode the electrical alarm system on the steel doors of the source vaults and cut through the retaining padlocks. RiH climbed down into the first section of the vaults, found the metal container with the source, and passed it up to his brothers. While he was doing this a metal cylinder fell out of the open tube in the container. This cylinder, approximately 18 cm long and 1.5 cm in diameter, was thrown back into the pit by RaH. A shorter cylinder of similar diameter, which subsequent investigations showed to be the radioactive source, was picked up by RiH and placed in his coat pocket. The brothers also entered the liquid waste storage facility, which was not fitted with an alarm, and removed several aluminium drums, after emptying their contents into the facility. While they were doing this, one of the drums fell against RiH's leg, causing minor injury. The drums and the metal container were carried 50 m through the wood to the road, where they were left while the brothers collected a car. The container was then placed in the boot of the car and taken to Tallinn where it was sold as scrap metal. On the way to Tallinn the brothers stopped first at RaH's house and then at RiH's house in Kiisa. RiH had begun to feel ill shortly after entry into the repository and a few hours after the unauthorized entry vomited repeatedly. He continued to feel unwell in the evening and went to bed. The other occupants of the house were the man's stepson (RT), the boy's mother (BK) and the boy's great-grandmother (AS).

In the subsequent investigations, the two surviving brothers maintained that they had entered the facility looking for scrap metal to sell and were not aware that the metal container contained a radioactive source. It is not known why RiH kept the source cylinder that fell out of the container. The record of periodic dose rate measurements made around the disposal facility by the technical staff both before and after the theft indicates that the cylinder that was thrown back into the pit was probably an inactive spacer in the source assembly (see Annex I). From the similarity between the dose rates measured close to the source, both at the time of its discovery at EMEX and at the time of its eventual recovery, it can also be inferred that the container contained only the one radioactive source, and hence nobody has been at risk from the subsequent handling of the container, which has never been recovered.

At some stage in the day the source was placed in a drawer in the kitchen of RiH's house.

*25 October 1994*

RiH was hospitalized with a severe injury to his leg. He did not reveal that he had entered the waste disposal facility or that the injury to his leg had been partly caused by a drum falling on him during the theft. He claimed that he had received the injury while working in the forest, and hence he was diagnosed as suffering from a 'crush injury'.

*2 November 1994*

RiH died. The cause of death was not associated at that time with radiation exposure.

*8 November 1994*

The next disposal of radioactive waste at Tammiku was carried out by the operators of the facility. The workers noticed that the securing padlocks on the steel doors to the waste vaults had been cut through, and replaced them. The dose rates measured after the disposal were two orders of magnitude lower than after the previous disposal on 30 September, i.e. 0.015–0.02 mGy/h as against 1.5–2 mGy/h respectively. The decrease of dose rate was explained by the shielding factor of the recently disposed waste. However, the supervisory authorities were not notified about this reduction in dose rates, the signs of forced entry or the theft of waste drums from the liquid waste storage facility.

*9 November 1994*

The stepson, RT, came in contact with the source while he was working on his bicycle and briefly handled it. A possible brief earlier contact can also be supposed (on the basis of his clinical symptoms, despite his failure to recall the chronology of events).

*16 November 1994*

The family's pet dog (4 months old, staying mainly in the kitchen) died. Previously the dog had been vomiting and blood was present in the dog's urine.

*17 November 1994*

RT was admitted to hospital with severe burns to his hands. These were diagnosed as radiation induced and the police were notified.

#### **4. RECOVERY OF THE SOURCE**

As soon as radiation induced injury was diagnosed, the police were notified of the evident loss of control over a radioactive source. The police in turn notified the Estonian Rescue Board, which immediately dispatched staff to Kiisa, who arrived at approximately 23:30 on 17 November 1994. Initial dose rate measurements confirmed that high gamma dose rates existed close to RiH's house, and neighbours within 200 m of the house and in areas where the dose rate exceeded  $0.4 \mu\text{Gy/h}$  were evacuated. This evacuation took place early in the morning of 18 November, and 15 houses in total were evacuated. Members of the Rescue Board briefly entered RiH's house to locate the source and measured a dose rate of  $1.2 \text{ Gy/h}$  close to the surface of a drawer in the kitchen. The measured dose rates in the other rooms were around  $50 \text{ mGy/h}$  (see Section 6.1).

Representatives from a number of authorities participated in a meeting to discuss the source recovery operation. The goals of the recovery were determined to be:

- (a) protection of the workers and the inhabitants;
- (b) recovery of the source and its safe disposal;
- (c) assessment of any residual contamination.

A shielded box with 3.5 cm thick lead walls was obtained for the recovery of the source. The house was entered twice by two Rescue Board staff for source

recovery purposes (on the first occasion for 2 minutes 35 seconds and on the second for 2 minutes 15 seconds). The staff involved wore lead aprons and thin rubber gloves but did not have any adequate remote handling tongs. During the first entry the drawer of the kitchen cupboard was emptied onto the floor and the source was identified. The dose rate at 5–7 cm from the unshielded source was measured to be 1.5–1.8 Gy/h. The source was a cylinder approximately 1.5 cm in diameter and 3 cm long. During the second entry the lead box was carried into the kitchen and the source was placed into the box. The person carrying out the recovery briefly picked up the source by hand (approximately 2 seconds contact) to place it in the box. The dose rate close to the surface of the lead box was measured to be 100 mGy/h.

The container was placed in the boot of a van suitable for the transportation of radioactive waste and taken straight to the repository, where it was disposed of, still in the shielded box, in the central section of the vaults. A dose rate of approximately 0.7–0.8 mGy/h was measured on the surface of the steel door of the central section of the vaults after the disposal.

The house was subsequently checked by the Rescue Board for radioactive contamination. None was found. The recovery operation was completed by 14:30 on 18 November 1994 and neighbours were permitted to return to their houses. On the same day a radiological inspection of all the scrap metal stockyards in the country was carried out by the rescue and environmental authorities. No other sources were found.

## 5. SUBSEQUENT ACTIONS

### 5.1. INVESTIGATIONS AND INTERNATIONAL ACTIVITIES RELATED TO THE ACCIDENT

Estonia has close ties with the neighbouring countries of the Russian Federation, Sweden and Finland, and all three countries offered to provide assistance following the accident. Several delegations visited Tallinn in the subsequent months and provided advice and assistance to the Estonian authorities.

*25 November 1994*

A Russian medical delegation (one physician and one health physicist from the Institute of Biophysics, Moscow) visited Tallinn to evaluate grades of severity of radiation injuries and to give advice on the treatment of the exposed individuals. The

invitation came from the physician in charge at the hospital to which the adult victims were taken.

*1 December 1994*

The Estonian Ministry of the Environment held a meeting for the IAEA team and representatives from the Swedish and Finnish radiation protection authorities.

*1 to 8 December 1994*

An extensive radiological investigation was made by the Estonian and the invited foreign experts.

*13 December 1994*

An Estonian governmental commission was set up to investigate a number of issues relevant to the accident.

*13 January 1995*

A second source was found by members of the governmental commission on the highway between Tallinn and Narva (see Section 5.3).

*January–February 1995*

Sweden provided assistance in the proper handling of the second source by contracting the Studsvik Radwaste AB. A concrete container was transported to Tammiku for the storage of the source. Meanwhile security procedures were considerably strengthened at Tammiku and the repository was permanently guarded by the police.

*January 1995*

The Estonian Medical Society sent out information to all medical personnel in the country about the characteristics of radiation induced injuries, and urged all such personnel to be vigilant.

*May 1995*

Following an exchange of information between Estonian and Swedish medical experts, it was decided to offer the injured boy, RT, medical treatment at the Huddinge

hospital, Stockholm, in order to reduce possible future risks to his health. The boy, his mother and the physician in charge were invited to Stockholm, where stem cells from the boy's bone marrow were collected. These cells were stored cryogenically and could be used in an autologous transplantation if he should develop leukaemia or a haemopoietic deficiency in the future.

*May–June 1995*

A search for possible additional sources out of control commenced in Estonia. This was done by car-borne surveys covering the highways, roads, waste dumps, etc., in northern and western Estonia. This undertaking was carried out with help in the form of equipment and training in measuring techniques from Finland and Sweden.

## 5.2. DETERMINATION OF THE RADIONUCLIDE AND ITS ACTIVITY

The absence of portable gamma spectrometry equipment in Tallinn made it very difficult for the Estonian authorities to determine the precise nature of the radioactive source. Attenuation measurements made by members of the Estonian Health Board shortly after the discovery of the source at EMEX suggested that the radionuclide was cobalt-60 with an activity of about 7 TBq, but the dose rate profiles made by the Rescue Board at the time of the source recovery at Kiisa indicated that the source was more likely to be caesium-137. The profile of the deterministic injuries to the hands of the exposed individuals also indicated that the source was more likely to be caesium-137 than cobalt-60.

This uncertainty was resolved by the use of a portable gamma spectrometer, provided by the IAEA, at the Tammiku facility in December 1994. Although the gamma radiation from the returned source was significantly reduced by the lead walls of the container in which it had been disposed, the radiation field close to the disposal vaults was still strong enough to obtain a gamma spectrum of the source. The disposal records for the facility also indicated that no other gamma emitting sources of significant activity were present. The gamma spectrum obtained confirmed that the radionuclide was caesium-137.

The activity of the source was more difficult to determine. A series of dose rate measurements were made by the Estonian Rescue Board at the time of the source recovery at Kiisa; these were made at various locations inside the house and at other locations up to 230 m away. These dose rates may be used together with distances from the source to estimate the source activity, and from these data an activity range of 150 GBq to 7.4 TBq caesium-137 was obtained. Other estimates based on dose rate measurements made around the waste repository after the source had been returned also give varying activities that fall within this range.

### 5.3. THE SECOND SOURCE

The events in Kiisa evoked considerable public interest and were described in detail in the Estonian news media. The Estonian authorities gave schematic drawings of the missing metal source container to the police and urged members of the public to report any information they had.

A governmental commission was set up on 13 December 1994 with the task of assessing the environmental consequences of the radiological accident. The main objectives for the commission were:

- (a) to assess the environmental consequences of the accident;
- (b) to update the inventory of radiation sources and equipment in Estonia;
- (c) to inspect the storage conditions of the radiation sources and substances in institutions and enterprises;
- (d) to develop basic guidelines for radioactive waste management in Estonia.

The commission commenced its work immediately after the New Year holidays in 1995. On a routine inspection trip to a company in the Narva region on 14 January, elevated dose rates (about 2  $\mu\text{Gy/h}$ ) were detected at a point along the highway between Tallinn and Narva by the members of the commission. The finding was made by chance by one of the radiation experts, who during the trip had turned on one of the survey instruments carried in the car. Because of snowcover and high levels of radiation in the vicinity of the source, it was not possible to locate the source accurately on the same day.

On the following day the radiation source in a metal container was located, and the radionuclide caesium-137 was identified. On 15 January the source was transported to the Tammiku radioactive waste repository by the Estonian Rescue Board and the Chemical Accident Response Group.

The dose rate at a distance of 1 m from the object was measured to be 0.14 Gy/h, which corresponds to a caesium-137 activity of 1.6 TBq. On this occasion the radiation source was not disposed of in the repository pit but stored in a temporary container. The source was subsequently placed into a special waste container, provided by Studsvik Radwaste AB, Sweden, on 15 February 1995. According to the information provided by members of the Rescue Board this source container had similar dimensions but was not identical with the metal container recovered one year earlier in the EMEX company stockyard.

The finding of a second source strengthened the suspicions of the authorities that more sources of a similar kind might be out of control in Estonia. Different strategies to scan Estonian territory were discussed. A car-borne survey was judged to be the most suitable and economical method.

