

**DEVELOPMENT OF AN IMPROVED DOSE RECONSTRUCTION  
SYSTEM FOR THE GENERAL POPULATION AFFECTED BY THE  
OPERATION OF THE MAYAK PRODUCTION ASSOCIATION**

**Submitted to the Office of International Health Programs, U.S. Department of Energy  
for the  
US–Russia Joint Coordinating Committee on Radiation Effects Research**

**Progress Report on Project 1.1**

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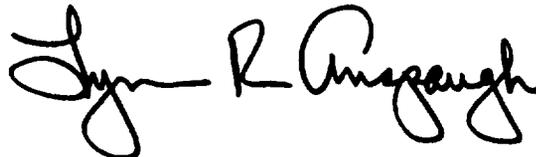
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**October 1998**



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## **BACKGROUND**

### **History of the Mayak Complex**

The Mayak Production Association (MPA) was the first facility in the former Soviet Union for the production of plutonium for nuclear weapons (Fetisov 1996; Fetisov et al. 1993; Akleyev and Lyubchansky 1994) and is located north of the City of Chelyabinsk. Construction of the MPA facility began in 1945; startup of the plant occurred in June 1948 with the operation of the initial uranium-graphite reactor for the production of plutonium. The first radiochemical plant was placed on-line in December 1948, and the production of enough plutonium for an initial weapons-related test soon followed. There was also a plant for "standard plutonium production" (Koshurnikova et al. 1996). Over the intervening years a total of seven reactors have operated at this site; five uranium-graphite reactors have now been decommissioned. The two remaining reactors are operated mainly for the production of isotopes. Since 1977 the radiochemical plant has been used extensively to reprocess fuels from power reactors and from transport and research reactors. The radioisotope plant dates back to 1962 and is now one of the major suppliers of sources and preparations of radionuclides. Other major activities include the Instrument Engineering Plant, the Repair and Machine Shop, the Central Research Laboratory (CRL), and the Experimental Scientific Research Centre.

Significant worker and population exposure in the nearby Urals region occurred as a result of failures in the technological processes in the first facilities of the MPA in the late 1940's and early 1950's. Members of the public were exposed via major discharges to the Techa River and to the atmosphere. In addition one of the waste-storage tanks exploded in 1957 with another major release. The Mayak CRL undertook early radiation monitoring and determination of exposures. The major work that has been done in reconstructing the identity and quantity of material released to the Techa River was performed by staff of the CRL during the 1950's.

### **Environmental Releases**

The major sources of environmental radioactive contamination were the discharges of about  $10^{17}$  Bq of liquid wastes into the Techa River (1949–1956); an explosion in the radioactive waste-storage facility in 1957 (the so-called Kyshtym Accident) that formed the East Urals Radioactive Trace (EURT) due to dispersion of  $7.4 \times 10^{16}$  Bq into the atmosphere; and gaseous aerosol releases (about  $2 \times 10^{16}$  Bq of  $^{131}\text{I}$  in total) within the first decades of the facility's operation.

The majority of the releases occurred in the first years of facility operation, when the waste-management facilities were still being developed. Low- and intermediate-level liquid wastes were routinely released; these routine releases, however, were not the only source of environmental contamination. The Mayak Facility C (radioactive waste-storage facility) was the site of both the Techa River releases and the EURT release. The first available description of the Techa River releases is in the report of the Alexandrov Commission, a special governmental panel created after the discovery of high environmental contamination in 1951. (This report itself is still classified.) In mid-1951, high levels of contamination were found in canals



returning supposedly clean cooling water to the Techa River. Studies showed a correspondence of pulses of large amounts of radioactive materials with application of cooling to a particular series of tanks. These tanks were operated in a manner somewhat analogous to those at the U.S. Hanford Site: Liquid wastes came from multiple sources within the Mayak processes, wastes cascaded from one tank to another, and complex chemical interactions produced solids that scavenged certain radionuclides in some tanks and freed them in others. Thus, a fraction of the Techa River release was soluble and another fraction was associated with particles. Therefore, the mixture of radionuclides released is very difficult to determine. Soon after the discovery of the uncontrolled and unmonitored releases the low- and intermediate-level discharges, and the other cooling water, were diverted into Lake Karachai.

### **Exposure of the Population**

There were many villages on the Techa River that were downstream from the MPA when the discharges occurred to the river. Villagers were exposed according to a variety of pathways. (A more complete discussion of all pathways that have been considered is detailed in Degteva et al. 1996a.) The more significant pathways included drinking of water from the Techa River, external gamma exposure due to proximity to the Techa River bottom sediments and shoreline, and use of the Techa River water for irrigation of food crops. In addition some members of the Techa River Cohort may have been exposed to the gaseous emissions from the MPA, although this is believed to be a relatively minor source of exposure. After the extent of the major contamination of the Techa River became known, several villages on the upper part of the Techa were evacuated. Villagers on the lower part of the Techa have remained in their homes up through the present time.

### **Unique Opportunities**

Both the Mayak Worker and the Techa River Cohorts are unique in that members have received unusually high doses, but at low-to-moderate-dose rates. It is likely that study of these populations will provide the best opportunities to determine whether a dose-rate-reduction factor exists for the induction of cancer in human populations. The Techa River Cohort is one of a few that represents an unselected population; the presence of two distinct ethnic groups also provides the opportunity to examine the population variability of risk factors.

The foundation of the dose reconstruction for the Techa River Cohort is also unique. *Over half of the members of the cohort have had whole-body counter measurements of direct relevance to the dose reconstruction.*

### **Preliminary Studies**

Studies of the possible effects of radiation on those exposed to the releases to the Techa River were started in Russia in the 1950s. Russian and United States scientists have been involved in collaborative research programs since 1995.



**Epidemiologic studies.** Medical checkups of people living in the Techa Riverside communities had been started by 1951. In 1955 a specialized medical institution, known as Specialized Dispensary No. 1, was established in order to determine the health status of the exposed population. This institution became known later as Branch No. 4 of the Institute of Biophysics and is now the URCRM. In 1968 the Techa River Registry was created with the goal of including residents of the Techa Riverside villages who lived there during the periods of high exposure from 1949 through 1952. The registry includes data on 26,500 such residents; the registry also contains data on 29,700 persons exposed in utero and/or the progeny of exposed parents and on 7,800 persons who were late entrants exposed after 1952

A preliminary report on the status of the follow-up of the Techa River Cohort has been published by Kossenko et al. (1997). It is reported that, despite a number of limitations, there does appear to be an increasing risk of mortality from leukemia and other cancers with increasing radiation dose.

**Dose-reconstruction studies.** Systematic measurements of radioactive contamination in and near the Techa River started in the summer of 1951. The contamination of the river water, bottom sediments, flood-plain soils, vegetation, fish, milk, and other food stuffs, and external gamma-exposure rates were measured.

The population of the contaminated territories was chronically exposed to external and internal irradiation. In addition to medical examinations, individual data on the conditions of contact with the contaminated river (the distance of the house from the water's edge, the source of drinking water, fishing, etc.) were collected. Also, radiometric measurements of bioassay and autopsy samples were performed. All places and terms of residence inside the contaminated area were collected for the members of this registry for the purposes of individual-dose reconstruction. Also, extensive measurements of  $^{90}\text{Sr}$  content in teeth were performed beginning in 1960 and in forehead bone beginning in 1976; whole-body counting for  $^{90}\text{Sr}$  has been performed since 1974; at this time over half of the members of the Techa River Cohort have had at least one whole-body count.

The basis of the past dose-reconstruction efforts for the Techa River Cohort has been summarized in several publications, including Degteva and Kozheurov (1994); Degteva et al. (1994, 1996b, 1997a); Kozheurov (1994); Kozheurov and Degteva (1994). The absorbed doses due to external exposure were estimated on the basis of systematic measurements of gamma-exposure rate along the banks of the river and the typical life-style patterns of the inhabitants of the riverside villages. This approach has given the average annual absorbed doses from external sources for different age groups in each village. A major activity of the current project is to provide this information on an individual basis.

Several efforts have been undertaken in an effort to validate the external doses. One study was undertaken in the now evacuated Village of Metlino. Samples of bricks from abandoned buildings were collected, the quartz was extracted from the bricks, and dose was assessed by using the quartz as a thermoluminescent dosimeter (Bougrov et al. 1995). The results were doses ranging from 0.76 to 5.28 Gy in the outer cm of the bricks. The highest dose



was from a brick wall located near the Techa River and the Metlinsky Pond. Another study (Romanyukha et al. 1996) was performed using electron paramagnetic resonance (EPR) measurements of teeth collected from current or former residents. Teeth were collected from 86 inhabitants of the town of Kamensk-Uralsky, which is not located on the Techa River, in order to determine the age-dependent contribution of background to tooth-enamel dose. EPR measurements of teeth and  $^{90}\text{Sr}$  whole body counts were performed on 22 residents of the middle and lower Techa Riverside communities, and absorbed doses were measured in teeth from five residents of the upper Techa.

Results of the above mentioned studies and historical evidence indicates that the main contributor to internal exposure among the radionuclides released into the Techa River was  $^{90}\text{Sr}$ , which is accumulated in bone tissues and retained for many years. In vivo beta-ray measurements on teeth, which have been performed since 1960, and a large number of  $^{90}\text{Sr}$  measurements in whole body have been the basis of internal dose reconstruction (Kozheurov 1994). The reconstruction of internal dose depends on both estimates of the intake and models for metabolism of ingested radionuclides. Beta-ray measurements on teeth are utilized to deduce the annual levels of intake of  $^{90}\text{Sr}$  in the different villages in the different age cohorts. The ingestion of other radionuclides ( $^{89}\text{Sr}$  and  $^{137}\text{Cs}$  predominantly) occurred mostly with water in the first three years of the river contamination. The intake rates of  $^{89}\text{Sr}$  and  $^{137}\text{Cs}$  were therefore derived from estimates of the ingestion of  $^{90}\text{Sr}$  scaled in terms of the radionuclide composition of the river water. These data were used to estimate age-dependent intake rates for all Techa River villages (Kozheurov and Degteva 1994). Calculation of absorbed doses in tissues due to radionuclide incorporation is based on age-dependent metabolic and dosimetric models and the corresponding ingestion rates. A large number of measurements of  $^{90}\text{Sr}$ -body content made with a whole-body counter (WBC) has been utilized for the validation of the metabolic model for strontium retention in human bone (Degteva and Kozheurov 1994). Absorbed doses in red bone marrow (RBM) and bone surfaces (BS) have been calculated for all age cohorts. The absorbed doses in RBM and BS are substantially higher than those in other tissues, because  $^{90}\text{Sr}$  was the main radionuclide of interest and strontium is a bone-seeking element. The upper limit of total doses absorbed in RBM is estimated as about 3 Gy.

## THE CURRENT STUDY

This project is a comprehensive program to develop improvements in the existing dosimetry system for the members of the Techa River Cohort by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results. This current project is the result of the first year's pilot study (Degteva et al. 1996a) and extensive meetings and discussion among the participants in the dosimetric and epidemiologic studies. The details of the project have been specified in the proposal document (Degteva et al. 1996c). Following approval of the proposed work it was necessary to provide a revised list of Tasks and Milestones. This latter list was reproduced as Appendix 1 in the March 1998 Progress Report (Degteva et al. 1998).



The specific aim of this project is to enhance reconstruction of external and internal radiation doses for approximately 26,500 individuals in the Techa River Cohort. The purpose of the enhanced dose reconstruction is to support companion epidemiologic studies of radiogenic leukemia and solid cancers (NCI-RERF-URCRM Project and JCCRER Project 1.2). The current database of preliminary individual doses will be expanded and upgraded, and the uncertainty in the doses reconstructed will be evaluated.

The most recent joint meeting of the Russian and American team members took place in April 1998 in Chelyabinsk. About half of the time was spent in preparing for and participating in a review meeting for the Russian and US Scientific Review Groups. The remainder of the time was spent in completing preparation of publications for submission to peer-reviewed journals, review of progress on various tasks, and initiation of the task on analyzing the uncertainty in the doses now being calculated and the doses that will be calculated in the future.

### **Progress on Task 1:**

*Task 1. Feasibility analysis of the development of a special system for obtaining tooth samples from the Techa River residents (M. Degteva).*

*Subtask 1. Examine the feasibility of establishing a system of tooth collection for the members of the Techa River Cohort. This system should be oriented toward the collection of teeth as they are being extracted for dental health purposes. A special aim would be the collection of teeth from former residents in the Upper Techa River locations that have long since been evacuated.*

This work was completed with delivery of the Milestone 5 Final Report (Degteva et al. 1997b), which was included as Appendix 3 in the March 1998 Progress Report (Degteva et al. 1998).

### **Progress on Task 2:**

*Task 2. Source-term and environmental data analysis (M. Vorobiova).*

*Subtask 2. Check the consistency of available source-term and historical monitoring data in order to verify and confirm limited available information on the source term.*

*Subtask 3. Develop a simple empirical river model in order to verify distance dependence of radionuclide composition of river water and to link radionuclide concentrations in water and bottom sediments with  $\gamma$ -exposure rates on the banks near the river.*

This work was completed with the delivery of the Milestone 1 Final Report (Vorobiova et al. 1997), which was an Attachment to the March 1998 Progress Report (Degteva et al. 1998). In addition, two manuscripts based upon this Milestone Report have been prepared and have been submitted to *Health Physics*. These two manuscripts are reproduced here as Appendices 1 (Vorobiova et al. 1998) and 2 (Vorobiova and Degteva 1998). Both of these manuscripts were distributed to the members of the Scientific Review Groups in April 1998, but the manuscripts have not previously been included in a Progress Report.



**Progress on Task 3:**

*Task 3. External dose reconstruction (D. Burmistrov)*

*Subtask 4. Verify  $\gamma$ -exposure rates on the banks near the river and validate accumulated doses calculated on the basis of verified  $\gamma$ -exposure rates in specific points of the upper and middle Techa by the results of thermoluminescent measurements in bricks from the same sites.*

*Subtask 5. Enter into computer and analyze available data on the outdoor and indoor  $\gamma$ -exposure rates on the territories of the Techa Riverside settlements in order to determine the dependence of  $\gamma$ -exposure rate from the distance from the edge of the water and to verify the contributions to the total external dose from the sources of radiation exposure in the streets, gardens, and houses.*

*Subtask 6. Evaluate organ-specific absorbed doses from external exposure (based on literature data) and include these values into the TRDS system code.*

*Subtask 7. Validate absorbed doses in individuals from external sources of radiation by the results of electron paramagnetic resonance measurements in teeth.*

Routine work on this task is continuing. Some results were included in the Milestone 1 Report (Vorobiova et al. 1997). Most of the work to be done on this task is scheduled for a later period with completion in February 1999.

**Progress on Task 4:**

*Task 4. Internal dose reconstruction (V. Kozheurov and E. Tolstykh).*

*Subtask 8. Verify radionuclide-ingestion levels derived from the results of river modeling (Subtask 3) and include into TRDS Module 1 the ingestion levels of other radionuclides from the releases (Zr, Nb, Ru, Ce, etc.).*

*Subtask 9. Include into TRDS Module 2 appropriate metabolic and dosimetric model calculations (based on literature data) for other radionuclides according to the results of Subtask 8.*

Work on this task was completed with the submission of the Milestone 3 Final Report (Tolstykh et al. 1998), which is included as an Attachment to this report.



## **Progress on Task 5:**

### *Task 5. Uncertainty analysis (M. Degteva, L. Anspaugh, and B. Napier)*

*Subtask 10. Enter into computer "interview data" (source of drinking water, the distance of the house from the edge of water etc.) for the residents of two Techa Riverside villages: Metlino and Muslyumovo.*

*Subtask 11. Develop an algorithm of classification (grouping) of the persons belonging to "families" (households) according to the source of drinking water and/or the distance from the edge of water based on individual-strontium measurements, family-member lists (available in data base MAN) and interview data (Subtask 10).*

*Subtask 12. Perform the grouping of persons according to the algorithm from Subtask 11 for the residents of Metlino and Muslyumovo villages. Evaluate "inside group" and "between groups" contributions into variation for internal dose (based on whole-body counter measurements). Evaluate the feasibility of estimating the contributions to external dose uncertainties from the distance of the house from the river and individual variability in behavioral regimes*

*Subtask 13. Evaluate the contributions to internal dose uncertainties from the source of drinking water (river or wells) and individual variability in diet habits and metabolic parameters on the basis of Metlino and Muslyumovo data.*

*Subtask 14. Evaluate the feasibility of the reconstruction of household specific doses for the entire Techa River Cohort on the basis of the results of Subtasks 12 and 13. Develop a plan for further study on dose reconstruction.*

Accurate quantification of the dose-response function for populations exposed to ionizing radiation requires appropriate treatment of the uncertainty in the dosimetry and the uncertainty in measurements of the response. Uncertainty in the determination of either the dose or biological effect (cancer mortality, for example) obviously can influence the derivation of the dose-response function for an exposed cohort. Until recently, uncertainty in dosimetry has been given limited attention in risk analysis. A dose estimate for any specified individual may be affected by two types of errors: random and systematic. Random errors are those that vary independently among exposed individuals. Systematic errors are those that cause an uncertain bias to an entire subgroup of an exposed cohort. The examination of sources and magnitudes of uncertainty in radiation doses to individuals in an exposed cohort is an important task of any dosimetric study.

Work on Subtask 10 was completed and reported upon in the March 1998 Progress Report (Degteva et al. 1998).

Work on the remaining tasks has begun, and it has been agreed that the American investigators will make this a major focus of their contributions to Project 1.1. During the April 1998 visit the computer system that is the heart of the Techa River Dosimetry System was examined thoroughly with the view of how best to incorporate uncertainty. This work will be



extensive and is not scheduled for completion until the delivery of Milestone 11 in February 2000.

**Progress on Task 6:**

*Task 6. Whole-body-counter calibration and modification (V. Kozheurov and A. Kovtun).*

*Subtask 15. Manufacture an anthropomorphic physical phantom of the body of an adult with an uniform distribution of  $^{90}\text{Sr}$  in the skeleton. This phantom will be used for the recalibration of whole-body counter SICH-9.1. The design of this phantom was described in the Final Report of JCCRER Project 1.1 (Degteva et al. 1996a).*

*Subtask 16. Prepare the protocol and perform the calibration of whole-body counter SICH-9.1 for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  using special phantoms. Evaluate instrumental errors for SICH-9.1 in order to provide better verification of the thousands of whole body counts available at the URCRM.*

*Subtask 17. Develop a mathematical phantom to simulate the spectral and angular distribution of photon radiation, including bremsstrahlung, at the surface of the phantom resulting from the radioactive decay of incorporated radionuclides in order to study the effects of non-uniform distribution of  $^{90}\text{Sr}$  in the different bones and structures of the skeleton and to study the effects of variations in individual-body geometry.*

*Subtask 18. Develop the design for modification of SICH-9.1 and perform the upgrade of SICH-9.1 in order to replace obsolete detectors and electronics and provide continuity of whole body measuring.*

**Subtasks 15 and 16.** Work on these two subtasks has been completed and is documented in the Milestone 2 Final Report (Kozheurov et al. 1998), which is included as an Attachment to this Progress Report.

**Subtask 17.** Now that subtasks 15 and 16 have been finished, more attention will be devoted to Subtask 17. It is our intent to transfer an existing mathematical phantom from the US to the URCRM. This will be accomplished during a visit of Dr. Kozheurov to the US within the next several months.

**Subtask 18.** Work on upgrading WBC SICH-9.1 consists of two parts:

- To specify and purchase a new set of detectors and electronic system for spectrum analysis; and
- To replace computer controlling scanning bed system (mechanical and electronic parts).

The University of Utah sent out a request for bid package in early 1998. One bid was received from "Pribory Oy," an official distributor of EG&G ORTECH. However, this bid included a Value Added Tax of 20%, which put the bid over the amount of money available for



the purchase. As agreements between the governments of the US and the Russian Federation were interpreted as exempting the payment of such tax, requests were made to EG&G ORTECH to secure a letter of exemption from this tax. This letter of exemption was not received, and the existing bid expired.

The University of Utah has gone out for bid again. We anticipate that a response will be received this time directly from EG&G ORTECH.

Other work to upgrade the WBC, called "computer controlled scanning bed system," was reported on in the March 1998 Progress Report (Degteva et al. 1998). This work has been completed.

**Progress on Task 7:**

*Task 7. Electron paramagnetic resonance (EPR) measurements (A. Romanyukha).*

*Subtask 19. Perform about 90 measurements of teeth for the residents of the middle Techa in order to evaluate age dependence of EPR signal for exposed people and compare individual external doses based on these measurements with the results of calculations (Subtask 7).*

*Subtask 20. Perform about 90 measurements of teeth for the residents of non-contaminated areas of the Ural Region in order to evaluate age dependence and the nature of background EPR signal.*

Progress on this Task was reported on extensively in the March 1998 Progress Report (Degteva et al. 1998) and at the Scientific Review Groups' meeting in April 1998 in Chelyabinsk. In July 1998 work on EPR dosimetry at the University of Utah was cancelled by the DOE (Appendix 3), and support was also withdrawn for work to be done in Russia.

The Final Report (Haskell 1998) on EPR dosimetry that includes work performed by the Russian investigator is included as an Attachment to this Progress Report.

**Progress on Task 8:**

*Task 8. Luminescence measurements (N. Bougrov).*

*Subtask 21. Perform TL measurements for about 7-10 sites in Muslyumovo Settlement in order to evaluate the distribution of radiation fields and to compare external doses based on these measurements with the results of calculations (Subtask 4).*

Progress on this Task was reported on extensively in the March 1998 Progress Report (Degteva et al. 1998). In July 1998 work on thermoluminescence dosimetry at the University of Utah was cancelled by the DOE (Appendix 3), and support was also withdrawn for work to be done in Russia.



The status of work that had been performed on this Task is summarized in Appendix 4, which is a pre-print of a manuscript (Bougrov et al. 1998) that has been accepted for publication in *Health Physics*.

### ACKNOWLEDGEMENTS

This work has been funded by the US Department of Energy's Office of International Health Studies, the US Environmental Protection Agency, the US National Aeronautics and Space Administration, and the Federal Department of the Ministry of Health of the Russian Federation.

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## APPENDIX 1

### Pre-Print

**Vorobiova, M. I.; Degteva, M. O.; Burmistrov, D. S.; Safronova, N. G.; Kozheurov, V. P.; Anspaugh, L. R.; Napier, B. A. "Review of historical monitoring data on Techa River contamination." *Health Phys.* (submitted, 1998).**



# REVIEW OF HISTORICAL MONITORING DATA ON TECHA RIVER CONTAMINATION

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**Abstract**—The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. The extensive increase in plutonium production during 1948–1955, as well as the absence of reliable waste management technology, resulted in significant releases of liquid radioactive waste into the rather small Techa River. This resulted in chronic external and internal exposure of about 20,000 residents of riverside communities; these residents form the cohort of a major epidemiologic investigation. This paper presents main historical data on Techa River radioactive contamination: The operating history of the Mayak complex and available information on source-term and environmental monitoring data. Analysis of the available historical monitoring data indicates that the following reliable data sets can be used for reconstruction of doses received during the early periods of operation of the MPA: Temporal pattern of specific beta activity of river water for several sites in the upper Techa region since July 1951; average annual values of specific beta activity of river water and bottom sediments as a function of downstream distance for the whole river since 1951; gamma-exposure rates near the shoreline as a function of downstream distance for the whole Techa River since 1952; and gamma-exposure rate as a function of distance from the shoreline for several sites in the upper and middle Techa since 1951.

## INTRODUCTION

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. This plant began operation in 1948, and during its early days there were technological failures that resulted in the releases of large amounts of radioactive waste into the rather small Techa River. The residents of the riverside communities were exposed to chronic external and internal irradiation. Extensive monitoring efforts for the environment and the population at this site began in 1951. The "Techa River Cohort" (TRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM), and an increase in both leukemia and solid tumors with plutonium exposure has been noted (Kossenko et al. 1997). This finding suggests that, with continuing improvements in the quality of the follow-up and dosimetry, study of the TRC has the potential to provide quantitative estimates of the risks of stochastic health effects produced by chronic low-dose-rate radiation exposure for the general population. Study of this population affords an unique opportunity to address the question of the existence of a dose-rate-reduction factor for the induction of stochastic effects in an unselected general population. A definitive answer to this question would have relevance to the regulation of radiation exposure throughout the world.

The current dose-reconstruction system (known as the Techa River Dosimetry System or TRDS) for the TRC, which numbers about 30,000 people, is grounded firmly on whole-body counts for half of the members of the cohort (for the evaluation of internal dose, which was mainly due to incorporated  $^{90}\text{Sr}$ ) and on direct measurements of external gamma-exposure rates (Kozheurov 1994; Degteva et al. 1998). A joint Russian-American project has been now been implemented under the auspices of the US-Russian Joint Coordinating Committee on Radiation Effects Research (JCCRER). This project is concerned with a comprehensive program to

develop improvements in the existing dosimetry system for the TRC members by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results.

A set of conceptual models that defines the relationships, pathways, and parameters that form the basis of the dose-reconstruction efforts has been described (Degteva et al. 1996). The hierarchy of information required for calculating doses to people who lived along the Techa River also has been described. One of the important tasks of the project is the analysis of available historical data on source terms of the releases and on environmental contamination.

The purposes of this paper are the following:

- To describe historical data regarding the operating history of the MPA and available measurements of releases;
- To present accumulated historical Techa River data on hydrology, sediment loading, and dam-construction history;
- To compile and evaluate available data on radionuclide concentrations in river water and sediments and gamma-exposure rates on the banks.

#### **THE MAYAK COMPLEX AS A SOURCE OF TECHA RIVER CONTAMINATION**

The MPA was created in 1948 in the Southern Urals for the production of plutonium for nuclear weapons (Fetisov 1996). This complex consisted of a series of uranium-graphite reactors operating with thermal neutrons and using direct flow water-cooling loops (Complex A); a radiochemical plant for the extraction of  $^{239}\text{Pu}$  from uranium irradiated in the reactors (Complex B); a chemical-metallurgical plant for metallic plutonium production and machining (Complex V); and radioactive waste-management and storage facilities (Complex C). Over the

intervening years six reactors operated at the MPA for the production of weapons-grade plutonium. Of these, five were graphite moderated while the sixth was originally heavy-water moderated (Novosselov et al. 1995). The graphite-moderated reactors have now been shut down (Fetisov 1996); the heavy-water reactor was later modified to a light-water reactor and remains in operation today. A seventh reactor is operational for the production of isotopes for civilian uses. The operating histories of the plutonium-production reactors from 1949 through 1967 are shown in the Appendix. Since 1977, the radiochemical plant has been used extensively to reprocess fuels from power, transport, and research reactors.

The extensive increase in plutonium production during 1955, as well as the absence of reliable waste-management technology, resulted in significant releases of liquid radioactive waste into the Techa River. The chronology of major events connected with the evolution of the exposure situation on the Techa River is shown in the Appendix. These data were extracted from several sources (Marey et al. 1953, 1954, 1956, 1965; Ilyin 1956; Alekseeva et al. 1957; Borovinskikh et al. 1958; Anikin et al. 1959; Marey 1959; Makhonko 1994; Fetisov 1996; Novosselov et al. 1997).

In 1949, high-level wastes were routed to the tank farms in Complex C, and low-level wastes only were released to the Techa River (Fig. 1). However, in 1950 in order to reduce the volume of material going to the tanks in Complex C, a process for "decontamination" of high-level wastes was introduced, with a portion of the radioactivity directed to the tanks and a portion released to the river. In July 1951, it was discovered that this process did not work as intended, and that during this period high concentrations of radionuclides had been released into the river. Also during this time, cooling water from the Complex C tanks was discharged into the Techa River at the same location as the technological wastes. Leaks in the tank-cooling lines caused

some of these discharges to be highly contaminated. These "wild releases" were unmonitored and unnoticed until 1951. Over this period, about half of the total release to the Techa River resulted from the technological releases and about half from the wild releases.

In late 1951, several activities were started to control the releases and to remediate the environmental contamination. The main technological releases were diverted into Karachay Lake. Large amounts of water were released into the Techa River from Lake Kyzyl-Tash in an effort to wash the contamination out of the area of Metlinsky Pond and reduce the exposure rates. Over the next several years, a series of dams were built in the upper Techa with bypass canals to halt the spread of contamination (Figs. 2a, b, c, and d). The construction of Reservoir 11 and the creation of the by-pass canals provided isolation of the most contaminated part of the river-bed from the lower parts of the Techa River where the population continued to live. Between 1952 and 1956, residents of all villages within 75 km of the site of the release were resettled in uncontaminated areas. Some of the remaining villages along the Techa River were also resettled between 1956 and 1960. About 7500 people in total were resettled.

It must be noted that in addition to radiochemical wastes that were the major source of the Techa River contamination, radionuclides entered the river with the water of Kyzyl-Tash Lake used for reactor cooling. Also, some surface contamination of the upper-Techa region occurred as a result of the Kyshtym accident in 1957 and wind transfer of contaminated silt of Karachay Lake in 1967 (Figs. 2c and d). And finally, in the period of January–April 1958, Berdyanish Lake waters, which were contaminated as a result of the plume from the Kyshtym accident, were artificially discharged to the Techa River (Fig. 2c). Since 1967, the Techa River system has been in a natural self-cleaning regime.

## HYDROLOGICAL CHARACTERISTICS OF THE TECHA RIVER

The Techa River (the right tributary of the River Iset) belongs to the basin of the Kara Sea. On the basis of the hydrological characteristics of the river (240 km in length, up to 2 m in depth, mean annual flow-rate in the outfall of about  $7 \text{ m}^3 \text{ s}^{-1}$ ), it can be assigned to the category of small rivers. Its main tributaries are the Mishelyak, Zyuzelka, Baskazyk, Borovaya and Shutishka Rivers (Fig. 1).

The Techa riverside area can be divided into two parts depending on its flood-land and bed characteristics:

- In the upper reaches of the Techa, a cascade of hydraulic engineering constructions are located (Fig. 2), including Reservoirs No. 3 (Koksharov Pond was created at the location of an old weir in August 1951), No. 4 (Matlinky Pond, which already existed in 1949), No. 10 (created in 1956) and No. 11 (created in 1964). The stretches of the river from Reservoir No. 11 up to the village of Muslyumovo are for the most part swampy, with a poorly marked winding bed overgrown with water plants. The width of the river bed is from 3 to 15 m and its depth is from 0.5 to 2 m; bed deposits consist of turf-silt or clay;
- In its middle and lower reaches (downstream of the village of Muslyumovo), the river has a well marked bed, its bottom consisting of layers of sand and slime, in some places clay, sand and gravel. The mean width and depth of the river during the summer time are 22 and 0.5–1 m, respectively.

As the Techa belongs to the type of plain rivers, it has few turnings: The mean value of winding coefficient is about 1.07. The longitudinal profile of the river bed is characterized by the slope of the average line of the bottom and the slope of the water surface along the river

course; the midstream and downstream reaches of the river have a slope virtually equal to that of the water surface, i.e., about  $0.6 \text{ m km}^{-1}$ .

The Techa River receives its supply of water from melting snow and intensive spring floods (Fig. 3). The main source of water supply to the Techa during the summer months is groundwater discharge from water-bearing horizons formed by atmospheric precipitation (Koloskov 1968). During the period of floods, a backwater phenomenon develops due to the river-water effluent that minimizes the amount of groundwater entering the river. On the average, the groundwater penetrating into the river makes up about 10% of the overall river runoff.

A comparison between the mean annual flow rate in the river and the annual level of precipitation in the area under observation (Fig. 4) has shown that the curve representing the flow rate is actually similar in shape to that referring to precipitation, with a year's delay in dynamics. The installation of the upstream reservoirs in the mid-1950s changed the flow patterns markedly, as seen in Fig. 4. The flow rate has ranged from 2 to  $10 \text{ m}^3 \text{ s}^{-1}$  in recent years. The minimum estimates of flow rate ( $<1 \text{ m}^3 \text{ s}^{-1}$ ) were noted in the drought-afflicted years 1975–1976. In terms of chemical composition the water of the Techa River is classified among the carbonate-sodium type;  $\text{pH} = 7.5\text{--}8.5$ ; mineralization is estimated to be about  $700 \text{ mg L}^{-1}$  at present. Ion concentration in the river water does not change significantly with time and averages about  $60 \text{ mg L}^{-1}$  for  $\text{Ca}^{++}$ ,  $30 \text{ mg L}^{-1}$  for  $\text{Mg}^{++}$ ,  $300 \text{ mg L}^{-1}$  for  $\text{HCO}^-$ , and  $40 \text{ mg L}^{-1}$  for  $\text{Cl}^-$ . The most clear-cut changes after the installation of the last dam (No. 11) were observed in the river-water mineralization level and the content of sulfates in it (Komissarova 1985).

The bed deposits in the upper reaches of the river between Reservoir No. 11 and Muslyumovo consist of layers of turf, silt and clay. There are flood swamps measuring 300 m to

2 km in width along the river shoreline; the most swampy areas are located between the villages of Nadyrov Most and Muslyumovo at the site where the Zyuzelka River enters the Techa. The central portion of the flood soils is composed of turf-bog soils that give way to meadow-turf ones along the boundaries of the swamps. The thickness of the turf layer ranges from 10 cm to 3 m, the turf contains a considerable amount of mineral inclusions and increased percentage of ash (10–35%, up to 60% in the near-to-bottom layers). Clay and sandy loam, less frequently sand, compose the underlying layer of turf.

Bed deposits in the middle and lower reaches of the river are of sandy-silt and sandy-gravel type; the dry flood land measures 200–500 m in width and is composed of turfy-meadow soils. Studies of the mechanical composition of the soils have shown that the sandstone bed deposits and sandy loam soils of the flood meadows are characterized by a higher content of large (0.25–1 mm) particles while the fine fraction (<0.01 mm) is uniformly distributed over the vertical profile with its content determined by the type of soils (Safronova et al. 1986).

