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ACCIDENT RETROSPECTIVE GAMMA-DOSIMETRY IN CHERNOBYL AREA AND IN ESTONIA

Summary. Accident dose reconstructions were made on samples from Chernobyl zone and from the place of radiological accident in Kiisa, Estonia. Natural quartz extracted from ceramics served as dosimeter. Relatively low doses were measured on different kinds of samples from the town of Pripyat using the pre-dose technique. High-temperature technique was used on samples from Kiisa, where high doses were measured. The location of radiation source during the accident is determined.

RETROSPEKTYWNA DOZYMETRIA WYPADKOWA PROMIENIOWANIA GAMMA W REJONIE CZERNOBYLA I W ESTONII

Streszczenie. Autor w swoim artykule przedstawia wyniki pomiarów dawek promieniowania gamma, jakie otrzymały próbki z okolic Czernobyla i miejsca awarii w Kiisi (Estonia). Jako dozymetru użyto naturalnego kwarcu wyekstrahowanego z próbek ceramiki. Pomiary, które wykonano na różnych rodzajach próbek ceramiki pochodzących z miasta Prypeć, wykazały względnie niskie dawki. Próbki z Kiisi mierzono metodą wysokotemperaturową i wykazały one duże dawki. Określono przypuszczalne położenie źródła promieniowania w czasie awarii.

1. Introduction

Accident on Chernobyl Nuclear Power Plant evidently revealed that either routine personnel dosimetry or any emergency radiation monitoring can not supply data necessary for dose assessment for population of wide area polluted in the case of nuclear accident. Developement of detailed stationary network for radiation monitoring is very costly. So, retrospective accident dosimetry using widely spread and easily available natural or artificial materials seems very reasonable way for dose assessment in that case.

On the other hand, it can be helpful in cases of local radiological accidents as well. Last time Estonia became a bridge for scrap metal reselling from the countries of former Soviet Union to the West. Cases of discovering of radiation sources or radioactive pieces of metal, especially in the scrap metal, are becoming more frequent last time in this country. 16 ^{137}Cs sources were stolen from different Estonian plants during 1992–1994 period. At the same time, 11 unknown radiation sources were found in Estonia in 1993 and 1994. The most popular became the accident in the village Kiisa near Tallinn in 1994, where strong ^{137}Cs source was discovered in the kitchen of dwelling house.

We have performed accident dose reconstructions in the Chernobyl zone measuring different kinds of samples from the town of Pripyat. The similar work was done after the radiological accident in Kiisa using ceramic samples from inside the house.

2. Method background

2.1. Retrospective dosimetry

Thermoluminescence (TL) detectors for routine dosimetry are well known. Besides synthetic materials, many natural ones posses also good dosimetric properties. Among them are most widely spread quartz and alkali feldspars. These minerals are in the content of sand and clay, used for production of construction and utility ceramics. Glass and vitreous construction materials which contain SiO_2 as a basis, also posses certain dosimetric properties (Brodski and Hütt, 1994). Porcelain reveals TL dosimetric properties very similar to those of quartz (Stoneham, 1984).

Thermoluminescence is very popular as a tool for archaeological dating (Fleming, 1976). The main idea implies that components of a ceramics loose TL, acquired during their geological history, in the course of pot firing. High temperature of the firing resets the TL level to zero. New pot is then exposed to natural radiation from radioactive isotopes, naturally present in ceramics and its surrounding. The dose accumulated by ceramics till now is proportional to its archaeological age. Having the natural dose rate measured, one can determine the age by dividing the accumulated dose by dose rate.

Methods used in archaeological dating were successfully applied after some modification in retrospective dosimetry. Doses acquired by population from radioactive fallouts due to nuclear tests in USA were evaluated (Bailiff and Haskell, 1984). This accident dosimetry technique was also applied in very careful study on retrospective gamma-dosimetry for survivors of nuclear bombings in Hiroshima and Nagasaki (US-Japan j. reass., 1987).

We continued these studies on Estonian materials (Hütt and Brodski, 1990) and then in the Chernobyl zone (G. Hütt et al., 1993).

2.2. Measurement techniques

The first step of laboratory procedure both for dating and for accident dosimetry is extraction of TL detectors from the ceramics. These are, particularly, quartz grains. The second step is measuring of TL. The sensitivity of natural minerals serving as detectors is not known initially. It is necessary to measure it for every new object. Calibration of TL material by laboratory radiation source with known dose rate is performed to determine the sensitivity to dose. Thus, it is possible to measure TL signal from the sample, then irradiate it by any known dose and measure TL response from it. This way, however, is not suitable in most cases because natural detectors usually change their sensitivity due to heating.