A radiological survey of the territory of Estonia was eventually carried out with the co-operation of Sweden and Finland. Finland provided a vehicle equipped



with a high pressure ionization chamber detector combined with an electronic global navigation system. Sweden provided additional equipment and training for the rescue and environmental departments of Estonian counties. Both types of equipment had sufficient sensitivity to identify a caesium-137 point source with an activity of 3.7 GBq from a moving car at least 50 m distant. Altogether more than 20 000 km of roads were surveyed in the summer of 1995, but no more radiation sources were found.

The place where the second source was found was frequently visited and used as a playground by children in the vicinity. Also adults used to visit the area in the autumn to pick mushrooms and berries. The authorities therefore sent out a request for all persons who had visited the area in recent months. In all, seven children and eight adults claimed that they had visited the place, and were medically examined. Blood samples of three children who regularly met close to where the source was found were sent to Finland for chromosomal analyses. However, neither radiation induced chromosomal aberrations nor symptoms of radiation sickness were found.

## **6. DOSIMETRY**

It was clear from the moment of the source recovery at Kiisa that several people had been exposed at high dose rates for an extended period of time, and that there had also been the potential for significant radiation exposure of many other people living in the neighbourhood. The members of the family that lived in the house were hospitalized shortly after the recovery of the source and several of these persons exhibited deterministic radiation injuries. There was a clear need for dose information which could be used both for the subsequent medical management of the patients and the scientific assessment of the accident. Arrangements were made for the doses of the exposed individuals to be assessed using biological dosimetry techniques, and the dose distribution in the house and its neighbourhood to be assessed by external dosimetry (electron paramagnetic resonance and thermoluminescence measurements on inert samples).

### **6.1. PRELIMINARY DOSE EVALUATIONS**

#### **6.1.1. Dose rate measurements by the Estonian Rescue Board**

The Rescue Board measured the dose rate with a scintillation G-M counter (type DP-5), aiming to localize and find the source in the village and in the house on 18 November 1994. The dependence of the dose rate on the distance from the house is

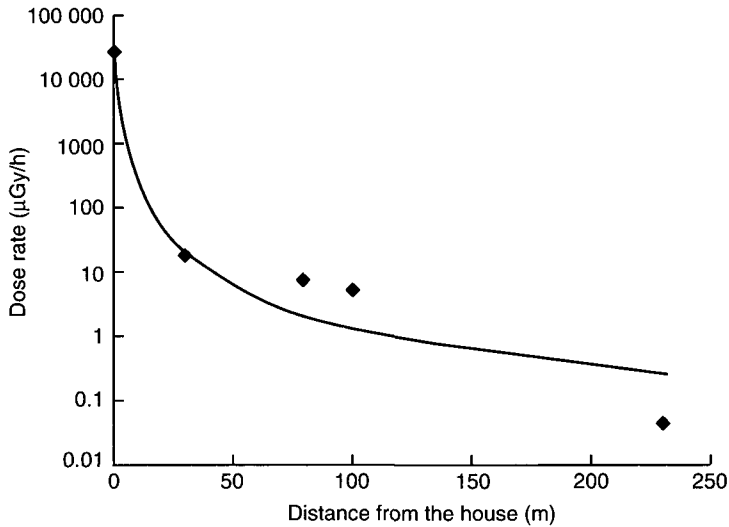


FIG. 2. Dose rate measurements by the rescue team around the house in Kiisa made on 18 November 1994.

illustrated in Fig. 2. There is an increase of the dose rate from the railway station towards the house. There were a few further measurements of the dose rate performed on the neighbouring streets around the house; however, they showed no elevated values above the background level because of the effect of shielding by buildings and fences. In the house the dose rate was measured at around 50 mGy/h in the bedrooms, 200 mGy/h at about 1 m from the kitchen cupboard, and 1.2 Gy/h on the top of the kitchen cupboard.

#### 6.1.2. Preliminary dose estimation by the experts from the Institute of Biophysics, Moscow

As was mentioned in Section 5.1, two Russian experts from the Institute of Biophysics in Moscow (I.A. Gusev and G.D. Selidovkin) visited Tallinn on 25 November 1994. As a result of this visit, first attempts to estimate the doses received by the victims of the accident were performed [6]. The dose rates provided by the Estonian Rescue Board, as well as calculated values for different points in the house, were used for dose calculations.

According to information given by the Estonian Rescue Board, the source was described as a rectangular metal assembly (200 mm  $\times$  420 mm  $\times$  40 mm) with five

cylinder channels, two of which had contained two radioactive metal cylinders ('pencils') about 18 cm in length and 1.5 cm in diameter. The total mass of the above assembly, which was possibly part of a radiation sterilization source assembly, was approximately 7 kg. The metal used for manufacturing the assembly and for the capsule of the sources is unknown. No referral inventory marks or trademarks were found. The dose rate at the surface of the assembly exceeded 2 Gy/h (the upper limit of measurement of the available dosimeters). According to dose rate measurements at different distances from the source, the calculated dose rate at the source surface (1 cm distance) was of the order of 2000–3000 Gy/h. Allowing for the attenuation of radiation emitted by the source placed in a standard lead container, the experts concluded that the radiation source contained caesium-137.

Afterwards, the team consulted the Radiological Department of the Mendeleev Institute of Standards in St. Petersburg, which has full information about all radiation sources produced in the USSR. To confirm the conclusion of the Russian experts about the caesium-137 source, the Department provided them with two ampoules containing 3.7 TBq and 2.6 TBq caesium-137, respectively. These ampoules helped the Russian experts to estimate the activity of the source. The activity was estimated to be 3.33 TBq.

It was suggested that the radiation source had been left until 9 November 1994 in a jacket pocket, hung in the entrance hall, and then put in a tool box in the kitchen cupboard. It was considered that the source of radiation was situated at distances of 7 to 4.5 m from the beds of the people in the house.

The preliminary results, which yield estimates of the effective and equivalent doses (obtained by the experts from the Institute of Biophysics, Moscow), are presented in Table I.

Owing to many uncertainties in the scenario of the accident, these first results gave only a rough estimation and a more detailed study was needed to clarify the situation.

## 6.2. RETROSPECTIVE DOSIMETRY USING CERAMICS AND NATURAL MATERIALS

### 6.2.1. Sampling

Solid state dosimetry techniques including thermoluminescence (TL), electron paramagnetic resonance (EPR) and chemiluminescence (CL) were applied for dose reconstruction in the house. These techniques can provide estimates of cumulative absorbed dose due to the accident when used with suitable materials that have remained in the house in fixed positions since the introduction of the source [7]. The potentially suitable samples within the house included: (i) for TL, ceramic plant pots and a vase, porcelain dishes, fuses and light fittings; and (ii) for EPR and CL, sugar, medicine and mollusc shells from a decorative casket.

TABLE I. EARLY ESTIMATES OF RADIATION DOSES TO VICTIMS OF THE ACCIDENT  
(Derived from the data obtained by the Russian group of experts)

Patient	Distance from the source to the specific body part		Dose rate (Gy/h)	Time of contact	Exposure doses and localizations	
	Distance (m)	Body part			Dose (Gy)	Localization
RiH	0.01	Thigh	$(2-3) \times 10^3$	55 min/single	~1830	Thigh
	0.5	Whole body	1.08	55 min/single	~4	Whole body
	7.1	Whole body	0.005	110 min/single	~20-30	Left hand
	0.01	Hands	$(2-3) \times 10^3$	15-25 s	~8-10	Right hand
AS	0.5	Whole body	1.08	30-40 min	~2.5	Whole body
	5.4	Whole body	0.009-0.015	99 h	~2-2.5	Whole body
	3.0	Whole body	0.03	27 h	~2-2.5	Whole body
	4.5	Whole body	0.013-0.015	153 h	~12-20	Right hand
RaH	0.01	Hands	$(2-3) \times 10^3$	15 s/single	~8-10	Left hand
BK	0.5	Whole body	1.08	15 s/single	<1	Whole body
	7.1	Whole body	0.005-0.008	72 h	~0.5	Whole body
IH	≥2	Whole body	<5	≤1 h/single	<1	Whole body

Samples were collected from the house by the laboratory of the Institute of Geology in Tallinn (see Fig. 3 and Table II); at a later stage, subsamples were also distributed to a number of other laboratories with the aim of performing an inter-comparison of results [8]. TL techniques were employed with ceramic samples and sugar samples were tested using EPR and CL techniques.

The house is made of wood, although some of the interior structure includes brick (which has the most reliable position history). Although it was not possible to sample evenly within the area of the house, samples from different distances from the two main possible locations of the source were obtained. In the kitchen, where the highest dose rates were found, the number of available ceramic samples was modest; in the bedroom of AS and RT only two samples were retrieved; unfortunately neither provided entirely reliable information.

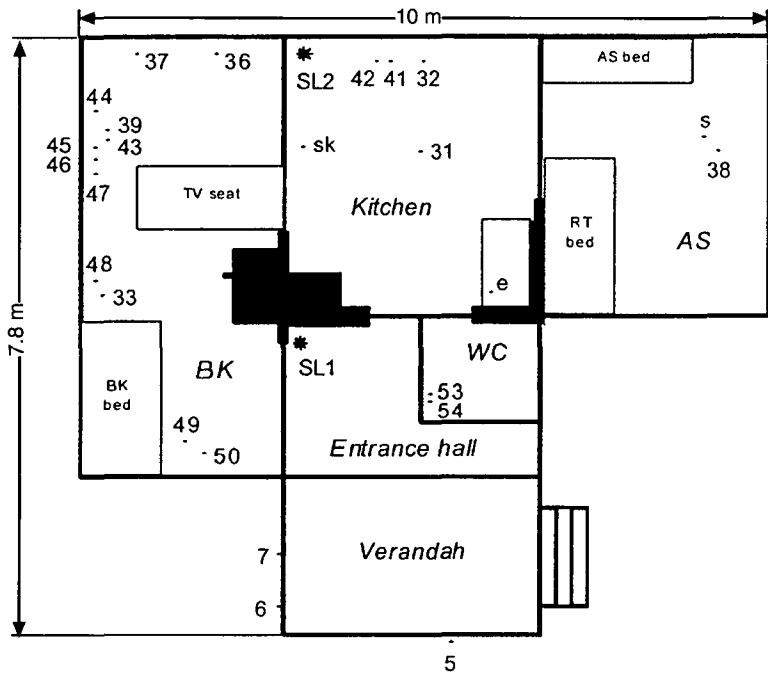


FIG. 3. The plan of the house in Kiisa. Possible source locations (SL1 and SL2) are indicated by the large asterisks. The sample codes are given in Table III.

TABLE II. LOCATION AND DISTANCE FROM THE RADIOACTIVE SOURCE OF THE SAMPLES TAKEN FOR EPR

Sample No.	Sample type	Location	Distance from SL2 <sup>a</sup> (m)
52	Porcelain fuse	Drawer	0–0.5
42	Ceramic ashtray	Kitchen	1.1
36	Mollusc shells	RT's room	1.3
41	Plant pot	Kitchen	2.0
sk	Sugar	Kitchen	1.4
51	Plate	Kitchen	1.5
32	Medicinal tablets	Kitchen	2.2
31	Porcelain light fitting	Kitchen	2.2
37	Plant pot	RT's room	2.5
39	Plant pot	RT's room	3.1
43	Plant pot	RT's room	3.2
44	Plant pot	RT's room	3.3
45	Plant pot	RT's room	3.4
46	Plant pot	RT's room	3.5
47	Plant pot	RT's room	3.6
e	Egg shell	Kitchen	3.9
48	Plant pot	RT's room	4.5
33	Plant pot	RT's room	4.6
53, 54	Bricks	WC	5.3
49	Plant pot	RT's room	5.9
50	Plant pot	RT's room	6.0
s	Sugar	AS's room	6.1
38	Plant pot	AS's room	6.2
7	Bricks	House perimeter	7.3
2	Plant pot	Shed	7.5
1	Ceramic vase	Shed	7.6
6	Plant pot	House perimeter	8.1
3	Bricks	Shed	8.2
5	Plant pot	House perimeter	8.9
4	Plant pot	Shed	9.6

<sup>a</sup> The distances in the last column are measured from source location SL2 in the kitchen (see Fig. 3).