#### **AVAILABLE INFORMATION ON DISCHARGE OF RADIOACTIVE MATERIALS INTO THE TECHA RIVER**

Information on radioactive releases into the Techa River was analyzed and presented in URCRM Technical Reports (Kozheurov 1985; Shvedov et al. 1990). The primary data on the releases for these reports were extracted from MPA Technical Reports, 1951–53, and the doctoral thesis of D. I. Ilyin (1956) (these documents are kept in MPA archives and are still classified). The extracted information contains data on release dynamics and isotopic composition of wastes. The other sources of information were Technical Reports of the Biophysics Institute (Marey et al. 1953, 1954, 1956, 1965; Alekseeva et al. 1957; Borovinskikh et al. 1958; Anikin et al. 1959), and the doctoral thesis of A. Marey (1959).

Radioactive wastes were released into the Techa River beginning in March 1949. Initially, high-level wastes were concentrated and kept in specially equipped containers (the tanks of Complex C) and only low-level wastes (after passing through special absorbers) were released into the river. In January 1950, a special facility for the decontamination of high-level liquid radioactive waste was put into operation and the construction of additional Complex C tanks was stopped. Later it was found that this technology of radioactive waste management was poor and could not give the necessary level of decontamination. As a result of this failure, release rates into the Techa sharply increased. And, in addition to pre-planned releases, some unexpected releases (so-called "wild overflows") were discharged episodically into the river with the cooling waters of Complex C tanks, and the average daily release between March 1950 and November 1951 reached  $1.6 \times 10^{14}$  Bq (4,300 Ci). Starting 28 October 1951, the release of all technological wastes from process operations was routed to Karachay Lake and only low-level wastes (from laundry, laboratory, and septic systems) continued to enter the Techa River.

All information on the releases may be divided into three parts:

- Mean-in-period activity and isotopic composition of releases (main part). The data contain average values of alpha-, beta- and gamma-emitting activities released and mean percentage of some radionuclides for the time periods March–December 1949, January–February 1950, March 1950–October 1951, and November 1951–December 1952. It is believed that these data were reconstructed by Dr. D. Ilyin (the Head of the Mayak Central Laboratory) based on knowledge of technological processes and some measurements that were made in 1951–56. The methods of reconstruction were not described in the available documents.

- Mean-in-year activity and isotopic composition of releases in 1953–56. The data contain average values of beta activities released and mean percentage of main radionuclides and/or groups of radionuclides in 1953–56. These data were estimated by Dr. A. Marey on the basis of Moscow Biophysics Institute research expeditions (1952–1959).
- Primary data on activity released in various time periods from 1951 to 1956 (including the information about “wild overflows”). This information contains results of measurements of daily volume and specific beta activity of releases and/or daily activity released, radionuclide composition and percentage of activities of the solids in some days (periods) of the release time. The character of primary data on the release dynamics is presented in Table 1.

Average release rates evaluated by the Mavak experts under supervision of Dr. D. Ilyin are presented in Fig. 5. The total amount of fission products released into the Techa River was about  $10^{17}$  Bq of beta emitters (about 98% of this value was released during 1950–1951). The ratio of gamma-equivalent to beta-activity was about 0.24 gram-equivalent of radium\* per curie (a reporting scheme in use at the time relative to a radium standard). This corresponds to an average holdup time (the time of cooling of irradiated uranium blocks) of about 150 days. The total amount of alpha emitters discharged to the river with wastes was evaluated as less than  $1.85 \times 10^{12}$  Bq (50 Ci). Available data of daily measurements permitted evaluation of the unevenness of releases in the initial period of monitoring (Fig. 6) and showed also irregularities in rate after the main technological releases were routed to Karachay Lake (Fig. 7).

It was found in 1951 that the releases consisted of suspensions ( $\text{pH} = 7\text{--}8$ ) with  $3\text{--}5 \text{ g L}^{-1}$  of sodium nitrate and sodium acetate. Suspended particles mainly consisted of iron hydroxide

and organic matter. On average, about 70% of beta activity entered the Techa River with the suspended particles and the remaining 30% were in soluble form.

Radionuclide composition of the releases from the radiochemical plant is presented in Table 2. The values before 1952 should be considered as evaluated theoretically, because the first measurements of total activity were started in July 1951, and radiochemical techniques for some radionuclides were not developed until later (in 1953 for Zr + Nb, and in 1956 for Ru).

There was only one experimental analysis of radionuclide composition (on 24–25 September 1951), and this should not be interpreted as representative for the whole period of acute exposure. The results of this analysis, presented in Table 3, show that almost all cesium, 75% of strontium, and about 50% of zirconium and niobium were in soluble form, but about 98% of rare earth elements entered the river absorbed on suspended particles. The measured radionuclide composition is different from theoretical values. The ratio of gamma-equivalent to beta-activity for the composition calculated using a holdup time of 150 days gives the value of about 0.15 gram-equivalent of radium per curie. The measurements of beta- and gamma-activity for the release on 25 September 1951 gave the ratio equal to 0.48; that is three times more than the calculated value. Such a discrepancy suggests the existence of some systematic errors in the technique for measuring gamma activity. But, in spite of such discrepancies, it is possible to conclude that releases of high-level wastes consisted of a mixture of materials from various stages of processing and with varying times of cooling. Further, it seems obvious that the radionuclide composition and physico-chemical character of the released material fluctuated widely.

In 1952–1955, an additional source of contamination became significant against the background of the decrease of releases from the radiochemical plant. This source was reactor

cooling water released into Kyzyl-Tash Lake and which subsequently flowed into the Techa River (Fig. 2). For example, during seven months in 1953, the activity released from the reactor plant was five times the release from the radiochemical plant (Fig. 8). The water of Kyzyl-Tash Lake entering the Techa was also contaminated by such activation radionuclides as  $^{32}\text{P}$ ,  $^{35}\text{S}$ ,  $^{45}\text{Ca}$  and had a specific activity of about 7–30 kBq L<sup>-1</sup> (according to measurements of 1953). For comparison, the concentration of activation products in the Columbia River downstream of the analogous Hanford plutonium-production reactors during peak operations in the early 1960s reached an annual average of 2 kBq L<sup>-1</sup> ( $5 \times 10^{-8}$  Ci L<sup>-1</sup>) and even exceeded 10 kBq L<sup>-1</sup> ( $3 \times 10^{-7}$  Ci L<sup>-1</sup>) during low water flows (Wilson 1964; Napier 1993). The primary activation products at Hanford included  $^{32}\text{P}$ ,  $^{51}\text{Cr}$ ,  $^{56}\text{Mn}$ , and  $^{65}\text{Zn}$ . Radiation doses to downstream individuals from these radionuclides in the Columbia River averaged only a few tens of  $\mu\text{Sv y}^{-1}$  (Napier 1993).

#### AVAILABLE ENVIRONMENTAL MONITORING DATA

Systematic measurements of radioactive contamination in and near the Techa River started at the same time as release control (July 1951). The contamination of the river water, bottom sediments, flood plain soils, vegetation, fish, milk, and other food stuffs, and external gamma-exposure rates were measured. Several settlements and specific sites in the upper reaches were selected as points for routine sampling. These points with historical numeration are shown in Fig. 1 (numbers 21–48) and in Fig. 9 (numbers 1–20). The most common technique was measurement of beta activity with an end-window counter calibrated with an uranium-oxide standard. The exposure-rates were measured with a “MAK” device, which had an ionization chamber with aluminum walls as the sensing element. Descriptions of these techniques have been published (Gussev et al. 1959).

In the 1990s, historical data of importance for dose-reconstruction purposes were arranged in the special computer database ENVIRONMENT (Degteva et al. 1996). This database now includes more than 10,000 records of measurements of radioactive materials in the environment for the period 1951–1990: Specific alpha, beta, and gamma activity; concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in river water, concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in bottom sediments and flood-plain soils; and external exposure-rate measurements. The most representative data set was on specific beta activity of the Techa River water. Figure 10 presents the time patterns of specific beta activity of river water at four sites in the upper reaches of the river. Significant fluctuations in concentration observed near the site of release became less pronounced after the water passed through Koksharov and Metlinsky Ponds. These ponds played a very important role: They served as settling reservoirs and dampers for release peaks. As seen from Fig. 10, radionuclide concentration in water decreased one order of magnitude from 1952 to 1956.

The measurements of gamma- and alpha-emitter concentrations in the river water were limited in comparison with measurements of beta activity. The concentration of alpha emitters in the water of the ponds in 1952–1954 was about  $4 \text{ Bq L}^{-1}$  and decreased to  $0.4 \text{ Bq L}^{-1}$  in river water downstream of the ponds. The results of measurements of gamma activity in the river water had large fluctuations and were not correlated with beta-activity (as noted above, some errors in the technique for measuring gamma activity could have existed in the early 1950s). There were also limited radiochemical analyses of the river water in 1951–1956, but these results were incomplete and sometimes discrepant. It was impossible to make any conclusions about radionuclide composition of river water on the basis of these sparse and contradictory data.

Fig. 11 (a, b, c) illustrates the sets of data on specific beta activity of water and bottom sediments as a function of downstream distance. Such data sets were available for 1951–1955,

and in all cases the slope of concentration versus distance for the water was significantly lower than the slope for the bottom sediments. This means that the waterlogged bed deposit in the upper reaches of the Techa actively accumulated some of the radionuclides from the releases. Also, it is clearly apparent that the difference between contamination levels of water and sediments increases with time within the period 1951–1953: The sharp decrease of water contamination was not accompanied by a similar decrease of the contamination in the sediment, which had become a repository for long-lived radionuclides.

The first measurements of external gamma-exposure rates were performed in 1951 at several specific sites on the upper reaches of the Techa River. Starting in 1952, such measurements were performed along the entire Techa River. The results of exposure-rate measurements near the shoreline as a function of downstream distance are presented in Fig. 12. The external gamma-exposure rate did not change significantly from 1952 to 1956; this indicated rather strongly that the responsible radionuclides were long-lived (presumably primarily  $^{137}\text{Cs}$ ). Exposure rates measured as a function of distance from the shoreline at several sites (Fig. 13) suggested that the main source of gamma radiation was the contaminated silt, with no appreciable shielding by the water layer near the bank strip. Fig. 14 shows that there is a correlation between parallel measurements of exposure rate and beta activity of bottom sediments. It is possible to use this empirical dependence for the reconstruction of exposure rates near the shoreline for cases of known beta activity of the bottom sediments.

#### **ASSESSMENT OF AVAILABLE DATA**

A brief description of the exposure situation on the Techa River and the data available on the source term and environmental contamination could be completed with the following

considerations. Several specific periods should be considered in the evolution of the exposure situation on the Techa River (Table 4).

These periods are determined by the character and completeness of data available and by the changes in the configuration of the river system. Important criteria for the determination of such stages are the magnitude of release rates and their significance for later radioactive contamination of the river system and population exposure. It is most important for the purposes of dose reconstruction to know radionuclide concentrations and exposure rates during the "acute period" of the exposure situation. Especially important was the first phase of the exposure situation, when the contact of people with the contamination was unlimited. As seen from Table 4, environmental monitoring data are very limited for this period. The only technique available to solve this problem is development of a model describing radionuclide transport from the site of release along the river and the accumulation of radionuclides by bottom sediments. The period after 1956 was less important for dose reconstruction, because some countermeasures, including the relocation of people from the upper-Techa region, the ban of the use of river water for drinking and other household needs, etc., were introduced. Nevertheless, the results of measurements of radionuclide concentrations in bottom sediments and flood-plain soils could help to validate such a river model, because the contamination of the river system was caused mostly by the massive releases of 1950–1951. If the consistency between the release rates reconstructed by the Mayak experts and the environmental monitoring data could be confirmed using a more or less realistic river model, this also could serve as a test of the validity of the estimates of radionuclide releases.

## CONCLUSIONS AND SUMMARY

The relevant operating history of the MPA is presented. Historical data are described regarding available measurements of releases of radionuclides to the Techa River and concentrations of radionuclides in the Techa River water and sediments. The history of dam construction and watercourse changes is also described, as these alterations have had a significant impact on the movement of radionuclides within the Techa River system.

The releases during the early years (1949–1951) of operation of the MPA were the most significant and account for more than 95% of the total releases. Analysis of the available historical monitoring data indicates that the following reliable data sets can be used for reconstruction of doses received during the early periods of operation of the MPA:

- Temporal pattern of specific beta activity of river water for several sites in the upper Techa region since July 1951;
- Average annual values of specific beta activity of river water and bottom sediments as a function of downstream distance for the whole river since 1951;
- Gamma-exposure rates near the shoreline as a function of downstream distance for the whole Techa River since 1952; and
- Gamma-exposure rate as a function of distance from the shoreline for several sites in the upper and middle Techa since 1951.

Further, analysis of the historical data also demonstrates that environmental modeling must be used to fill in the gaps in monitoring data.

## ACKNOWLEDGMENTS

This work was supported as Project 1.1 of the U.S.–Russian Joint Coordinating Committee on Radiation Effects Research. Financial support during 1997 was provided by the

U.S. Department of Energy, the U.S. Environmental Protection Agency, the U.S. National Aeronautics and Space Administration, and the Federal Department of the Ministry of Health of the Russian Federation. Two of us (M. V. and M. D.) would particularly like to thank A. Suslov, E. Drozhko and E. Ryzhkov (Mayak PA), S. Malyshev (Ministry of Atomic Energy of the Russian Federation), and M. Kiselev (Federal Department of the Ministry of Health of the Russian Federation) for providing an opportunity to work with original reports in archives of the MPA and the Federal Department.

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**FOOTNOTE**

\* “Gram-equivalent of radium” is an old method used to specify the amount of gamma-emitting materials present in a sample. Thus, one “gram equivalent of radium” is that amount of gamma-emitting radionuclide (or mixture of radionuclides) that produces the same amount of ionization as does one gram of radium in equilibrium with its short-lived progeny. One gram-equivalent of radium is approximately equal to  $0.5 \mu\text{Gy m}^2 \text{s}^{-1}$ .

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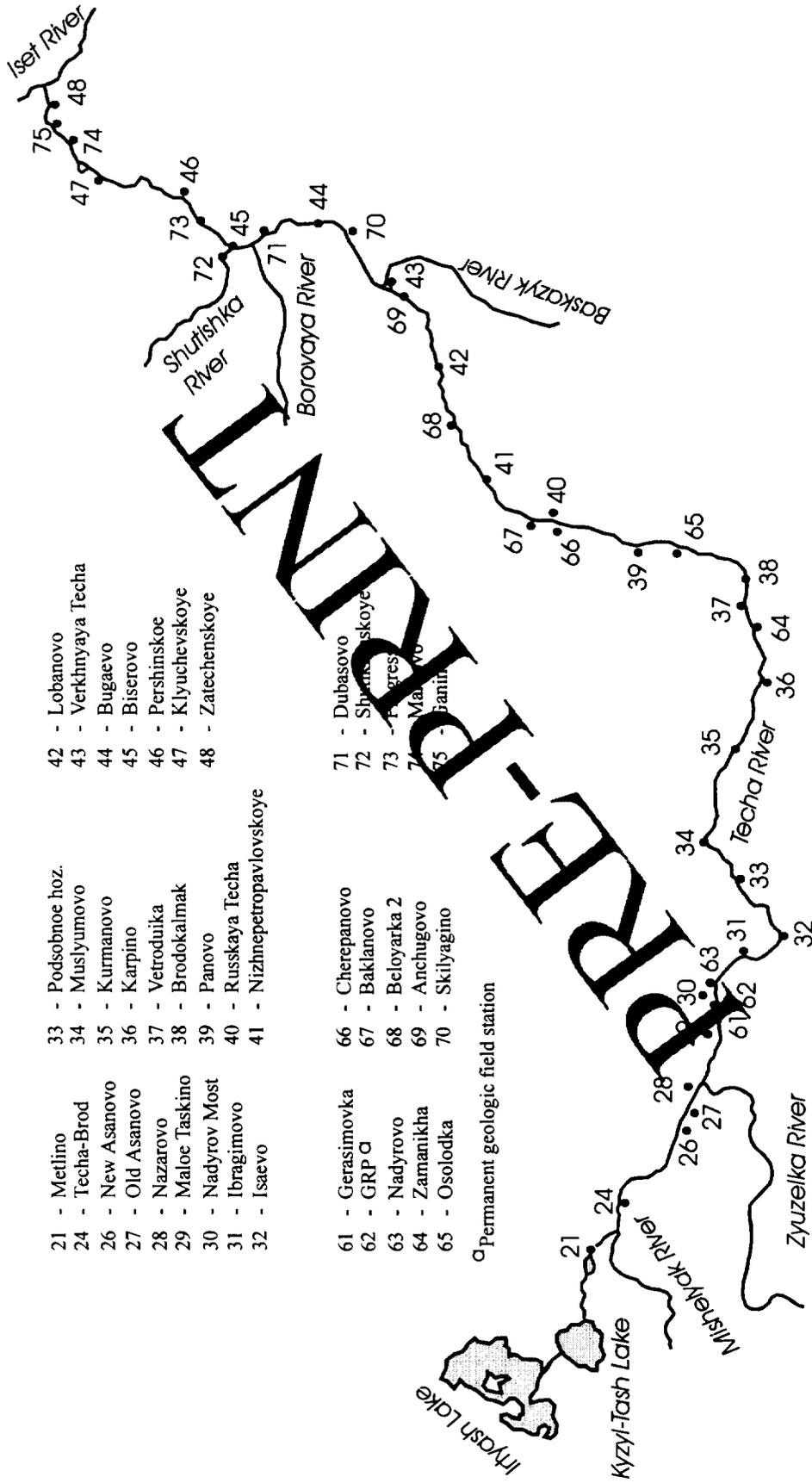


Fig. 1. Schematic map of the Techa River (approximate scale) and of the villages located on its banks before contamination occurred.

Villages No. 21-48 served as the sites of historical routine sampling.

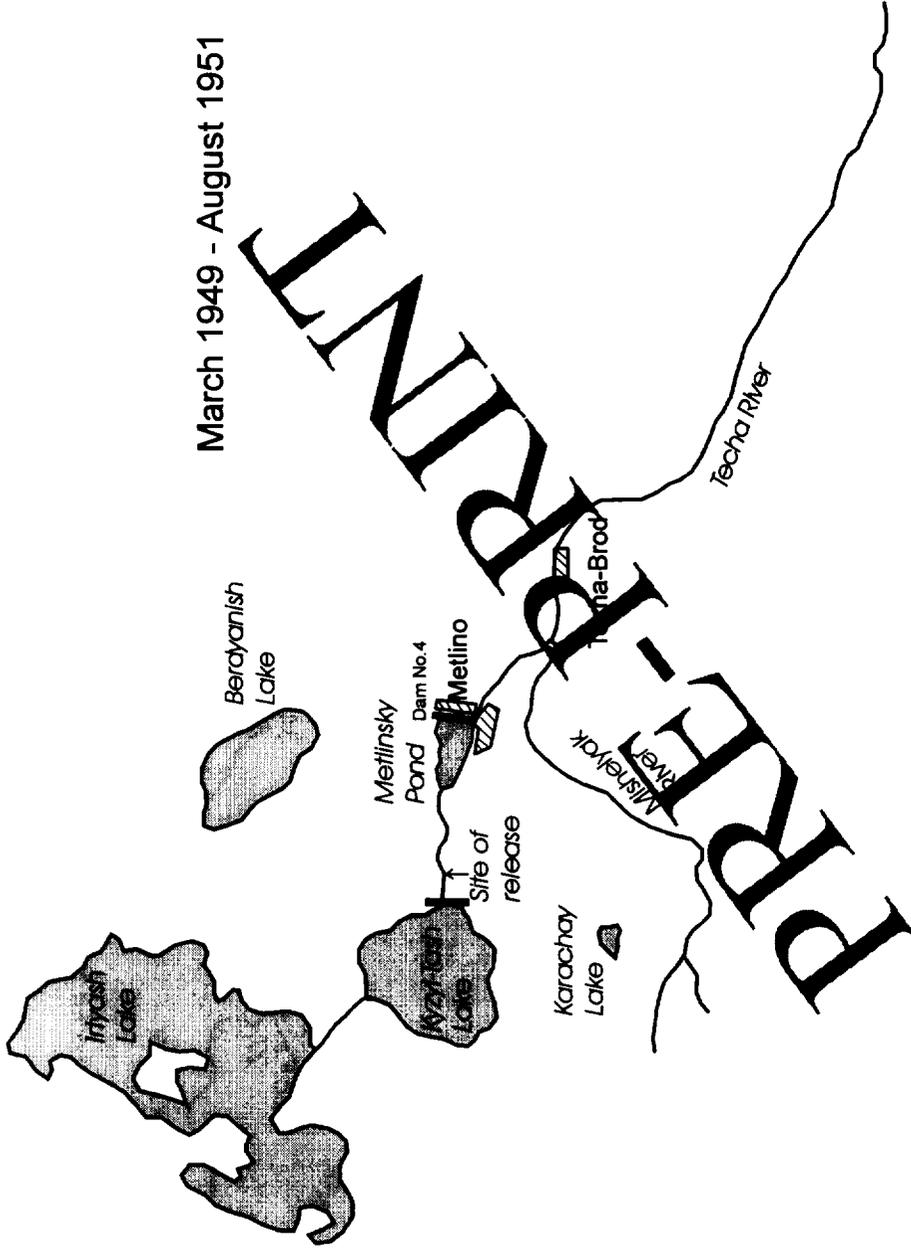


Fig. 2. Schematic maps of the upper reaches of the Techa River (approximate scale): a) before August 1951; b) 1951-1956; c) 1956-1964; and d) after 1964. Heavy bars indicate dams; hatched lines indicate canals; thin arrows indicate liquid releases; heavy arrows indicate atmospheric releases; the hatched areas in c and d indicates the region of  $^{90}\text{Sr}$ -deposition density greater than  $740 \text{ MBq m}^{-2}$  ( $20 \text{ Ci km}^{-2}$ ).

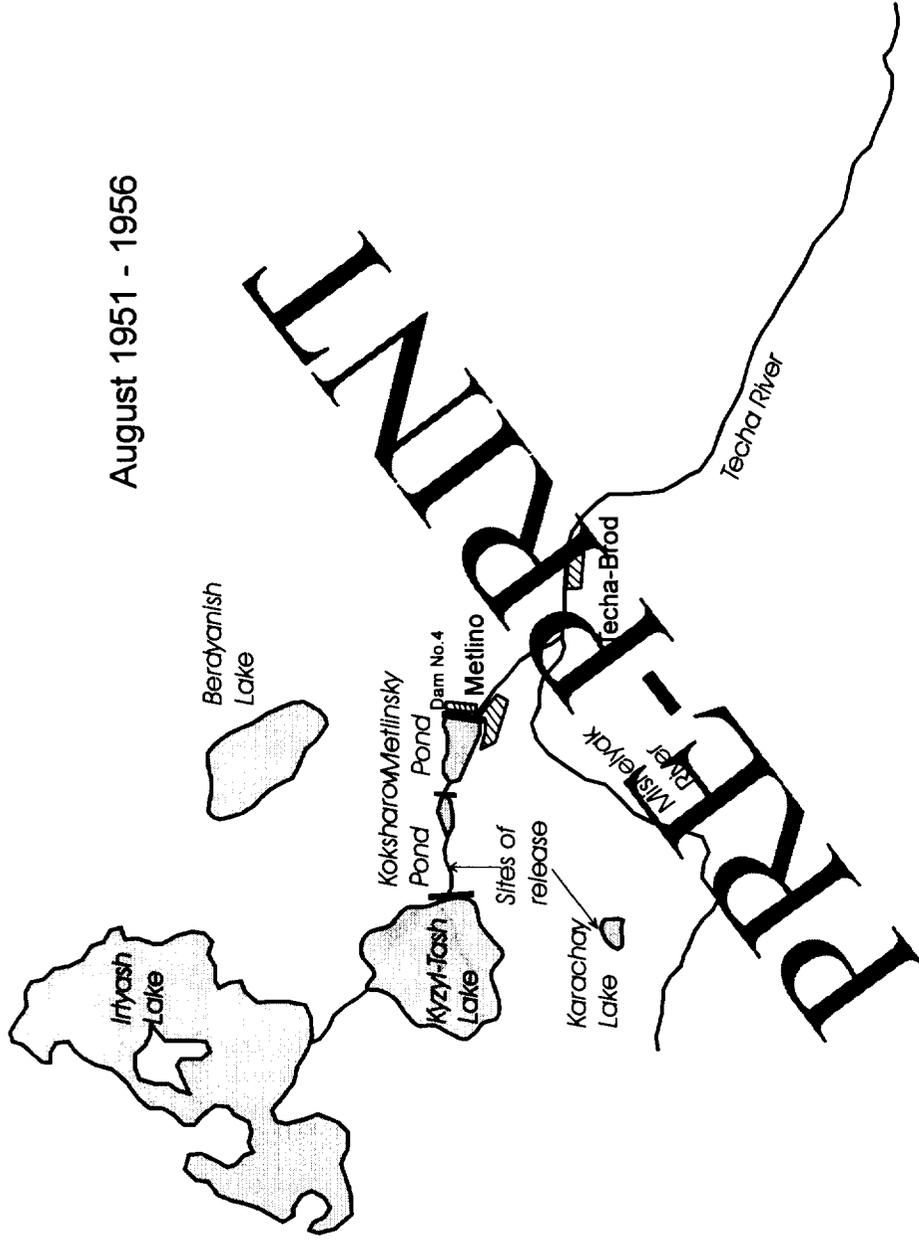


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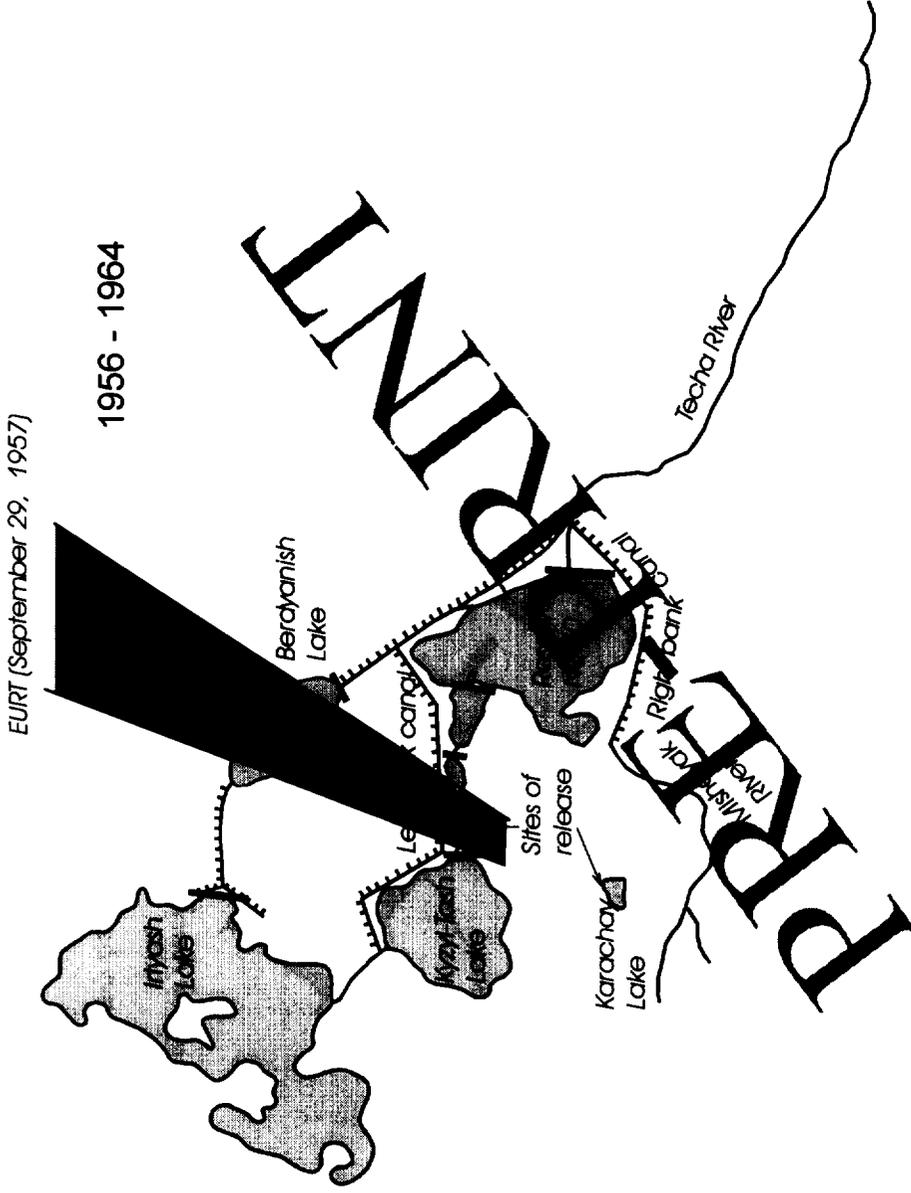


Fig. 2. Schematic maps of the upper reaches of the Techa River (approximate scale): a) before August 1951; b) 1951-1956; c) 1956-1964; and d) after 1964. Heavy bars indicate dams; hatched lines indicate canals; thin arrows indicate liquid releases; heavy arrows indicate atmospheric releases; the hatched areas in c and d indicates the region of  $^{90}\text{Sr}$ -deposition density greater than  $740 \text{ MBq m}^{-2}$  ( $20 \text{ Ci km}^{-2}$ ).

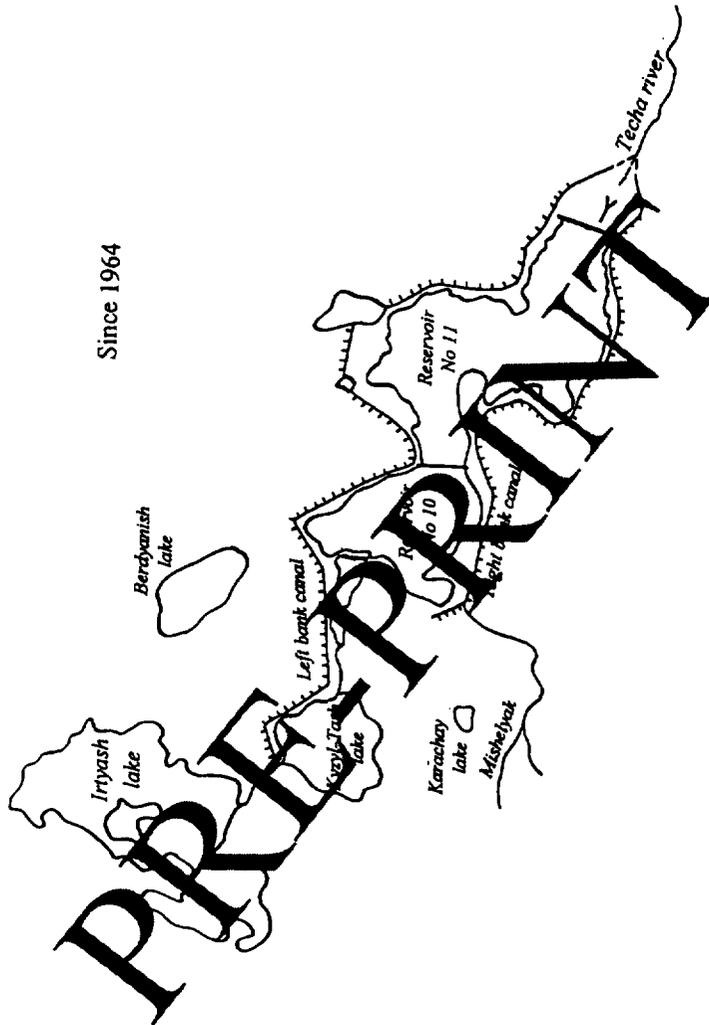
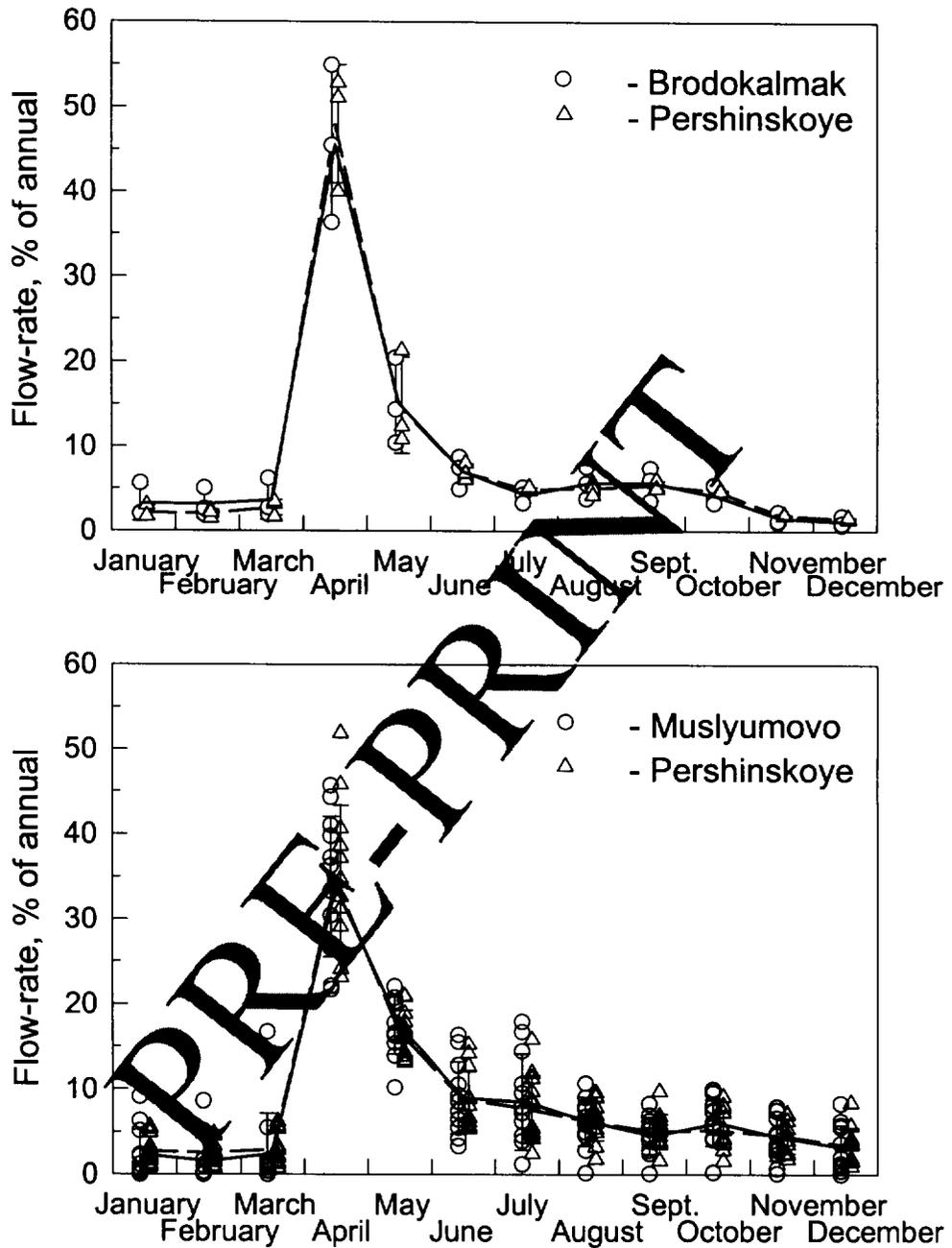
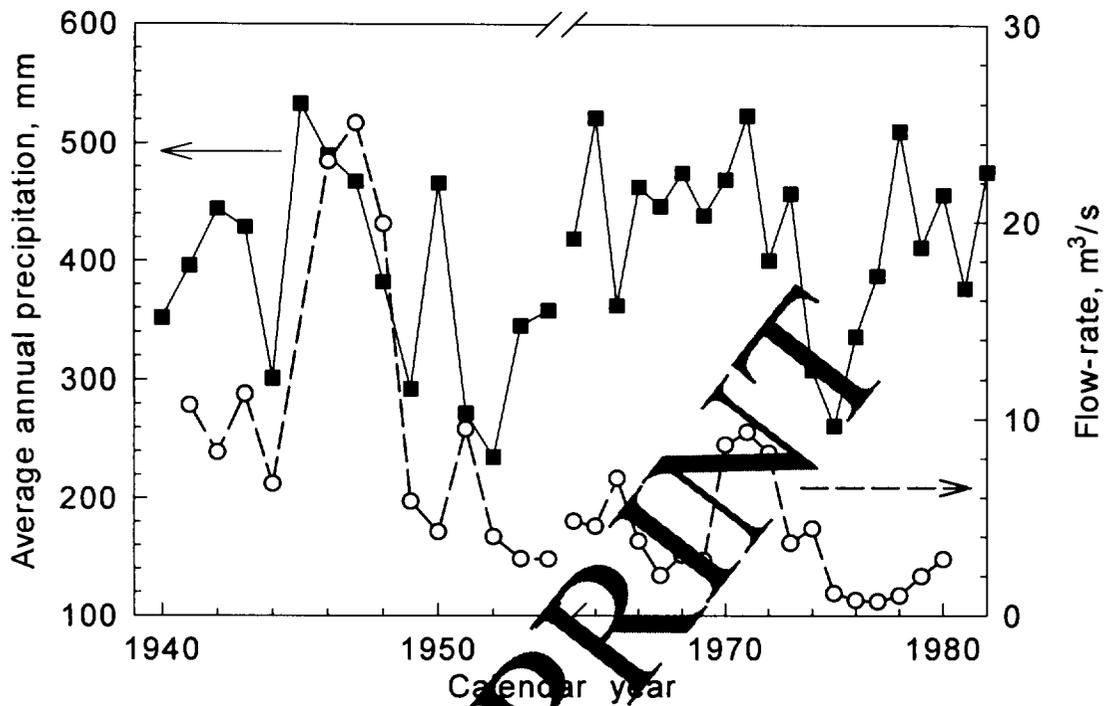


Fig. 2. Schematic maps of the upper reaches of the Techa River (approximate scale): a) before August 1951; b) 1951–1956; c) 1956–1964; and d) after 1964. Heavy bars indicate dams; hatched lines indicate canals; thin arrows indicate liquid releases; heavy arrows indicate atmospheric releases; the hatched areas in c and d indicates the region of  $^{90}\text{Sr}$ -deposition density greater than  $740 \text{ MBq m}^{-2}$  ( $20 \text{ Ci km}^{-2}$ ).