That is why dose reconstruction is made by additive dose technique (e.g. Aitken, 1985). TL signal is measured using a part of a sample. Some other portions of a sample are irradiated in a laboratory before any measurements by different doses of the same order as supposed accident (archaeological). TL from these portions is measured and plotted together with TL from unirradiated sample versus laboratory calibration dose. The interception of this dependence (dose response curve) with dose axis will give accident (archaeological) dose value. The example of such dose response curve will be given on fig. 1.

Pre-dose technique is based on the fact that the value of sensitization S_N - S_0 depends on the value of accumulated

Natural quartz is good detector of radiation, which means 1) that it has high sensitivity to dose and 2) that the same radiation-sensitive defect responsible for TL peak 340°C is present in every kind of quartz regardless of its genesis. TL glow curve (heating rate 2.5 deg/s) of freshly irradiated quartz is shown on fig. 1. 340°C peak is stable enough not only for accident dosimetry, but also for archaeological dating: lifetime of the information on this peak is more than 10 000 years (Hütt et al., 1979). However, low sensitivity of the peak enables to measure doses not less than 3 Gy.

Peak 100°C, also seen on fig. 2, is used widely in accident TL dosimetry.

Its sensitivity is much higher that that of 340°C peak, but because of low depth of corresponding trap, the fading of this peak is very fast. 50% of the information fades at room temperature during about 20 min. However, 100°C peak has a property which enables its usage in accident dosimetry: the sensitivity of this peak depends on the value of accumulated dose.

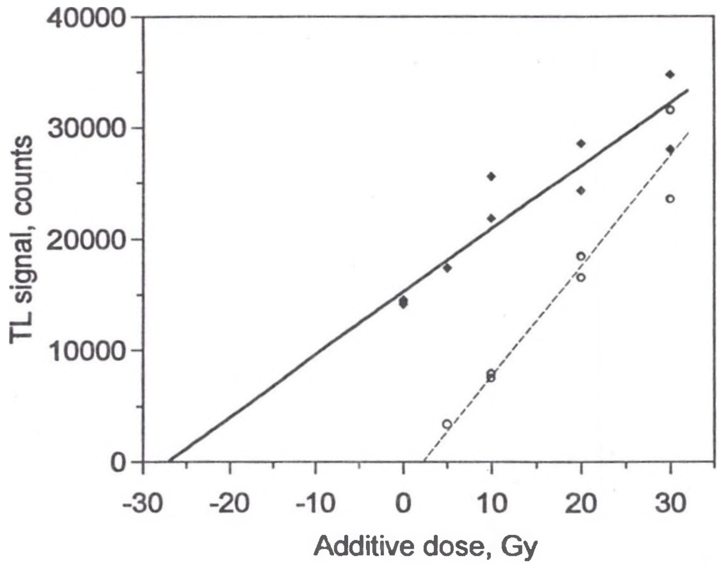

Fig. 1. Dose response curves for the sample 41. Solid line and diamonds – first glow; dashed line and circles – second glow. Second dose response curve does not come to zero due to nonlinearity at low doses

Rys. 1. Krzywe wzrostu TL dla próbki 41. Linia ciągła i pełne kwadraty – linia wzrostu pierwszego grzania; linia przerywana i puste kółka – linia wzrostu drugiego grzania. Linia wzrostu drugiego grzania nie przechodzi przez punkt 0 z powodu braku liniowości w zakresie małych dawek

2.3. Pre–dose technique

Pre–dose technique was initially used for archaeological dose determination by Fleming in 1969 and was acknowledged as a dating tool in early 70-s (Fleming, 1973).

This technique deals with sensitivity changes of 100°C peak of quartz. The sensitivity means TL signal (e.g. area under TL curve around a peak) divided by dose value. The procedure of sensitivity measurement on every step of the pre–dose technique implies 1) irradiation of a sample by a test dose and then 2) measurement of the TL signal. The test dose is 10 or 100 times less than reconstructed one, and therefore does not distort it.

The pre–dose technique generally involves following steps.