There is no established methodology for the use of solid state techniques in retrospective dosimetry and the work described in this report should be considered to be exploratory in terms of application to dose reconstruction, although there is considerable experience in the evaluation of absorbed dose with ceramics. The aim of using solid state techniques was:

- (a) To characterize the radiation field within the house to determine whether the source had been predominantly in one location; and
- (b) Using the data from (a), and also using measurements of the cumulative dose at different locations, to derive the average dose rates in air at various locations in the house.

A semi-quantitative analysis of the cumulative dose for a presumed pattern of occupancy behaviour by three individuals has also been attempted. Further details of the experimental work are given in Section I-2 in Annex I.

### **6.2.2. Location of the source**

Although the source was found in a drawer in the kitchen, the statements given by the members of the family suggested that it was left in a jacket pocket in the hall (location 1) and then subsequently moved to the kitchen drawer (location 2) when the boy repaired his bicycle.

The thermoluminescence (TL) measurements enable the cumulative absorbed dose for the ceramic samples to be determined. In a preliminary study performed by the Tallinn laboratory of the Institute of Geology, TL measurements were made with quartz extracted from the ceramic samples [8].

If the measured cumulative dose for the tested samples is plotted against distance from location 2 (Fig. 4), an inverse quadratic relationship is obtained, confirming location 2 as the predominant position of the source. The data for distances less than 5 m correspond to those obtained using TL with ceramics, and it has been assumed that the average dose in (thin) ceramic is approximately equivalent to kerma in air. On the basis that the source was at this location for a period of 28 days, the average dose rate at a distance of 2 m from the source is  $40 \pm 8$  mGy/h, corresponding to a caesium-137 source activity of  $2 \pm 0.4$  TBq (see Section I-2 in Annex I).

### **6.2.3. Dose reconstruction**

The dose received by the inhabitants of the house is of central interest in applying retrospective dosimetry techniques to this problem. An assessment of the external dose to each individual requires a detailed knowledge of their movements in time and space within the non-isotropic radiation field that existed in the house and

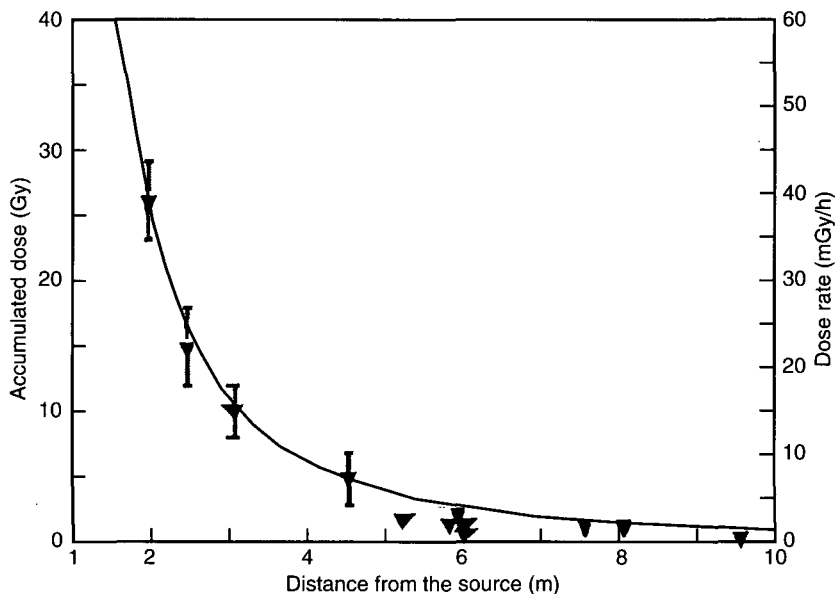


FIG. 4. Absorbed dose in quartz evaluated by TL for different samples versus distance of the samples from the source (location in the kitchen). Error bars present standard errors at 66% level of confidence. Error bars for values with small errors are removed for readability. The regression curve for the data was drawn using the formula  $y = k/x^2$ . The dose rate was calculated assuming the source was in the kitchen drawer for 28 days.

the use of computational modelling to assess the cumulative dose distribution within the body and the whole body dose. One factor that militates against a rigorous assessment based on currently available data is that only a rudimentary record of the movements of the individuals concerned is available — particularly for the kitchen, where the field was highly non-isotropic. Modelling calculations have not been commissioned so far. In the absence of a more sophisticated analysis, an ad hoc procedure has been employed to arrive at a semi-quantitative estimate of the cumulative absorbed dose (tissue kerma) for an occupancy pattern by specific individuals, based on the behaviour they reported and employing the experimental results. Using this approximation, further details of which are discussed in Annex I, estimated dose ranges for the individuals BK, AS and RT have been derived and are summarized in Table III. It must be stressed that this approach can only be approximate; the estimates should therefore be taken to be indicative.



The following steps were performed:

- (i) The average dose rate range in air at the main locations of occupancy was calculated taking into account differences in the radiation field at these locations (e.g. at either end of the bed and the television viewing seat) but assuming a low proportion of scattered radiation;
- (ii) The duration of occupancy at the main locations was estimated. The location of AS is uncertain for a substantial fraction of the day; no precise estimates of time spent in the kitchen are available and one hour is assumed;
- (iii) The cumulative dose for 28 days was calculated on the assumption of occupancy of each location as indicated by the individuals;
- (iv) The values of air kerma given in column 4 of Table III, and column 3 of Table V, are not directly comparable with the dose estimates provided by biological dosimetry, which are indicators of whole body average dose. The estimation of average cumulative whole body dose from the luminescence dosimetry when the individuals were in the locations reported is given in Table III. For example, in the case of the most prolonged occupancy in bed, the incident radiation was cranio-caudal or caudio-cranial with the longitudinal body axis subtending a very small angle with respect to the source. The results of such calculations, using the Monte Carlo method, were not available when preparing this report and Monte Carlo conversion factors available in the literature do not match the geometry given in this situation. Over and above these considerations, there is also significant uncertainty concerning the exact movements of individuals in the kitchen.

TABLE III. SUMMARY OF SEMI-QUANTITATIVE ESTIMATES OF CUMULATIVE DOSE TO BK, AS AND RT

Individual	Locations	Occupancy per day (h)	Cumulative 28 day air kerma range (Gy)
BK	Bed	8	0.7–1.5
AS	Bed,	8	1.0–2.6
	TV seat,	4	1.6–3.8
	Kitchen	1 (estim.)	0.5
	<i>Total</i>		3.1–6.9
RT	Bed,	8	1.1–1.7
	TV seat	4	1.6–3.8
	<i>Total</i>		2.7–5.5

Nevertheless, given the uncertainties prevailing in the information available and the semi-quantitative nature of the comparison, there is a reasonable degree of agreement between the estimates derived from physical and biological dosimetry measurements.

### 6.3. BIOLOGICAL DOSIMETRY

#### 6.3.1. Chromosomal aberration analysis

Chromosomal aberration analysis (CAA) was carried out at the Finnish Centre for Radiation and Nuclear Safety (STUK) at Helsinki, on the blood samples from 18 subjects. These subjects included the two surviving brothers involved in the removal of the source from the vault, neighbours and visitors to the house and seven persons involved in the source recovery. Additional samples were forwarded to the GSF Forschungszentrum für Umwelt und Gesundheit, Neuherberg, for analysis. All the persons with signs of deterministic radiation effects also showed high frequencies of dicentric chromosomes in their peripheral blood lymphocytes. The dose estimates were based on the frequency of dicentric chromosomes, and were calculated using standard models. However, as it was known that in all cases of high exposure the exposure was protracted over an extended period of time, dose estimates were also given for a protracted exposure pattern. In addition, since there was clear evidence that non-uniform irradiation of some persons had occurred, dose estimates for partial body exposure were calculated using the Qdr approach (see Annex II). A summary of the estimated doses to the five most exposed persons is given in Table IV.

TABLE IV. DOSE ESTIMATES OBTAINED BY CAA FOR THE FIVE MOST EXPOSED PERSONS

Subject	Estimated dose (Gy) (95% confidence interval)		
	Acute and homogeneous exposure	Protracted and homogeneous exposure	Protracted or partial body exposure (Qdr)
RT (stepson)	1.1 (0.7–1.3)	2.7 (1.0–4.5)	1.9 (1.2–2.4)
AS (great-grandmother)	1.0 (0.7–1.3)	2.7 (0.9–4.4)	2.9 (2.6–3.1)
IH (brother)	0.9 (0.6–1.0)	1.2 (0.7–1.6)	2.2 (1.6–2.6)
RaH (brother)	0.8 (0.5–0.9)	1.0 (0.5–1.4)	1.7 (1.1–2.2)
BK (mother)	0.3 (0.1–0.5)	0.5 (0–1.0)	n/a

Note: n/a — not applicable.

TABLE V. WHOLE BODY DOSE ESTIMATES (Gy) FOR THE INHABITANTS OF THE HOUSE, OBTAINED BY DIFFERENT DOSE CONSTRUCTION METHODS

Subject	Early estimates (Nov. 1994)	Physical dosimetry (estimated air kerma range)	CAA (protracted exposure)	GPA	EPR (tooth)
RT (stepson)	2.5	2.7–5.5	2.7	1.5	2.1
AS (great-grandmother)	2.0–2.5	3.1–6.9	2.7	1.2	—
BK (mother)	0.5	0.7–1.5	0.5	—	—

### 6.3.2. Other biodosimetric data

Glycophorin A (GPA) locus somatic mutation assays in erythrocytes were performed at the University of Pittsburgh on the blood samples of the great-grandmother (AS), the boy (RT) and the boy's friend (LJ). The doses were estimated to be 1.2 Gy (AS), 1.5 Gy (RT) and 0.02 Gy (LJ). Electron paramagnetic resonance (EPR) analysis was also carried out on a sample of tooth from RT, and this gave an estimated dose of 2.07 Gy.

## 6.4. CONCLUSIONS ON DOSIMETRIC DATA

A summary of the conclusions of these analyses is given in Table V. Details of the measurement techniques used are given in the following sections. Full details of the measurement techniques, together with the results, are given in Annexes I and II.

## 7. MEDICAL ASPECTS

### 7.1. EXPOSED INDIVIDUALS

The caesium-137 source was in RiH's house for the period 21 October to 17 November 1994, i.e. about 4 weeks. The investigation into the sequence of events,

and the dose assessment provided in Section 6, showed that the whole family and some relatives and other visitors were exposed at varying dose rates, which were especially high in the kitchen (of the order of 10–100 mGy/h).

The overexposed individuals exhibited injuries of various types, depending mainly upon the source–subject geometry and the length of exposure. Some individuals were never close to the source and, as a consequence, received whole body exposures which can be considered to be relatively homogeneous. Some others touched the source and developed radiation burns. A third group received both whole body and localized overexposures, and developed acute radiation syndrome complicated by localized injuries.

Medical findings for those five individuals who were exposed to radiation doses above the threshold for deterministic effects are discussed in the following section in more detail.

Some other individuals required medical and biological investigations, as they had visited or lived in the house during this period. However, none of them exhibited any related clinical effects that were attributable to radiation:

- BK — female, 35 years, RT's mother, dose estimate 0.5 Gy (received lower doses than the other family members owing to shorter periods of exposure).
- LJ — male, 12 years, friend of the boy RT, dose estimate 0.1 Gy.
- AN — female, 12 years, friend of the boy RT, no chromosomal aberration detected.
- MM — female, 78 years, friend of the great-grandmother AS, dose estimate 0.1 Gy.
- EM — male, 67 years, physician, no chromosomal aberration detected.
- HH — male, 47 years, physician, dose estimate <0.1 Gy.
- IP — female, 56 years, nurse, dose estimate 0.13 Gy.