*Fig. 3. The Techa River flow-rates measured in the following ranges: Brodokalmak-Pershinskoye in 1951–1953, and Muslyumovo-Pershinskoye in 1963–1976. Data were extracted from Shakhov (1967); Starostina (1973, 1974, 1975, 1976); Agapitova (1975); and Katomina (1977, 1979).*



*Fig. 4. Mean annual water discharge in the Techa River and annual level of precipitation according to data of Shakhov (1967); Starostina (1973, 1974, 1975, 1976); Agapitova (1975); and Katomina (1977, 1979).*

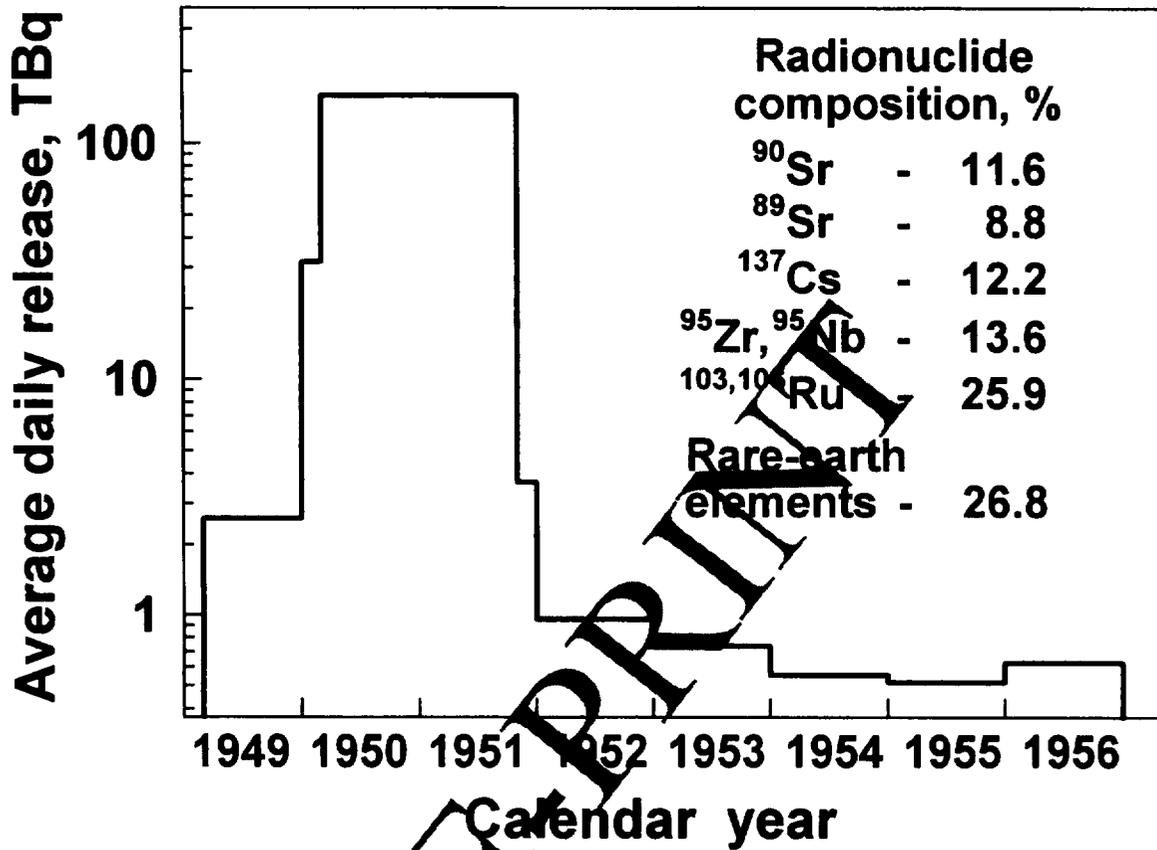
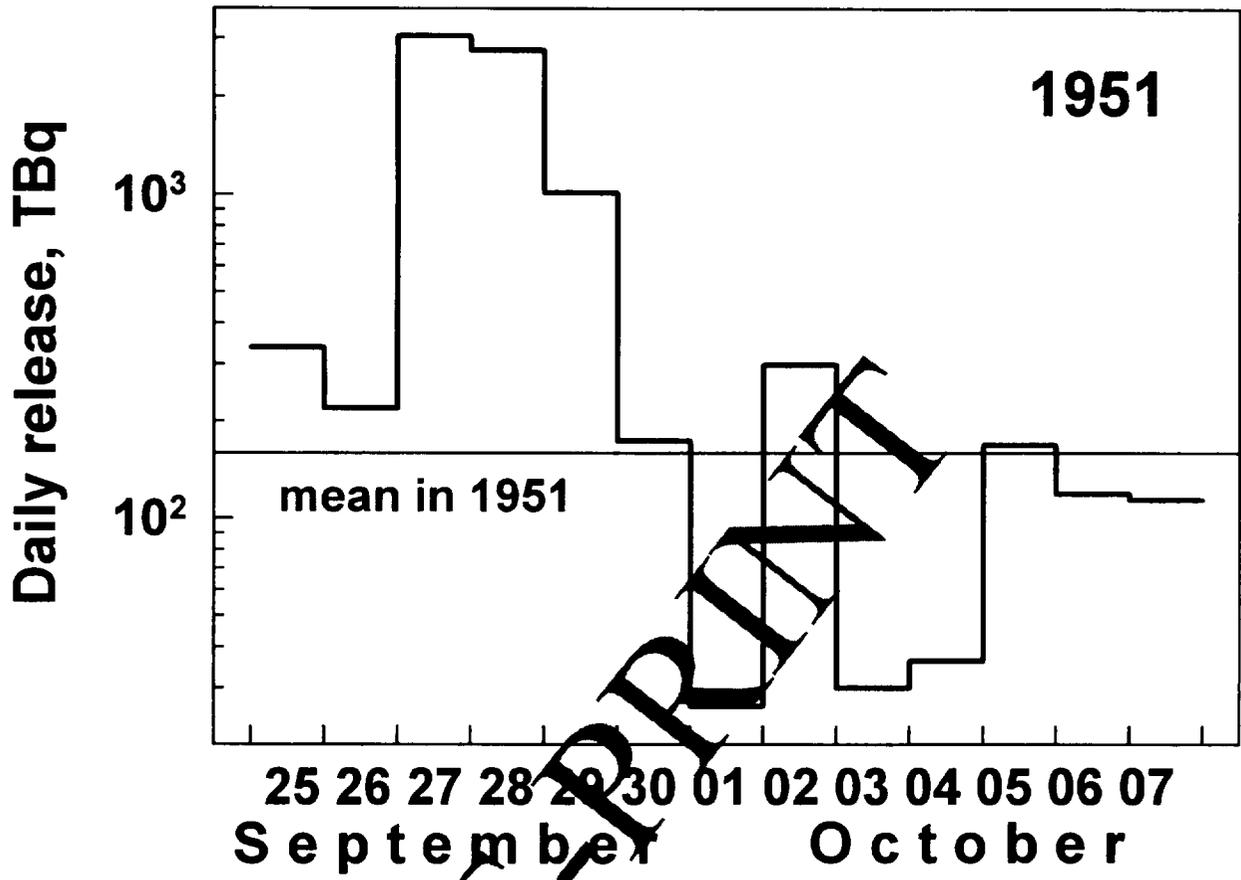


Fig. 5. Average amount of radioactivity released per day into the Techa River between 1949 and 1956, and radionuclide composition of the release (according to the data of the Mayak Central

Laboratory, Project Director Dmitry Ilyin).



*Fig. 6. Fluctuations in release rates observed in Autumn 1951.*

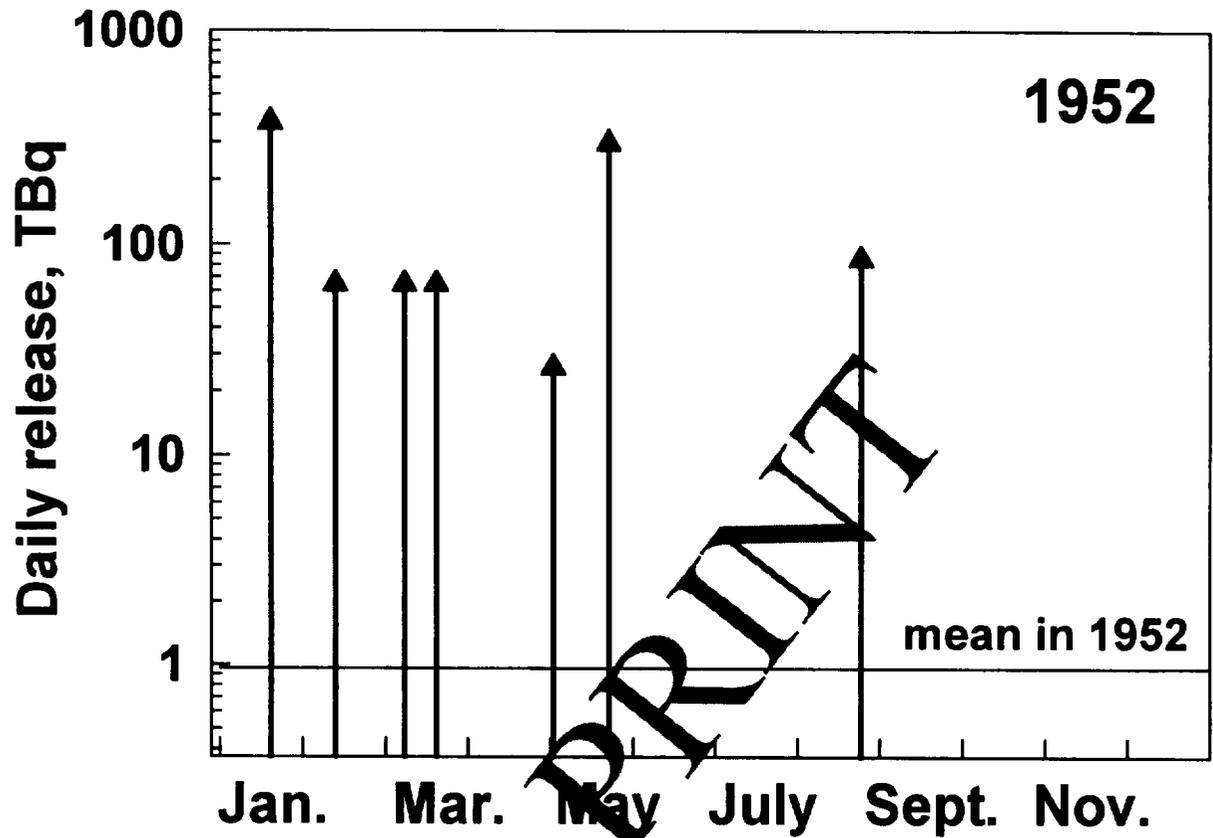
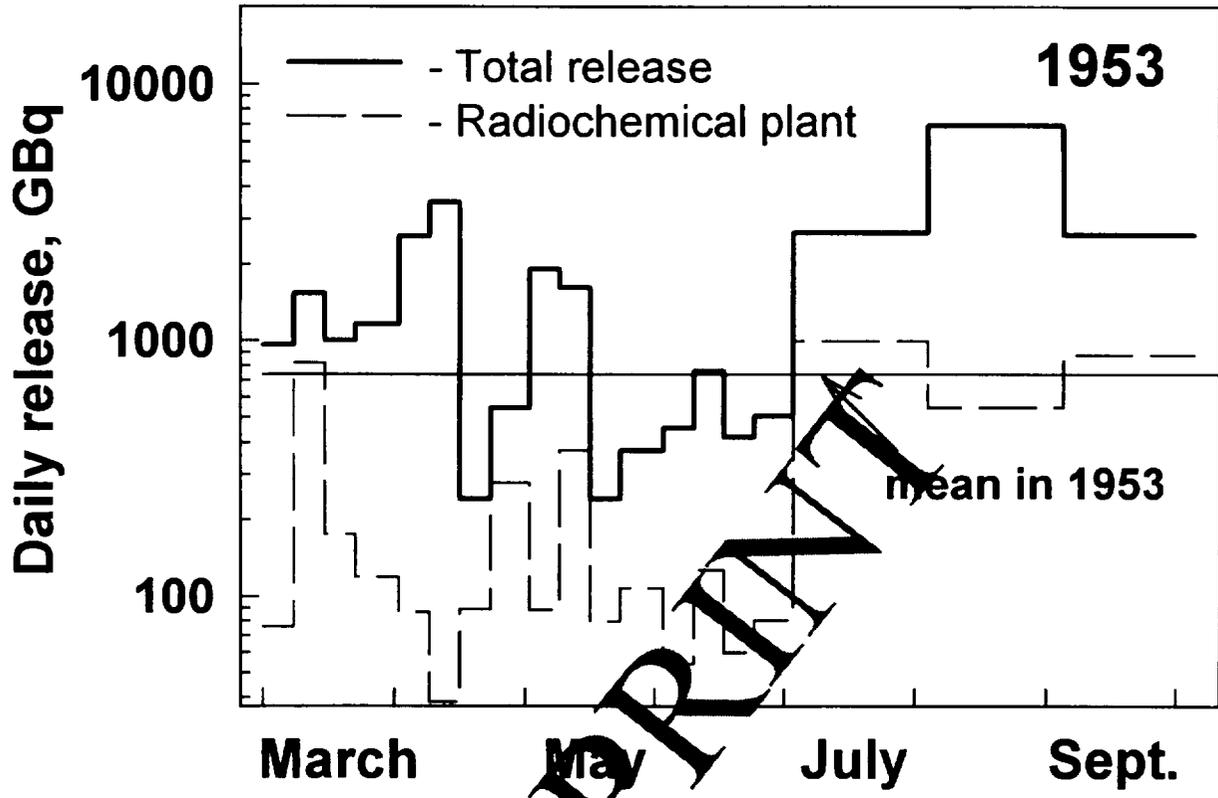
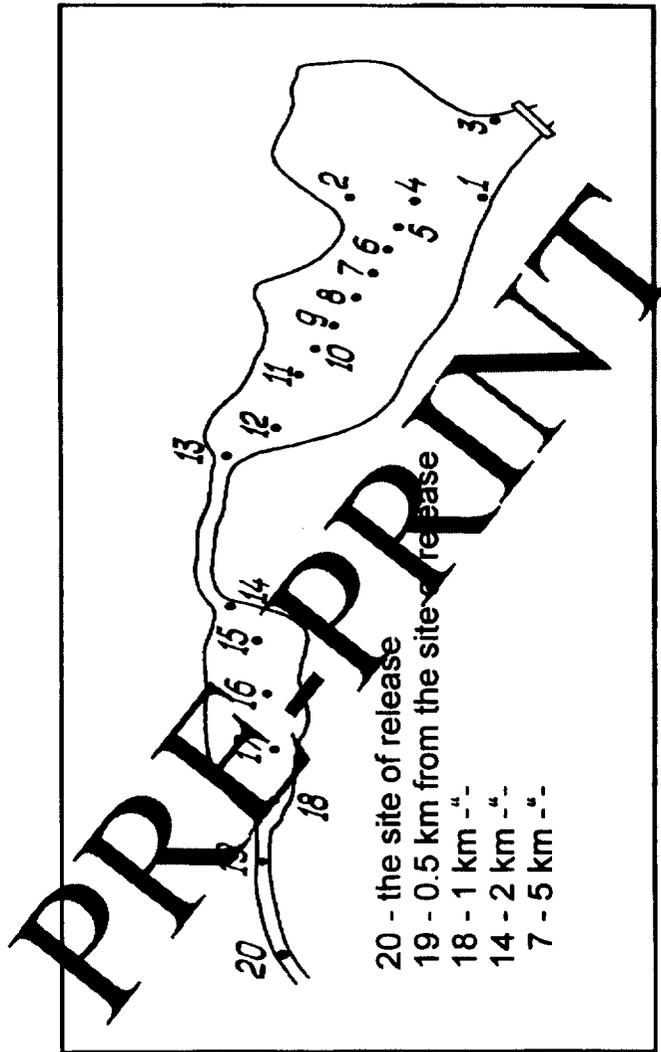


Fig. 7. Peaks of radionuclide releases observed in 1952 relative to annual mean values.



*Fig. 8. Total release rates in 1953 in comparison with the release rates from the radiochemical plant.*



*Fig. 9. Schematic map of Koksharov and Metlinsky Ponds with the sites of historical routine sampling.*

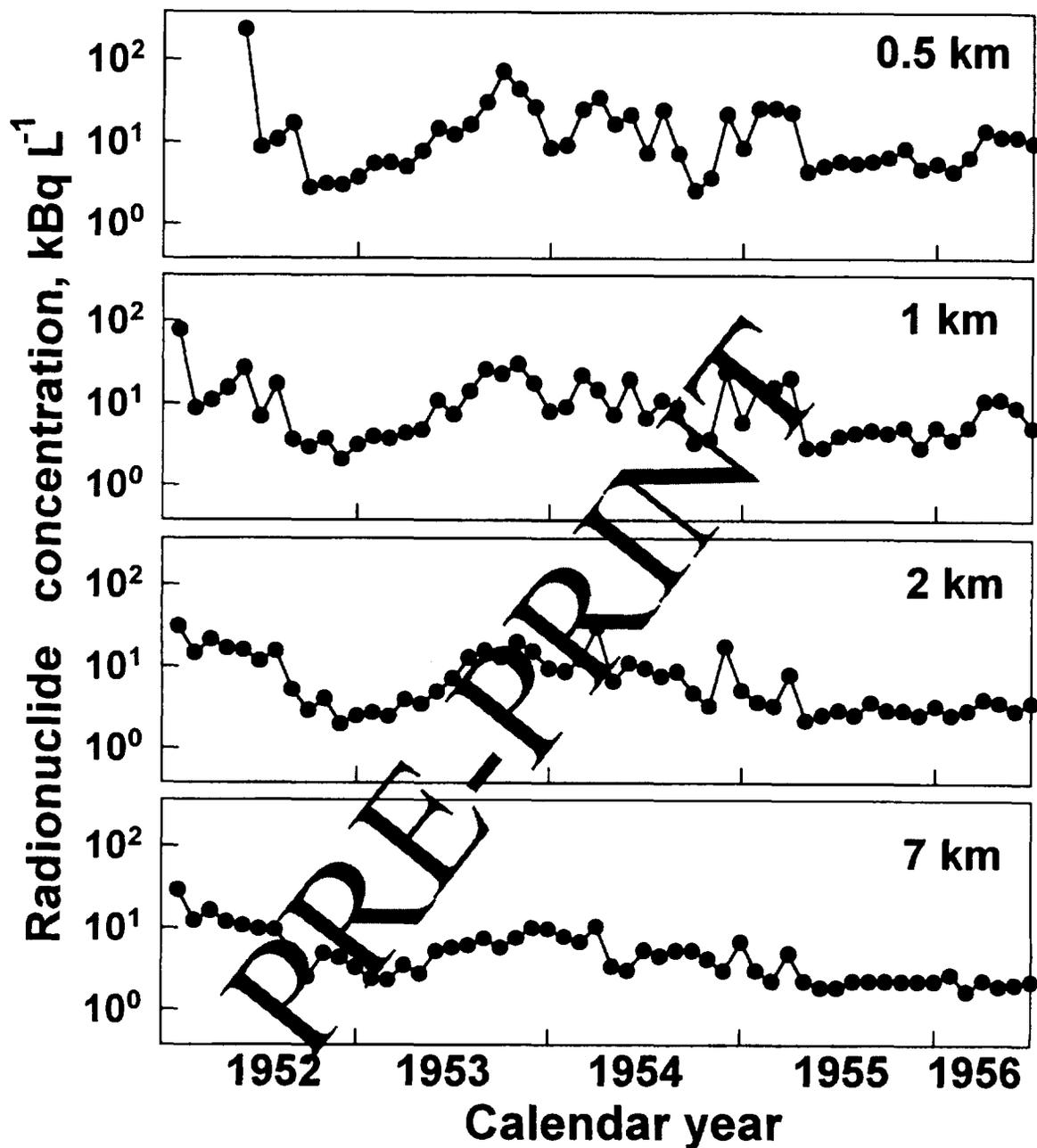


Fig. 10. Time pattern of beta activity of the Techa River water measured in the early period of contamination at several sites of the upper reaches (the range of downstream distance is from 0.5 to 7 km from the site of release).

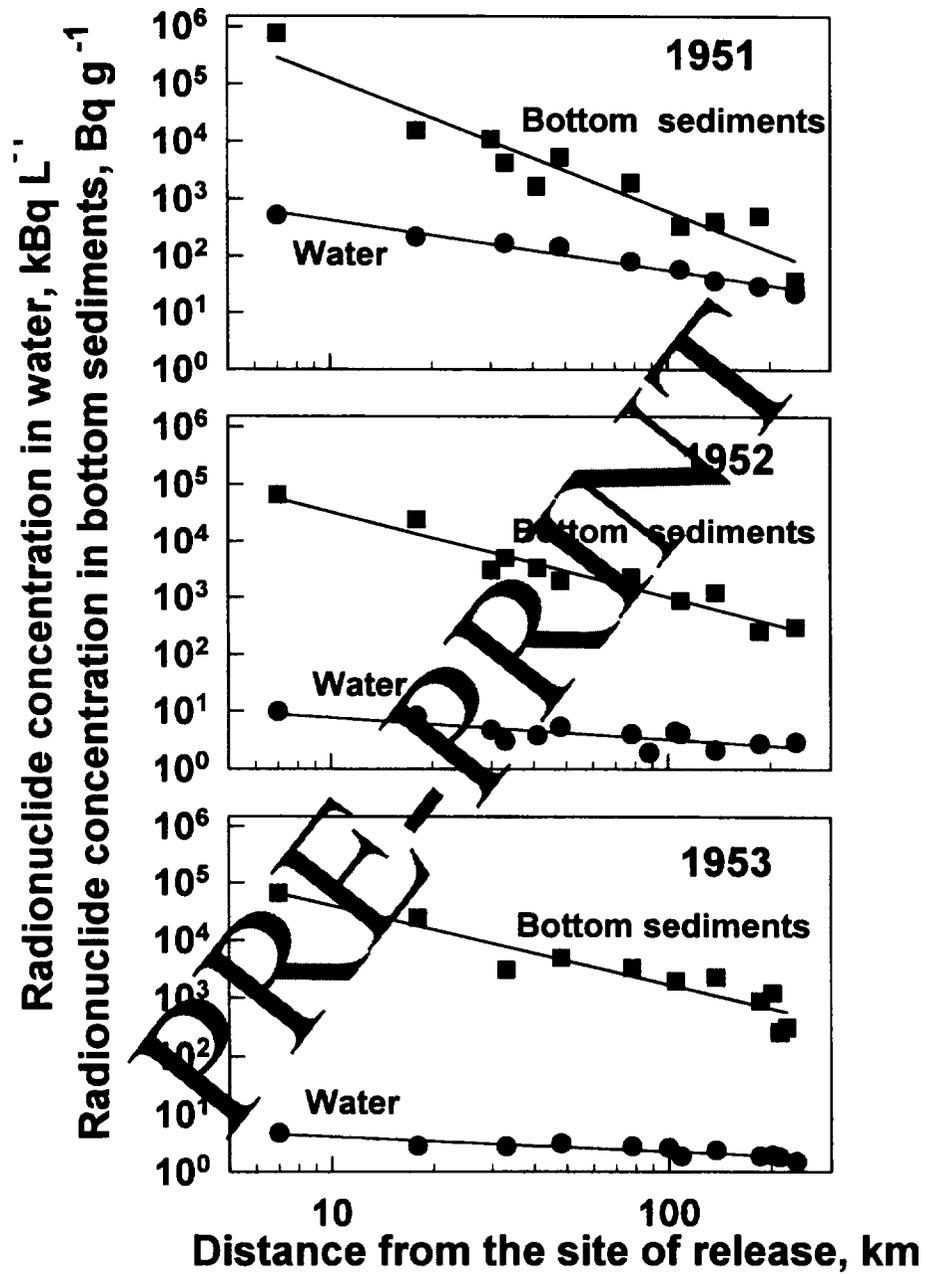


Fig. 11. Total beta activity of the river water and the bottom sediments as a function of downstream distance measured in 1951, 1952, and 1953.

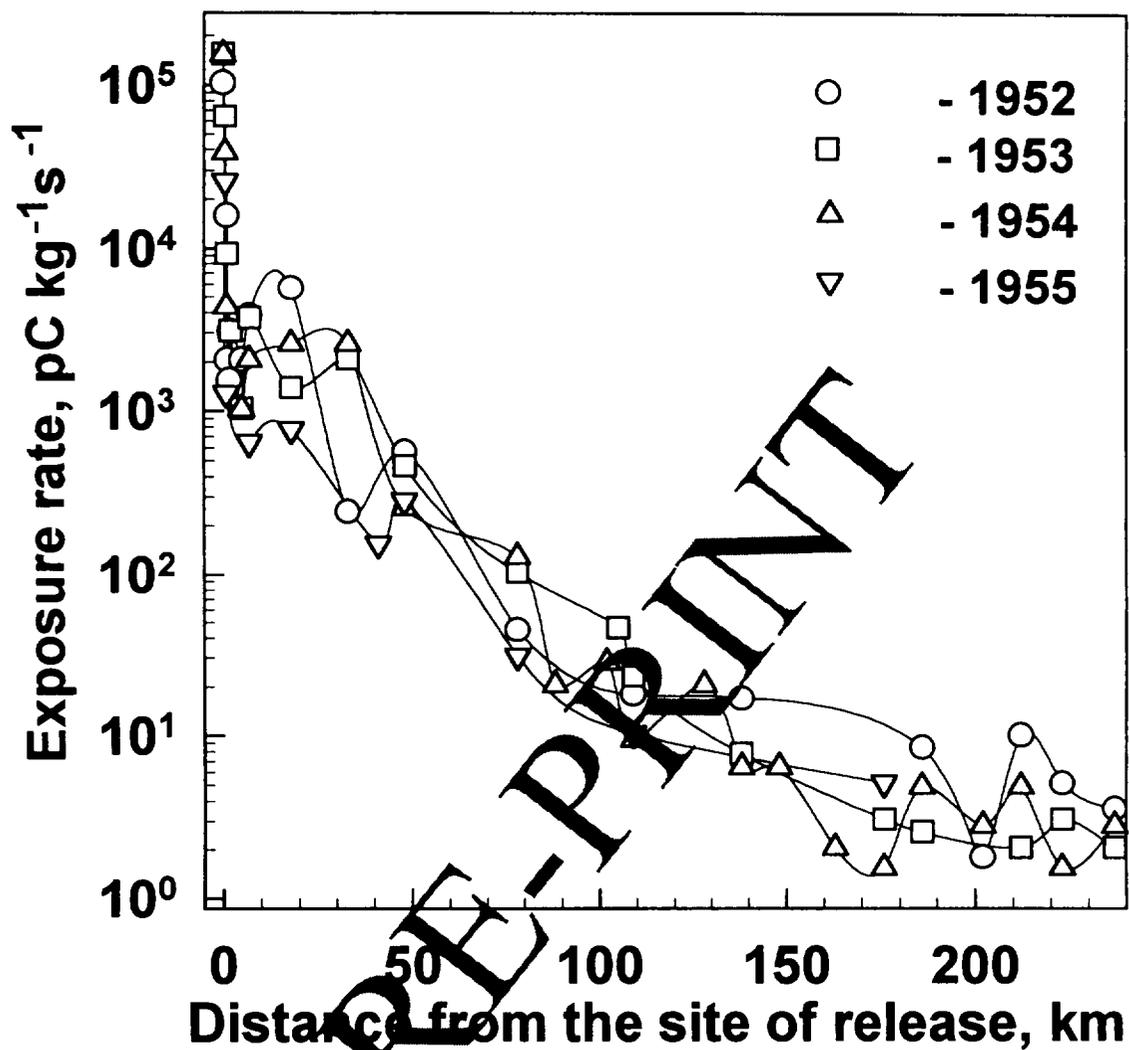
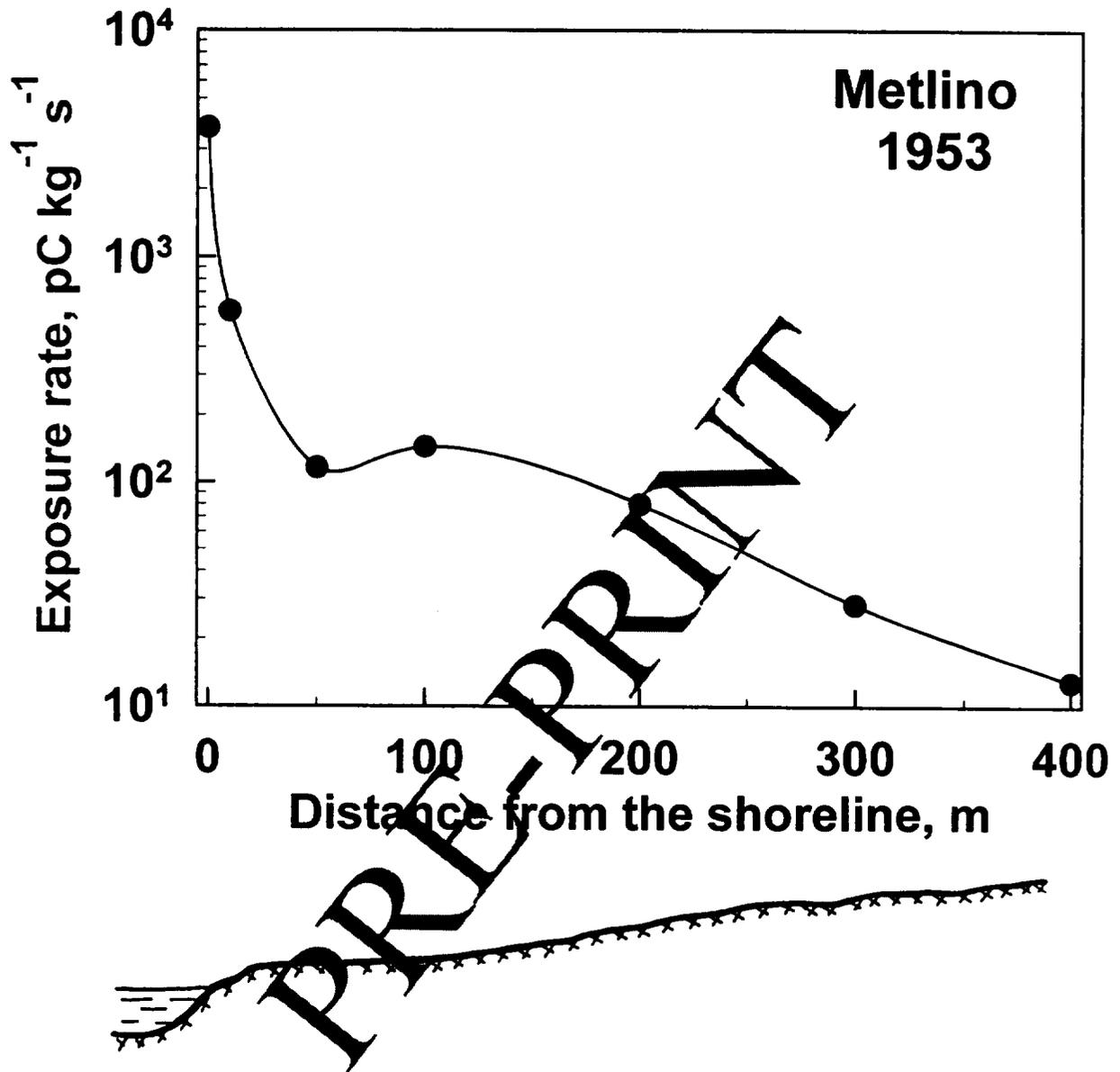


Fig. 12. Results of gamma-exposure-rate measurements near the shoreline as a function of downstream distance, as measured in 1952–1955.