1. Initial 100°C peak sensitivity S0 is measured

2. The sample is heated to some high temperature Tact (e.g. in the span 300–500°C) and cooled

3. The sensitivity S ($S > S_0$) is measured again

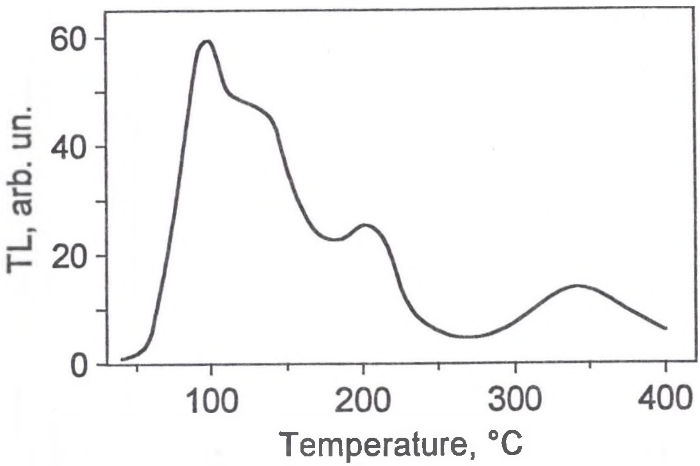

Fig. 2. TL glow curve of freshly irradiated quartz (heating rate 2.5 deg/s)

Rys. 2. Krzywa jarzenia TL średnio napromieniowanego kwarcu (szybkość grzania 2.5°C/s)

Step 2 is called thermal activation, and the temperature T_{act} – activation temperature. The sensitivity S measured after the thermal activation depends on activation temperature value T_{act}. The shape of this dependence (thermal activation characteristic – TAC) is specific for quartz extracted from different objects. As an example, TAC of quartz from a wall tile (from the town of Pripyat) is given on the fig. 3.

The typical feature for the TAC is that during temperature rise the sensitivity reaches its maximum SN and then goes down. The value that the temperature of TAC maximum is different for quartz from every object.

Pre-dose technique is based on the fact that the value of sensitization S_N - S_0 depends on the value of accumulated dose. This dependence generally has exponential behaviour. As typical, dose response curve for quartz from Pripyat sample „2 SE to 15" is presented on fig. 4.

It is necessary to use additive dose technique again to reconstruct accumulated dose, which means following. Sensitization is made for few portions of a sample, irradiated (additionally to reconstructed dose) by some different doses of the same order. The dependence of the value of sensitization S_N - S_0 is plotted against the added dose, giving dose response curve. The interception of the curve with the dose axis gives, finally, the value of the reconstructed dose. Different modifications of a measurement technique for the pre-dose method are given in papers US – Japan j. Reass., Chapter 4 (1987) and Haskell and Bailiff (1985).

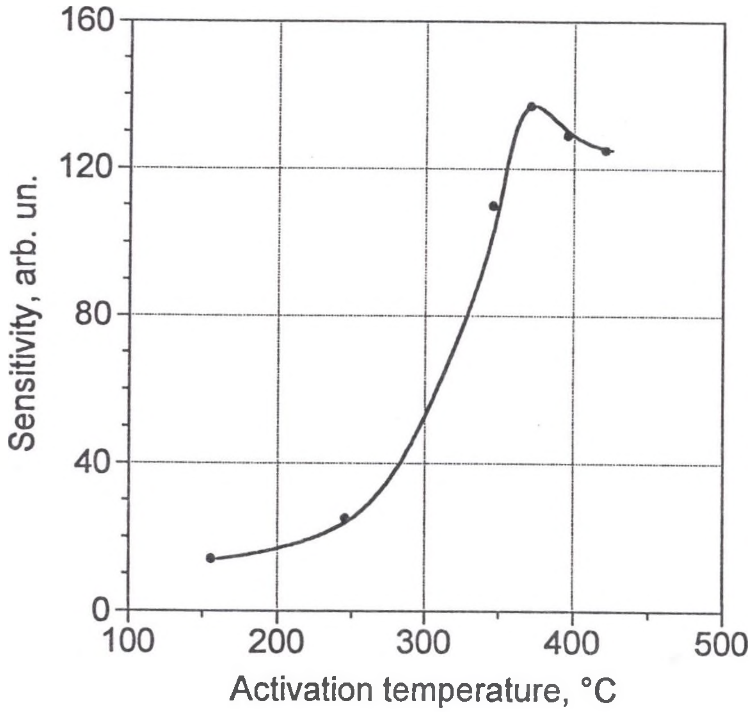

Fig. 3. Thermal activation characteristic (TAC) for quartz extracted from wall tile „2SEto15"