The Estonian Rescue Board workers (seven males, aged between 23 and 59 years) were also clinically and biologically investigated. They did not receive high doses as the recovery action was short (dose estimates below or at 0.13 Gy). Even the man who picked up the source with his hand to place it in a lead container did not exhibit any local injury.

## 7.2. CLINICALLY AFFECTED INDIVIDUALS

The medical descriptions cover five individuals who exhibited deterministic effects:

**RiH**, male, 25 years, who kept the source in his pocket for several hours with a dose rate estimated to be 2000 to 3000 Gy/h at the skin surface. He showed a rapid evolution combining local severe injury and deterioration of his general state. He died

12 days after the beginning of exposure (acute radiation syndrome, grade V, combined with local radiation injury, grade IV).

RiH developed an acute radiation syndrome very soon after entering the repository area. He felt unwell and after some hours he vomited several times. He thereafter stayed bedridden with a progressive functional deficiency of his right leg together with a worsening of his general state. During the first three days at home and thereafter in hospital, a physical trauma to his right thigh while working in the forest was given as an explanation to the physicians and nurse. Exposure to ionizing radiation was not suspected at that time. On the fourth day after exposure, the patient was hospitalized because of a severely altered general state. He was drowsy, and progressively developed a generalized shock syndrome together with a functional incapacity of both legs. Locally, oedema and heavily injured tissues were noted in the region of the right hip, the right thigh and the lower part of the pelvis, together with blisters that rapidly haemorrhaged. Despite an active treatment relevant to crush syndrome (haemodialysis and peritoneal dialysis, antibiotics, repeated massive blood transfusions), the general state continued to worsen in the following days, with the appearance of peritonitis symptoms. His haemoglobin and leucocyte dynamics (under intensive transfusional supportive care) from 25 October to 2 November 1994 are presented in Fig. III-1 in Annex III. The patient died on 2 November 1994 (day 12), in a context of acute renal failure and severe anaemia.

The autopsy found signs of acute necrosis of the right thigh and hip. In the gastrointestinal tract, a massive haemorrhage was observed in the small intestine and the colon and a decrease of the thickness of the intestinal wall was detected. This latter indicates a direct exposure of the abdominal region from the source carried in RiH's pocket. Signs of bilateral bronchopneumonia and septic shock were also found. Besides the lesions described in the autopsy report, a direct injury to the kidneys that could have participated in the fatal evolution has also been indicated.

**RaH**, male, 28 years, who had been in the vicinity of the source for a few hours and had handled the source, developed typical radiation induced lesions in the right hand (radiation sickness, grade I, combined with local radiation injury, grade III).

RaH received a dose estimated at 1 Gy, related to a few hours of heterogeneous overexposure on 21 October 1994. He presented no symptom of acute radiation sickness but he developed radiation burns in the right hand after eight days with blisters which started to heal following local treatment.

Upon the discovery of the source, RaH was hospitalized on 21 November 1994, one month after exposure, in the Tallinn Magdalena Hospital. His general state was good with no clinical or haematological disorder. Radiation induced lesions at that time were essentially localized on the thumb and the adjacent part of the right hand, with secreting wounds (wet desquamation). Seven weeks after his short contact with the source, a wet ulcerative lesion developed on his right thumb (Photo 3). Under vitamin therapy, corticotherapy and local treatment, the general evolution and healing



*Photo 3. Right palm of RaH (28 years old) 7 weeks after a short time contact with an approximately 2 TBq caesium-137 source. Deep wet ulcerative lesion on the thumb; the healing has started on the thenar. [W. Paile, STUK, Finland]*

of the lesions were satisfactory in spite of residual signs of local inflammation. The long term evolution was complicated by the reappearance of an ulcerative lesion on the right thumb one year later.

**IH**, male, 27 years, had also been in the near vicinity of the source for a few hours. He subsequently developed a moderate degree of radiation sickness (radiation sickness, grade II).

IH was heterogeneously overexposed on 21 October 1994 with a resulting dose estimated at 0.9 Gy. No alarming symptom was noted in the evolution. Upon admission at the Tallinn Magdalena Hospital one month after exposure, his general state was good. Nevertheless, he presented a mild thrombocytopenia ( $54 \times 10^9 \text{ L}^{-1}$ ) and mild leucopenia ( $(3-4) \times 10^9 \text{ L}^{-1}$ ) that rapidly recovered. Under vitamins, corticoids and antibiotics, the initial alteration of the myelogram normalized after two months. On 16 November, small ulcers appeared on the joints of the third and fourth fingers of the right hand, which rapidly became covered with a crust. The origin of these lesions was uncertain because no evidence of direct contact with the radioactive

source could be found on interrogation. The medical follow-up between October 1994 and January 1996 showed a good long term evolution.

**RT**, male, 13 years, was the most exposed among the survivors. He suffered from a severe and prolonged bone marrow aplasia complicated by radiation burns predominating on the left hand. These localized injuries were related to direct source–skin contact. He needed intensive care and recovered after a few months, while his burns necessitated some further surgical excisions. (Severity grading is not given owing to the protracted exposure for 4 weeks; see Section 8.2.)

RT was subject to a protracted, moderately low dose rate exposure, considered to be relatively homogeneous, from 21 October 1994 over a four week period. In addition, on 9 November 1994 (referenced as day 0 for the following description of haematological evolution), he also received an acute heterogeneous overexposure while repairing his bicycle. In addition, he had physical contact with the radioactive source, most probably on 22–23 October 1994 since blisters appeared on his left palm as early as 10 November 1994. Subsequently, mild pain appeared on movement of each palm that later extended to the elbows, probably indicating a bilateral lymphangitis. On 14 November 1994, he was sent to the Tallinn Children's Clinic where ordinary medication for burns was prescribed. In addition to the blisters on both hands, nausea and moderate diarrhoea were reported at that time.

On 17 November 1994 (day 8), RT was hospitalized at the Tallinn Children's Clinic, where the radiological origin of the lesions was suspected. Both his haematological state and the radiological burns were of concern. Despite a good general state, the boy was subfebrile and looked pale and subicteric. Minimal cutaneous hemorrhagic signs on the forearms and the limbs were also noted. The haematological follow-up showed a mild anaemia, but severe and prolonged granulocytopenia ( $<0.5 \times 10^9 \text{ L}^{-1}$  from day 19 to day 40) and thrombocytopenia (below or around  $50 \times 10^9 \text{ L}^{-1}$  from admission (day 8) until day 40 (see Fig. III–2 in Annex III).

Because of two epistaxis episodes, platelets were transfused on 18 and 24 November 1994. The recovery started on day 40 for platelets, i.e. about 1.5 months after the acute overexposure. The recovery for granulocytes was slower, as subnormal values were reached only after 2.5 months. Bone marrow cytological and histological examinations were performed repeatedly to certify the central origin and then follow up the recovery of the aplasia. Despite this, RT developed a moderate fever. During the whole length of the aplasia, no severe infection complicated the evolution. The treatment during this period associated trimethoprim sulfamethoxazole, fluconazole, nystatin and vitamins. Cytokines as a haemopoietic stimulatory therapy were not used, as it was judged that spontaneous recovery could be expected.

On May 1995, Estonian and Swedish physicians discussed the possible complications that could arise in the long term evolution of RT, especially the development of late bone marrow radiation induced diseases such as leukaemia or



*Photo 4. Left hand of RT (13 years old) taken 6 weeks after a short time contact with an approximately 2 TBq caesium-137 source. Ulcerative lesions on the thumb and index finger. [J.-C. Nenot, IPSN, France]*

aplastic anaemia. It was decided to collect haemopoietic stem cells from the patient and to store them frozen so that they could be used in the future as an autologous graft if needed. After a classic protocol for mobilization from the bone marrow, the blood stem cells were collected twice.

The local radiation lesions affected both hands. After the handling of the source (most probably on 2 November 1994), blisters developed on day 8 on the left hand and these subsequently evolved to ulcers on day 12. They extended on the palmar surfaces of the thumb and index finger. On the right hand an erythema and moist desquamation extended to the first three fingers. The local evolution under antiseptic bandages was good. Photos 4 and 5 were taken 6 and 7 weeks after the presumed contact with the source. Even for the more injured left hand, the lesions were virtually completely healed (although with a certain degree of cutaneous atrophy) in early December 1994. The nails of two burnt fingers were replaced by new ones in January 1995, indicating nail matrix conservation. Unfortunately, in December 1995, this good initial evolution turned to recidivist necrosis of the distal phalanx of the left thumb with signs of underlying osteoporosis, leading to amputation.





*Photo 5. Hands of RT (13 years old) taken 7 weeks after a short time contact with an approximately 2 TBq caesium-137 source. Ulceration of the thumb and rapid healing of the index finger. [W. Paile, STUK, Finland]*

AS, female, 78 years, RT's great-grandmother, had a prolonged exposure owing to her long stay in the house. She developed a moderate bone marrow syndrome from which she recovered (see Fig. III-3 in Annex III). She died from cardiovascular failure in December 1995. (Severity grading is not given owing to protracted exposure — see Section 8.2.)

AS used to stay mostly in the kitchen where the source was discovered. Her dose was assessed to be 2.7 Gy after dose protraction had been taken into account.

On 18 November 1994, she was hospitalized at the district hospital in Keila and then at the Tallinn Magdalena Hospital, Department of Surgery. Her general state was satisfactory upon admission, although she complained of exhaustion. Her haematological evolution was characterized by a mild anaemia accentuated by vaginal bleeding on 24 November. This necessitated red blood cell transfusions on 25, 28 and 30 November. Fluctuating but pronounced thrombocytopenia and granulocytopenia continued until the beginning of December 1994 and were subsequently followed by a progressive recovery of blood cell counts (Annex III, Fig. III-3). This improvement was confirmed

by the recovering profile of the myelogram in mid-December. The treatment included preventive therapy with antibiotics, antifungal compounds as well as corticoids.

Subsequently, AS was hospitalized at the Tallinn Magdalena Hospital twice during the following year, in May and November 1995, for a decompensation of a chronic cardiovascular ischemia (already known before the radiological accident) as well as an anaemia that the medical team related to iron and vitamin B<sub>12</sub> deficiency. AS died on 31 December 1995 with the diagnosis of cardiac failure.

## **8. GENERIC LESSONS LEARNED**

This accident graphically illustrates the risks associated with insufficient and/or inadequate source control, and the information presented in this report provides a basis for reaching conclusions about the causes of the accident and how it was dealt with. Some of these conclusions are specifically relevant to the facilities in Estonia, and shortly after the IAEA investigation the Estonian authorities implemented actions to rectify the weaknesses identified. There are also several conclusions which are of relevance and interest to the radiation protection community generally, and it is these general 'lessons to be learned' which are considered here. Some of these are of relevance to regulatory authorities and waste disposal operators, while others will be of direct relevance to those in the medical community who may need to identify and treat persons who have received radiation injuries. The conclusions and recommendations are accordingly divided into these two categories.

### **8.1. LESSONS FOR REGULATORY AUTHORITIES AND WASTE DISPOSAL OPERATORS**

The conclusions reached for each aspect of the accident are presented, followed by the associated measures *for prevention (in italic type)*.

#### **8.1.1. Lessons from the history of the source**

The source and associated metal container could not be identified by the authorities as a source which had been used and registered in the country. However, the source activity and the nature of the container in which it was initially housed indicate that it might have been part of an irradiator assembly. No gamma irradiators have ever been operated in Estonia and hence it is possible that the source and metal container were brought into Estonia from the Russian Federation with miscellaneous

scrap metals for export to western Europe. Such a sequence of events is a cause of concern, implying that there is significant potential for other sources to reach the public domain by similar pathways.