*Fig. 13. Exposure rate as a function of the distance from the shoreline measured in several specific sites: (a) Metlino, 1953; (b) Techa-Brod, 1953–1954; (c) Muslyumovo, 1953–1954; and (d) Kurmanovo, 1956. In the lower part of each drawing the shore topography is indicated.*

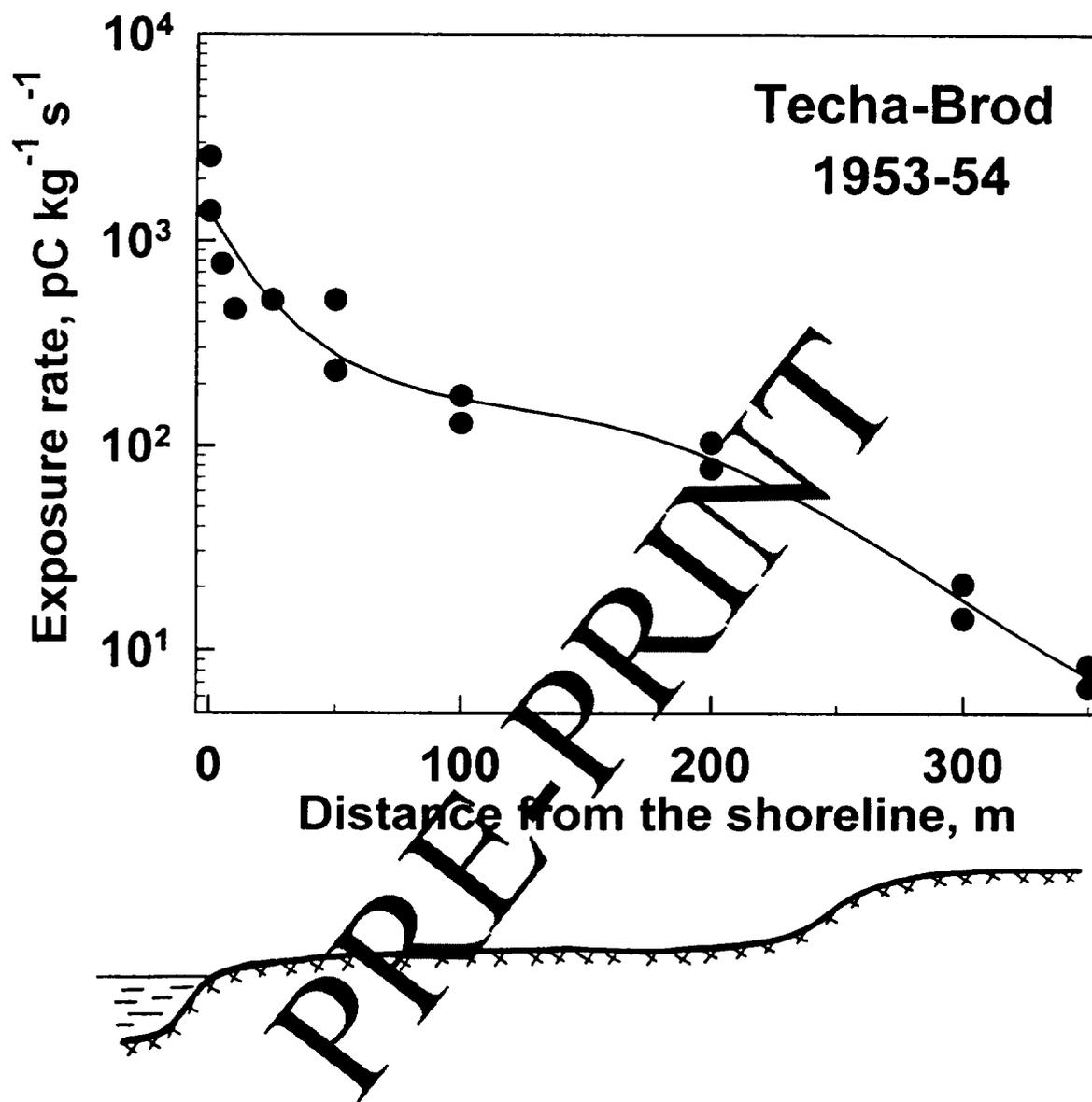


Fig. 13. Exposure rate as a function of the distance from the shoreline measured in several specific sites: (a) Metlino, 1953; (b) Techa-Brod, 1953–1954; (c) Muslyumovo, 1953–1954; and (d) Kurmanovo, 1956. In the lower part of each drawing the shore topography is indicated.

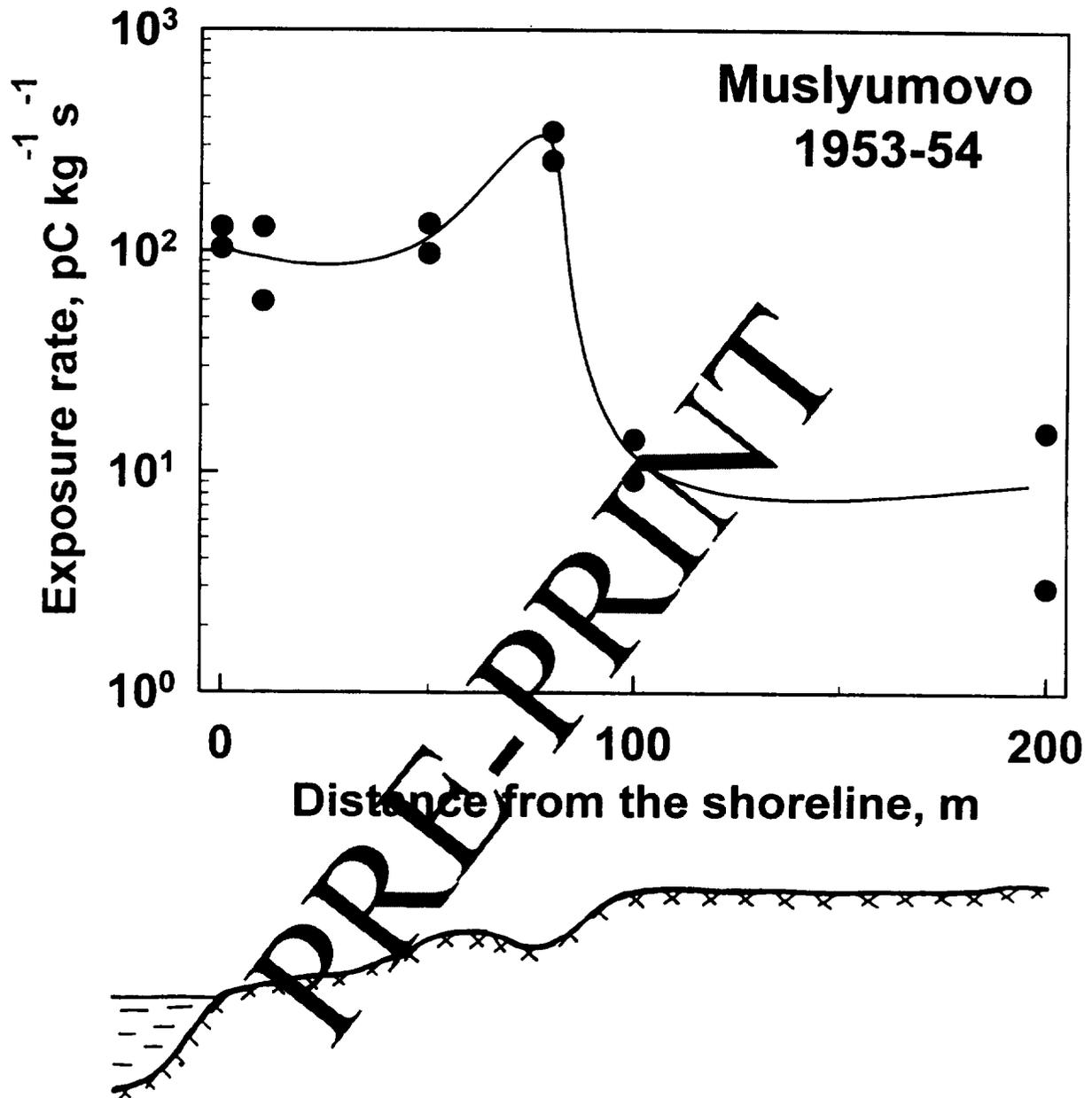


Fig. 13. Exposure rate as a function of the distance from the shoreline measured in several specific sites: (a) Mellino, 1953; (b) Techa-Brod, 1953–1954; (c) *Muslyumovo*, 1953–1954; and (d) Kurmanovo, 1956. In the lower part of each drawing the shore topography is indicated.

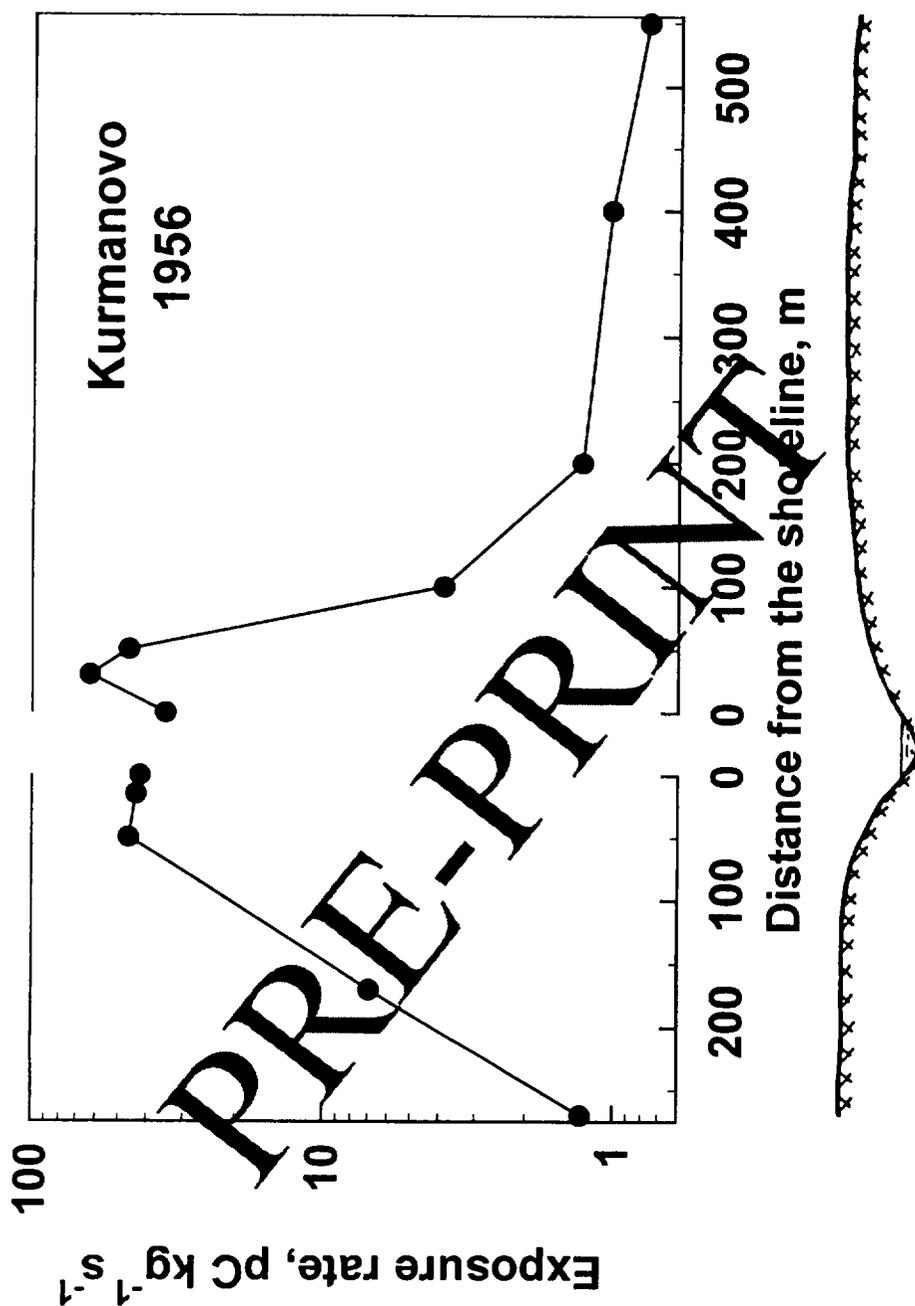


Fig. 13. Exposure rate as a function of the distance from the shoreline measured in several specific sites: (a) Metlino, 1953; (b) Techa-Brod, 1953–1954; (c) Muslyumovo, 1953–1954; and (d) Kurmanovo, 1956. In the lower part of each drawing the shore topography is indicated.

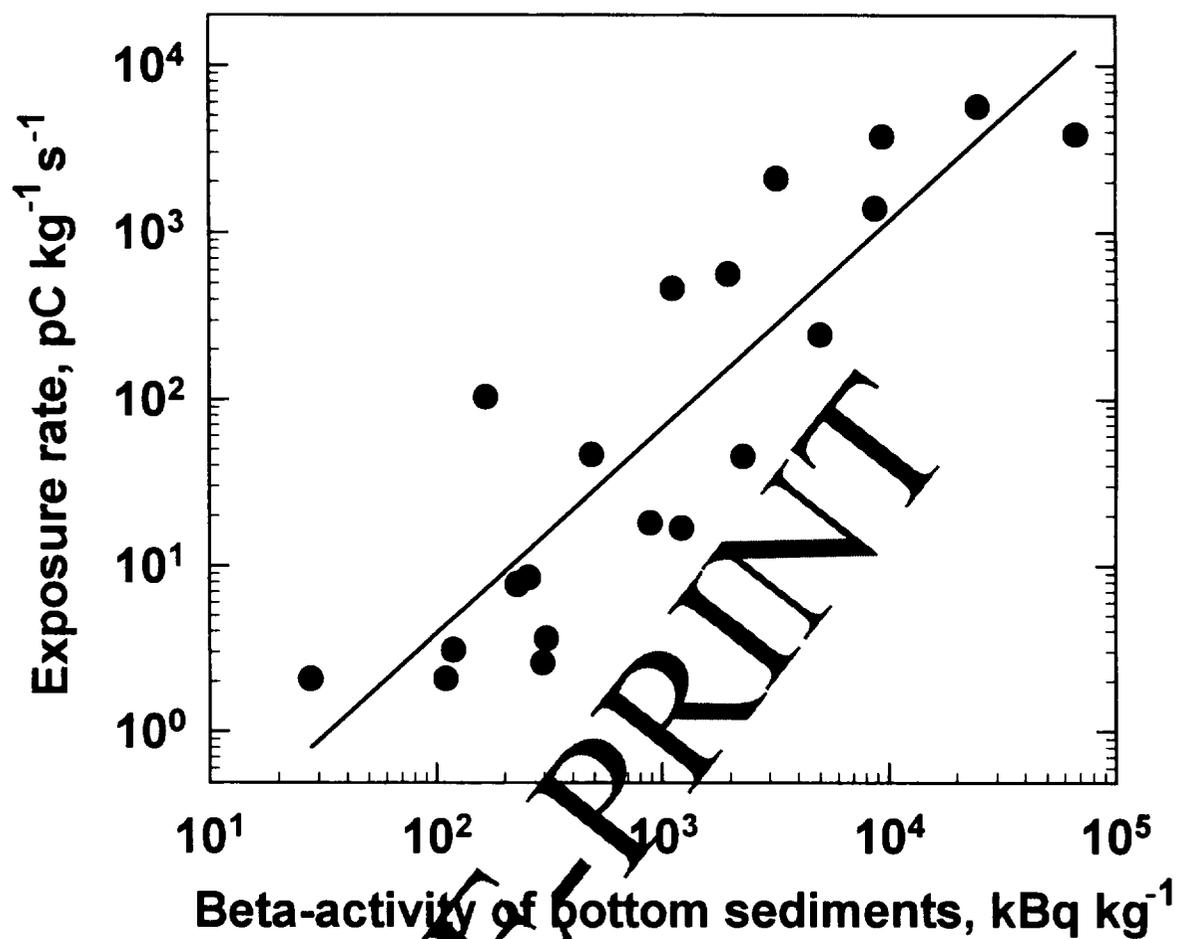


Fig. 14. Correlation of external gamma-exposure rate near the shoreline with beta-activity of bottom sediments (measurements of 1952–1953).

Table 1. Characteristics of primary data on release dynamics.

Period	Values	Character of data	Sources of release
25 September– 7 October 1951	Volume and specific beta- and gamma- activity released	Daily measurements	Determined partially
1952	Volume and/or specific alpha-, beta- and/or gamma-activity released	Episodic measurements	Not determined
March–June, 1953	Total beta- and/or gamma-activity released	Weekly mean	Determined
July–September, 1953	Total beta- and/or gamma-activity released	Monthly mean	Determined

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Table 2. Radionuclide composition of the releases in different years according to different authors.

Time period	Radionuclide composition, %						Reference	
	<sup>89</sup> Sr+ <sup>90</sup> Sr	<sup>89</sup> Sr	<sup>90</sup> Sr	<sup>137</sup> Cs	REE	<sup>103,106</sup> Ru		<sup>95</sup> Zr+ <sup>95</sup> Nb
1949		1.8	4.1	11		55.6	30	Ilyin (1956)
January–February 1950		6.9	15.3	21.2	5.7	45.3	9	Ilyin (1956)
March 1950–October 1951		8.8	11.6	12.2	26.8	25.9	13.6	Ilyin (1956)
November 1951–December 1953	25.6-58.0			4.5-15	10-	1	8-25	Ilyin (1956)
1954	44.0			10.0			1.0	Marey (1959)
1955	39.0			12.0				Marey et al. (1956)
1956	53.5			23.8	12.5	7.3		Marey et al. (1957)

- Notes: 1) All data presented correspond exactly to the original references;  
 2) REE - rare-earth elements;  
 3) Radionuclide composition for the period 1949–51 - assessed data;  
 4) For the period November 1951–December 1953 - minimal-maximal percentage;  
 5) For 1954–1956 - data of radiochemical analyses.

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Table 3. Radionuclide distribution in liquid waste, which was sampled on 25 September 1951,  
before and after centrifugation

Type of sample	Beta activity, kBq L <sup>-1</sup>	Concentration of radionuclides, %				
		<sup>89,90</sup> Sr	<sup>137</sup> Cs	REE	<sup>103,106</sup> Ru	<sup>95</sup> Zr+ <sup>95</sup> Nb
Native suspension	44.4	20.2	2.8	47.4	7.6	22.0
Solution (supernatant)	13.7	49.2	9.0	3.5	8.3	30.0
Sediment	30.7	7.3	0	67.0	7.3	18.4

Note: REE - rare-earth elements.

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Table 4. Main stages of the evolution of the exposure situation

Period	Main sources of contamination	Availability of source-term data	River configuration	Availability of environmental monitoring data
March 1949– October 1951 - first phase of “acute period”	High-level wastes of radiochemical plant; “wild overflows”	Releases are reconstructed theoretically by the Mayak experts.	Two ponds in upper reaches	Beta activity since July 1951
November 1951– November 1956 - second phase of “acute period”	Low-level wastes of radiochemical plant; Kyzyl-Tash Lake water	Release monitoring data	Two ponds in upper reaches	Beta activity, exposure rates
December 1956– 1963 - isolation of the upper reach	Berdyanish Lake water; washing off EURT area	Total beta activity is evaluated.	Cascade of three reservoirs in upper reaches	Beta activity, exposure rates, $^{90}\text{Sr}$ and $^{137}\text{Cs}$ since 1963
After 1963 - natural self- cleaning regime	-	-	Cascade of four reservoirs and by-pass canals in upper reaches	Beta activity, exposure rates, $^{90}\text{Sr}$ and $^{137}\text{Cs}$ , plutonium since 1991

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APPENDIX

Chronology of the main events 1948–1967: Stages in the development of the Mayak site; human interference in the Techa River system; important changes of meteorological and hydrological parameters.

Year	Time period	Event	Notes
1948	June 19	The first industrial reactor for plutonium production (A) begins operation at full capacity of 100 MW	
1948	December 22	The beginning of radiochemical plant operations	
1949	January 20 — March 26	Overhaul of shutdown Reactor A	Releases of radioactive materials into the environment
1949	March	Start of liquid radioactive releases from the radiochemical plant into the Techa River	Average daily release about 2.6 TBq (70 Ci) (low-level wastes after decontamination)
1950	January	Increase of radioactive releases into the Techa River	Average daily release was about 32 TBq (860 Ci)
1950	March	Sharp increase of radioactive releases into the Techa River	Average daily release reached about 159 TBq (4300 Ci): <ul style="list-style-type: none"> <li>• Low-level wastes without decontamination;</li> <li>• High-level wastes after treatment and decontamination;</li> <li>• Episodic “wild overflows” up to 3,700 TBq (100,000 Ci) day<sup>-1</sup></li> </ul>
1950	May 15	Reactor AV-1 was put into operation	
1951	April 6	Reactor AV-2 was put into operation	
1951	July 5	First measurements of river-water activity	
1951	July	Water discharge from Kyzyl-Tash Lake	Flow-rate about 7.5 m <sup>3</sup> s <sup>-1</sup>
1951	August 11	Dam No. 3 (Koksharov Pond) was constructed	Capacity of pond was increased 4 fold and reached about 400,000 m <sup>3</sup>
1951	October 13-22	Washing of Koksharov and Metlinsky Ponds. More than 15 million m <sup>3</sup> of water were discharged from Kyzyl-Tash Lake	Part of radioactive contaminated sediments was moved with water to the lower part of the Techa River. Essential decrease of gamma-dose rates was observed near Metlinsky Pond
1951	October 28	Main technological wastes were diverted into Karachay Lake	The total radioactive releases into the Techa River decreased to about 3.7 TBq (100 Ci) day <sup>-1</sup>
1951	November	Creation of external dosimetry group in the Central Research Laboratory of MPA	
1951	November 17	HW-Reactor was put into operation	
1951	December 22	Reactor AI begins operation	
1952	May	Increasing of the water level in Koksharov Pond by 80-90 cm	Sharp decrease of dose rate on pond shoreline

1952	September 15	Reactor AV-3 put into operation	
1953	May 30– June 25	Cessation of water discharge from Kyzyl-Tash Lake	Decrease of the water level and increase of gamma dose rates in the upper Techa region
1954	March 13– April 17	Discharges of Kyzyl-Tash Lake water into the Techa River	
1954	July	High air temperatures and drought	2–3 times decrease of river flow-rate in comparison with June 1954.
1955	January– March	Discharge of Kyzyl-Tash Lake water with the purpose of exchange of water in the lake	Discharge rate to the Techa was about 2.6 TBq (70 Ci) day <sup>-1</sup> , and total activity about 174 TBq (4,700 Ci)
1956	October– November	Dam No. 10 was constructed	Creation of Reservoir No. 10 (Shubinsky Pond). Decrease of radionuclide entrance to the river system
1957	September 29	Chemical explosion of high-level waste-storage tank (Kyshtym accident)	Formation of the East Urals radioactive trace (EURT)
1958	January 1– April 25	Berdyanish Lake (volume 7.9 million m <sup>3</sup> with average activity 15 kBq L <sup>-1</sup> ) was released into Reservoir No. 10. After that more than 1.8 million m <sup>3</sup> of clean water from Irtyash Lake were passed through Berdyanish Lake to Reservoir No. 10	Total beta-activity released was about 127 TBq (3,440 Ci). The surface level of Reservoir No. 10 increased by 107 cm. Beta activity of water increased from 1.5 up to 15 kBq L <sup>-1</sup>
1958	April	Strong filtration through Dam No. 10 was found .	Strengthening of Dam No. 10 on April 29–30
1964		Dam No. 11 and by-pass canals were constructed .	Creation of Reservoir No. 11 (capacity about 217×10 <sup>6</sup> m <sup>3</sup> ). Isolation of more contaminated upper Techa from rest of river system.
1967	April 10– May 15	Wind transfer of radioactive materials from Karachay Lake	Formation of so-called New trace

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**APPENDIX 2**

**Pre-print**

**Vorobiova, M. I.; Degteva, M. O. Simple model of radionuclide transport in the Techa River. *Health Phys.* (submitted, 1998).**



## **SIMPLE MODEL OF RADIONUCLIDE TRANSPORT IN THE TECHA RIVER**

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*Abstract*—The Techa River (Southern Urals, Russia) was contaminated in 1949–1956 by liquid radioactive wastes from the Mayak complex, the first Russian facility for the production of plutonium. A simple model describing radionuclide transport from the site of release along the river and the accumulation of radionuclides by bottom sediments is presented. This model successfully correlates the release rates of radionuclides as reconstructed by the Mayak experts, hydrological data, and available environmental monitoring data for the early period of contamination. The model was developed for dose reconstruction for the population that lived in riverside communities during the period of the releases and who were chronically exposed to external and internal irradiation. The model fills the data gaps and permits reconstruction of external gamma-exposure rates in air on the river bank and radionuclide concentrations in river water used for drinking and other household needs.

### **INTRODUCTION**

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. The extensive increase in plutonium production from 1948 to 1955, as well as the absence of reliable waste-management technology, resulted in significant releases of liquid radioactive waste into the rather small Techa River. This resulted in chronic external and internal exposure of about 30,000 residents of riverside communities; these residents form the cohort of an epidemiologic investigation (Degteva et al. 1996). Analysis of the available historical monitoring data indicates that the releases during the early years (1949–1951) of

operation of the MPA were the most significant and account for more than 95% of the total releases (which were equal to  $10^{16}$  Bq). The relevant operating history of the MPA is presented in Vorobiova et al. (1998). Historical data are described regarding available measurements of releases of radionuclides to the Techa River and concentrations of radionuclides in water and sediments. The history of dam construction and watercourse changes is also described, as these alterations have had a significant impact on the movement of radionuclides within the Techa River system.

Analysis of the available historical environmental monitoring data (Vorobiova et al. 1998) indicates that the following reliable data sets are available for reconstruction of doses received during the early periods of operation of the MPA: Average annual values of specific beta activity of river water and bottom sediment as a function of downstream distance (available since summer 1951) and external gamma-exposure rates in air near the shoreline as a function of downstream distance (available since summer 1952). It is most important for the purposes of dose reconstruction to know radionuclide concentrations and exposure rates during the first phase of the exposure situation (1949–1951), when contact of people with the contaminated river was not limited. Very few measurements are available for this period. The only information available for the first two years of contamination is release rates and major radionuclide contributions to the total released activity, as reconstructed by the Mayak experts. The only technique available to solve this problem is development of a model describing radionuclide transport along the river from the site of release and the accumulation of radionuclides by bottom sediments. If consistency between the release rates reconstructed by the Mayak experts and the environmental monitoring data can be confirmed using a realistic river model, this can serve as a test of the validity of the estimates of radionuclide releases.

## DESCRIPTION OF THE MODEL

The model was created to describe stationary (annual average) parameters of radionuclide transport in the Techa River in the early years from the beginning of contamination (1949–1951). The Techa River system consisted of two ponds (Koksharov and Metlinsky) in the upper reaches and the river itself during this period (Fig. 1).

We consider a situation with long-term releases and long-term averaged activity of river water. Radionuclide transport in water in this case is determined by processes of advection, sorption and radioactive decay and can be described by the equation:

$$V \frac{\partial A(R)}{\partial R} = -\lambda_s A(R) - \lambda A(R), \quad (1)$$

where  $A(R)$  is the activity transferred through a cross section of flow at downstream distance  $R$  (from Dam No. 4);  $\lambda_s$  is the fractional rate of loss of activity by sorption onto sediments;  $\lambda$  is the physical decay constant; and  $V$  is the river velocity.

The analytical solution of this equation is given by

$$A(R) = A_0 \exp(-kR - \frac{\lambda}{V} R), \quad (2)$$

where  $A_0$  is the activity at  $R = 0$  and  $k = \lambda_s V^{-1}$  is the coefficient of exponential decrease of water activity due to sorption.

For radionuclide concentration in river water eqn (2) can be written as

$$C(R) = C_0 \cdot \frac{q_0}{q(R)} \exp(-kR - \frac{\lambda}{V} R), \quad (3)$$

where  $C(R)$  and  $C_0$  are radionuclide concentrations at distances  $R$  and  $R = 0$ , respectively; and  $q(R)$  and  $q_0$  are volume-flow rates of the river at distances  $R$  and  $R = 0$ .

Changes in activity of bottom sediments are determined by

$$\frac{\partial S(R,t)}{\partial t} = -kA(R) - \lambda S(R,t), \quad (4)$$

where  $S(R,t)$  is the content of radionuclide in bottom sediments per unit of river bed length at the distance  $R$  at time  $t$ .

The solution of eqn (4) can be written as

$$S(R,t) = \frac{A_0 k}{\lambda} \exp(-kR - \frac{\lambda}{V} R) [1 - \exp(-\lambda t)]. \quad (5)$$

As can be seen, the model gives a simple exponential dependence for the decrease of radionuclide concentration in water and sediments with downstream distance. Such dependence has only one unknown parameter ( $k$ ), which is specific for each radionuclide. The total beta activity for the mixture of radionuclides is described by the sum of exponential terms:

$$C_{\Sigma}(R) = \sum_i C_i(R) = \frac{q_0}{q(R)} \sum_i C_{0i} \exp(-k_i R - \frac{\lambda_i}{V} R). \quad (6)$$

For further analysis it is useful to introduce two relative quantities:  $a_i = k_i k_{Sr}^{-1}$ , the ratio of exponent coefficients for radionuclide  $i$  and for  $^{90}\text{Sr}$ ; and  $\Delta_{0i} = C_{0i} C_0^{-1}$ , the fraction of radionuclide  $i$  in the total initial activity. Substitution of these quantities gives the following expression describing the decrease of specific beta activity of river water with downstream distance:

$$C_{\Sigma}(R) = \frac{C_0 \cdot q_0}{q(R)} \sum_i \Delta_{0i} \cdot \exp(-k_{Sr} a_i R - \frac{\lambda_i}{V} R). \quad (7)$$

### INPUT DATA AND HYDROLOGICAL PARAMETERS

An estimate of the radionuclide composition of the releases in 1950–1951 given in the Doctoral Thesis of Dmitry Ilyin, who was the head of the Mayak Central Laboratory and knew the Mayak processes at that time, was used for model calculations (Table 1, Column 2). The

historical methods of radiochemical separation and analysis were sufficient to permit the separation of groups of radionuclides with similar chemical properties. Short-lived radionuclides were reconstructed from theoretical ratios for fission products of  $^{235}\text{U}$  irradiated by thermal neutrons for radionuclides within groups of similar chemical characteristics. From analysis of the data reported by D. Ilyin, it appears that the average age of fission products released to the Techa River in 1950–1951 was about one year.

A fraction of the release was soluble and another fraction was associated with particles: Almost all cesium, 75% of strontium, and about 50% of zirconium and niobium were in soluble form, but about 98% of rare earth elements entered the river adsorbed on suspended particles. To describe the changes in radionuclide composition during passage through Koksharov and Metlinsky Ponds, all solid particles suspended in the releases were assumed to have become sediment in these ponds. The radionuclide composition of the soluble fraction (Table 1, Column 3) was used as input data for the model of the free flowing Techa.

The hydrological parameters for the Techa River,  $q(R)$  and  $V$ , were obtained from hydrological measurements published in Agapitova (1975) and Marey (1959). The average river velocity was equal to 0.3 m s<sup>-1</sup> and distance dependence of volume-flow rate was described as  $q(R)q(0)^{-1} = 1 + 0.0136R$ .