Rys. 3. Charakterystyka aktywacji termicznej (TAC) dla kwarcu wydzielonego z płytki ściennej „2SEto15"

3. Dose reconstruction in the town of pripyat

3.1. Aims of the study

As a result of the accident on the Chernobyl Nuclear Power Plant on April 26, 1986 at 1:23 a.m., large densely populated areas were subject to radioactive pollution. First of all, this was the town of Pripyat with its 50,000 inhabitants, situated 3 km from the CNPP and in the direction of accident plume. Evacuation of the town was not started until 36 hours elapsed since the moment of the accident. During the first days, the control over radiation situation was random or entirely absent. As a matter of fact, strict systematic monitoring at 80 points over the Pripyat was set up only in 1987. It was of greatest concern to reconstruct the doses received by the staff at the nuclear power plant and local population during April 26 – 27. We attempted to reconstruct gamma-doses since

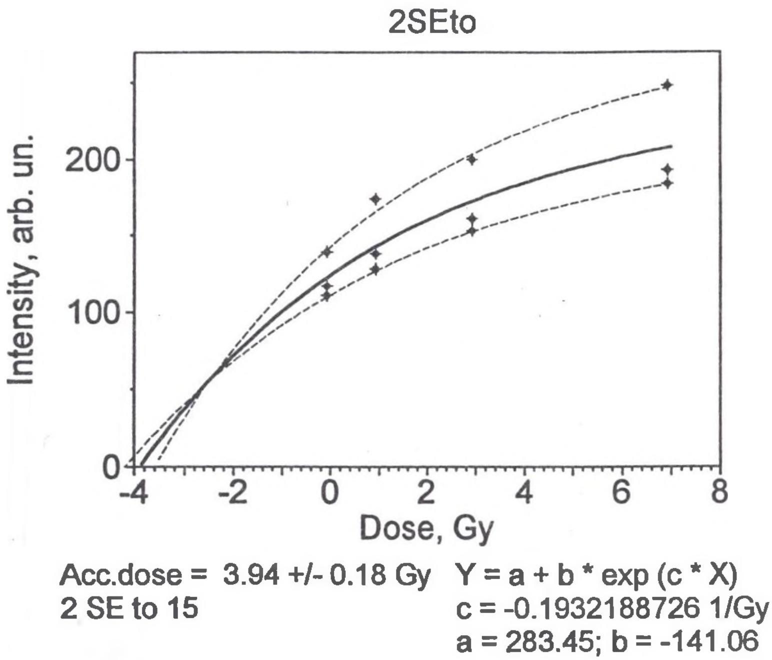

Fig. 4. Dose response curve for quartz extracted from sample 2 SE to 15. Error estimation using modification of jackknifing method is demonstrated: solid curve was built using mean values, dashed curves were obtained using only one aliquot per each dose and give minimal and maximal accumulated doses

Rys. 4. Krzywe wzrostu TL dla kwarcu wydzielonego z próbki „2SEto15". Przedstawiono zmodyfikowaną metodę „scyzoryka" do ustalania błędów: Linia ciągła odpowiada wartościom średnim, linie przerywane otrzymano dla pojedyńczych porcji ziaren dla każdej dawki i dają one wartości minimalne i maksymalne dawki zakumulowanej

the moment of the accident till now. Widely available artificial materials served as study objects: ceramic bricks, wall and floor tiles, plant pots, porcelain plumbing fixture.

The strategy of sampling in Pripyat has been described in considerable detail in Hütt et al (1993). Samples were taken from different kinds of constructions (dwelling houses, several institutions etc.) as well as from their surroundings. Sampling locations are presented on fig. 5 (Hütt et al., 1993).

Radiation situation in Pripyat is highly complicated due to nonhomogeneous spread of pollution. We tried to select most typical districts and buildings for our studies, with the aim of further extrapolation of results obtained for the area of Pripyat as a whole.