As opposed to the situation elsewhere, where sealed sources and devices are constructed to ISO or IEC standards, and where comprehensive documentation on source designs and uses is available, there appears to be little available information on the range of sources manufactured in the former USSR. The origin of the source assembly causing the accident and the type of equipment it belonged to are still unknown. With such uncertainty, it is not unlikely that other sources will appear in future.

*National regulatory authorities shall keep adequate records of the types of radioactive sources that are present in their countries.* The revised International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (BSS, [10]) require national authorities to set up a system of notification, registration and licensing of sources, and this system will provide the data needed to monitor the safety of sources in the country.

*National regulations shall require users of radioactive sources to keep comprehensive records of all the sources in their possession.* The BSS requires that “a periodic inventory of movable sources be conducted at appropriate intervals to confirm that they are in their assigned locations and are secure” (BSS, para. 2.34(c)). The legislation should also prohibit the unauthorized disposal of radioactive materials and should require that comprehensive records be kept describing the sources disposed of and the methods of disposal.

Regulatory authorities need to be aware of the possibility that sources will be found in scrap metal, and need to put in place procedures to identify such sources should they appear, and procedures to safely recover them. Suitable information and guidance should be provided to scrap dealers and exporters and secondary metal smelters.

### **8.1.2. Lessons from the operation of the waste disposal facility**

The waste disposal facility at Tammiku was not suitable for the storage and disposal of hazardous radioactive sources. Although a record of each consignment of waste was made, there was not sufficient segregation or preconditioning of sources prior to disposal, other than the placing of the more active gamma emitters in the highly shielded vault reserved for their disposal. Sources were placed into this vault via an S-shaped steel tube which penetrated the shielding, but this method of entry restricted the use of the vault to sources with relatively small dimensions. As a consequence, the caesium-137 source involved in this accident was on both occasions disposed of in the central, low activity section of the pit because the container was too large to drop through the S-tube. Thus, the source, with relatively high activity, was not in the shielded inaccessible part of the repository.

Some sources, including the one involved in this accident, had been disposed of into the facility without the radionuclide or the activity being known. This made it difficult to determine the original source from the dose rates measured around the pit.

The security of the facility was also inadequate. Although the entrance gate into the facility and the door into the disposal pit were fitted with an electrical intruder alarm, the alarm was very rudimentary and could be easily overridden. The fence around the facility was also in a poor condition and could be crossed very easily. Furthermore, the operators of the facility took no action when it was noted that a break-in had occurred and that the measured dose rates around the pit had decreased considerably.

Regulatory authorities should ensure that sufficient resources are made available for the construction of comprehensive source disposal facilities and the establishment of a radioactive waste management system. The IAEA Safety Standard on national systems for radioactive waste management [11] establishes the following:

*“The basic requirements of a radioactive waste management system are:*

- (a) Identification of the parties involved in the different steps of radioactive waste management, including waste generators and their responsibilities;*
- (b) A rational set of safety, radiological and environmental protection objectives from which standards and criteria may be derived within the regulatory system;*
- (c) Identification of existing and anticipated wastes, including their location, radionuclide content and other physical and chemical characteristics;*
- (d) Control of radioactive waste generation;*
- (e) Identification of available methods and facilities to process, store and dispose of radioactive waste on an appropriate time-scale;*
- (f) Taking appropriately into account interdependencies among all steps in radiological waste generation and management;*
- (g) Appropriate research and development to support the operational and regulatory needs;*
- (h) The funding structure and the allocation of resources that are essential for radioactive waste management, including decommissioning and, where appropriate, maintenance of repositories and post-closure surveillance.”*

As part of the establishment of this management programme, it is recommended that regulatory authorities ensure that appropriate guidelines are in place for the categorization of radioactive wastes. The mechanism of disposal will then depend on the type of waste and its potential hazard. The disposal of liquid scintillation vials containing trace levels of tritium, for example, need not be subject to the same rigorous standards of disposal as gamma radiography sources, and the disposing of such low level waste in the same facility as high level waste will result

in the unnecessary filling of valuable space in the waste facility. To ensure the economical use of space, the waste disposal operator should also compact waste, where practicable, prior to disposal. In the case of the Tammiku accident the disposal of the source within the metal container, and then in a lead box, was an inefficient use of the limited space available.

The radionuclide content and activity of all radioactive sources should be identified prior to disposal. Ideally, suitable analytical equipment should be available for this (e.g. gamma spectrometry facilities), but if such instruments are not available, attempts should be made to identify the radionuclide by use of such techniques as half-value layer measurements, with an appropriate geometry and with provisions for the protection of personnel.

Legislation, or the conditions of the site authorization, should require the waste disposal operator to keep comprehensive accountancy records of all the radioactive material disposed of. These records should include radionuclide, activity, origin, date and location of disposal, and form of the source.

The waste disposal operator should ensure that a comprehensive monitoring programme is carried out around a radioactive waste disposal facility. This programme should include the periodic measurement of radiation dose rates and the sampling of any water run-off from the site. Notification and appropriate action levels should be determined for these measurements. The staff at the facility should also be encouraged to check the safety and security systems at regular intervals and report anything unusual to the site managers.

Appropriate staff training programmes should be established to secure the necessary competence of staff, to foster the necessary dedication to quality and safety, and to keep the staff up to date with changes in relevant technology and regulations [11].

### **8.1.3. Lessons from the source recovery**

The Estonian Rescue Board responded promptly to the discovery of the source at Kiisa, and rapidly recovered the source. However, although the members of the Board had some experience of source recoveries, and had also been provided with basic training in radiological protection, the lack of specialist knowledge, of well prepared emergency plans and of suitable emergency equipment was apparent in some of the actions that they took. The decision to evacuate the area was taken on a rather arbitrary basis, since no intervention criteria of dose or dose rate were available. The absence of any tools for the remote handling of the source necessitated a member of the Board picking up the source in his hand, and this action was also indicative of insufficient training. While the Board had satisfactory radiation dose rate monitors, their contamination monitoring facilities were very limited, and it is doubtful that they would have detected any radioactive contamination had it occurred.

*Regulatory authorities need to ensure that suitable emergency plans are in place for dealing with incidents involving radioactive sources. These plans would include intervention levels that can be used as an input to decisions on sheltering and evacuation. The tasks and responsibilities of the organizations specified in the emergency plans would be well defined to facilitate the response of governmental and intergovernmental organizations should an emergency occur. Requirements on the content of emergency plans and the derivation of intervention levels are given in the BSS [10].*

*The organization responsible for the implementation of the emergency plans needs to be provided with suitable training in radiological protection and source recovery procedures. It also needs suitable emergency equipment, including long handling tongs, spare lead pots of different sizes for source recovery purposes, and appropriate radiation monitoring equipment for both dose rate and radioactive contamination. Rehearsals of the emergency plans should be carried out at regular intervals.*

## 8.2. LESSONS FOR MEDICAL STAFF

Lost or stolen sources represent one of the highest potential radiation hazards to the general population. Accidents which have actually occurred with such sources could easily have been misinterpreted and the cause not recognized. The actual cause was often discovered by chance and it should be recognized that, in the accident in Tammiku, only the awareness of the physician who examined RT on 17 November 1994 led to the radiation aetiology of the injury being recognized. This medical reaction was crucial in the sense that it initiated the rescue action and limited the extent of the consequences to the members of the family. It also prevented significant accumulated doses to the population living in the vicinity. This underlines the need for general medical practitioners to be aware of the effects of radiation in order to provide the victim(s) with appropriate care and to prevent the escalation of such an accident.

In this accident, several features can be singled out from the case of RT. The pancytopenia he presented was an aplasia as defined by granulocyte counts below  $0.5 \times 10^9 \text{ L}^{-1}$  and platelet counts below  $50 \times 10^9 \text{ L}^{-1}$ . This aplasia was prolonged until day 40 after the acute exposure, i.e. day 60 after the beginning of the exposure. Had the exposure been acute and homogeneous, such a severe and prolonged haematological deficiency could be expected for a dose in the range of 4–6 Gy [12].

Granulocyte and platelet counts in this range can, however, be seen during a few days in cases of acute exposures of 2 Gy, according to experience from the Chernobyl accident and other accidents in the former USSR [13].

From this point of view, the whole body dose estimate (1.1 Gy) based on the acute calibration curve for dicentric chromosomes was clearly an underestimation.

The protracted dose estimation (2.7 Gy), on the other hand, was more realistic in respect to the haematological data and is not in conflict with the estimates received by physical dosimetry or EPR analysis (Table V). In fact, despite the ill defined parameters (dose rate, mixed pattern, spatial heterogeneity) in the present case, the different methods applied gave fairly consistent dose estimates (Table V). However, it should be borne in mind that the extrapolation for protracted exposure gives merely an indicative dose estimate owing to the assumptions required for the model. To limit the uncertainties that are evident in the protraction method, each biodosimetry laboratory should establish a dose–response curve for protracted low dose rate irradiation.

So, whereas the dosimetry based on chromosomal aberrations can give satisfactory assessment of the absorbed dose in the case of acute homogenous exposure, the interpretation of the results must be more cautious in other situations. The higher the number of unknown or uncertain parameters (dose rate, mixed pattern, spatial heterogeneity), the more difficult the interpretation of the results becomes. Moreover, as the clinical picture is known to vary greatly even among people that have received the same doses, it is advisable to plan individual treatment on the basis of symptoms, not only on the basis of estimated doses. Such conclusions have already been drawn following the assessments of the accidents in Algeria (1978) and in Japan (1971) [14], for which the exposures were also reported to be protracted over long periods (days or weeks).

Therefore, it has to be stressed that if a protracted exposure is expected (such as when a radioactive source is lost), medical personnel should be prepared to deal with a more severe and a more prolonged haematological depression than after a comparably estimated acute exposure.

The combined analysis of the dose estimates obtained by different methods partly overcomes the limitations of each method alone. This underlines the importance and the need for continuing research in order to find new markers that can contribute to a better assessment of the actual cellular or functional damage, for better diagnosis, prognosis and treatment.

Accidents due to the loss of control over radioactive sources often result in casualties. As medical handling of such patients involves several medical specialities and may require intensive medical care, the capabilities of some countries may be insufficient. In these instances, international collaboration, which needs to be prepared and planned, can ensure that the best possible medical care is available to the victims.

The overexposure pattern is not always clear or simple and the radiation dose can induce several types of lesion in the body. Furthermore, the higher the number of unknown or uncertain parameters (dose, dose rate, heterogeneity), the more difficult becomes the interpretation of the results of the chromosomal aberration dosimetry. This dosimetry gives global and integrated values for various parameters with reference to a mean value of DNA repair capabilities instead of that of the patient.

The acute effects of radiation overexposure may be followed and complicated by long term effects, which may only become apparent after a long period of time. These effects concern all organs and tissues, and exist for whole body as well as localized exposures. Examples are effects on the bone marrow, such as aplastic anaemia, and on the skin and underlying tissues, such as fibrosis, ulceration and late necrosis. These secondary conditions also include neoplastic transformation such as leukaemia and skin cancer. The prevention of such effects has been purely speculative until now and was restricted to follow-up in order to detect secondary illnesses as soon as possible. As new techniques become available, such as banking of frozen haemopoietic cells from individuals in order to be able to later perform an autologous transplantation, it might be reasonable to construct a realistic and global approach for the prevention and treatment of secondary diseases. This approach should not differ from the one used for other comparable diseases caused by cytotoxic agents. With regard to secondary diseases after chemical or radiological injury, the value of autologous transplantation is still being debated by specialists, notably because the collected haemopoietic stem cells could have been injured. Furthermore, the probability of secondary disease should be evaluated, in order to optimize the operation, thus balancing the total cost to society and the total benefit to the victim.