### EVALUATION OF MODEL PARAMETERS

Two sets of data were used to evaluate model parameters. The values of  $a_i$  were estimated on the basis of special experiments carried out under the supervision of Nickolay Timofeev-Ressovsky in 1953–1954 in the Urals and published later (Agafonov et al. 1960; Timofeeva-Ressovskaya et al. 1962; Ivanov et al. 1965). The aim of these experiments was physical modeling of radionuclide behavior in different kinds of flowing and stagnant reservoirs.

The Mayak authorities initiated these experiments specifically to clarify the situation of the contaminated Techa River and its lakes (Novosselov et al. 1997). The results of two series of experiments with artificial flowing reservoirs (Agafonov et al. 1960) are presented in Fig. 2 and Table 2. It can be seen that the sorption of radionuclides by bottom sediments decreases in the order  $^{137}\text{Cs} > ^{144}\text{Ce} > ^{106}\text{Ru} > ^{90}\text{Sr}$ . These results also showed that the radionuclide composition changed with downstream distance as a result of the difference in sorption coefficients.

Absolute values of  $k_i$  are determined not only by the nature of the particular nuclide but depend also on the various characteristics of the watercourse (e.g., river velocity). We suggest that all  $k_i$  change proportionally with the changes in the parameters of river bed or hydrological parameters. Therefore,  $a_i = k_i k_{\text{Sr}}^{-1}$  for the Techa River are equal to respective values for  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$  and  $^{106}\text{Ru}$  derived from the data of Table 2. The second experimental series is used, since it was longer and thus more representative.

Other radionuclides of interest (such as  $^{90}\text{Zr}$ ,  $^{95}\text{Nb}$  and  $^{91}\text{Y}$ ) were studied together with  $^{90}\text{Sr}$  in other experiments with stagnant reservoirs (Timofeeva-Ressovskaya et al. 1962). We suggest that it is possible to evaluate  $a_i$  for these radionuclides on the basis of ratios of sorption coefficients obtained in these experiments (Table 3).

The values of  $k_{\text{Sr}}$  and  $C_0$  were evaluated by fitting the model results to the data on specific beta activity in the Techa River for 1951 (Fig. 3). Fig. 3 illustrates that the model describes well the decrease of beta activity in water for distances up to 100 km from Dam No. 4 and underestimates values for the lower reaches. This may be caused by the different characteristics of the river's bed (turf-silt and clay in the upper and mid-Techa region against sand and gravel in the lower reaches of the river). Probably, it would be reasonable to improve this model in the

future by introducing two compartments according to the bed-deposit characteristics. Table 4 Column 2 presents the results of evaluation of the parameters for major radionuclides.

### VALIDATION OF THE MODEL

First, the results of parameter evaluations were compared with literature data. As seen from Table 4, the values for the Techa River are compatible with the values for an irrigation canal of the Rhone River (Schaeffer 1975). Small differences in parameter values could be explained by the difference in characteristics of the watercourse. The second data set used for model validation was beta activity of the bottom sediments measured in 1951 in the Techa River. The actual density of bottom sediments and the width of the river bed were taken into account in these calculations. As seen in Fig. 4, the model calculation is in good agreement with the experimental data for this quantity.

The measurements of long-lived radionuclides, performed in the 1960s and 1970s served as an additional data set for model validation. For  $^{90}\text{Sr}$ , the data on concentrations in flood-plain soils and humans were used (Table 5). This set was used because the contamination of flood-plain soils occurred as a result of an extraordinary flood in April to May 1951, and  $^{90}\text{Sr}$  content in humans (measured by whole body counter) was largely the result of the consumption of river water in 1950–1951 (Degteva et al. 1998). Therefore, the distance dependencies for these quantities must reflect the decrease of  $^{90}\text{Sr}$  concentration in river water during the early period of contamination. For body content, “maximal in village” values were used because “average in village” values depended also on the number of wells available in each settlement in 1950–1951 (Degteva et al., 1998). As seen from Table 5, the rate of change of concentration as a function of distance for calculated water concentration, measured soil concentration, and human-body burdens is in reasonable agreement.

$^{137}\text{Cs}$  is sorbed tightly to sediments, so it is possible to compare calculated and measured concentrations in bottom sediments over a long time period (Koloskov 1968). The comparison of model results for  $^{137}\text{Cs}$  and statistical evaluation of the slope for this nuclide in bottom sediment and flood-plain soils is given in Table 6. Again, estimates of parameter values are consistent with experimental data. The data of Fig. 5 show that the model gives slightly elevated levels of  $^{137}\text{Cs}$  concentration in bottom sediments in comparison with measured values. This may be caused by the process of desorption, which was not taken into account in the model. The desorption coefficient for  $^{137}\text{Cs}$  is very low, but the desorption process would result in a decreased concentration over long periods.

In general, the data presented suggest that the Techa River model and its evaluated parameters are reliable for describing the exposure situation on the Techa River during the earlier period of contamination.

## RESULTS OF MODELING

The main purpose of river modeling is to reconstruct radionuclide concentrations in water and exposure rates on the river bank for the early period of contamination, when there were no environmental monitoring data. Fig. 6 presents modeled concentrations of radionuclides in the Techa River for the period 1949–1951. As can be seen, the beta activity of the river water increased two orders of magnitude in 1950 compared with 1949. The subsequent slight decrease of concentration in 1951 was caused by increased amounts of water discharged. Fig. 7 illustrates the concentrations of different radionuclides relative to  $^{90}\text{Sr}$  for the period 1950–1951. Significant changes occurred in radionuclide composition of water with downstream distance:  $^{90}\text{Sr}$  and  $^{106}\text{Ru}$  became dominant from the mid-Techa to the lower part of the river. It is possible to derive absolute concentrations of different radionuclides in the settlement locations along the

river using the data of Figs. 6 and 7. Also, the river model allows reconstruction of intake levels for  $^{137}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{95}\text{Zr}$ , and  $^{144}\text{Ce}$  using the ratios of these nuclides to  $^{90}\text{Sr}$  and  $^{90}\text{Sr}$ -intake rates reconstructed independently on the basis of multiple measurements of beta count on front teeth of the Techa riverside residents (Kozheurov and Degteva 1994).

Another important result of the modeling is the possibility to reconstruct external gamma-dose rates in air (DRA) on the river bank in the early years, when there were no appropriate measurements. Exposure rates measured in 1952–1953 as a function of distance from the shoreline at several sites suggested that the main source of gamma radiation was from the contaminated sediments, with no appreciable shielding by the water layer near the bank strip. It is possible to use the empirical dependence of exposure rates on beta activity of bottom sediments derived from the parallel measurements of both values carried out in 1952–1953 on the Techa River and described in Vorobiova et al. (1978). Another possibility is the calculation of DRA on the basis of modeled radionuclide concentrations in bottom sediments using coefficients obtained by Monte Carlo simulations of air kerma for contaminated soil with a dose-reduction factor for river shorelines (Bakerman and Ryman 1993). For this purpose, it was assumed that the sediments were uniformly contaminated to a depth of 5 cm (that is close to the depth distributions of beta activity measured in 1951–1952). Also, it was assumed that free-in-air kerma, at a distance of one meter above ground surface, is approximately equal to absorbed dose (electron-equilibrium approach).

The results of DRA calculations by both methods are shown in Fig. 8. The results of measurements of beta activity in bottom sediments and modeled radionuclide compositions were used for dose-rate reconstruction for 1951 and 1952 (Figs. 8b, c) and modeled concentrations were used for 1950 (Fig. 8a). The results of measurements (available only for 1952) are also

presented for the comparison. The comparison of data for different years shows that the accumulation of activity in bottom sediments resulted in a five-fold increase of exposure rates in the upper reaches of the river in 1951 compared to 1950. DRA values for 1951 and 1952 did not change significantly, this indicates that the responsible radionuclides were long-lived (primarily  $^{137}\text{Cs}$ ). The close agreement of the values calculated using both approaches and measurement results provides assurance that our understanding of the historical situation is correct.

### CONCLUSIONS

The most important result of retrospective modeling of the Techa River system is that for the first time it has been possible to elaborate a relatively simple river model that successfully correlates the release rates of radionuclides as reconstructed by the Mayak experts, hydrological data, and environmental monitoring data. This suggests that the levels of river-system contamination in 1949–1950 and the external gamma dose rates reconstructed using this model are reliable and can be utilized for a soundly based assessment of external and internal doses for the Techa riverside residents.

PRE-PRINT

*Acknowledgments*—This work was supported as Project 1.1 of the U.S.–Russian Joint Coordinating Committee on Radiation Effects Research. Financial support during 1997 was provided by the U.S. Department of Energy, the U.S. Environmental Protection Agency, the U.S. National Aeronautics and Space Administration, and the Federal Department of the Ministry of Health of the Russian Federation. The authors would particularly like to thank L. R. Anspaugh (University of Utah, Salt Lake City UT, USA) and B. A. Napier (Battelle, Pacific Northwest Laboratories, Richland WA, USA) for useful discussions and editorial contributions.

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Table 1. Radionuclide composition of the releases and input data for model calculations.

Radionuclide	Fraction in the total released activity	Fraction in the total input activity
<sup>89</sup> Sr	0.088	0.149
<sup>90</sup> Sr	0.116	0.210
<sup>91</sup> Y	0.03*	0.001
<sup>95</sup> Zr	0.044*	0.042
<sup>95</sup> Nb	0.092*	0.085
<sup>103</sup> Ru	0.047*	0.037
<sup>106</sup> Ru	0.212*	0.189
<sup>137</sup> Cs	0.122	0.294
<sup>144</sup> Ce	0.241*	0.013

\*Calculated from theoretical ratios for one-year-old fission products and the fractions of elements with similar chemical properties given by Dyllyin.

Table 2. Values of parameters  $k_i$  for experiments with flowing reservoirs.

Experimental Series	Radionuclide			
	<sup>90</sup> Sr	<sup>96</sup> Ru	<sup>137</sup> Cs	<sup>144</sup> Ce
I	0.38	0.41	1.04	0.62
II	0.46	0.51	1.48	0.83

Table 3. Values of parameters  $a_i$  for experiments with stagnant reservoirs.

Radionuclide	$a_i$
<sup>91</sup> Y	1.81
<sup>95</sup> Zr	1.74
<sup>95</sup> Nb	1.37

Table 4. The results of parameter evaluation for the Techa River, and comparison with analogous parameters for an irrigation canal of the Rhone River (according to Shaeffer 1975).

Radionuclide	$k_i, \text{km}^{-1}$	
	Techa River	Irrigation canal of Rhone River
$^{90}\text{Sr}$	0.007	0.01
$^{106}\text{Ru}$	0.01	0.025
$^{137}\text{Cs}$	0.03	0.05
Average river velocity, $\text{m s}^{-1}$	0.3	0.4

Table 5. Comparison of rate-of-change of  $^{90}\text{Sr}$  concentration in water with the rate-of-change of flood-plain soil concentration and human-body burden.

Quantity	Period of measurements	Slope, $\text{km}^{-1}$	Correlation coefficient	Distance downstream from Dam No. 4, range in km
<i>Model evaluation</i>				
$^{90}\text{Sr}$ concentration in water	1951	0.007		0-230
<i>Statistical evaluation of measurements</i>				
$^{90}\text{Sr}$ concentration in flooded soil	1979	0.007	0.26	71-230
Maximum $^{90}\text{Sr}$ content human body	1974-79	0.008	0.79	0-230

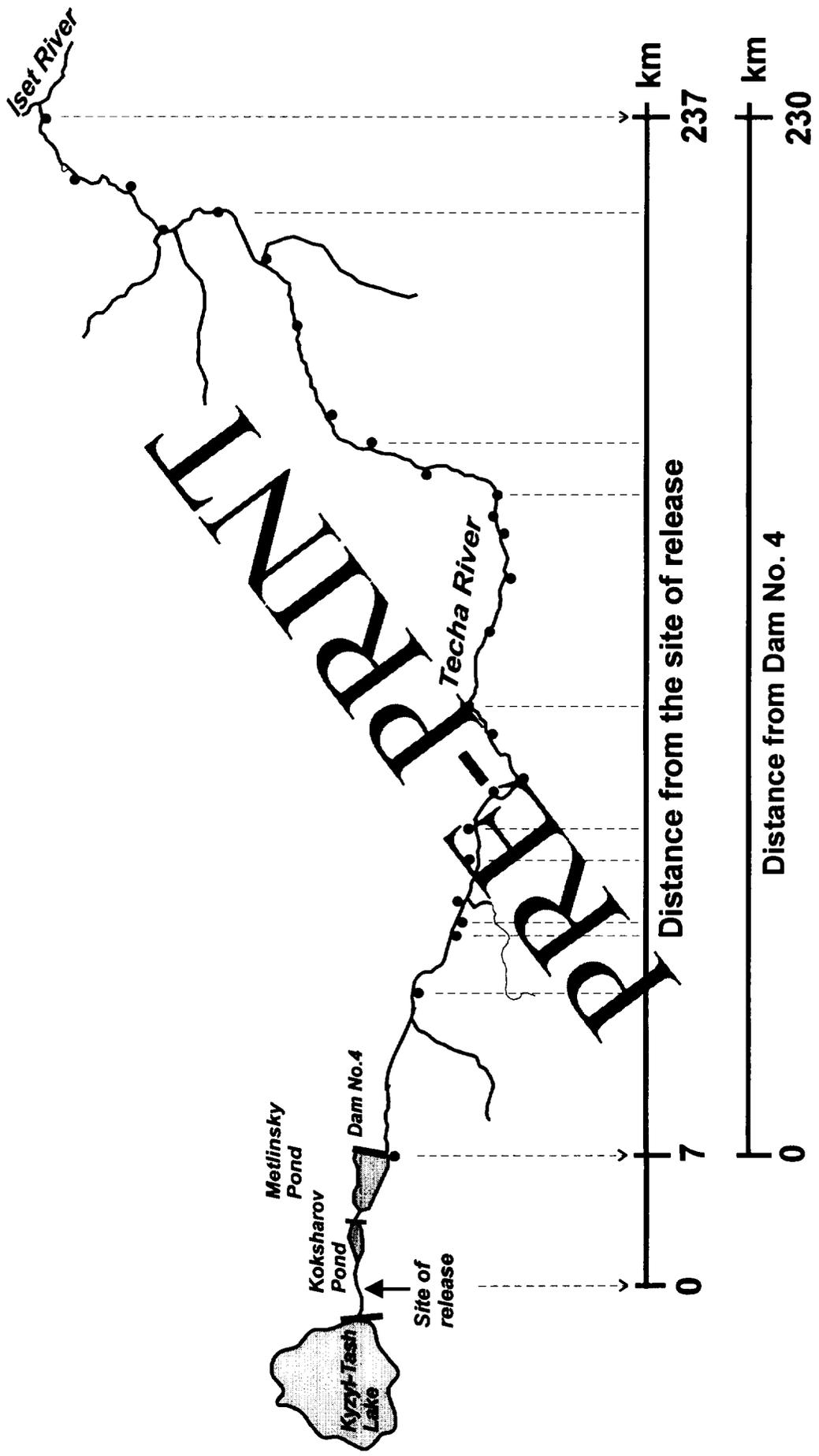
Table 6. Comparison of calculated rate-of-change of  $^{137}\text{Cs}$  concentration in water with rate-of-change of measured bottom sediment and flood-plain soil concentrations.

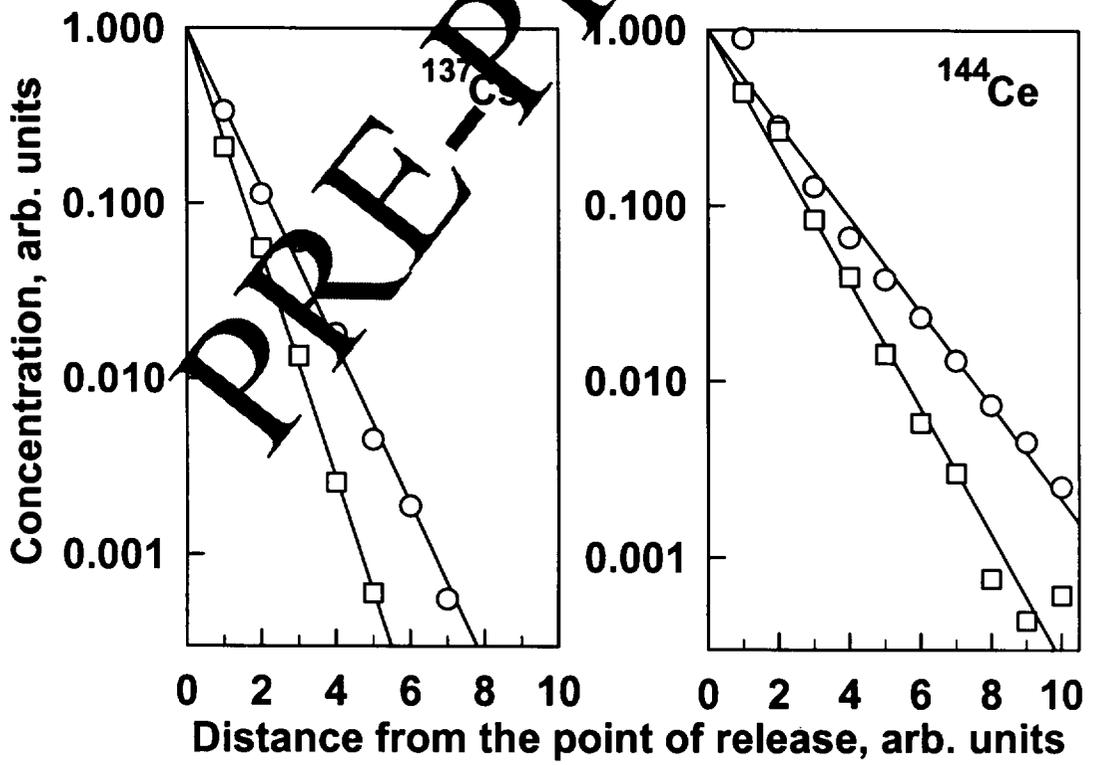
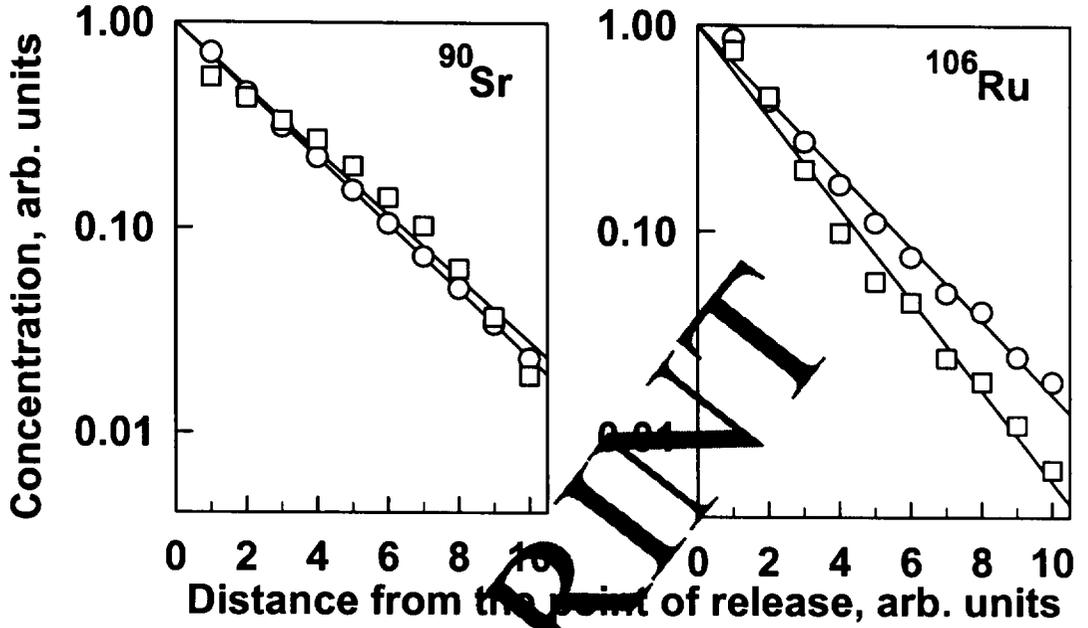
Quantity	Period of measurements	Slope, $\text{km}^{-1}$	Correlation coefficient	Distance downstream from Dam No. 4, range in km
<i>Model evaluation</i>				
$^{137}\text{Cs}$ concentration in water	1951	0.03		0-230
<i>Statistical evaluation of measurements</i>				
$^{137}\text{Cs}$ concentration in bottom sediments	1963-64	0.033	0.98	26-230
$^{137}\text{Cs}$ concentration in flooded soil	1979	0.03	0.81	71-230

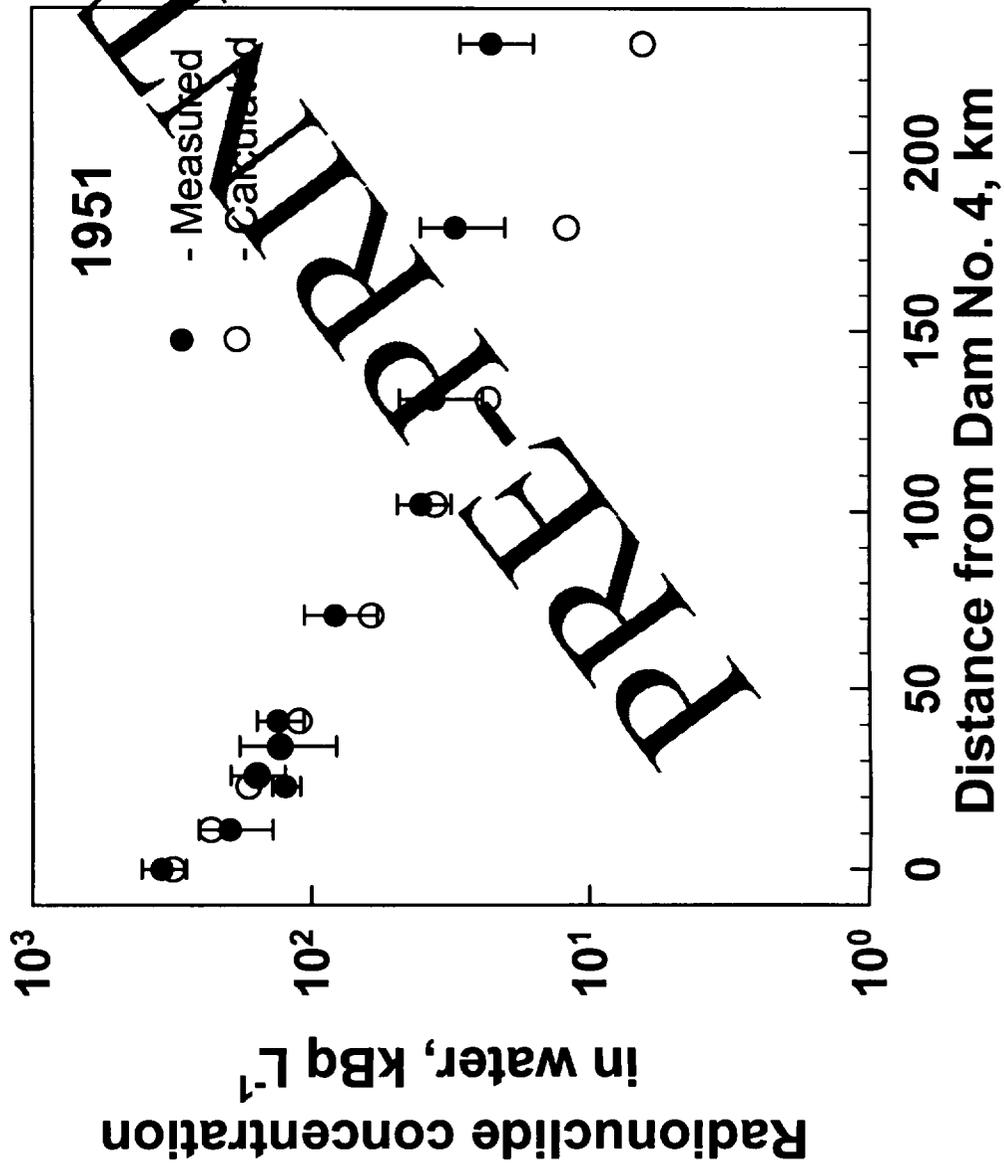
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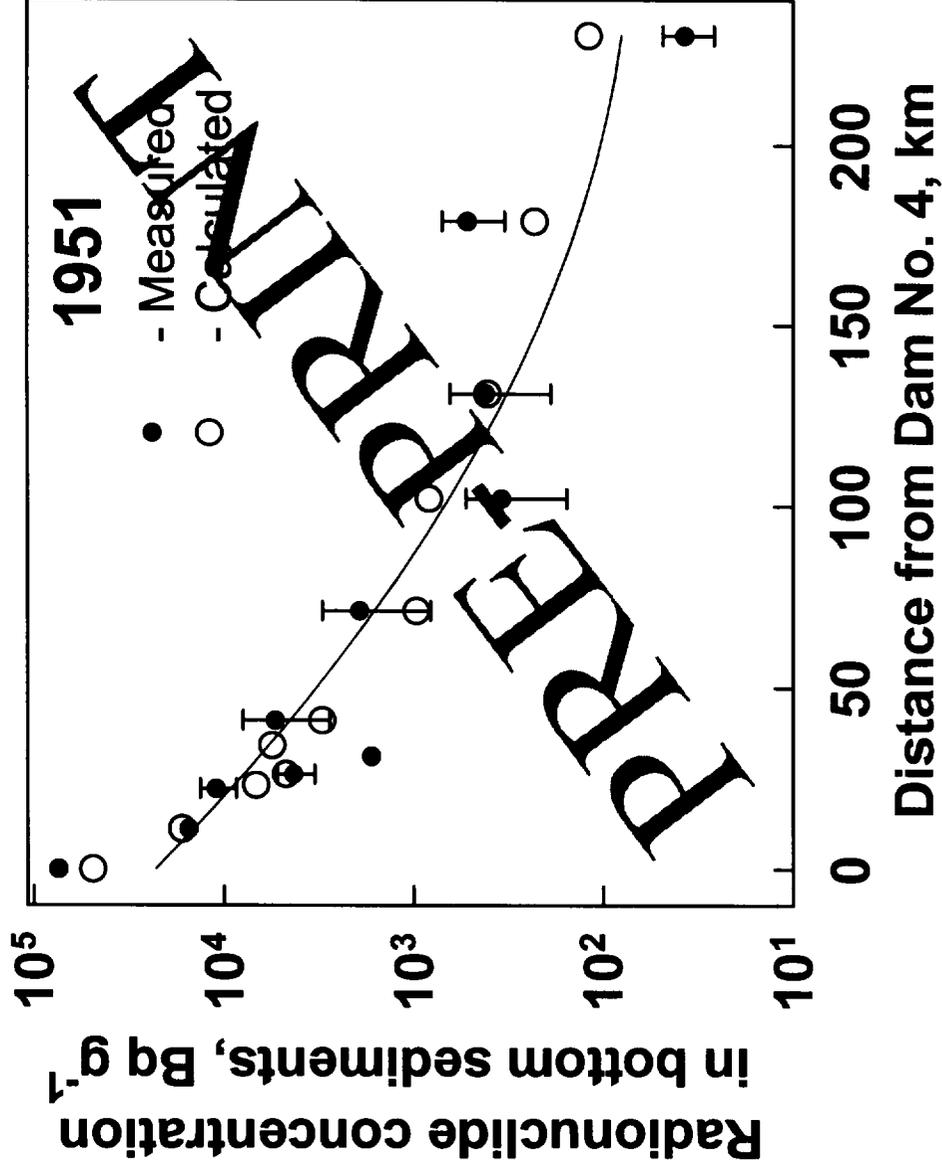
## FIGURE LEGENDS

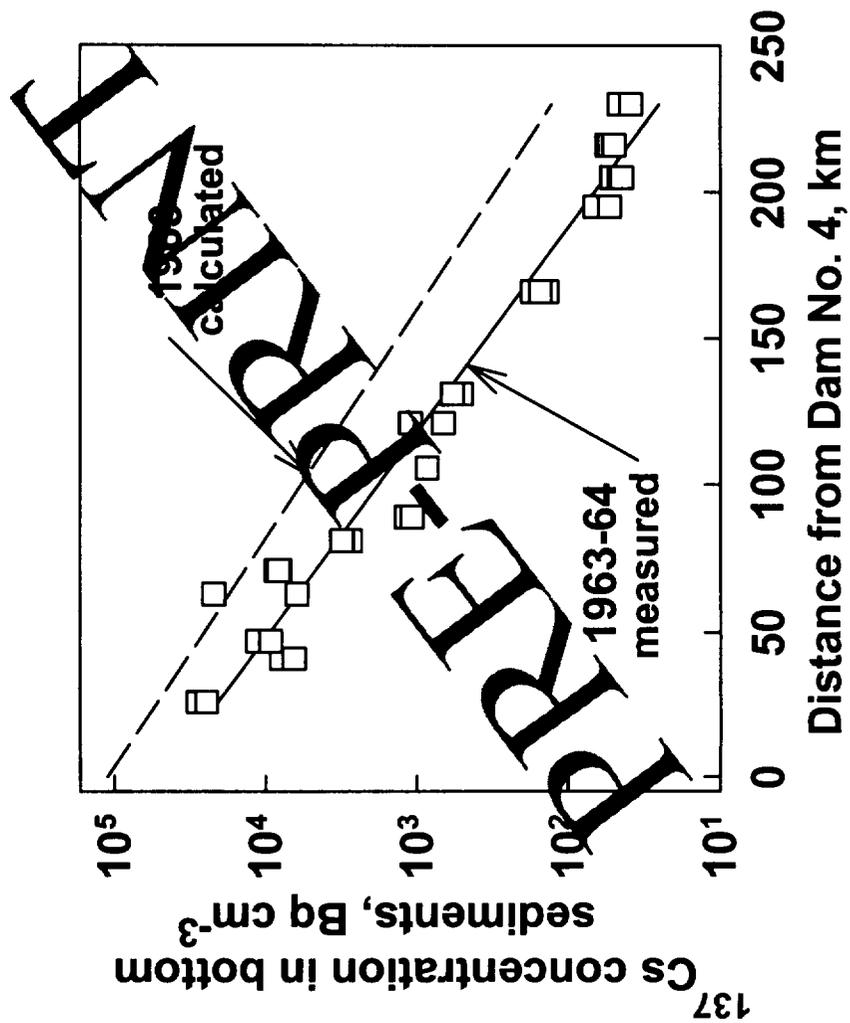
- Fig. 1. Schematic map of the Techa River (approximate scale) and its ponds. The points correspond to historical monitoring sites. Dashed lines indicate sites of routine sampling in 1951–1952.
- Fig. 2. Concentrations of different radionuclides in water as a function of downstream distance obtained in two series of experiments with flowing reservoirs (circles correspond to the first series; squares correspond to the second series).
- Fig. 3. Measured and modeled radionuclide concentrations of Techa River water in 1951 as a function of downstream distance.
- Fig. 4. Measured and modeled radionuclide concentrations of Techa River bottom sediments in 1951 as a function of downstream distance.
- Fig. 5. Measured and modeled  $^{137}\text{Cs}$  concentrations of the Techa River bottom sediments in 1963–1964 as a function of downstream distance.
- Fig. 6. Modeled radionuclide concentrations of the Techa River water in 1949, 1950, and 1951 as a function of downstream distance.
- Fig. 7. Modeled concentrations of different radionuclides relative to  $^{90}\text{Sr}$  in the Techa River water in 1950–1951 as a function of downstream distance.
- Fig. 8. Inferred dose rate in air in (a) 1950, (b) 1951 and (c) 1952 as a function of downstream distance. Triangles correspond to DRA values (black = measurements; grey = Monte-Carlo simulations; open = calculation from empirical curve). Squares correspond to beta-activity of bottom sediments (black = measurements; open = model calculations).