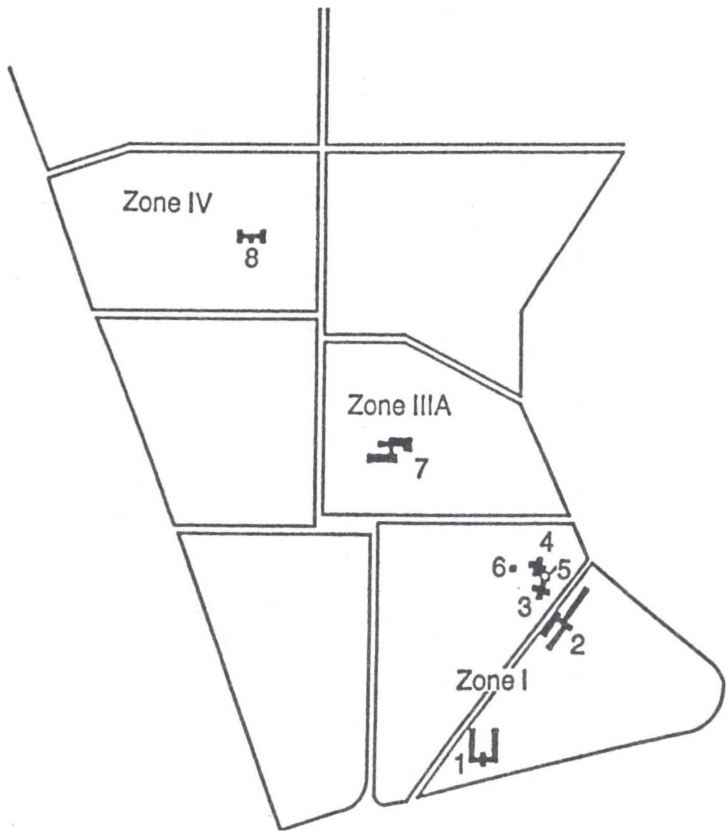

Fig. 5. Plan of Pripyat showing sampling locations. 1–Hostel No. 4; 2–Hospital, 3–15 Druzhby Narodov St.; 4–1 Kurchatova St.; 5–Playground; 6–Electrical sub-station; 7–Palace of Culture; 8–Kindergarten

Rys. 5. Plan miasta Prypeć z zaznaczonymi miejscami poboru próbek. 1 – hotel pracowniczy Nr 4; 2 – szpital; 3 – ul. Przyjaźń Narodów 15, 4 – ul. Kurczatowa 1, 5 – plac zabaw; 6 – podstacje elektroenergetyczne; 7 – Pałac Kultury; 8 – przedszkole

3.2. Measurement technique

Preparation technique was a variant of that described in Bailiff and Haskell (1984). The outer ca. 3-mm thick layer of ceramics was removed. Thus, the dose reconstructed on the basis of the remaining of the sample is an averaged gamma-component of the dose. The sample was grinded. The fraction ¡ 0.400 mm was separated by sieving, fine fraction

(¡ 0.050 mm) was removed by washing. Then the sample was treated with HCl to remove carbonates. Thereafter it was subject to treatment with hydrofluoric acid HF in steps with different HF concentrations (5, 10, 20, 40%). The sample was rinsed and dried for sieving out the fraction 0.125 - 0.200 mm. The last step was the treatment with 40% HF in ultrasonic bath during 40 minutes.

The purity of quartz was checked by the presence of high-temperature peak (340°C at heating rate 2.5 deg/s) on the TL curve. Additive-dose variant of the pre-dose method (US – Japan j. reass., Ch. 4, p. 168) was used in reconstruction of gamma-dose. Thermoluminescence was studied by means of the TL-reader constructed in Tallinn. Dose response curves were obtained in the following regime:

Photomultiplier	THORN EMI type 9514SA
Filter	C3C-22 (460^{+70}_{-80}nm FWHM, $\tau_{max} = 0.93$)
Initial temperature	40°C
Annealing to	250°C
heating rate	2.5 deg/s
Measurement	
integration interval	60 – 90°C
heating rate	2.5 deg/s
Activation to	380°C
heating rate	12.5 deg/s
halt	3 min
Test dose	5.4 mGy (beta-source ^{90}I – ^{90}Sr)
dose rate	1.8 mGy/min
Calibration dose	2.00 - 10.00 Gy
dose rate	0.44 Gy/min

3.3. Results of measurements

Figure 4 presents, as an example, a dose response curve for the sample „2 SE to 15"" with deviations reflecting error limits of the reconstructed dose (error estimation will be discussed below).

Basic results obtained are presented in the table 1

3.4. Treatment of study results

Dose response curves were built according to the less-square method using exponential fitting (program composed by V. Polyakov – Polyakov and Hütt, 1990), error calculation was carried out by means of jackknifing method (Fox et al., 1980). This method enables one to compile a statistically independent set of a searched value, which may be then treated by common statistic methods. In building dose dependencies by the additive-dose technique (see fig. 4) 3 – 5 portions of a sample were taken for each dose. In this case following modification of the jackknifing method was applied. Dose response curve was built using the whole set of data. Then several dose response curves were built taking into account only one portion for each dose: first, the portions marked with number 1, then those with number 2, etc. As a result we got a statistically independent set of reconstructed doses. The number of doses in the set was n+1, where n is the number of portions per each dose. Then the medium value and error were determined, proceeding from the assumption that the set is governed by Gauss distribution, with confidence interval 0.66.