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## Annex I

### PHYSICAL DOSIMETRY

#### I-1. DOSE RATE MEASUREMENTS IN THE TAMMIKU RADWASTE REPOSITORY (NEAR KIISA) IN JANUARY–NOVEMBER 1994

The site operators at Tammiku were required to measure and record the radiation dose rate above the disposal pit immediately after every disposal. The recorded measurements for the period January–November 1994 are as follows:

Date of measurement	Measured dose rate (mGy/h)
15.01.94	5.0–6.4
01.02.94	5.0–7.0
18.02.94	5.0–6.0
20.04.94	4.0–5.0
27.05.94	4.0–5.0
30.09.94	1.5–2.0
08.11.94	0.015–0.020
18.11.94	0.7–0.85

#### Notes:

- (1) The high measured dose rate on 15 January was attributable almost entirely to the disposal of the unshielded source recovered from EMEX.
- (2) The progressive reduction in dose rate between January and October was caused by the addition of shielding, in the form of other batches of low level waste, on top of the EMEX source.
- (3) The very low measured dose rate on 8 November was consistent with the removal of the EMEX source.
- (4) The source was returned to the disposal pit on 18 November in a shielded box with 4 cm thick lead walls.

#### I-2. DOSE RECONSTRUCTION ON THE BASIS OF ENVIRONMENTAL SAMPLES TAKEN IN KIISA

##### I-2.1. Persons and laboratories involved

- (a) G. Hütt, L. Brodski, V. Polyakov — Radiometric Laboratory, Institute of Geology, Tallinn, Estonia.

- (b) I.K. Bailiff — Luminescent Dating and Dosimetry Laboratory, University of Durham, Durham, United Kingdom.
- (c) L. Bøtter-Jensen — Risø National Laboratory, Health Physics Section, Roskilde, Denmark.
- (d) Y. Göksu, A. Wieser — Institut für Strahlenschutz, GSF Forschungszentrum, Neuherberg, Germany.
- (e) E. Haskell — University of Utah, Salt Lake City, Utah, United States of America.
- (f) L. Heide — Institut für Strahlenhygiene, Bundesamt für Strahlenschutz, Salzgitter, Germany.
- (g) H. Jungner — Dating Laboratory, University of Helsinki, Helsinki, Finland.
- (h) D. Stoneham — Research Laboratory for Archeology, Oxford University, Oxford, United Kingdom.

## **I-2.2. Introduction**

Retrospective accident dosimetry using thermoluminescence (TL) of natural materials is well established and has been applied in dose assessments in Hiroshima and Nagasaki, in nuclear test sites in Nevada, in areas of the former USSR which were contaminated with radioactive wastes, and in the town of Pripyat after the accident at the Chernobyl nuclear power plant. The technique has also been shown to be very helpful in localized radiological accidents. Optically stimulated luminescence (OSL) techniques differ from TL techniques mainly by the method of stimulation of the useful signal [I-1, I-2].

Chemiluminescence (CL) has also been used to measure absorbed doses in materials such as sugar and medical tablets following accidental exposures.

In recent years, electron paramagnetic resonance (EPR) dosimetry has also been applied to accident dose reconstruction. The localized doses to exposed persons can be assessed by measurements on samples of tooth enamel. Some common household materials, such as sugar or medicines, have also been found to be useful for dose reconstruction by EPR spectroscopy.

In this study a range of materials was collected from various locations in the house for solid state luminescence (TL and OSL) and EPR and CL measurements. It was hoped that the results of these measurements could be used to determine the likely location of the source in the house, and to provide further information on doses and dose rates.

## **I-2.3. Thermoluminescence and optically stimulated luminescence**

### *I-2.3.1. Physical background*

Irradiation causes new defects to be produced in the crystal structure of minerals. The stability of these defects can be quite high (about  $10^4$ – $10^5$  years), and

this physical effect is the mechanism by which palaeodosimeters are used in radiometric dating methods. The concentration of defects, which is correlated with the accumulated dose, can be revealed by either heating the minerals or exposing them to light at a certain wavelength. Minerals under such stimulation emit light by TL (when heated) or OSL (when illuminated). The intensity of the light emitted is proportional to the concentration of defects and, hence, correlates with the accumulated dose.

High temperature annealing clears all TL and OSL information from a crystal, which then commences a new dose accumulation. Ceramic materials begin to accumulate dose information from natural background radiation immediately after production. This dose accumulation is proportional to the object's lifetime. When a ceramic material (e.g. brick or terracotta) is exposed to transient ionizing radiation, it receives an additional dose that can be determined by luminescence techniques. The accident dose information recorded in materials collected in Kiisa was large enough for the radiation dose received from natural sources to be ignored as negligible in comparison.

### *1-2.3.2. Measurement techniques*

Bricks, plant pots and other ceramic objects were measured using several techniques:

- (a) *Fine grain technique.* Powdered samples with a grain size in the range 2–8  $\mu\text{m}$  were obtained by either drilling or crushing and then separating by precipitation. The resultant grains were deposited onto aluminium or stainless steel discs for measurement. Measurements were made using high temperature TL.
- (b) *Quartz inclusion technique.* Quartz grains of approximately 100  $\mu\text{m}$  diameter were extracted from the crushed sample by etching in hydrofluoric acid and/or by density separation. Measurements were made using the high temperature peak (usually 360°C) of quartz TL.
- (c) *Pre-dose technique.* This technique uses the sensitivity changes of the 110 or 220°C TL peak of quartz after high temperature annealing (400–600°C). It is performed on approximately 100  $\mu\text{m}$  quartz grains prepared as in the quartz inclusion technique.
- (d) *Technique using porcelain.* Porcelain was prepared by either coring with a 1 cm diamond core drill and then slicing into 200–300  $\mu\text{m}$  thick slices using a slow speed diamond wheel cutter (Oxford University), or by crushing to coarse grains and acid etching (GSF).

The laboratories involved in this investigation have participated in several inter-laboratory comparisons and source calibrations. Laboratory procedures used in this

study have been described in various other publications concerning retrospective dosimetry.

An additive dose technique was used for dose reconstruction. Measurements were performed with a TL–OSL reader (Risø, Denmark). The illustration of dose reconstruction by TL using the quartz inclusion technique (Tallinn) is shown in Fig. 4 (in Section 6.2.2).

## **I-2.4. Chemiluminescence**

### *I-2.4.1. Background*

Every luminescent effect involves light emission from atoms or molecules in excited electronic levels. In CL, this state is formed during a chemical reaction. When solid substances previously exposed to ionizing radiation are dissolved in water, light is emitted. The light yield depends on the radiation dose and, in many cases, increases with the dose until a saturation value is reached.

Enhancement of the light yield is achieved if the substances are dissolved in solutions of luminol, a cyclic hydrazide of 3-aminophthalic acid which is able to dissociate two protons from the hydrazide structure in alkaline solution. The molecule disintegrates during oxidation, creating CL. Luminol reacts to 3-aminophthalate, via an unstable intermediate product, by nitrogen separation and light emission. In aqueous solutions the emission spectrum achieves a maximum at a wavelength of 424 nm. The minimum detectable dose for sugar was found to be about 0.4 Gy.

### *I-2.4.2. Measurement technique*

Aliquots of the sugar were irradiated with additive doses of 0/2/4/8/12/16/24 Gy from a cobalt-60 source (20.4 Gy/min) and preheated at 60°C for 2 days before CL measurement. The CL reaction was initiated by adding 0.2 mL luminol solution (0.7 mM luminol, 3.8 µM hemin and 11.8 mM sodium carbonate at pH11) to 15–17 mg sugar. The emitted light was measured with an AutoLumat LB953 from Berthold and the CL intensity integrated in the range 0.2–4.0 s after starting the reaction. All tests were performed at room temperature.

To estimate the accidental dose, the CL intensity was plotted against the added dose. Assuming a linear dose response, linear extrapolation of the curve onto the dose axis gives the accident dose of  $1.5 \pm 0.1$  Gy.

An attempt was also made to assess the absorbed dose in different medical tablets such as adelphane, cinnarisin, ebuprophene, intercordin and paracetamol. The

radiation induced signal could not be separated from the background signal. Therefore evaluation with the CL method for tablets was not possible.

## **I-2.5. Electron paramagnetic resonance spectroscopy**

### *I-2.5.1. Background*

When materials are exposed to ionizing radiation, chemical bonds are broken in molecules or crystals. As a result, unpaired electrons with a resulting paramagnetic moment remain in a radical or crystal defect. They can be trapped in solids for long times and used for dose reconstruction. Their concentration is proportional to the absorbed dose and can be measured by electron paramagnetic resonance (EPR) spectroscopy.

### *I-2.5.2. Measurement technique*

EPR spectra acquisition was performed on sugar samples with an EPR spectrometer (model ESR 221, manufactured in the former German Democratic Republic), equipped with a standard rectangular cavity. The spectrometer operated in the X radiation band. The spectra were obtained with a magnetic field sweep of 10 mT, modulation amplitude of 0.2 mT and time constant of 0.3 s at 4.5 mW microwave power. For low doses an accumulation regime of up to 10 magnetic field sweeps was used to increase the signal to noise ratio. An added dose was administered using a cobalt-60 radiation source with an exposure rate of 0.81 Gy/min. Sugar sample aliquots of 400 mg were used for the EPR measurements. To obtain the dose response curves, 400 mg aliquots were irradiated in a dose range up to 44 Gy.

The value of the reconstructed accident dose was 1.53 Gy.

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## Annex II

### BIOLOGICAL DOSIMETRY

Blood samples of accident victims and other people possibly exposed during the course of the accident were shipped to the Finnish Centre for Radiation and Nuclear Safety (STUK), Department of Research, and to the GSF Forschungszentrum für Umwelt und Gesundheit, Neuherberg, for biological dose assessment by chromosomal aberration analysis.

In addition to the chromosomal aberration analyses, other biological dosimetry methods such as glycophorin A (GPA) mutations of erythrocytes (University of Pittsburgh, Department of Environmental and Occupational Health) and EPR analysis of tooth enamel (Institute of Chemical Physics and Biophysics, Tallinn) were performed.

#### II-1. CHROMOSOMAL ABERRATION ANALYSIS

Eighteen subjects were studied for chromosomal aberrations at STUK. The subjects included the two brothers involved in the removal of the caesium-137 source from the radioactive waste repository (IH, RaH), three residents of the private house where the source was located for four weeks (RT, AS, BK), three neighbours and visitors to the house (LJ, AN, MM), two doctors and a nurse visiting the house (EM, IP, HH) and seven members of the rescue personnel (HS, PS, KT, BKu, KM, IP and TU). Eight persons (IH, RaH, RT, AS, BK, LJ, AN and MM) were sampled on 22 November, and the remaining ten on 13 December 1994. Whole blood cultures were established and chromosomal aberration analyses performed using standard methods accepted by the IAEA [II-1].

Mitotic indices from all subjects were comparable to those from control individuals, giving the first impression that the radiation doses were less than 3–4 Gy. The dose estimates were based on the frequency of dicentric chromosomes and calculated using the standard linear quadratic model for cobalt gamma radiation (0.25 Gy/min). Moreover, as it was known that in all cases with high exposures the radiation exposure was protracted over a minimum of seven hours (RaH, IH), and in many cases the dose was delivered during several days or weeks (AS, BK, RT), dose estimates were also given for a protracted exposure pattern using a time dependent function (see Section II-3).

Where the whole body exposure had been uniform, the incidence of aberrations among lymphocytes followed a Poisson distribution. Deviation from a Poisson distribution could indicate a non-uniform (partial body) exposure. Since evidence was

present for partial body exposure in some cases, the Qdr approach was applied (see Section II-3). Chromosomal analyses were also performed at GSF (Neuherberg), where blood cultures from five subjects (RT, LJ, KN, VN, AN) were set up in the evening of 24 November 1994. First dose estimates were available by 28 and 29 November 1994 from STUK and GSF, based on 50–100 cells analysed from the highly exposed individuals. The high dose group has been sampled repeatedly to follow up the aberration rates in lymphocytes at different times after the accident.