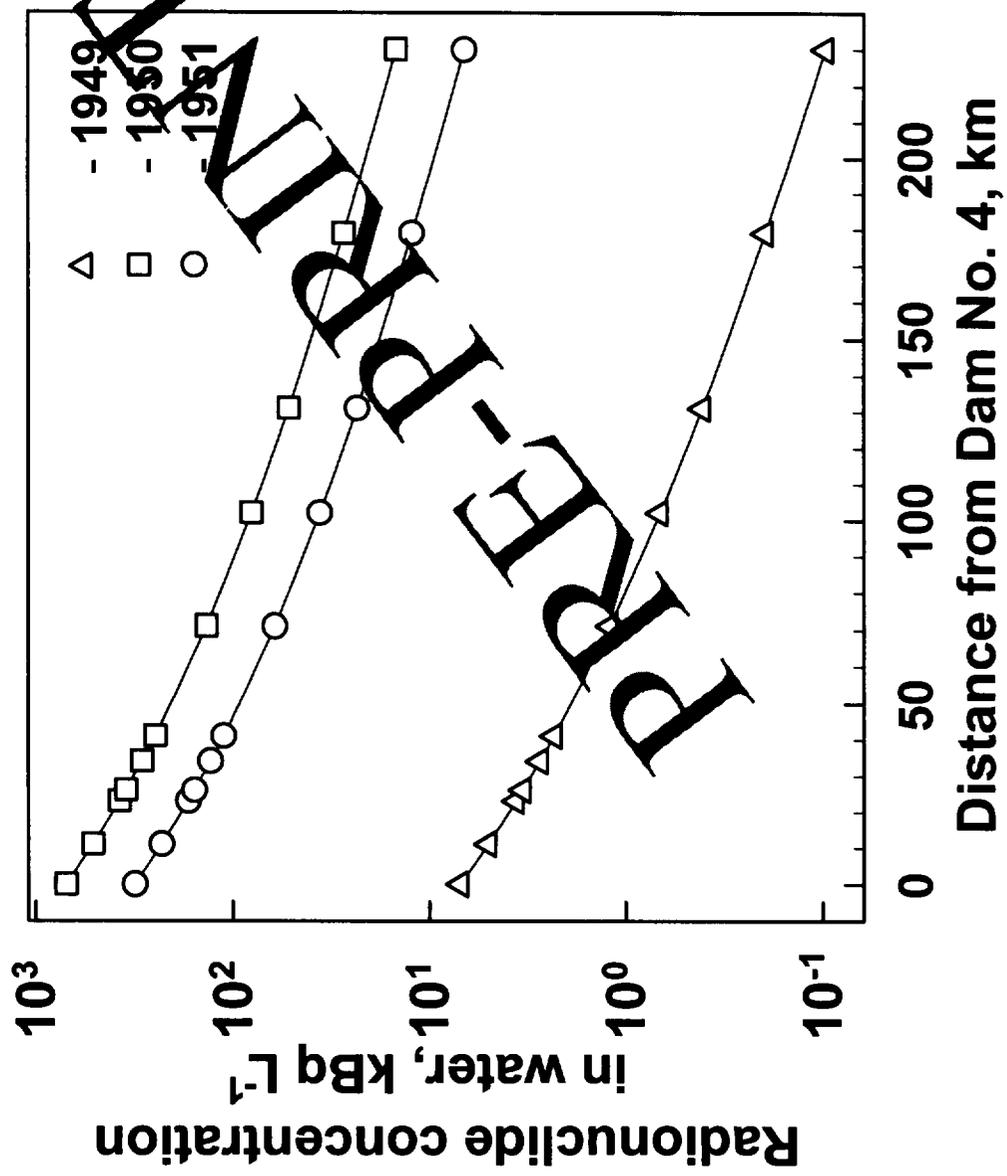


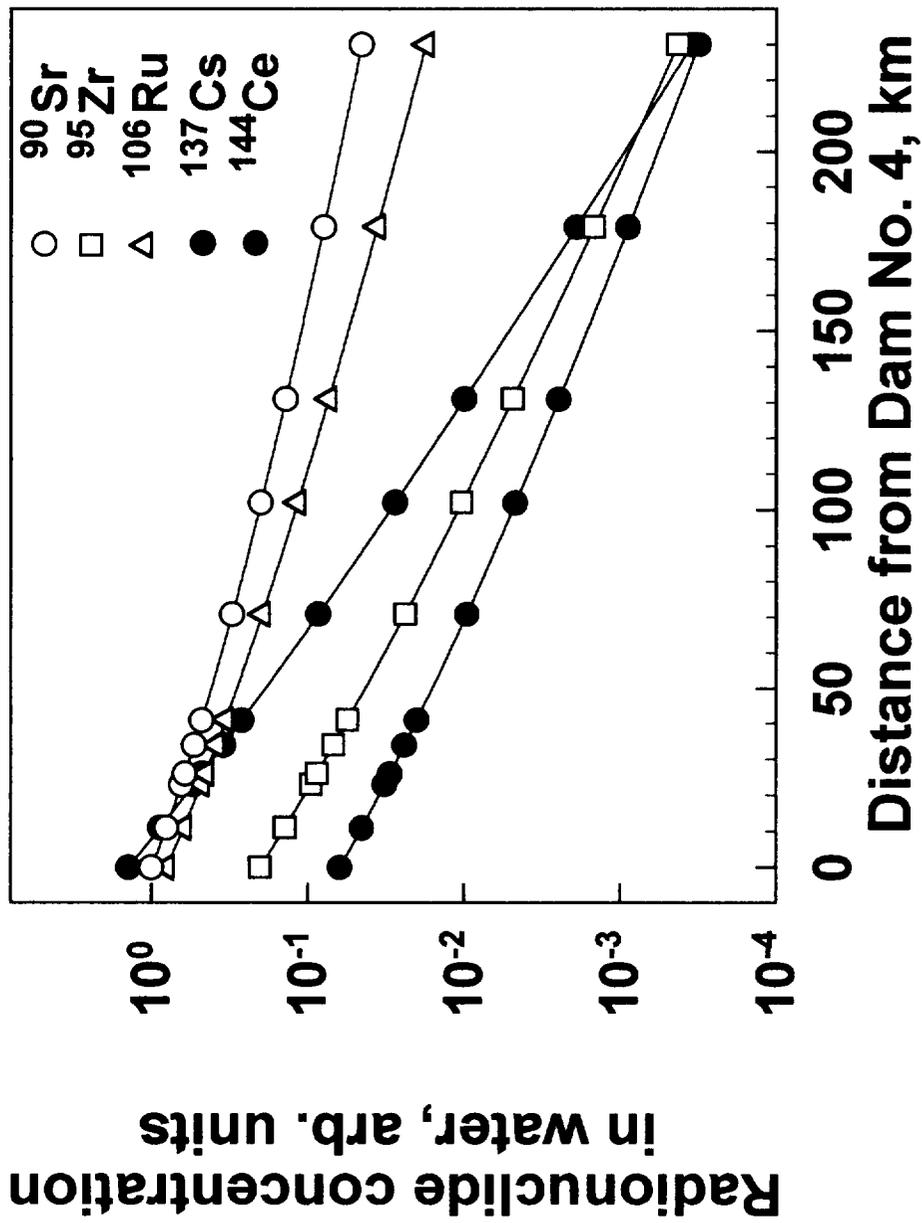


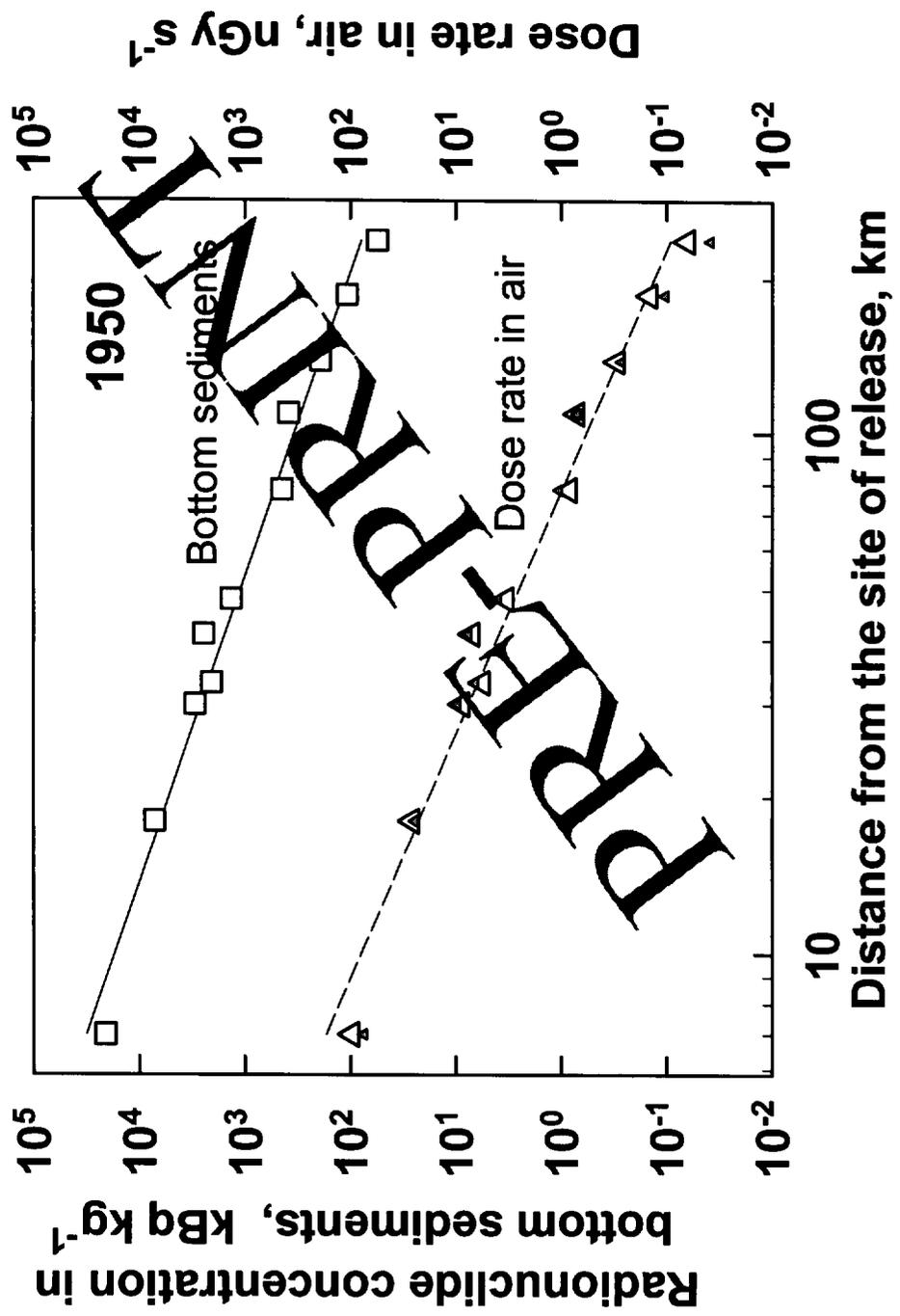


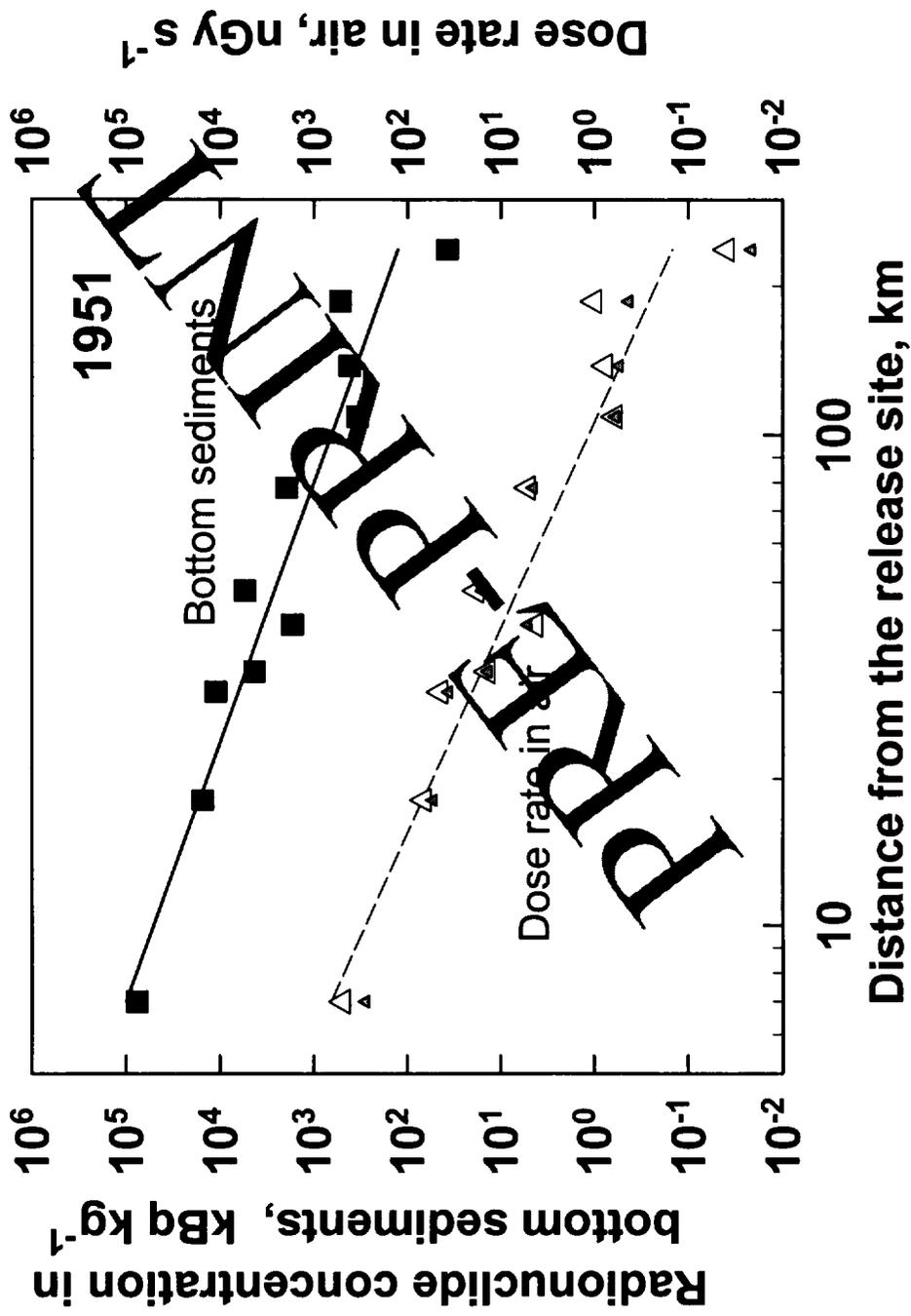


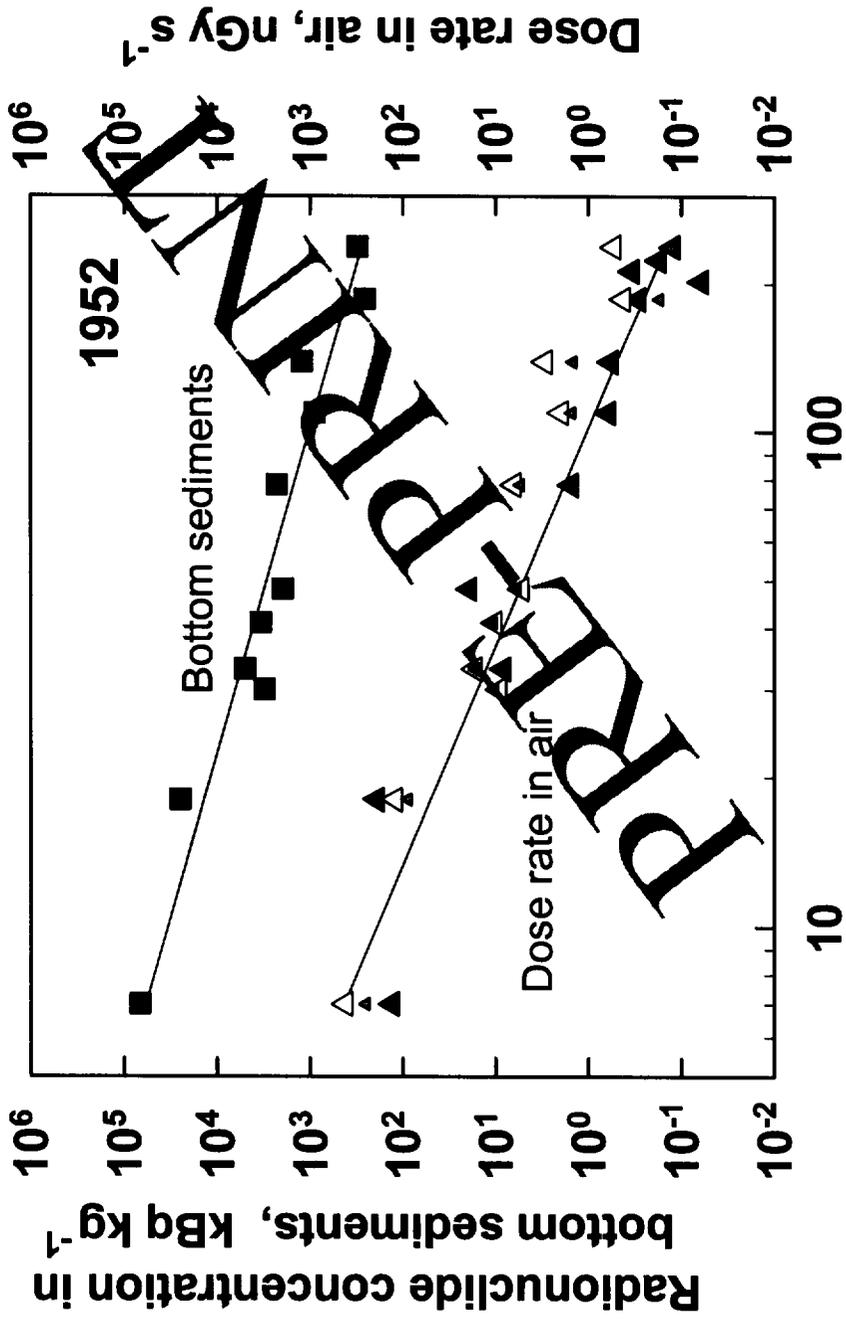






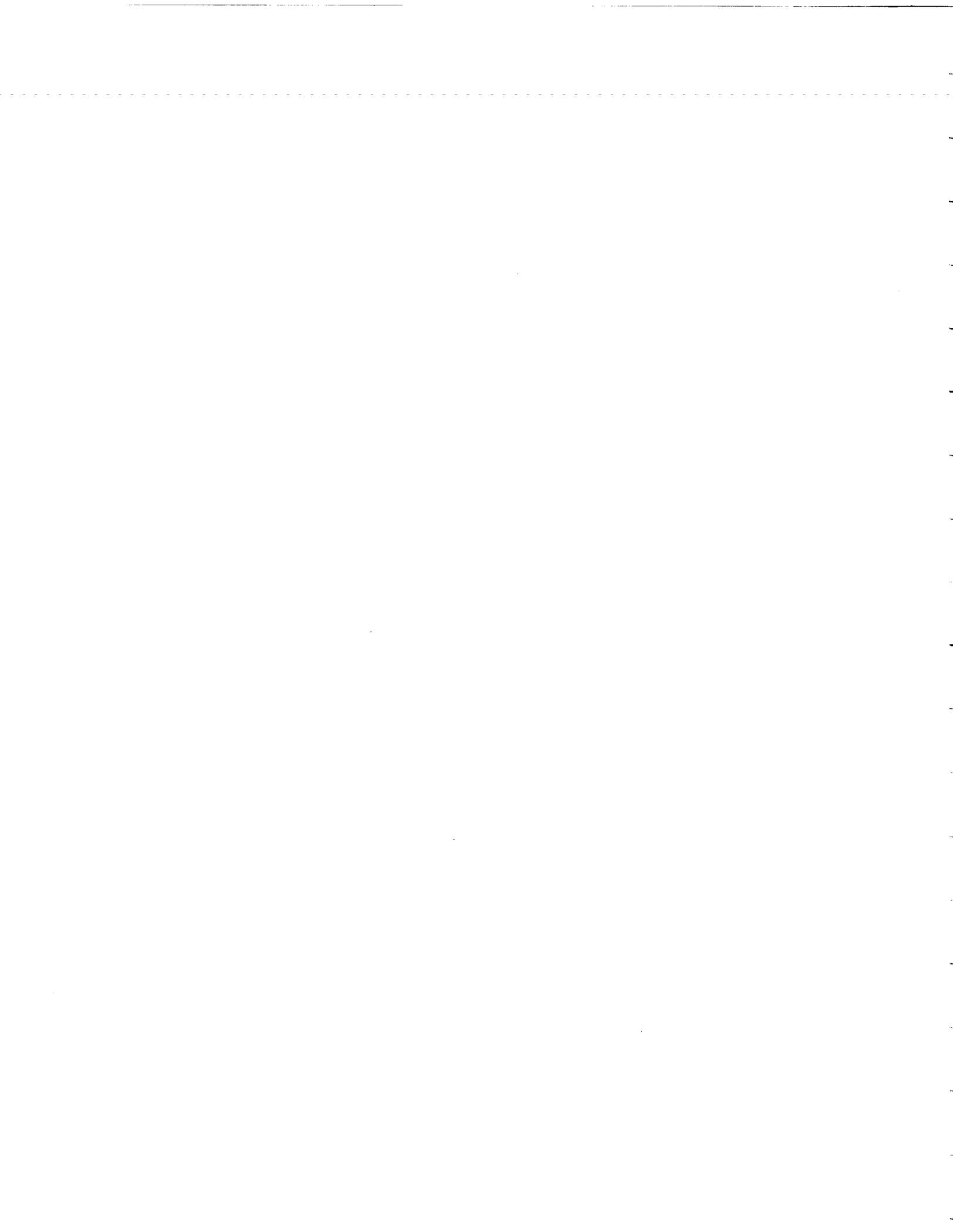






**APPENDIX 3**

**Letter to Scott Miller from Frank Hawkins, July 15, 1998**





**Department of Energy**  
Germantown, MD 20874-1290

JUL 15 1998

Scott Miller, Ph.D.  
Director  
Radiobiology Division  
University of Utah  
Building 586  
Salt Lake City, Utah 84112

Dear Dr. Miller:

I am sending this letter to provide guidance for the continuation application you will be submitting for the FY 99 budget period for cooperative agreement DE-FC03-97SF21354. This letter is also a follow-up to the call we held today regarding Task 3 of the cooperative agreement.

An updated statement of work and associated budgets should be provided for the continuation of Tasks 4 and 5.

Funding for Task 3 of the cooperative agreement is being discontinued as of September 30, 1998. In order to conclude the activities under task 3, we ask that the University submit a final report to the Department of Energy, Office of International Health Programs by September 30, 1998, detailing the number of electron paramagnetic resonance and thermo-luminescence samples collected and measured and findings to date. We also ask that all tooth, brick, and tile samples collected from the Techa River area be returned to Marina Degteva at the Urals Research Center for Radiation Medicine by September 30, 1998.

Please feel free to contact me on (301) 903-2476 with any questions.

Sincerely,

Frank Hawkins  
Director  
Office of International  
Health Programs

cc: Lynn Anspaugh, Univ. of Utah  
Ed Haskell, Univ. of Utah  
Amy Hofheins-Sikalas, Univ. of Utah

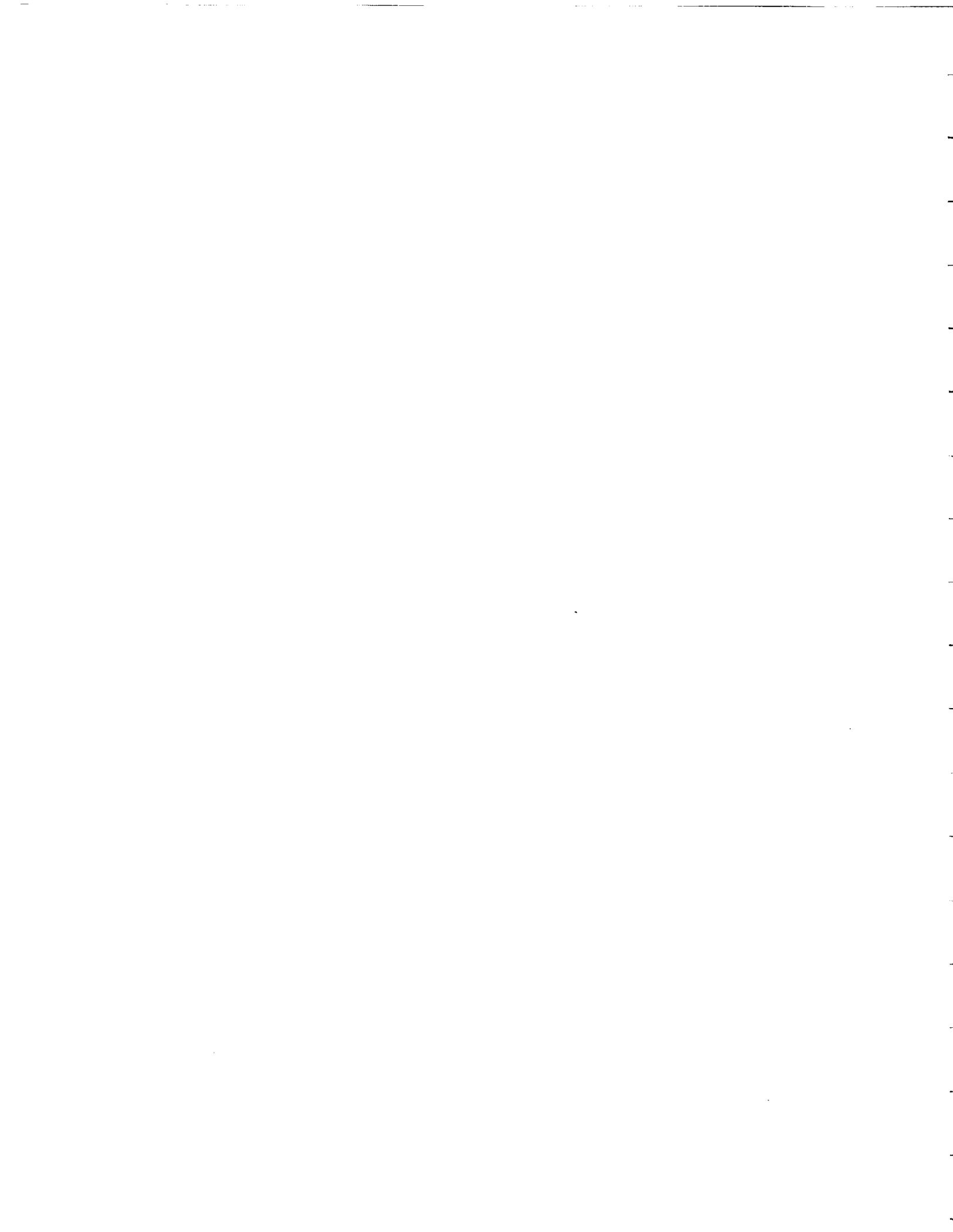




## APPENDIX 4

Pre-print

**Bougrov, N. G.; Göksu, H. Y.; Haskell, E.; Degteva, M. O.; Meckbach, R.; Jacob, P. Issues in the reconstruction of environmental doses on the basis of thermoluminescence measurements in the Techa Riverside. *Health Phys.* (accepted, 1998)**



**ISSUES IN THE RECONSTRUCTION OF ENVIRONMENTAL DOSES ON THE BASIS OF  
THERMOLUMINESCENCE MEASUREMENTS IN THE TECHA RIVERSIDE**

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*Abstract* - The potential of thermoluminescence (TL) measurements of bricks from the contaminated area of the Techa river valley, Southern Urals, Russia, for reconstructing external exposures of affected population groups has been studied. Dating of background samples was used to evaluate the age of old buildings available on the river banks. The anthropogenic gamma dose accrued in exposed samples is determined by subtracting the natural radiation background dose for the corresponding age from the accumulated dose measured by TL. For a site in the upper Techa river region, where the levels of external exposures were extremely high, the depth-dose distribution in bricks and the dependence of accidental dose on the height of the sampling position were determined. For the same site, Monte Carlo simulations of radiation transport were performed for different source configurations corresponding to the situation before and after the construction of a reservoir on the river and evacuation of the population in 1956. A comparison of the results provides understanding features of the measured depth-dose distributions and height dependencies in terms of the source configurations and shows that bricks from the higher sampling positions are likely to have accrued a larger fraction of anthropogenic dose from the time before the construction of the reservoir. The applicability of the TL dosimetry method to

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environmental dose reconstruction in the middle Techa region, where the external exposure was relatively low, was also investigated.

Key Words: thermoluminescence; dosimetry; TL dating; radiation accident; radiation exposure; dose reconstruction.

## INTRODUCTION

The Techa river and its adjacent territories (Southern Urals, Russia) were contaminated as a result of the releases of radioactive wastes by the Mayak plutonium facility from 1949 through 1956. The residents of the Techa riverside settlements who lived in the period of massive releases were exposed to both external and internal radiation. Long term epidemiological studies of the exposed population suggest that the risks of mortality from leukemia and other cancers increase with increasing radiation dose (Kossovskiy et al. 1997). The Techa River Dosimetry System Project was established in the Ural Research Center for Radiation Medicine (URCRM) to perform individual dose assessments by taking into account all available data sets on human and environmental contamination, as well as new data to be obtained by electron paramagnetic resonance (EPR) and thermoluminescence (TL) methods (Degteva et al. 1996). The TL technique is planned to be used mainly for the validation of calculated doses at specific sites along the banks of the Techa river within several meters of the water. Such validation is very important because the main source of gamma radiation was the contaminated silt along the river banks (Degteva et al. 1994).

TL methods have been used earlier for assessments of external gamma doses in Hiroshima and Nagasaki (Maruyama et al. 1987; Haskell et al. 1987), in the areas of the Nevada Test site (Haskell et al. 1994), in the town of Pripjat and in the 30 km exclusion zone of the

Chernobyl event (Hütt et al. 1993; Stoneham et al. 1996). However, all these situations were significantly different from the Techa river contamination, and the study of the Ural samples could also be of importance for the further development of TL as a retrospective dosimetry method.

A pilot TL study of brick samples from the village of Metlino, located at 7 km from the site of radioactive release, has demonstrated the applicability of this technique for dose reconstruction in the upper Techa river area, where external dose rates near the river were extremely high (Bougrov et al. 1995). Further, Monte Carlo simulations of radiation transport can be used to relate source distributions of gamma radiation to depth-dose profiles in brick walls (Meckbach et al. 1996), as well as to the doses accrued in bricks at different sampling heights. For a brick sample from Metlino, the comparison of a depth dose profile determined by TL measurements with the results of Monte Carlo simulations for an assumed source distribution (Göksu et al. 1996) showed the potential of a combination of TL measurements with Monte Carlo simulations for obtaining information about the past distribution of radiation sources. These studies also help to formulate the issues that should be solved in the further investigations of the Techa valley.

The first issue is the evaluation of the accumulated doses due to the natural radiation in the samples. This task is important because the majority of the houses of the inhabitants of the upper Techa were demolished after the evacuation of the population in 1956, and only several massive old brick buildings, constructed a long time before the accident, are available now. Therefore, the natural radiation dose accumulated in the bricks is relatively high and special attention must be taken to evaluate carefully this contribution to the total TL dose.

The second issue is related to the changes in radiation source configuration which occurred at the Metlino sampling site as a result of the construction of an artificial reservoir in the upper Techa (the so called reservoir No. 10). This reservoir was created in 1956 after the evacuation of the inhabitants of Metlino. In order to obtain information for the reconstruction of external doses of the population from the results of TL measurements, it is necessary to find ways to determine the component of dose accrued in bricks before 1956 due to the radionuclides released by Mayak, separating it from the component accumulated after the reservoir was built. The source distribution was different during these two periods. The dependence of accumulated doses on the height of the sampling position and depth of the distributions can be determined by TL measurements. After subtraction of doses due to natural radiation, a comparison with Monte Carlo simulations could give indications on the contribution of different source configurations to doses accrued in the bricks.

All former preliminary TL studies have been carried out only for the Metlino village in the upper Techa region, at a short distance from the site of radioactive release. A further issue is to check the applicability of TL methods for the lower parts of the Techa where external dose rates were relatively low (Degteva et al. 1994). The population of this region has received substantial doses to bone tissues due to ingestion of  $^{90}\text{Sr}$  with river water, but the evaluation of external doses for these people is also important in order to estimate the risk of solid tumors.

## MATERIALS AND METHODS

### Method of dose assessment

Some minerals show thermoluminescence after having been exposed to ionizing radiation, *i.e.* they emit light during heating. When a recently fired material is exposed to an

anthropogenic ionizing radiation field it acquires an excess dose over that which is due to natural radiation sources. The total dose accumulated in a building brick can be assessed using minerals like quartz and feldspar incorporated in it. The external gamma-dose component of the anthropogenic dose  $D_{Ant}$  can be estimated by use of the following equation:

$$D_{ant} = D_{TL} - D_{Nat}$$

$$D_{nat} = A \times (R_{\alpha} + R_{\beta} + R_{\gamma} + C) \quad (1)$$

where:  $D_{TL}$  = total accumulated dose as measured by TL (mGy);  $D_{nat}$  = total accumulated dose due to natural radiation;  $A$  = age of the building in years;  $R_{\alpha}$  = contribution of the  $\alpha$ -radiation of the uranium and thorium in the brick to the dose rate. This value includes a correction factor for TL efficiency of  $\alpha$ -particles and is termed the *internal effective alpha-particle dose rate* (mGy  $y^{-1}$ );  $R_{\beta}$  = internal beta-particle dose rate due to the uranium, thorium and potassium content of the brick (mGy  $y^{-1}$ );  $R_{\gamma}$  = dose rate due to  $\gamma$ -radiation of uranium, thorium and potassium in the brick and in the environment (mGy  $y^{-1}$ );  $C$  = dose rate due to cosmic rays (mGy  $y^{-1}$ ).

### Description of the sampling sites

The samples were collected at two sites at the Techa river: 1) At the upper Techa, in the former village of Metlino, located at 7 km from the site of release; and 2) at the middle Techa, in the village of Muslyumovo, at a distance of 78 km from the site of release. Bricks from three buildings located on the banks of the river were investigated, the mill of Metlino and the mill and the waterworks of Muslyumovo. For each building, two kinds of samples were collected, exposed samples taken from the outside walls facing the river, and background samples taken from the inner walls of the buildings or from walls opposite to the river. The background samples

were used for an assessment of the age of the sampling site. The exposed samples were used for the reconstruction of the anthropogenic dose. A description of the samples is given in Table 1.

The Metlino mill site is shown in Fig. 1, where the sampling positions on the brick wall facing the reservoir No. 10 are indicated. The samples No. 26 and 33 were taken from the wall at heights of respectively 2 m and 4 m above the water surface and at a distance of about 4 m from the shore of the reservoir; and samples No. 32 and 34 were taken at a height of 6 m, at distances of respectively 2 m and 4 m from the shore. Also indicated in Fig. 1 is the position of sample No. 16, investigated in a previous publication (Göksu et al. 1996) and of samples No. 23, 25 and 27, investigated in (Bougrov et al. 1995). The background level sample No. 31 was taken from the middle of a partially crushed 1 m thick inner wall of the Metlino mill.