3.5. Discussion

The data obtained allow the following conclusions to be drawn; these support preliminary results (Htt et al., 1993):

1. Ceramic objects (bricks, wall tiles, flower pots) can be used in retrospective dosimetry.
2. The trend of considerable decrease in doses with height is revealed. It is proved by the fact that pollution of soil is considerably higher than that of other surfaces.
3. Walls and windows of buildings have remarkable shielding properties.
4. The data obtained must be used in interlaboratory checking of results.

4. Radiologicalaccident in Kiisa

4.1. History

21 October, 1994 three brothers broke into the Tammiku repository of radioactive wastes, some 20 km from Tallinn, with the aim to stole some metal and then sell it. During this „operation" a radiation source fell down from one metal block. It had an appearance of small metal cylinder, about 1.5 cm in diameter and 3 cm long. One of the brothers, R.H. took the cylinder and placed it into the pocket of his jacket. He came

Table 1

Accident dose reconstructed on samples from the town of Pripyat (downwind of Chernobyl NPP). Column D – description of the sample: B – brick, P – flow pot, T – toilet tank top, F – floor tile, W – wall tile, D – drainage pipe, L – porcelain base of light source

Name of the sample	D	Location		Dose, Gy
2SE to 15	W	Druzhby narodov, 15	Playground	3.94 ± 0.18
3N/E	W	Druzhby narodov, 15	Playground	2.17 ± 0.15
5SE to 15	W	Druzhby narodov, 15	Playground	1.65 ± 0.07
PG NF (1) @ 1m40	W	Druzhby narodov, 15	Playground	1.88 ± 0.40
PG WF @ 2m 2	W	Druzhby narodov, 15	Playground	0.97 ± 0.46
HR15.10[2]LI Nwall	L	Druzhby narodov, 15	Above the entrance	0.96 ± 0.06
HR 15 roof A1	B	Druzhby narodov, 15	Roof	0.69 ± 0.24
TP B ventilation tube	B	Druzhby narodov, 15	Roof	0.54 ± 0.07
HR 15 roof D	B	Druzhby narodov, 15	Roof	0.62 ± 0.12
HR 15 roof C	B	Druzhby narodov, 15	Roof	0.32 ± 0.10
HR 15 balcony of 8 floor side C	P	Druzhby narodov, 15	Balcony	0.11 ± 0.23
5W	T	Druzhby narodov, 15	5-th floor	0.04 ± 0.02
6N	T	Druzhby narodov, 15	6-th floor	0.19 ± 0.04
9S	T	Druzhby narodov, 15	9-th floor	0.18 ± 0.04
1.7 HR15.1I[1-10]FT	F	Druzhby narodov, 15	1-st floor	0.08 ± 0.02
1 Kurchatov st-door playground	W	Kurchatova, 1	Porch wall	1.94 ± 0.26
Greenhouse	P	Druzhby narodov, 15	Greenhouse	0.20 ± 0.08
GRN.H.	D	Druzhby narodov, 15	Greenhouse	1.09 ± 0.32
Electricity	B	Druzhby narodov, 15	Transformer building	0.47 ± 0.04
School to reactor 2 floor	P	School No.1	East sill, 2 floor	0.19 ± 0.16
Hosp. DN roof tile	W	Pripyat, hospital	Roof	0.14 ± 0.14
Hospital DN GF LBY1 skirting ground	W	Pripyat, hospital	Lobby, 1st floor	0.18 ± 0.06
Hospital DN F N4 Ext. facade	W	Pripyat, hospital	Facade, 4 floor	1.10 ± 0.58
Hospital DN 4F #2	W	Pripyat, hospital	Facade, 4 f	0.68 ± 0.16
Hostel # 4, 4	W	Pripyat, hostceel N4	South corner	1.53 ± 0.50
12.8.91 hostel # 4	W	Pripyat, hostel N4	South corner	2.51 ± 0.58
Pot 2	P	Dental hospital	Window cill	0.31 ± 0.14
Pot 3	P	Dental hospital	Window cill	0.78 ± 0.13
Pot 4	P	Dental hospital	Window cill	0.14 ± 0.02
C.P. #3 stage door	B	Culture Palace	Internal gate	0.08 ± 0.03
Door	B	Culture Palace	Door to the roof	0.09 ± 0.04
Bus stop front #1	B	Red forest	Bus stop	—
Red Forest	B	Red forest	Road police house	0.40 ± 0.09