Table II-1 gives the results of the chromosomal aberration analyses of the 18 subjects studied at STUK and Table II-2 those of the 5 subjects studied at GSF. Of the subjects studied at GSF, three (RT, LJ and AN) were also studied at STUK. The additional 2 subjects (KN and VN) were next door neighbours of the house where the source was located. The dose estimates are expressed as average uniform equivalent whole body doses. Data on the exposure patterns to the 18 subjects studied at STUK and their estimated durations of exposure are given in Table II-3 together with dose estimations considering acute, protracted and partial body exposure.

All those subjects that showed signs of acute radiation effects (blood count changes and/or skin burns) also showed high frequencies of dicentric chromosomes in their peripheral blood lymphocytes. The whole body doses to the two brothers that entered the waste repository at Tammiku together with their fatally irradiated brother were of the order of one gray: 0.9 Gy (0.6–1.0) for IH and 0.8 Gy (0.5–0.9) for RaH. When the exposure time of 7 hours was taken into account, the G function (see Section II-3) yielded estimates of 1.2 Gy (0.7–1.6) and 1.0 Gy (0.5–1.4), respectively. Both showed a slight overdistribution of dicentric chromosomes, pointing to a non-uniform exposure and showing that they had been close to the source. The Qdr approach which was used to estimate partial body doses gave values of 2.2 Gy (1.6–2.6) and 1.7 Gy (1.1–2.2) for IH and RaH, respectively.

All residents of the house where the source was located had highly elevated frequencies of dicentric chromosomes in their lymphocytes. The dose estimated for the boy (RT) was 1.1 Gy (0.7–1.3), and 2.7 Gy (1.0–4.5) taking into account the dose protraction (STUK). The biological dose estimates for the boy by GSF were 1.2 Gy (0.9–1.5) using a high dose rate calibration curve and 1.3 Gy (1.0–1.6) using a lower dose rate curve. Both laboratories scored 45 dicentric chromosomes in 500 cells. The frequency of dicentric chromosomes did not show a Poisson distribution, reflecting the fact that the exposure had been non-uniform. This also fits the medical data as RT had severe skin burns on his hands after handling the source while repairing his bicycle. The dose to the great-grandmother (AS) was very similar to that of the boy: 1.0 Gy (0.7–1.3) by direct comparison with the calibration curve and 2.7 Gy (0.9–4.4) when the dose protraction was taken into account. However, the great-grandmother had been uniformly exposed, as evidenced by the fact that the dicentric chromosomes in her lymphocytes followed a Poisson distribution. This is in line with the knowledge of her exposure pattern. Also, the exposure of the mother (BK) had been uniform, and



a dose estimate of 0.3 Gy (0.1–0.5) was derived from the comparison with the calibration curve, and 0.5 Gy (0.1–1.0) when dose protraction was taken into account. The lower dose to the mother was consistent with the fact that she had spent less time in the house (especially in the kitchen) because of long work shifts. The partial body dose estimate, using the Qdr method, for the boy (RT) was 1.9 Gy (1.2–2.4), and for his great-grandmother (AS) 2.9 Gy (2.6–3.1).

The biological dose estimates for the other subjects studied (neighbours, visitors, medical and rescue personnel) were either much lower or the subjects did not show signs of radiation exposure detectable by the chromosomal aberration analysis. The detection level of dicentric chromosome analysis is about 0.1 Gy for gamma radiation. Of the persons that had visited the house, four subjects had indications of radiation exposure. The boy (LJ) who visited the house while RT was repairing his bicycle and handling the source was estimated to have received a dose of about 0.1 Gy (0–0.22). However, no dicentric chromosomes were scored in 500 cells of the same boy at GSF. The friend of the great-grandmother (MM) frequently visiting the house was estimated to have received a dose of the same order as LJ, 0.1 Gy (0–0.22). A nurse (IP) visiting the house to see the fatally ill patient and a member of the rescue team (KT) were both estimated to have received doses of 0.13 Gy (0–0.27). A neighbour of the family (KN) who was checked at GSF was estimated to have received about 0.18 Gy (0.002–0.510). Four other persons (HH, HS, PS and KM), a physician visiting RaH at the house and three members of the rescue personnel, each had one dicentric chromosome in the 500 cells analysed, which may be taken as an indication of minor radiation exposure (<0.1 Gy). No dicentric chromosomes were observed in the lymphocytes of the rescue man wearing rubber gloves who picked up the source and put it into the container (the dose to the hand was calculated by the Rescue Board to be 2 Gy).

## II-2. OTHER BIODOSIMETRIC DATA

Electron paramagnetic resonance (EPR) analysis of the tooth enamel of the boy (RT) was performed at the Institute of Chemical Physics and Biophysics, Tallinn, giving an estimate of 2.07 ( $\pm 0.32$ ) Gy to the tooth. The EPR signal is potentially a very powerful cumulative dose indicator and it has the advantage that it is not affected by the dose rate or fractionation, unlike the methods based on chromosomal aberration and mutation induction. A drawback is that it gives an indication only of the local dose in the head area (tooth). The tooth enamel EPR estimate obtained for the boy is in fair agreement with the cytogenetic estimate based on the time dependent G function (2.7 Gy to the whole body).

Glycophorin A (GPA) locus somatic mutation assays in erythrocytes were performed at the University of Pittsburgh on the blood samples of the great-grandmother (AS), the boy (RT) and the boy's friend (LJ). The MN blood groups of the

TABLE II-1. CAA DATA AND DOSE ESTIMATIONS BY STUK, FINLAND

Subject	No. of cells	Dic	Ace	Rc	Estimated acute dose (Gy)	Estimated dose of protracted exposure (Gy)
					(95% confidence interval)	(95% confidence interval)
RaH	500	27	21	3	0.8 (0.5;0.9)	1.0 (0.5;1.4)
IH	500	35	24	6	0.9 (0.6;1.0)	1.2 (0.7;1.6)
RT	500	45	32	4	1.1 (0.7;1.3)	2.7 (1.0;4.5)
AS	500	44	12	2	1.0 (0.7;1.3)	2.7 (0.9;4.4)
BK	500	9	12	0	0.3 (0.01;0.5)	0.5 (0;1.0)
LJ	500	2	3	0	0.1 (0;0.22)	
AN	500	0	5	0	0	
MM	500	2	7	0	0.1 (0;0.22)	
EM	500	0	0	0	0	
HH	538	1	1	0	<0.1	
IP	500	3	3	0	0.13 (0;0.27)	
HS	500	1	1	0	<0.1	
PS	524	1	1	0	<0.1	
KT	500	3	3	1	0.13 (0;0.27)	
Bku	500	0	3	0	0	
KM	509	1	1	0	<0.1	
IP	501	0	0	0	0	
TU	501	0	0	0	0	

TABLE II-2. CAA DATA AND DOSE ESTIMATIONS BY GSF, GERMANY

Subject	No. of cells	Dic	Ace	Rc	Estimated dose (Gy)	
					(95% confidence interval)	
					Curve A	Curve B
RT	500	45	37	6	1.2 (0.9–1.5)	1.3 (1.0–1.6)
JL	500	0	5	0	—	—
KN	500	2	3	0	0.170 (0.001–0.0045)	0.180 (0.002–0.510)
VN	500	0	1	0	—	—
AN	500	0	2	0	—	—

eight persons with presumably the highest doses were checked, and three of them proved to be heterozygous (MN instead of NN or MM) and suitable for the flow cytometric mutation rate determination. For the GPA dosimetry, a linear dose response function derived from the survivors of the atomic bombing of Hiroshima and victims of the Chernobyl accident was applied after background correction for age. The dose estimates were calculated from three replicate samples taken in May, September and November 1995 (95% confidence intervals in parentheses). The great-grandmother (AS) was estimated to have received 1.2 Gy (0.9–1.4), the boy (RT) 1.5 Gy (0.8–2.1) and the boy's friend (LJ), who was present while the bicycle was repaired, 0.02 Gy (0–0.2).

### II-3. BIODOSIMETRY METHODS USED FOR DOSE ESTIMATION IN PROTRACTED AND PARTIAL BODY EXPOSURE

Eighteen subjects were studied for chromosomal aberrations at the Finnish Centre for Radiation and Nuclear Safety (STUK) in Helsinki [II-2]. Eight persons (IH, RaH, RT, AS, BK, LJ, AN and MM) were sampled on 22 November 1994 (cultures set up on 24 November) and the remaining ten on 13 December 1994 (cultures set up on 14 December). Chromosomal aberration analyses were performed on Giemsa stained slides. With the culture method used at STUK (MEM medium, 48 hours), usually more than 95% of the metaphases are in the first division.

For the material analysed at STUK, the dose estimates were based on the frequency of dicentric chromosomes and calculated using the linear quadratic model for cobalt gamma radiation (0.25 Gy/min), where  $C = 1.0 \times 10^{-3}$ ,  $\alpha = 3.26 + 0.82 \times 10^{-2} \text{ Gy}^{-1}$  and  $\beta = 4.92 + 0.32 \times 10^{-2} \text{ Gy}^{-1}$ . Dose estimates were given as average equivalent whole body doses with 95% confidence intervals. However, it was known that in all cases with high exposures the radiation exposure was protracted over a minimum of seven hours, and in many cases the dose was delivered during several days or weeks. The time dependent factor known as the G function permits the modification of the dose squared coefficient and thus allows for the effects of dose protraction. The linear quadratic equation of the mutagenic yield may be modified to

$$Y = \alpha D + \beta G(x) D^2$$

where

$$G(x) = 2/x^2 (x - 1 + e^{-x})$$

$x = t/t_0$  and  $t$  is the irradiation time and  $t_0$  the mean lifetime (about 2 hours) of the breaks [II-3].

The G affects affects the quadratic component of the equation: as the time of irradiation increases, the curve approaches the linear response. At low doses (<0.3 Gy) and low dose rates the dose response in dicentric chromosomes is linear ( $Y = C + \alpha D$ ). The G function (protracted dose) estimate gives merely an indicative dose estimate owing to the assumptions required for the model. The Poisson distribution of dicentrics was tested to clarify whether the radiation exposure was uniform (dicentrics follow the Poisson distribution) or non-uniform (overdispersion of dicentrics).

Since some evidence was present for partial body exposure in some cases, the Qdr method was applied. This method is based only on the presence of damaged cells, whereas undamaged cells can be ignored. It is thus not influenced by normal cells from the non-irradiated part of the body or by those produced after irradiation. Qdr is the frequency of dicentrics and ring chromosomes among unstable cells, and follows the equation

$$Qdr = Y_{dr} / (1 - \exp(-Y_{dr} - Y_{ace}))$$

where  $Y_{dr}$  and  $Y_{ace}$  are the frequencies of dicentrics plus ring chromosomes and acentrics, respectively, determined from the gamma ray calibration curves established in the laboratory.

At GSF (Neuherberg), blood cultures from five subjects were set up in the evening of 24 November and chromosome preparations were established on 26 November 1994. Chromosome analysis was carried out exclusively on complete first division metaphases after FPG staining. Biological dose estimation was based on the frequency of dicentrics. Standard linear quadratic ( $Y = C + \alpha D + \beta D^2$ ) calibration curves from *in vitro* experiments with cobalt-60 gamma rays with a dose rate of 0.5 Gy/min (A) and 0.017 Gy/min (B) were used [II-3].

The parameters for curve A were:

$$C = 3.9 \pm 1.3 \times 10^{-4}, \alpha = 1.07 \pm 0.41 \times 10^{-2} \text{ Gy}^{-1}, \beta = 5.55 \pm 0.28 \times 10^{-2} \text{ Gy}^{-2}$$

and for curve B:

$$C = 3.8 \pm 1.4 \times 10^{-4}, \alpha = 0.90 \pm 0.40 \times 10^{-2} \text{ Gy}^{-1}, \beta = 4.17 \pm 0.28 \times 10^{-2} \text{ Gy}^{-1}$$

At the time the dose estimations were defined at GSF, there was no information on the length of exposure time for subject RT; therefore the estimates were expressed as uniform equivalent whole body doses with 95% confidence intervals.