Fig. 2 shows the Metlino site in a schematic view from above (A) and a vertical cross section (B); the sampling positions on the brick wall are indicated. Before the construction of reservoir No. 10 in 1956, the Techa river passed from Metlinsky pond through a lock (partially visible at the left side of Fig. 1) and flowed at a distance of about 10 m from the wall from which samples were taken. At that time, presumably the main sources of radiation were the contaminated sediments in the river bed and its contaminated shores. After 1956, the construction of reservoir No. 10 raised the water level by about 1 m, the river flowed directly through the lock into the reservoir, and the mill stood partially in the water. Presently, the reservoir has a depth of about 1 m in the vicinity of the buildings. Close to the wall of the mill there is a narrow fringe of shallow water with a depth between 20 cm and 50 cm. Gamma dose rate measurements were performed in 1997 above the water at a distance of about 3 m from the shore at different distances from the wall, as indicated in Fig. 2 (B). At 1 m distance from the wall, a dose rate of about 5  $\mu\text{Gy/h}$  was measured; at 3 m distance the dose rate was only about

0.6  $\mu\text{Gy/h}$ . Farther away, the dose rates were at a level of about 0.4  $\mu\text{Gy/h}$ , an order of magnitude less than near the wall. On the shore of the reservoir next to the wall the contemporary dose rates range between 9  $\mu\text{Gy/h}$  close to the wall and about 3  $\mu\text{Gy/h}$  near the lock. The dose rate measurements indicate that the contaminated shore and the ground of the reservoir near the wall, where the water is more shallow, could have given the dominant contribution to the anthropogenic doses accrued in the bricks since 1956.

At the Muslyumovo mill, exposed samples No. 1 and 3 were taken from the outside wall facing the river bank (Fig. 3). Background sample No. 6 was collected at about 1 m from the outer surface of a 1.5 m thick external wall opposite to the river. At the Muslyumovo waterworks, exposed sample No. 7 was taken from the outside wall facing the river (Fig. 4), at a height of 1 m above ground level and about 5 m above the level of the Techa river. Background sample No. 9 was extracted from the middle of the crushed round wall of the waterworks directed away from the river (Fig. 4).

#### **Sample preparation for TL measurements**

Two methods were used for sample preparation. Samples No 9, 32, 33 and 34 were prepared using the TL quartz inclusion technique (Zimmerman 1971; Haskell et al. 1987) in the Center for Applied Dosimetry, University of Utah under controlled lighting conditions. The first 3 - 5 mm of the outer surfaces of each brick sample was removed using a water-cooled diamond saw, and then the samples were prepared by crushing in a hydraulic press. Particles in the size range of 106-150  $\mu\text{m}$  were washed in concentrated HCl at temperature 30°C for one hour in an ultrasonic bath, and then washed in distilled water. Then the grains were etched in 49% HF for a period of 30 minutes in order to remove alpha-irradiated regions of the grains, washed in distilled

water, rinsed acetone, and dried in an oven at 70°C for several hours. If precipitate was observed during microscope exam the sample was additionally treated in HCl for 60 min. The dry crystals were separated from iron-containing particles with a magnetic separator. The non-magnetic portion of the grains was used for the measurements.

Samples No. 1, 3, 6, 7, 26 and 31 were prepared using fine-grain, additive dose or pre-dose techniques (Aitken 1985) in GSF - Institut für Strahlenschutz. The fine grain samples were prepared under laboratory red light using the Lee filter No. 106 (primary red). The outer 3 mm from all surfaces of the fragment of the brick were removed with a water-cooled diamond saw. Brick No. 26 was cut into 13 segments (1 cm thick) for the additive dose distribution.

#### **TL measurements**

At GSF the TL glow curves were measured using an automatic reader (TL-DA12, RISØ) with heating rate of 5°C s<sup>-1</sup> in nitrogen flow of 1 min<sup>-1</sup>. The heat-absorbing filter HA-3 was used together with Blue (Corning) BG-39 or Hoya U-340 and at the low doses Corning BG-12. TL dose evaluation was made using the additive-dose method. Additive doses were given using a <sup>90</sup>Sr-<sup>90</sup>Y beta ray source which was calibrated with respect to <sup>60</sup>Co gamma ray source at the Secondary Standard Dosimetry Laboratory in GSF. The procedures of calibration are described elsewhere (Göksu et al. 1995).

The samples were stabilized at 100°C for 100s after irradiation and before TL measurements. The short term stability of the signal was tested with the "plateau-test" (Aitken 1985) as shown in Fig. 5 for exposed sample (A) and background sample (B). The procedures of determining the accumulated dose are described elsewhere (Göksu et al. 1996).

TL measurement at the University of Utah were carried out on a Daybreak/Utah 100 TL reader (Custom-manufactured to University of Utah specifications by Daybreak Nuclear and Medical Systems, Inc., 50 Denison Drive, Guilford, CT 06437) equipped with a 9635QA photomultiplier tube and a 40 mCi  $^{90}\text{Sr}$ - $^{90}\text{Y}$  beta source (Isotope Products Laboratories, 1800 North Keystone St., Burbank California, 91504). TL emission was filtered with a 4-69 filter and a 7-59 filter (Corning glasses, available from Kopp Glass Inc., 2108 Palmer Street, Pittsburgh, PA 15218)

#### **Assessment of dose rate due to natural radionuclides**

*Internal effective alpha particle dose rate ( $R_\alpha$ )*. The uranium and thorium content of the bricks was measured using a 4.5 cm diameter ZnS screen with the thick-sample alpha counting method, calibrated by using the US Geological Standard (BCR-1). The internal effective alpha particle dose rate was calculated using the  $L$  value system developed by Bowman and Huntley (1984), which takes into account the efficiency of the alpha-particles independent of their energy for producing TL. The irradiation was performed with 6 plaque sources with a nominal  $^{241}\text{Am}$  activity of 6.66 GBq each and calibrated individually in vacuum.

*Internal beta-particle dose rate ( $R_\beta$ )*. The Berthold LB770  $\beta$ -counter was used to measure the  $\beta$ -particle dose rate due to the uranium, thorium and potassium content of the bricks. The thick-source beta-counting method was used for measurements and assessment (Sanderson, 1988). Samples of known uranium, thorium and potassium content were used as standards. The potassium content of the samples was also measured by gamma-spectrometry using Canberra bore whole detector of pure germanium (relative efficiency 55%) calibrated with

CaCO<sub>3</sub> of the same geometry as a second control. Total beta-dose rates were once more calculated using uranium, thorium and potassium contents of the samples.

*Gamma dose rate (R<sub>γ</sub>)*. The internal gamma dose rate was calculated using the uranium, thorium and potassium contents of the bricks, assuming an infinite brick media. This assumption is valid for bricks sampled from the inner parts of the 1 m thick interior walls. For brick samples taken from outside walls, however, the internal gamma dose rate is reduced due to the air-brick interface. On the other hand, for these samples there is a contribution to the gamma dose rate from gamma radiation originating from natural radionuclides in the environment, and one expects a partial compensation of both effects. These data require further validation. Gamma ray spectrometry measurements were carried out in Utah according to the method of Lloyd (1976) with the sealed sample placed between two 20cm (8inch) NaI detectors.

### **Monte Carlo simulations**

For the Metlino site, Monte Carlo simulations of photon transport from different source configurations to the brick sample positions were performed using the code SAM-CE (Lichtenstein et al. 1979). The code allows for the simulation of complex three-dimensional geometry through a combinatorial geometry technique. By assigning materials of given atomic composition and densities to definite regions in space, the environment under consideration can be defined. Spectral photon fluences are scored in appropriate detection regions by an expected track length scoring method. Doses in brick are computed from the scored spectral photon fluences using mass energy absorption coefficients for photon interactions in bricks.

It is assumed that radiation originating from <sup>137</sup>Cs has given the main contribution to the anthropogenic doses accrued in the brick samples, and correspondingly the simulations were

made for a source photon energy of 661.6 keV. In the simulation, the bricks were taken to have a density of  $1.8 \text{ g cm}^{-3}$ . Scoring regions were defined at the positions of the brick samples, extending into the brick wall to the same depth as the samples used for TL measurements. Also, spectral photon fluences were determined in scoring regions corresponding to the layers at different depths in the brick used for the experimental investigation of the dependence of dose on depth.

Separate Monte Carlo calculations were made for radiation originating from different sources, corresponding to the probable contamination patterns before and after the construction of reservoir No. 10 in the year 1956. For the time before 1956, it was assumed that the Techa river flowed at a distance of 10 m from the sampling wall and that the radiation originated from its contaminated shore and from the sediments of the river bed (see Fig. 2). Accordingly, two source regions were defined: two 1 m wide strips on both sides of the river with the radionuclides distributed homogeneously in the ground to a depth of 10 cm, and a 5 m wide strip with radionuclides distributed to a depth of 10 cm in the river bottom sediments, below an (effective) water level of 50 cm.

For the time after the construction of reservoir No. 10, several separate source regions were defined: (1) a strip with the radionuclides distributed to a depth of 10 cm in the ground, corresponding to the contaminated shore of the reservoir next to the wall, (2) a 1 m wide strip extending next to the wall below 30 cm of water, corresponding to the contaminated ground of the reservoir at the shallow water close to the wall (see Fig. 2) and (3) the rest of the ground of the reservoir, simulated with radionuclides distributed on the ground 60 cm below the water level.

It should be pointed out that for the conclusions to be derived in the present investigation from the results of the Monte Carlo simulations it is not necessary to make specific assumptions on the absolute or relative source strengths of the different source regions.

## RESULTS AND DISCUSSION

### Assessment of natural background dose rates

The content of natural radionuclides and the resulting internal alpha, beta and gamma natural background dose rates are presented in Table 2 for the samples analyzed using the fine-grain technique. For the samples prepared with the use of the quartz inclusion method, in which there is no alpha ray contribution to the measured TL dose, it is only necessary to include the internal beta and gamma dose rates, and the results are shown in Table 3. For both cases it is estimated that the internal dose rates are assessed with a 5% error. The natural background dose rates were calculated according to the Bell conversion tables (Bell 1979) and the revised data of Nambi and Aitken (1986).

For brick samples at positions at the air-brick interface there is a further uncertainty in the contribution of gamma radiation to the natural dose rate due to the difference of dose rate from photons originating from natural sources in the environment and the dose rate due to photons originating in the brick. It is estimated that this leads to a 10% error for the gamma component of the natural dose rate. The contribution of cosmic radiation to background dose rate is relatively small, and was taken to have a value of  $0.28 \text{ mGy y}^{-1}$  (Prescott and Stephan, 1982). The value may be modified by the shielding effect of the buildings, and an uncertainty of 40% was assumed.

### TL dating of background samples

The total accumulated absorbed dose measured using TL in the so-called background bricks is assumed to be due only to natural exposure. This assumption is valid for the samples collected from the mills at Metlino and Muslyumovo, where the walls at an height of 1 meter are about 1-2 m thick and the samples were taken from the interior of the walls. Therefore, the background samples were heavily shielded from external exposures. Possible internal contamination of the mills were not taken into account in this analysis. As a result, the ages of samples were determined using equation (1) where ( $D_{Nat}$ ) is measured by TL and the natural dose rates are calculated as shown in Tables 2 and 3. As can be seen in Table 4, the TL age of the Metlino Mill is found to be  $132 \pm 17$  years with TL additive dose technique and  $125 \pm 14$  years using TL pre-dose technique, where the alpha dose rate is ignored due to low efficiency of alpha particles producing pre-dose effect. These results are in good agreement with historical data of the buildings. The first description of the mill in Metlino was found in the book by Choupin published in 1873 (Choupin 1873), which means that the age of this mill at the time of measurement could not be less than 123 years.

For the Muslyumovo Mill a similar evaluation yielded an age of  $105 \pm 10$  years. This is also found to be consistent with the historical data. The first mention of the mill in Muslyumovo was found in (Vershova 1899), which indicates that the age is more than 97 years. For the water works tower building the quartz extraction method yielded an age of  $55 \pm 10$  years. No written document has yet been found about the age of the building but, according to residents of this village, the waterworks in Muslyumovo was built before 1940, which indicates an age of more than 57 years.

The ages obtained in the background samples are used to assess the total natural background dose in the respective exposed samples.

### **TL dose assessment of exposed samples**

The results of TL measurements and of anthropogenic doses obtained by subtracting the dose due to natural radiation are given in Table 5. It can be seen that the anthropogenic doses for the samples from Metlino are one to almost two orders of magnitude larger than for the Muslyumovo samples. A sharp decrease of anthropogenic dose levels with distance from the site of release was initially observed with dose rate measurements made along the river in the early 1950s (Degteva et al. 1994) and also later obtained in a pilot study of the Techa riverside population by tooth electron paramagnetic resonance dosimetry (Romanyukha et al. 1996). The present results of TL measurements confirm this dependence.

For the samples collected from Muslyumovo Mill (samples 1 and 3), the accumulated dose was measured by TL with about 10% accuracy using the fine grain additive dose technique. However, for this site the anthropogenic dose could not be resolved with an uncertainty of less than 80% due to the relatively high contribution of the natural radiation to the totally accumulated dose. A smaller uncertainty could eventually be obtained using the quartz inclusion method where about 1/3 of the annual dose due to the alpha dose rate would be eliminated. These measurements need to be repeated using quartz extraction to obtain better accuracy. The difference in doses between samples from the mill (No. 1 and 3) and sample from the waterworks (No. 7) can be explained by different distances from the river bank strip, which is the major source of radiation (7 m and 2 m respectively). This information demonstrates that TL methods could provide data to reconstruct the external doses to the population of the middle Techa.

## **Anthropogenic dose distributions at the Metlino site - comparison of measurements and simulations**

The village of Metlino was the most unfortunately situated, as it was the closest settlement to the site of radioactive release, and its approximately 1,200 inhabitants received considerable levels of external dose. In order to analyze the exposure situation at the Metlino sampling site, the results of TL measurements of the present study are supplemented with data obtained in our previous studies (Bougrov et al. 1995; Göksu et al. 1996) and combined with the results of Monte Carlo simulations.

For the wall of the Metlino mill facing reservoir No. 10 (see Figs. 1 and 2), the dependence of accrued anthropogenic dose on height above the water level as obtained by TL measurements is shown in Fig. 6. Several of the samples measured in the Radiochemistry Laboratory of Moscow State University (MSU) and published in (Bougrov et al. 1995) are included in the figure. For sample No. 26, an agreement within 10% was obtained for the results of measurements performed in two different laboratories. One observes in Fig. 6 that the bricks sampled at the closer distance of about 2 m from the shore of the reservoir have accrued substantially higher anthropogenic doses than the bricks sampled at a distance of about 4 m from the shore. This could be due to a larger contribution of radiation from the contaminated shore, which is not so strongly shielded by the water. The dependence of dose on height is somewhat steeper for the samples taken at the larger distance of about 4 m from the shore; the brick sampled at a height of 2 m has an anthropogenic dose almost a factor of two higher than the brick sampled at a height of 6 m.

It is now of interest to investigate whether the dependence of accrued anthropogenic doses on the height of the sampling position can be understood in terms of what is known about

the radiation source configurations before and after the building of reservoir No. 10, using the results of Monte Carlo simulations. For the samples taken at a distance of 4 m from the shore of the reservoir Table 6 gives the ratios of anthropogenic doses measured at heights of 6 m and 4 m relative to the dose measured at an height of 2 m in comparison with results for the corresponding ratios obtained by Monte Carlo simulations for different sources. The sources considered for the time before the construction of reservoir No. 10 (river sediments and shore of the river) lead to higher doses in the upper bricks than in the lower bricks. In order to reach the lower bricks, the radiation has to transverse effectively thicker layers of sediments and water, and is therefore more strongly attenuated. The sources corresponding to the situation after the construction of the reservoir either lead to higher doses in the lower bricks than in the upper bricks (reservoir close to the wall and shore of reservoir) or to approximately equal doses (rest of the reservoir). The dependence of accrued dose on height is most pronounced for radiation from the ground of the reservoir below the shallow water close to the wall, for which the dose at an height of 6 m is a factor of five lower than at 2 m height. Two factors determine this strong dependence on height: first, the strip of shallow water along the wall is narrow, so that the intensity of radiation from this source decreases nearly inversely to the height; second, at the higher sampling position the photons effectively have to transverse thicker brick layers to irradiate the samples than at the lower sampling positions.

The results of the Monte Carlo simulations show that the source configuration before the construction of the reservoir is likely to lead to the opposite dependence of dose on height than the source configuration after the construction of the reservoir. Eventually, this feature could be used in combination with the measurement results to obtain information on doses accrued before and after the construction of the reservoir in 1956. The preliminary conclusion from comparing

measured and calculated dose-height dependencies is that the anthropogenic dose in the lower bricks has a large contribution from the time after the construction of reservoir No. 10. Furthermore, the contribution to the dose from the time before the construction of the reservoir is likely to be larger in the upper bricks than in the lower ones.

The depth-dose distribution of anthropogenic dose was determined for brick sample No. 26 by TL measurements of 13 thin layers up to a depth of approximately 14 cm and subtraction of the natural radiation background dose. Results of analogous measurements for sample No. 16 have been published before (Göksu et al. 1996); for this sample, the natural radiation background dose has been reassessed, obtaining for the sample age 2100 ± 117 years. Brick No. 26 has been sampled at a height of 2 m above the water level, sample No. 16 at a height of 1 m (see Fig. 1). The respective depth-dose distributions are shown in Fig. 7. One can see that in both cases the dependence of anthropogenic dose on depth is basically exponential beyond a depth of about 2 cm. However, the dependence is considerably steeper for sample No. 16; at a depth of 12 cm the decrease in dose is about 50% larger than for sample No. 26. The effect seems to be larger than what could be attributed to the error in the determination of the natural radiation dose background.

A qualitative understanding of this effect can be obtained by investigating the results of Monte Carlo simulations of the depth-dose distributions at the sample positions for different radiation source configurations. Also shown in Fig. 7 are depth-dose distributions calculated for sample No. 16 for contamination of the river shores and of the river sediments at 50 cm below the water level, corresponding to the likely source configuration before the construction of reservoir No. 10, and the depth-dose distribution resulting from sources on the ground of the reservoir below the shallow water close to the wall. One can see from Fig. 7 that the reservoir

source close to the wall gives rise to a very steep depth-dose profile. On the other hand, as discussed before, for radiation from this source there is a strong dependence of the accrued dose on the height of the sampling position (see Table 6). By Monte Carlo simulations, one finds that its contribution to sample No. 16, located at a height of only 1 m above the water level, is about twice as large as its contribution to sample No. 26, which is located at a height of 2 m. This could explain the steeper depth-dose profile measured for sample No. 16. Furthermore, one can see from Fig. 7 that the measured depth-dose profiles would be consistent with adequately weighted superpositions of the profiles obtained by Monte Carlo simulations for the different sources.

### CONCLUSIONS

The results of the first steps in the Joint Russian-German-US TL studies in the Tcha river region are the following. Ages determined for old buildings located on the banks of the river by TL dating of background samples are in good agreement with available historical documents. Feasibility has increased for the potential use of TL methods for environmental dose reconstruction in the middle Tcha region where external exposure was relatively low. For the Metlino site on the upper Tcha river, anthropogenic dose accrued in bricks versus sampling height as well as depth-dose distributions in bricks were determined by TL measurements. Several features of these distributions could be understood in terms of the past and present configurations of the radiation sources by comparisons with the results of Monte Carlo simulations. It remains to be investigated whether more refined simulations, combined with further data on depth-dose distributions and with measurements of contemporary dose rates would allow for a quantitative determination of doses accrued at the Metlino site before the building of reservoir No. 10 and the evacuation of the population in 1956. If so, it would then be

possible to reconstruct external doses in air at the river banks, and use that information which could then be used to reconstruct external doses to the population.

*Acknowledgments* – The work discussed in this paper is supported by European Commission under Contract No ERBIC-15CT960305 within INCO-COPERNICUS program; European Dose Reconstruction project No. FI P-CT 97-0011 under EU; and by US Department of Energy, US Environmental Protection Agency, and US National Aeronautical and Space Administration under Russian-US Joint Coordinating Committee for Radiation Effects Project 1.1 “Dose Reconstruction for the Ural Population.” The authors would like to thank V. Baturin, V. Zhebel, N. Safronova (URCRM, Chelyabinsk) for the assistance in sample collection; L. Heide (BfS, Munich) for the assistance in some of the sample preparation; E. Tolstykh, D. Burmistrov, M. Vorobiova (URCRM, Chelyabinsk) for the fruitful discussions.

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Table 1. Description of the samples.

Sample code	Location	Building	Position	Height m	Exposed or background
1	Muslyumovo	mill	outside wall	1	exposed
3	Muslyumovo	mill	outside wall	1	exposed
6	Muslyumovo	mill	inner wall	1	background
7	Muslyumovo	waterworks	outside wall	1.4	exposed
9	Muslyumovo	waterworks	partially destroyed wall	4.5	background
26	Metlino	mill	outside wall	2	exposed
31	Metlino	mill	inner wall	3	background
32	Metlino	mill	outside wall	6	exposed
33	Metlino	mill	outside wall	4	exposed
34	Metlino	mill	outside wall	6	exposed

Table 2. Natural uranium, thorium, and potassium content and respective components of background dose rate for the samples prepared using fine-grain technique.

Sample No.	Uranium ppm	Thorium ppm	Potassium (by weight) %	Alpha dose rate mGy y <sup>-1</sup>	Beta dose rate mGy y <sup>-1</sup>	Gamma dose rate mGy y <sup>-1</sup>
1	1.66	4.41	1.44	0.64	1.34	0.71
3	2.48	4.61	1.52	0.84	1.52	0.83
6	2.98	6.29	1.59	1.05	1.69	0.99
7	2.04	7.57	2.00	0.92	1.87	1.03
26	2.16	4.59	1.45	0.76	1.22	0.72
31	2.38	3.50	1.22	0.74	1.27	0.86

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Table 3. Internal components of background dose rate for the samples prepared with the use of quartz inclusion method.

Sample code	Beta dose rate mGy y <sup>-1</sup>	Gamma dose rate mGy y <sup>-1</sup>
9	1.31	0.80
32	1.08	0.58
33	1.89	0.50
34	1.31	0.80

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Table 4. The age of background samples for three investigated buildings.

Sample No	Building	Annual dose rate mGy y <sup>-1</sup>	Measured TL dose mGy	Calculated age years
6	Muslyumovo Mill	4.01±0.22	420±31	105±10
9 <sup>a</sup>	Water-works	2.39±0.18	132±20	55±10
31	Metlino Mill	3.15±0.19	417±25	132±17
31 <sup>a</sup>	Metlino Mill	2.41±0.16	313±26	125±14

<sup>a</sup> Alpha dose rates are not included due to techniques used in these measurements

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Table 5. The results of TL investigations of exposed samples.

Sample No	Location	TL dose mGy	Age of building years	Background dose rate mGy y <sup>-1</sup>	Background dose mGy	Anthropog. dose mGy
1	Muslyumovo	380±40	105±10	2.97±0.20	312±36	68±54
3	Muslyumovo	416±32	105±10	3.46±0.22	364±42	52±53
7	Muslyumovo	519±28	55±10	4.10±0.25	246±44	273±52
26	Metlino	4070±160	129±17	2.98±0.20	383±57	3690±170
32	Metlino	2180±186	129±17	1.94±0.16	249±39	1960±190
33	Metlino	2920±164	129±17	3.13±0.21	402±60	2550±175
34	Metlino	3910±505	129±17	2.39±0.18	307±47	3630±507

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Table 6. Ratios of anthropogenic doses in bricks at sampling heights of 6 m and 4 m above the water level of reservoir No. 10 to the dose in brick at an height of 2 meters as obtained by TL measurements in comparison with the corresponding ratios obtained by Monte Carlo simulations for different sources (see text). The source photon energy is 662 keV.

Height of sampling	Ratio of doses in brick					
	Measured	Simulations:				
		river sediments	shore of river	reservoir close to wall	shore of reservoir	rest of reservoir
6 m	0.5	1.8	1.3	0.2	0.8	1.1
4 m	0.7	1.4	1.2	0.4	0.9	1.0
2 m	1.0	1.0	1.0	1.0	1.0	1.0



Fig. 1. The wall facing reservoir No. 10 of the mill in the village of Metlino on the Techa river. (7 km from the site of release). The numbers correspond to the codes of the investigated samples. Also indicated are samples No. 16, 23, 25 and 27 which have been described in Bougrov et al. (1995) and Cöksu et al. (1996). The positions of the samples extracted from the wall are visible near the numbers.

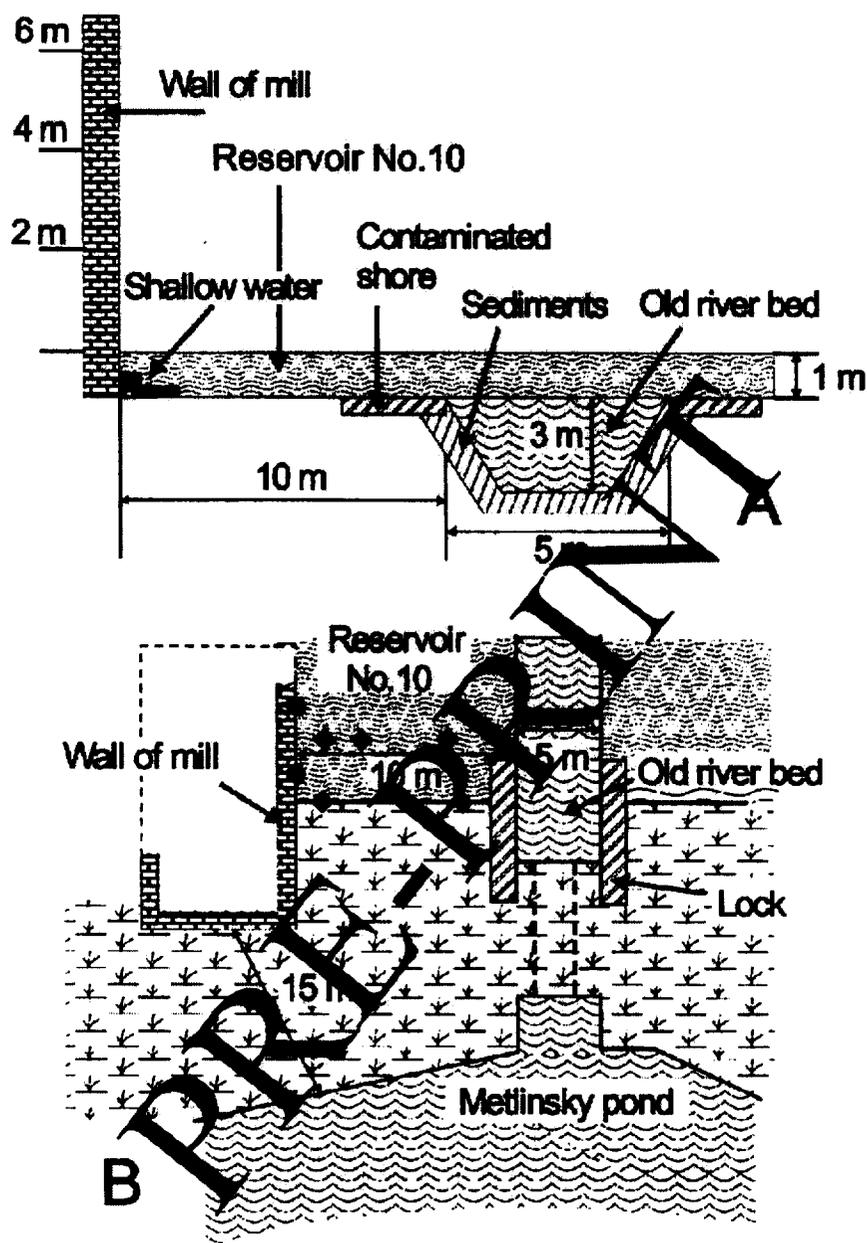


Fig. 2. Schematic view of the Metlino site, showing the position of the wall of the mill facing the reservoir (see Fig. 1) relative to the old Techa river bed: vertical cross section (A) and view from above (B). Also indicated in (B) are the sampling positions of bricks (full circles) and the locations where dose rate measurements were performed (full diamonds).

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Fig. 3. The mill in the village of Muslymovo on the Techa river (78 km from the site of release). The positions of exposed samples No. 1 and 3 are indicated.

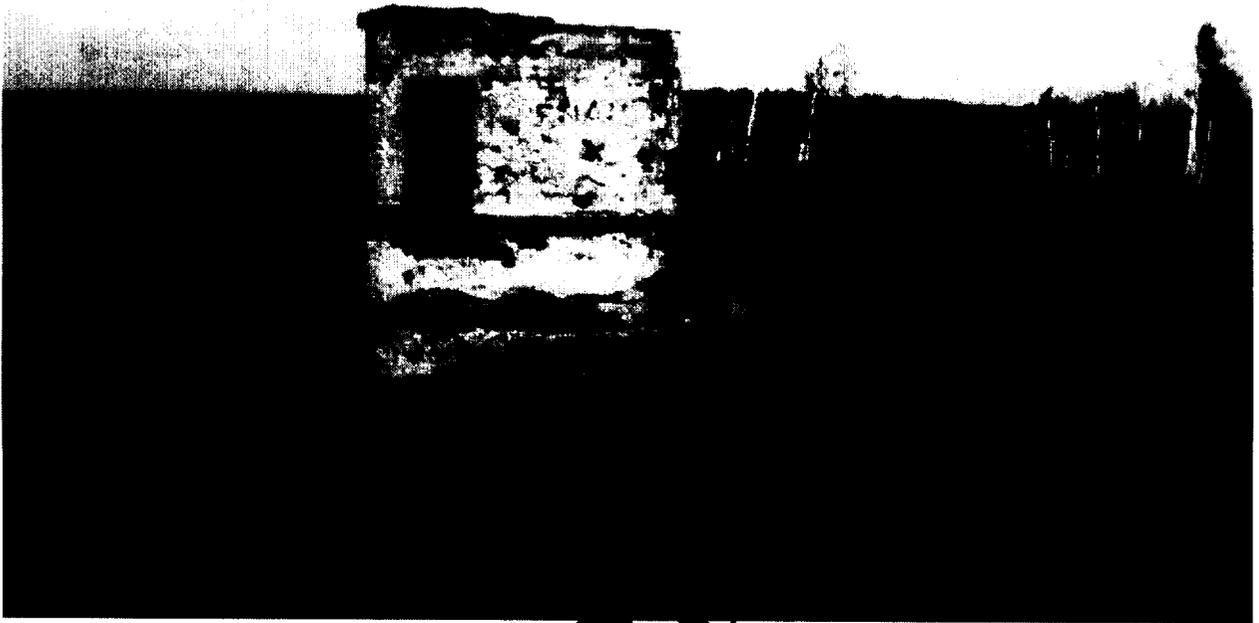


Fig. 4. The waterworks in the village of Muslyumovo on the Techa river (78 km from the site of release). The positions of exposed sample No. 7 and background sample No. 9 are indicated.

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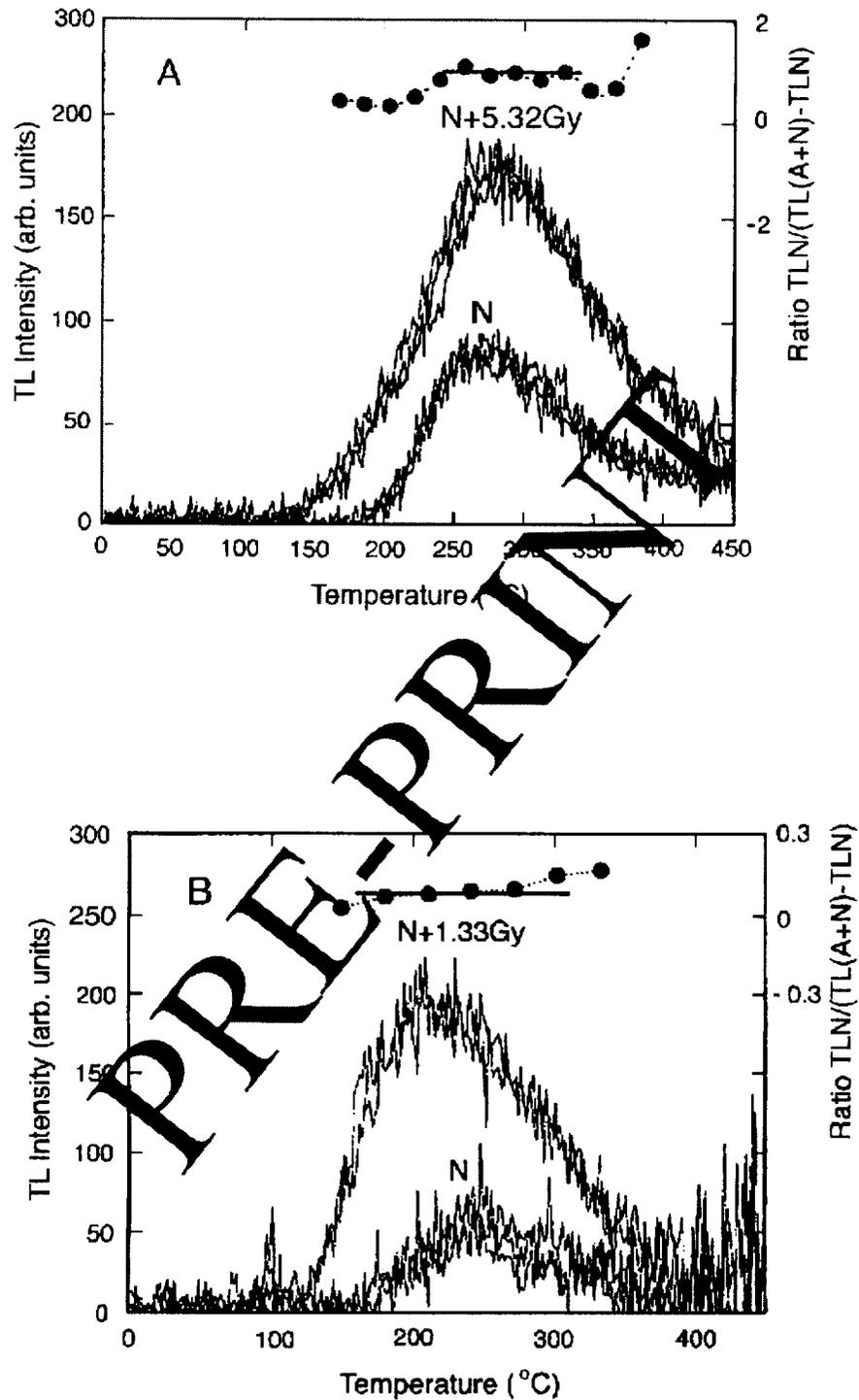


Fig. 5. TL glow curves and plateau tests for exposed (A) and background (B) samples.