home in the village Kiisa (about 30 km from Tallinn) next morning and put the jacket, supposedly with the source inside, onto the dress-hanger in the entrance hall. Not feeling well, he went to bed and probably did not ever put on his jacket. He was hospitalized 25 October and died 2 November due to acute radiation sickness. Nevertheless, the diagnosis was kidney failure, and nobody knows about the source.

Besides R.H., 3 members of his family lived in the house. His wife P.K., 13-year old stepson R.T. and boy's grandmother A.S. 9 November R.T. was repairing his bicycle. According to official version, he found the source in the pocket of the jacket, put it into the drawer with tools and then returned the drawer to the kitchen. 17 November the boy was hospitalized with severe burns to his hands. These were recognized as radiation ones, and police was notified.

18 November a staff of Rescue Board found the source in the drawer in the kitchen. The source was lately identified as ^{137}Cs. During the rescue operation, the dose rate of 20 R/h was roughly measured on the distance of 1 m from the source. The source was then irretrievably buried in the same repository. No more data about it are ever available.

4.2. Sampling

The calculation could not estimate exact doses acquired by inhabitants of the house. Because of disagreements in the words of witnesses, it was not known accurately when the source was transferred from the jacket to the drawer. The dose rate from the source was measured very roughly. Biological dosimetry, made using blood of the most injured persons, gave whole–body doses. However, the rate of accumulation for these doses remained unknown and the doses itself need proofing. The individual and collective doses for less injured persons (such as neighbours visited the house) are not known as well.

This was a reason for performing dose reconstruction using TL. Detailed sampling was conducted in the house (Fig. 6). The samples were mostly plant pots.

4.3. Measurement technique and results

The quarter of each plant pot, most probably faced the source, was used for extraction of quartz. First of all, outer layer of the ceramics about 1 mm, exposed to sunlight, was removed by grinding. The following quartz extraction was made in the same way as for Chernobyl samples. TL was measured using RISØ reader TL/OSL-DA-12 in Dating Laboratory of Helsinki University. Measurements were performed with high-temperature technique. Heating rate was 2.5 deg/s, beta-dose rate for calibration - 0.04 Gy/s. After the first heating to 450°C, the aliquots were irradiated to the same calibration doses and measured secondarily to evaluate correction for non-linearity of dose response curves.

Integration of TL curves were made in the interval 270–400°C. Linear fitting by the

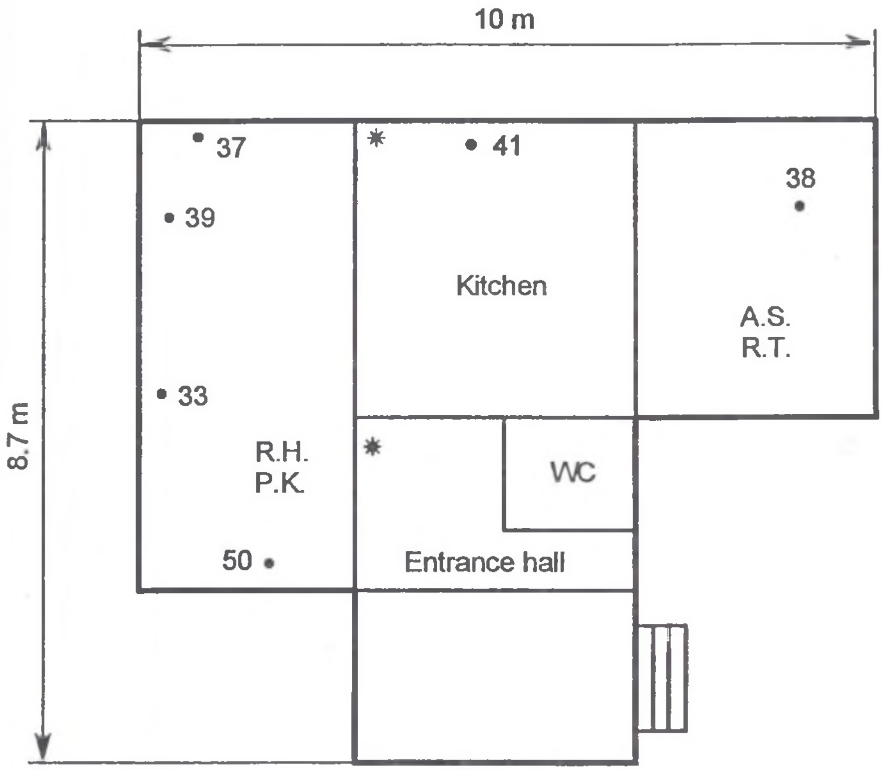

Fig. 6. Plan of the house in the village Kiisa (Estonia) with locations and numbers of the samples