Standard linear quadratic calibration curves are based on dicentrics analysed from *in vitro* irradiated whole blood using a high dose rate and short exposure time. Therefore, the dicentric method gives most reliable dose estimates in situations where

TABLE II-3. DOSE ESTIMATES OBTAINED BY CHROMOSOMAL ABERRATION ANALYSIS

Subject	Status	Exposure		Estimated dose (Gy) (95% confidence interval)		
		Characteristics	Estimated duration	Acute and homogeneous	Protracted only	Partial or protracted
RaH	Brother	Heterogeneous	7 h	0.8 (0.5-0.9)	1.0 (0.5-1.4)	1.7 (1.1-2.2)
IH	Brother	Heterogeneous	7 h	0.9 (0.6-1.0)	1.2 (0.7-1.6)	2.2 (1.6-2.6)
RT	Inhabitant stepson	Complex pattern: high dose rate heterogeneous + low dose rate homogeneous	?	1.1 (0.7-1.3)	2.7 (1.0-4.5)	1.9 (1.2-2.4)
			28 d			
AS	Inhabitant great- grandmother	Low dose rate homogeneous	28 d	1.0 (0.7-1.3)	2.7 (0.9-4.4)	2.9 (2.6-3.1)
BK	Inhabitant mother	Low dose rate homogeneous	28 d	0.3 (0.1-0.5)	0.5 (0-1.0)	n/a
LJ	Visitor	Low dose rate	?	0.1 (0-0.22)	n/a	n/a
AN	Visitor	Low dose rate	?	0	n/a	n/a
MM	Visitor	Low dose rate	?	0.1 (0-0.22)	n/a	n/a
EM	Physician	Low dose rate	?	0	n/a	n/a
HH	Physician	Low dose rate	?	<0.1	n/a	n/a
IP	Nurse	Low dose rate	?	0.13 (0-0.27)	n/a	n/a
HS	Rescue man	Low dose rate	?	<0.1	n/a	n/a
PS	Rescue man	Low dose rate	?	<0.1	n/a	n/a
KT	Rescue man	Low dose rate	?	0.13 (0-0.27)	n/a	n/a
BKu	Rescue man	Low dose rate	?	0	n/a	n/a
KM	Neighbour	Low dose rate	?	<0.1	n/a	n/a
IP	Nurse	Low dose rate	?	0	n/a	n/a
TU	Rescue man	High dose rate	?	0	n/a	n/a

Note: n/a, not applicable.

exposure has been acute, the irradiation has been uniform over the entire body, and the blood sample is taken soon after the exposure. The numerical values of the coefficients in the calibration curves have been shown to vary between laboratories, not least owing to different curve fitting methods. But in spite of the fact that the  $\alpha$  coefficients of the calibration curves differed considerably, the acute dose estimates for RT were almost equal in STUK and GSF.

The situation becomes more complicated as soon as the exposure is known to have been protracted. The acute calibration curve is no longer valid. In the linear extrapolation, where only the  $\alpha$  coefficient determines the dose estimate, the statistical uncertainties of this coefficient will result in unreliable dose estimates. This became evident also from the large confidence intervals in subjects RT and AS. To achieve more reliable dose estimates in protracted exposure cases, a calibration curve for long period, low dose rate irradiation would be necessary.

The differences between calibration curves for different durations of radiation exposure deserve further investigation.

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## **Annex III**

### **MEDICAL FINDINGS**

The dynamics of RiH's (25 years old) haemoglobin level shows a permanent decrease despite the intensive transfusional supportive care during his hospitalization from 25 October to 2 November 1994, the date of death (Fig. III-1).

The dynamics of the thrombocytes of RT (13 years old) from the day of his admission to the hospital (18 November 1994) for about 40 days reveals a moderate thrombocytopenia (around 50 G/L). He developed a severe granulocytopenia (<0.5 G/L from 11 November to 18 December 1994), showing a slow recovery for about a month with normalization of the number of segmented granulocytes by the end of January 1995 (Fig. III-2).

AS (78 years old) developed a moderate bone marrow syndrome from which she recovered. The minimum values of her thrombocytes were detected on 21-22 November and also on 2 December 1994, while the maximum granulocytopenia was observed from 29 November to 2 December 1994, from which date there was a rapid recovery (Fig. III-3).

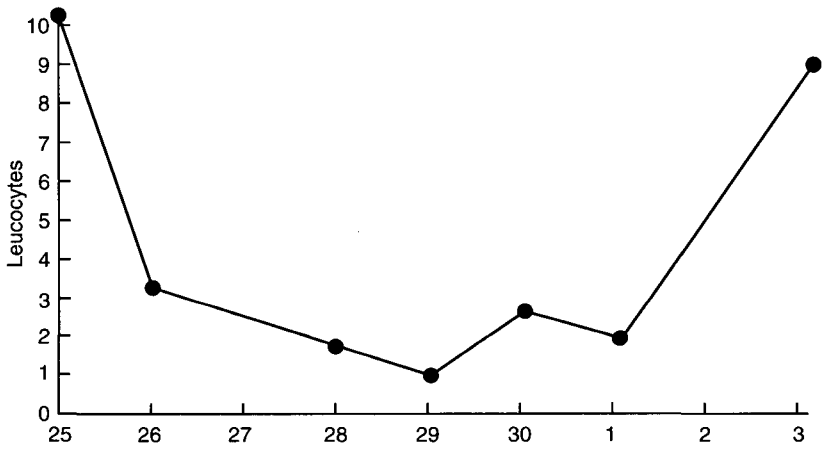
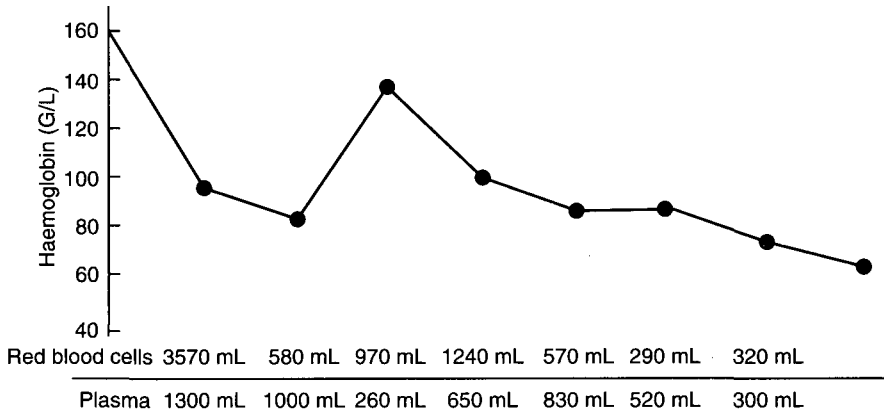


FIG. III-1. Dynamics of haemoglobin and leucocytes of RiH (under intensive transfusional supportive care from 25 October to 2 November 1994, the date of death).



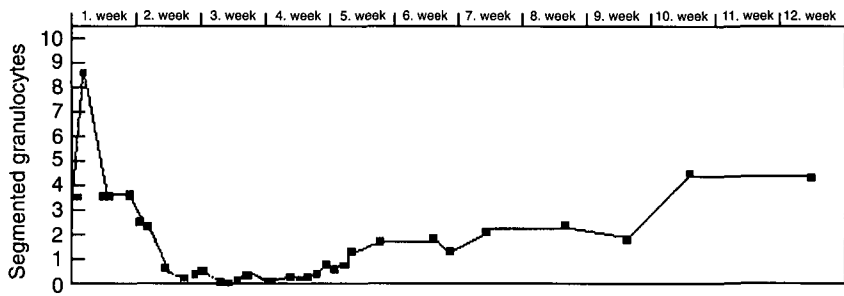
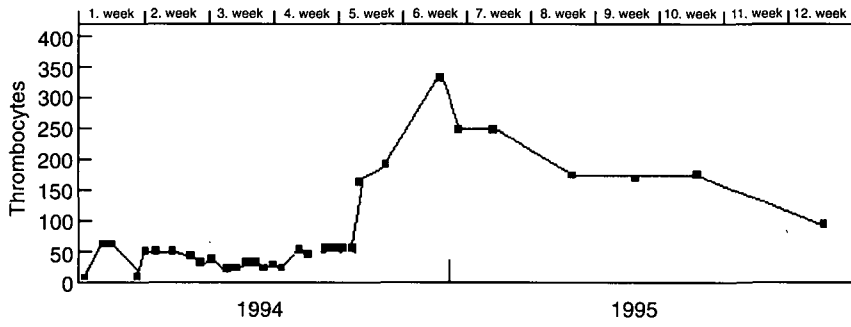


FIG. III-2. Dynamics of thrombocytes and granulocytes of RT from 18 November 1994 to 10 February 1995.

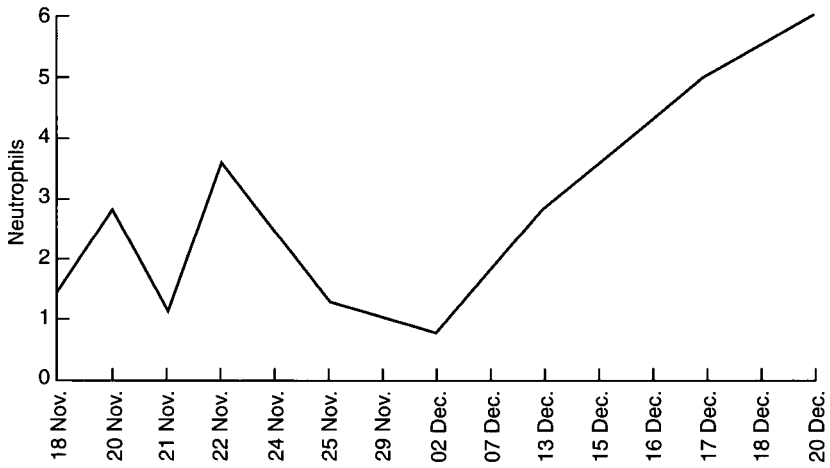
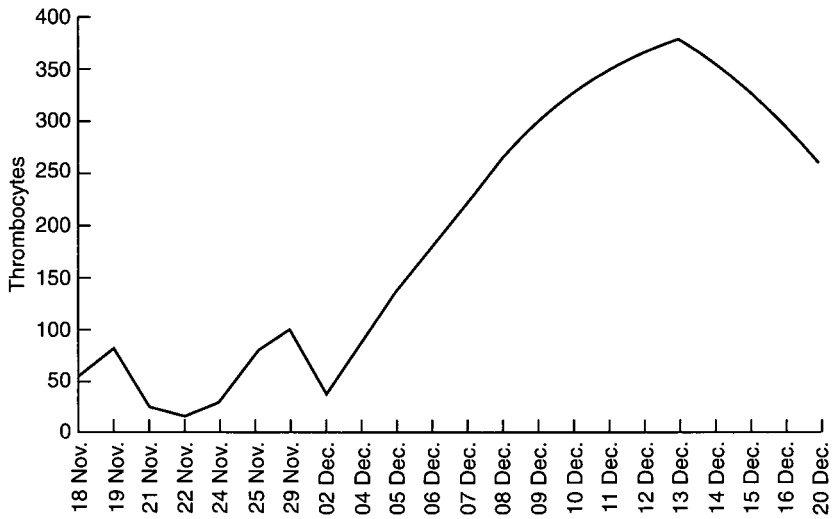


FIG.III-3. Dynamics of thrombocytes and neutrophil granulocytes of AS from 18 November to 20 December 1994.



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