Rys. 6. Plan domu we wsi Kiisa w Estonii z zaznaczonymi miejscami i numerami próbek

less squares method was applied. Accumulated dose was determined as $D = D_{rmI} - D_{rmII}$, where D_{rmI} was evaluated from the interception of first dose response curve with dose axis, and D_{rmII} (with corresponding sign) - from that of second dose response curve. As an example, dose response curves for one of the samples are given on fig. 1. The reconstructed doses are presented in the table 2.

4.4. Discussion

The calculation of accumulated doses to the samples measured was performed starting from the supposition that the source was most of the time in the kitchen, where it was found, and dose rate was 20 R/h on the distance of 1 m. The distances from supposed source location together with values obtained are also presented in the table 2.

Table 2

Accident doses reconstructed on samples from Kiisa, Estonia. Distances are given from the supposed location of the source, and doses calculated from location

No.	Reconstructed dose Gy	Distance from the source m	Calculated dose, Gy
41	29.3	1÷2	100÷25
37	17.3	2.6	15
39	13.2	3.2	10
33	7.8	4.7	4.5
50	1.0	6.0	2.8
38	19.7	6.2	2.6

As it can be seen, results of dose reconstruction are consistent. They show that the source was most of the time in the drawer in the kitchen, not in the entrance hall, as it follows from the official version. On the other hand, results proved the dose rate, measured for the source.

The dose measured for plant pot 38 do not agree with the whole pattern. This issue requires additional investigation.

5. Conclusions

Results obtained in present study show wide range of applications of retrospective accident dosimetry techniques. While in Chernobyl zone with its relatively low doses the pre–dose technique was suitable, in local radiological accident with high accumulated doses the high-temperature technique proved fruitful. It was also showed that even in such modern town as Pripyat, enough ceramic materials can be found for TL studies. The same seems true for local accidents in urban environment as well. Methodological aspects of sampling and measurement were solved.

Results obtained in the town of Pripyat can be used for evaluation of doses acquired by population of the town and liquidators during the first, most dangerous days. It can be done by subtraction of accumulated doses, obtained by integration of radiation monitoring data, which are available since some later moment.

Results obtained for the accident in Kiisa clarified the history of presence of the source in the house and proved the dose rate measured. They can serve also as a base for dose assessment for less injured persons.

ACKNOWLEDGMENTS

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Streszczenie

Wypadek w Czernobylskiej Elektrowni Jądrowej wyraźnie pokazał, że zarówno rutynowe metody dozymetrii osobistej, jak też żadne awaryjne techniki pomiaru promieniowania nie mogą dostarczyć żadnych potrzebnych do ustalenia dawek pochłoniętych przez ludność na dużych obszarach skażonych w czasie wypadku jądrowego.

Stworzenie gęstej stacjonarnej sieci pomiarów promieniowania jest bardzo kosztowne. Z tego powodu retrospektywna dozymetria wypadkowa wykorzystująca pospolicie występujące i łatwo dostępne materiały naturalne lub sztuczne wydaje się być bardzo sensowną alternatywą przy ustalaniu dawek w takich przypadkach. Podobnie dozymetria retrospektywna może być przydatna w przypadku lokalnych wypadków radiologicznych, na przykład w razie częstych kradzieży źródeł lub materiałów radioaktywnych.

Wykonane badania pokazują szeroki zakres zastosowań retrospektywnej dozymetrii wypadkowej. W strefie Czernobyla, ze względnie małymi dawkami odpowiednia okazała się technika „pre-dose". Natomiast w analizowanym lokalnym przypadku radiologicznym związanym ze skradzionym źródłem ^{137}Cs, z dużymi wartościami termoluminescencyjnymi. Badania udowodniły też, że praktycznie w każdym otoczeniu, również zurbanizowanym, można znaleźć materiały nadające się do celów dozymetrii retrospektywnej.