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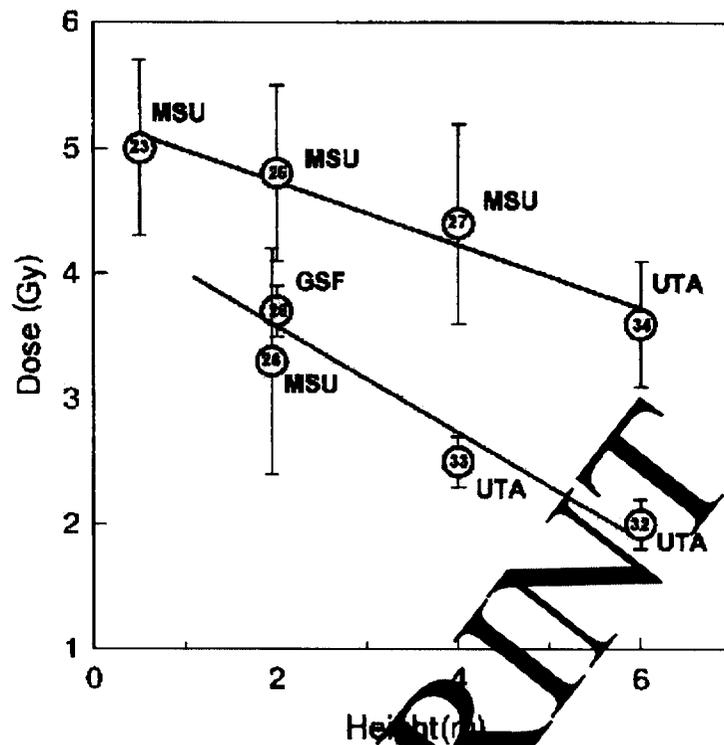


Fig. 6. Dependence of anthropogenic dose on the height of brick sampling positions for the wall of the Metlino mill (see Fig. 1). The numbers in circles correspond to the sample codes. The samples joined by the upper full line were taken at a distance of about 2 m from the shore of the reservoir; the samples joined by the lower line at a distance of about 4 m. The laboratories where the TL measurements were made are also indicated: Moscow State University (MSU); GSF-Institut für Strahlenschutz (GSF); University of Utah (UTA).

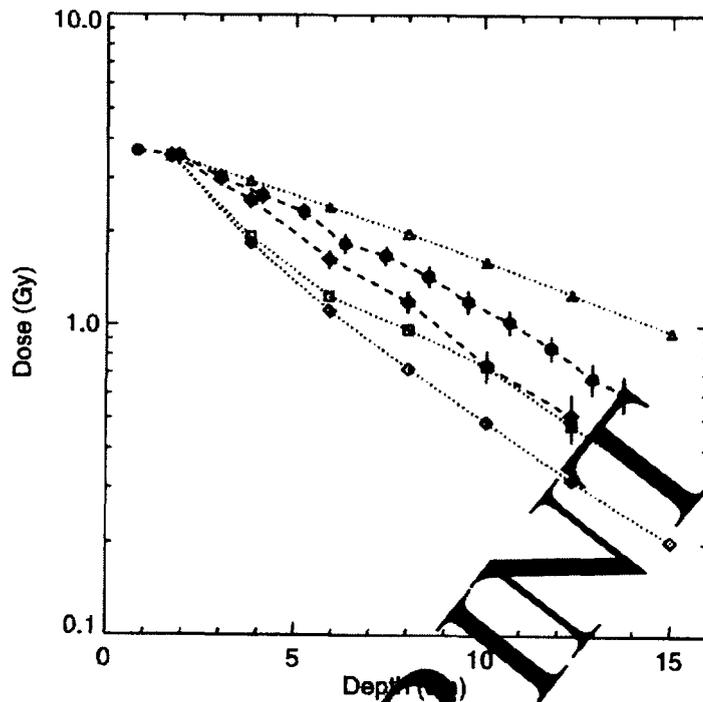


Fig. 7. Dependence of anthropogenic dose on depth as obtained by TL measurements for two bricks sampled from the wall of the Melino Mill (see Fig. 1): Sample No. 16 (full diamonds) and sample No. 26 (full circles). Also indicated are depth dose distributions obtained by Monte Carlo simulations for radiation from the contaminated shores of the old Techa river (open triangles), from the sediments of the old river bed (open squares) and from sources in the reservoir close to the wall (open diamonds). The simulated distributions are normalized to the value measured in the first layer of sample No. 16. The dashed and dotted lines are included to guide the eye.

**CALIBRATION OF WHOLE-BODY COUNTER SICH-9.1 FOR  
STRONTIUM-90, CESIUM-137 AND POTASSIUM-40  
USING SPECIAL PHANTOMS**

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**Final Report for Milestone 2**

**US-Russian Joint Coordinating Committee on Radiation Effects Research  
Project 1.1  
“Development of an Improved Dose Reconstruction System for the General  
Population Affected by the Operation of the Mayak Production Association”**

**October 1998**

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## 1. INTRODUCTION

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. This plant began operation in 1948, and during its early days there were technological failures that resulted in the release of large amounts of radioactive waste into the rather small Techa River. The residents of the riverside communities were exposed to chronic external and internal irradiation. Extensive monitoring efforts for the environment and the population at this site began in 1951. The "Techa River Cohort" (TRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM), and an increase in both leukemia and solid tumors with radiation dose has been noted (Kossenko et al. 1997). This finding suggests that, with continuing improvements in the quality of the follow-up and dosimetry, study of the TRC has the potential to provide quantitative estimates of the risks of stochastic health effects produced by chronic low-dose-rate radiation exposure in the general population. Study of this population affords an unique opportunity to address the question of the existence of a dose-rate-reduction factor for the induction of stochastic effects in an unselected general population. A definitive answer to this question would have relevance to the regulation of radiation exposure throughout the world.

The purpose of the US–Russia Joint Coordinating Committee on Radiation Effects Research (JCCRER) Project 1.1 is to define and implement a protocol for improvements in the dose-reconstruction system (known as the Techa River Dosimetry System or TRDS) for the TRC, which numbers about 30,000 people (Degteva et al. 1996a, b). The current dose-reconstruction system is grounded firmly on whole-body counts for half of the members of the cohort (for the evaluation of internal dose, which was mainly due to incorporated  $^{90}\text{Sr}$ ) and on direct measurements of external gamma-exposure rates. This project is concerned with a comprehensive program to develop improvements in the existing dosimetry system for the TRC members by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results.

The URCRM whole-body counter (WBC), which is identified as SICH-9.1, has been used since 1974 to measure  $^{90}\text{Sr}$  and other radionuclides in people exposed as a result of radioactive releases to the Techa River (Kozheurov 1994). More than 30,000 measurements were carried out during this period on more than 15,000 people. This data base of measurements is critical to the success of efforts to provide individual doses. The data of concern are of major interest to the reconstruction of dose to the bone marrow of the members of the Techa River Cohort and to the completion of the major study of strontium metabolism that is being undertaken at the URCRM (Tolstykh et al. 1997). The detectors and electronics of the SICH-9.1 are obsolete, and there are plans to update this WBC with modern detectors and electronics. The WBC will be used in the future for continuing measurements, and it is important to note that the WBC is now an integral part of the public-outreach program for the local residents. It is highly desirable to ensure that the old measurement system is once again calibrated in depth on the basis of

measurements of a specially constructed anthropomorphic phantom that contains  $^{90}\text{Sr}$  distributed through simulated bones and measurements of another set of phantoms that contain  $^{40}\text{K}$  and  $^{137}\text{Cs}$ . Also, the measurements of the same phantoms with the anticipated new system will ensure the comparability of the results with the old and new measurement systems.

In order to achieve the recalibration of SICH-9.1, the implementation of a mathematical phantom for further calibration, and the upgrading of SICH-9.1, Task 6 was formulated within the framework of Project 1.1 as "Whole-Body-Counter Calibration and Modification." To perform the first two parts of this four-part task it is necessary:

- To manufacture an anthropomorphic physical phantom of the body of an adult with a uniform distribution of  $^{90}\text{Sr}$  in the skeleton; and
- To prepare a protocol and perform the calibration of whole-body counter SICH-9.1 for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  using special phantoms.

This report is the Final Report for Milestone 2 from the Master List (Degteva et al. 1997).

## 2. DESCRIPTION OF WHOLE-BODY COUNTER SICH-9.1

The main goal of the SICH-9.1 WBC is to allow the in vivo measurement of  $^{90}\text{Sr}$  in the bones of the members of the TRC and other persons with potential exposure. As neither  $^{90}\text{Sr}$  (half life of 29.12 y) nor its daughter radionuclide  $^{90}\text{Y}$  (half life of 64 h) emit  $\gamma$  rays, the usual designs for WBCs are not adequate. In the URCRM application the detector system is designed to measure the presence of the  $\beta$  particle that is emitted by  $^{90}\text{Y}$  with an average energy of 930 keV. As  $\beta$  particles cannot be detected directly by an external counter, the detector system is designed to detect the bremsstrahlung that is emitted by the "braking" of the beta particle. The bremsstrahlung consists of very low energy photons. Unfortunately,  $\gamma$ -emitting radionuclides, such as  $^{40}\text{K}$  and  $^{137}\text{Cs}$ , also produce a "Compton continuum" of scattered radiation that includes a significant amount of low energy photons. Therefore, it is necessary to determine the presence of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  so that their contribution to the low-energy bremsstrahlung region can be subtracted.

The principal design of WBC SICH-9.1 is shown in Fig. 1. The shielding room is made of cast-iron rings with a wall thickness of 200 mm. The inner surface has linings of lead (5 mm), cadmium (1.5 mm), and copper (1 mm). Four phoswich detectors are used. The geometry of measurement is as follows: The detectors are fixed in the central vertical plane with a  $30^\circ$  inclination toward the vertical. The distance between the axes of each pair of detectors is 35 cm at the base. On the bed frame, a fabric is stretched which stretches under the weight of the body in such a way that the medium plane of the body is at a distance of 25 cm from the detectors.

During the measurement the person lying on the bed is moved through the detector array. The scanning length is 2 m. Counting is continued during stops at each end with counting times of 10% of the total measurement time at each end position. The motion is controlled by signals from the analyzer's real-time clock. The durations of the measurements are 2.5; 5; 10; 20 and 40 min.

For the original calibration of the WBC two surrogate-human structures were made by different laboratories. Both phantoms were made of natural human skeletons, paraffin imitations of soft tissues, and dry paper imitations of lung. Different methods of introducing  $^{90}\text{Sr}$  into the "bones" of the phantoms were used. In one of the phantoms the radionuclide was introduced by being dripped into uniformly distributed holes drilled into bones. The bones of the other phantom were impregnated with a  $^{90}\text{Sr}$  solution in a vacuum chamber. Each laboratory performed independent experiments aimed at determining bremsstrahlung-yield relations and the influence of human-soft tissues and the phantom paraffin on the absorption of bremsstrahlung. Independent activity measurements were carried out on each phantom. After scanning the phantoms and making the appropriate corrections, the difference in calibration coefficients was determined to be 6%. This value represents the estimate of the systematic error in  $^{90}\text{Sr}$  counting by means of the spectrometer. Water-filled phantoms made of plastic tanks laid out in such a way as to imitate a human body were used for the calibration of  $^{137}\text{Cs}$  and  $^{40}\text{K}$ . The length of the phantom could be changed by removing one or two tanks. Such calibration was done in 1974 and had not been confirmed during the following 20-year period of operation of the WBC. Of course, standard sources have been counted on a regular basis in order to monitor the performance of the detectors and their electronic systems.

### **3. DESCRIPTION OF THE ANTHROPOMORPHIC PHYSICAL PHANTOMS**

Work on manufacturing new phantoms was performed within the frame of Task 6 at the Research Institute of Industrial and Marine Medicine (RIIMM) in St. Petersburg. Phantoms of two types were manufactured: the  $^{90}\text{Sr}$  phantom and the gamma-emitter phantom.

#### **3.1. Solid whole-body phantom (UP-02T set)**

This phantom set is assembled of right-angled polyethylene units with masses of 1.0 and 0.5 kg and rod-radionuclide sources 6 mm in diameter inserted into them with two sources in each unit. The usual phantom set simulates the body characteristics of 2-, 6- and 14-year-old children and three adults with nominal body masses of 70, 90, and 110 kg; measurements can be performed with the phantoms in standing, sitting, lying, and sitting-bending positions (Fig. 2). These solid phantoms of persons of different ages are assembled from 130 or fewer polyethylene units and 260 or fewer rod sources included in the UP-02T set. Rod sources containing standard activities of  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  can be replaced without total dismantlement of the phantom. This set of radionuclides provides calibration of devices for gamma-radiation energies up to

1460 keV. Measurement results have proven that the yield of gamma radiation from the solid phantom agrees well with similar parameters of phantoms containing widely distributed liquid-radionuclide sources. Table 1 presents the main characteristics of the UP-02T set of phantoms, as somewhat modified for the URCRM application. Here, the weights of the adult phantoms have been reduced in order to make them more compatible with the body weights actually encountered among the members of the TRC.

A detailed description of this set of phantoms is presented in documents provided by the RIIMM. This set of phantoms with standard radioactive sources has been certified by the Russian General State Center for Measurement Standards, Mendeleev NPO VNIIM, St. Petersburg. These transformable solid phantoms are convenient for calibration and intercomparison of WBCs with different measurement geometries. The present version of the whole body phantom has been used during 1995-1996 for intercomparison of European whole body counters.

### 3.2. Anthropomorphic phantom with strontium-90 in the skeleton (FST-06T)

This physical phantom is an anthropomorphic model of the body of an adult with a uniform distribution of  $^{90}\text{Sr}$  in the skeleton (Fig. 3). The error in the muscle-tissue equivalent, lung-tissue equivalent and bone equivalent of the materials used is within 5% for the 15 to 60 keV range of photon energy that is typical of bremsstrahlung radiation. The activity error is less than 5%. The phantom consists of three main tissue substitutes designed to simulate bone, lung, and soft tissues (mass densities of these are equal to 1.3; 0.26 and 1.04 g cm<sup>-3</sup>, respectively). The materials are cold-cured epoxy resin products. The phantom imitates an adult man with height of 170 cm and body weight of 70 kg, which are the characteristics of the ICRP Reference Man (ICRP 1975). The phantom consists of sculptural models of body, head, and upper and lower extremities. The phantom imitates a prone position in order to use it in the scanning-bed geometry realized in SICH-9.1. Selected kinds of epoxy resin compounds were used for tissue substitutes. This phantom is an original design prepared especially for the SICH-9.1 calibration. The main characteristics of the FST-06T phantom are presented in Table 2. The RIIMM provided the URCRM with special documentation containing a detailed description of this phantom.

*Table 1. Main characteristics of UP-02T set of phantoms.*

Phantom type	Body mass, kg	Height, cm	$^{40}\text{K}$ content, Bq	$^{137}\text{Cs}$ content, Bq
F1	10.6	82.5	860	1,970
F2	20.9	121.0	1,420	3,960
F3	42.9	160.0	3,590	8,300
F4	61.5	170.5	5,000	11,500
F5	77.8	170.5	6,450	14,890
F6	95.2	170.5	7,880	18,190

*Table 2. Main characteristics of FST-06T phantom in comparison with ICRP-23 Reference Man (ICRP 1975).*

Parameter	FST-06T	Reference Man
Height, cm	170	170
Body mass, kg	68.6	70.0
Skeleton mass, kg	9.3	10
<sup>90</sup> Sr (and <sup>90</sup> Y) activity, kBq	91.0	-

#### **4. CALIBRATION OF WHOLE-BODY COUNTER SICH-9.1**

To perform the calibration it is necessary to measure the spectrum of background radiation in the empty shielding room, to investigate the change in the background spectrum with the presence in the room of a scattering body mass (without radioactive sources), and to measure the spectrum of radiation in the presence of the phantom containing standard radionuclide sources.

While an important goal is the recalibration of the existing SICH-9.1 detector system, it is important to point out that the SICH-9.1 contains out-of-date equipment that makes it very difficult to perform reliable measurements. For example, the amplitude analyzer, the high voltage supply, and the impulse discriminator were manufactured in 1968; 1970; and 1973, respectively. The use of this obsolete equipment has resulted in the instability of the energy scale during measurements due to drift of the zero-energy position and changes in amplification. In addition, there were shifts of photopeak positions in the spectra due to poor contacts in switches.

Each spectrum was measured during 20 min and was then examined for gross errors that sometimes occurred due to instability of the spectrometer electronics. Spectra containing such gross errors were excluded from further analysis. Several 20-min spectra were combined as necessary in order to achieve the required statistical accuracy.

##### **4.1 Background spectra in the empty whole-body counter**

There is crystalline surface rock (granite) with a high content of radium in close vicinity to the SICH-9.1 location. Therefore the influence of airborne radon-decay products on the accuracy of the measurements can be appreciable. Usually background spectra are measured three times per day, and the duration of measurements is 20 min under routine conditions. Radon-decay-product-concentration fluctuations in the air are the main cause of the background variations despite continuous aerosol filtration. The background spectra in the empty whole-body counter are presented in Fig. 4. These spectra are not directly used in the calibration, but they are necessary as baseline data to determine the influence of a scattering body mass on the background-radiation level.

## 4.2 Calibration for gamma emitters

Four phantoms (F3, F4, F5 and F6) from the modified UP-02T set were used for the calibration of SICH-9.1 for  $^{40}\text{K}$  and  $^{137}\text{Cs}$ . For each phantom measurements were made of the background spectrum with the phantom body alone (no radioactive sources in place), of the phantom with  $^{40}\text{K}$  sources in place, and of the phantom with  $^{137}\text{Cs}$  sources in place.

Phantom F3 has a mass of 42.9 kg and a height of 160 cm; it imitates the body of a 14-year-old adolescent. A background spectrum with this phantom in the counter is presented in Fig. 5 (the duration of measurements is 360 min). For the calibration of  $^{40}\text{K}$ , an activity of 3,590 Bq (97 nCi) was added to this phantom; this activity is equal to that contained in 117 g of natural potassium. The measured spectrum for  $^{40}\text{K}$  after subtraction of background and normalization to 1 g of natural potassium is shown in Fig. 6. For the calibration of  $^{137}\text{Cs}$  an activity of 8,300 Bq (224.3 nCi) was added. The measured spectrum for  $^{137}\text{Cs}$  after subtraction of background and normalization to 1 nCi is shown in Fig. 7. The duration of measurements of the phantom with radioactive sources was 520 min for  $^{40}\text{K}$  and 420 min for  $^{137}\text{Cs}$ .

Phantom F4 has a mass of 61.5 kg and a height of 170.5 cm; it imitates the build of a small adult. A background spectrum with this phantom in SICH-9.1 is presented in Fig. 8 (the duration of measurements is 780 min). For the calibration of  $^{40}\text{K}$ , an activity of 5,000 Bq (135 nCi) was added to this phantom; this activity is equal to that contained in 163 g of natural potassium. The measured spectrum for  $^{40}\text{K}$  after subtraction of background and normalization to 1 g of natural potassium is shown in Fig. 9. For the calibration of  $^{137}\text{Cs}$  an activity of 11,500 Bq (310.8 nCi) was added. The measured spectrum for  $^{137}\text{Cs}$  after subtraction of background and normalization to 1 nCi is shown in Fig. 10. The duration of measurements of the phantom with radioactive sources was 520 min for  $^{40}\text{K}$  and 480 min for  $^{137}\text{Cs}$ .

Phantom F5 has a mass of 77.8 kg and a height of 170.5 cm; it imitates the build of a medium-sized adult. A background spectrum with this phantom in the SICH-9.1 is presented in Fig. 11 (the duration of measurements is 720 min). For the calibration of  $^{40}\text{K}$ , an activity of 6,450 Bq (174.3 nCi) was added to this phantom; this activity is equal to that contained in 210 g of natural potassium. The measured spectrum for  $^{40}\text{K}$  after subtraction of background and normalization to 1 g of natural potassium is shown in Fig. 12. For the calibration of  $^{137}\text{Cs}$  an activity of 14,890 Bq (402.4 nCi) was added to the phantom. The measured spectrum for  $^{137}\text{Cs}$  after subtraction of background and normalization to 1 nCi is shown in Fig. 13. The duration of measurements of the phantom with radioactive sources was 800 min for  $^{40}\text{K}$  and 300 min for  $^{137}\text{Cs}$ .

Phantom F6 has a mass of 95.2 kg and a height of 170.5 cm; it imitates the build of a large adult. A background spectrum with this phantom in the SICH-9.1 is presented in Fig. 14 (the duration of measurements is 780 min). For the calibration of  $^{40}\text{K}$ , an activity of 7,880 Bq (213 nCi) was added to this phantom; this activity is equal to that contained in 256.6 g of natural potassium. The measured spectrum for  $^{40}\text{K}$  after

subtraction of background and normalization to 1 g of natural potassium is shown in Fig. 15. For the calibration of  $^{137}\text{Cs}$  an activity of 18,190 Bq (491.6 nCi) was added. The measured spectrum for  $^{137}\text{Cs}$  after subtraction of background and normalization to 1 nCi is shown in Fig. 16. The duration of measurements of the phantom with radioactive sources was 960 min for  $^{40}\text{K}$  and 240 min for  $^{137}\text{Cs}$ .

### 4.3 Calibration for strontium-90

Phantom FST-06T has a mass of 68.6 kg and a height of 170 cm; it was designed to imitate ICRP Reference Man (ICRP 1975). This phantom contains an activity of 91 kBq of  $^{90}\text{Sr}$  (1229.7 nCi, plus an equal amount of its short-lived daughter  $^{90}\text{Y}$ ) distributed uniformly in the skeleton. The background spectrum obtained by measuring phantom F4, which is also designed to imitate an adult man with parameters close to that of ICRP Reference Man, is used as the background spectrum for the FST-06T phantom. The spectrum of bremsstrahlung radiation from the FST-06T phantom, after subtraction of background and normalization to 1 nCi of  $^{90}\text{Sr}$ , is presented in Fig. 17. The duration of measurement was equal to 980 min.

### 4.4 Comparison of old and new calibration results

The so-called matrix method has been utilized to determine radionuclide-body burden on the basis of SICH-9.1 spectra. The essence of this method is the following:

- The total number of counts is calculated for the special ranges of the energy spectrum;
- The “ranges of interest” were the areas of the photopeaks for  $^{40}\text{K}$  and  $^{137}\text{Cs}$  (620-740 keV and 1400-1580 keV respectively) and the low-energy interval for bremsstrahlung (30-160 keV); and
- Count rates in these energy ranges normalized to the unit of activity in the special phantom are called “calibration coefficients.”

Comparison of the values of calibration coefficients for  $^{137}\text{Cs}$  and  $^{40}\text{K}$  obtained in the first (1973) and the second (1998) calibration of SICH-9.1 showed that the differences lie within 15%. Table 3 illustrates the values of calibration coefficients obtained for the phantom with parameters close to that of ICRP Reference Man.

Table 3. Old and new values of calibration coefficients for  $^{137}\text{Cs}$  and  $^{40}\text{K}$ .

Year of calibration	$^{137}\text{Cs}$ , counts $\text{min}^{-1} \text{nCi}^{-1}$		$^{40}\text{K}$ , counts $\text{min}^{-1} (\text{g K})^{-1}$	
	620-740 keV	30-160 keV <sup>a</sup>	1400-1580 keV	30-160 keV <sup>a</sup>
1973	5.36	3.78	0.38	0.38
1998	5.56	3.38	0.36	0.44
Difference	3.7%	10.8%	5.3%	15.8%

<sup>a</sup> In order to correct for the influence on the measurement of  $^{90}\text{Sr}$ .

To compare the results for  $^{90}\text{Sr}$  phantoms it is necessary to take into account the trend with time of the efficiency of the SICH-9.1 counter for the detection of bremsstrahlung from  $^{90}\text{Y}$ . Such a trend has been evaluated on the basis of daily measurements carried out since 1974 on a standard source of  $^{90}\text{Sr}$ - $^{90}\text{Y}$ . These measurements indicate that the efficiency of bremsstrahlung detection in 1998 is 22% lower than it was in 1974; this is believed to be due to detector aging and to changes in the parameters of pulse discrimination. The calibration coefficient for  $^{90}\text{Sr}$  obtained in 1998, corrected to the change of the effectiveness of bremsstrahlung detection, is equal to  $0.692 \text{ counts min}^{-1} \text{ nCi}^{-1}$ . This is almost the same value that had been obtained in 1973 ( $0.694 \text{ counts min}^{-1} \text{ nCi}^{-1}$ , the difference is 0.3%).

## 5. SUMMARY

The initial calibration of the SICH-9.1 was performed in 1973 with specially constructed phantoms of limited useful life. The results of this calibration have been used for the determination of  $^{90}\text{Sr}$ -body burdens in residents of the Techa Riverside during the past 25 years. After construction of new phantoms for this purpose, a new calibration of the SICH-9.1 was repeated in 1998. In general, the calibration factors obtained in 1973 and in 1998 agree quite well—the more important ones are within a few per cent and all factors agree within 16%.

The condition in 1998 of the original SICH-9.1 detectors and electronic equipment is poor. It is anticipated that this equipment will be upgraded with new detectors and modern electronics soon. At that time another extensive calibration will be performed. This will permit the continuity and intercalibration of measurement results over the period of past measurements and into the future. The “signal” from  $^{90}\text{Sr}$  is decreasing with time due to biological elimination and due to physical decay. This, plus the fact that the interference in this energy region from the scattered photons from  $^{40}\text{K}$  and  $^{137}\text{Cs}$  must be subtracted, makes it increasingly difficult to perform accurate measurements. The much improved stability of operation of new detectors and electronic equipment would greatly enhance the ability to measure current body burdens of  $^{90}\text{Sr}$ .

## ACKNOWLEDGEMENTS

This work has been funded by the US Department of Energy’s Office of International Health Studies, the Federal Department of the Ministry of Health of the Russian Federation, the US Environmental Protection Agency, and the US National Aeronautics and Space Administration.

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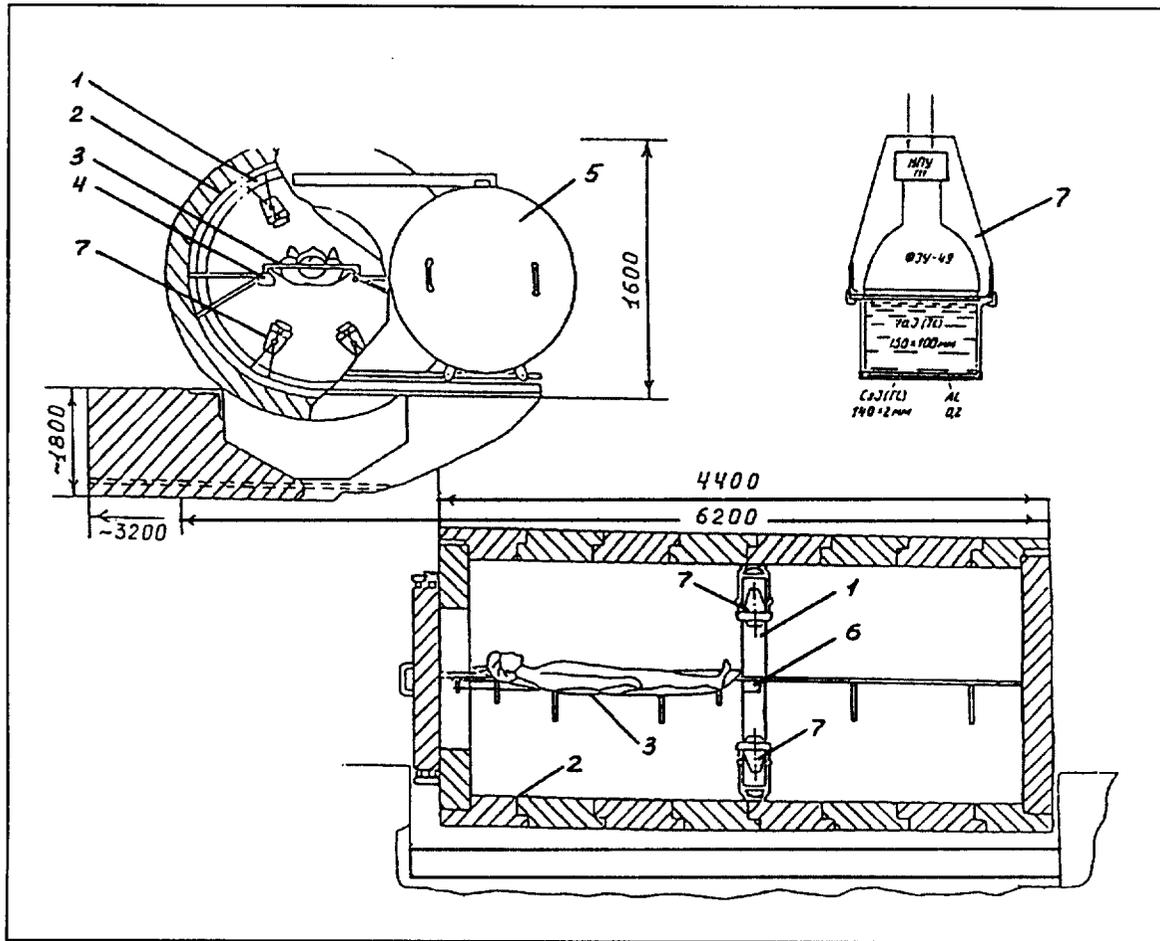


Fig. 1. A schematic sectional view of the SICH-9.1 whole body counter: 1) ring for mounting the detectors; 2) shielding, 200-mm cast iron and lining (lead, 5 mm; cadmium, 1 mm; and copper, 2 mm); 3) movable bed; 4) brackets for mounting the rails; 5) door; 6) motors; and 7) detectors (all measurements in mm).

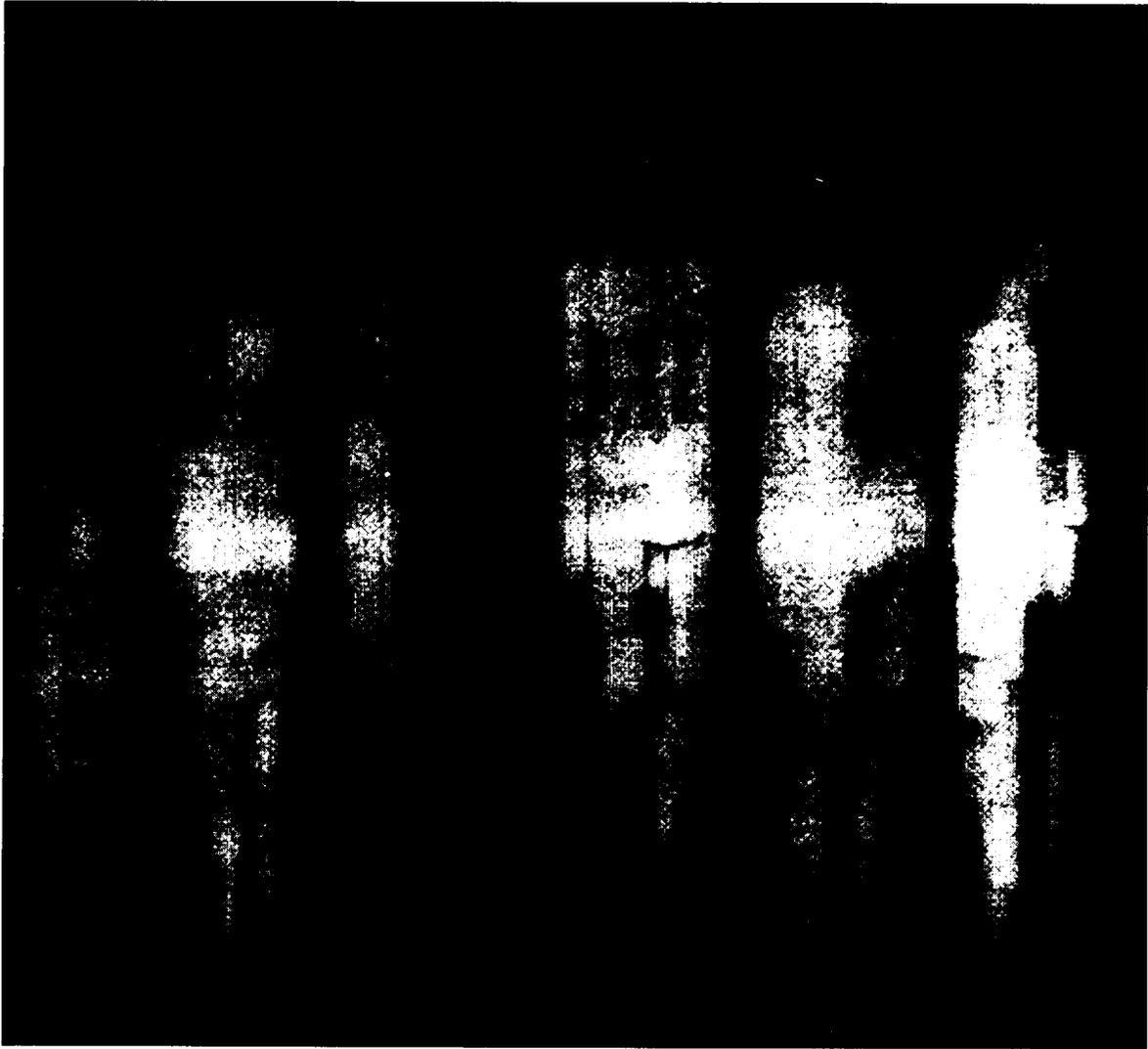


Fig. 2. Unified whole-body phantom (UP-02T set), standing position.



Fig. 3. Anthropomorphic phantom with  $^{90}\text{Sr}$  in the skeleton FST-06T; a - phantom skeleton; b - phantom body.

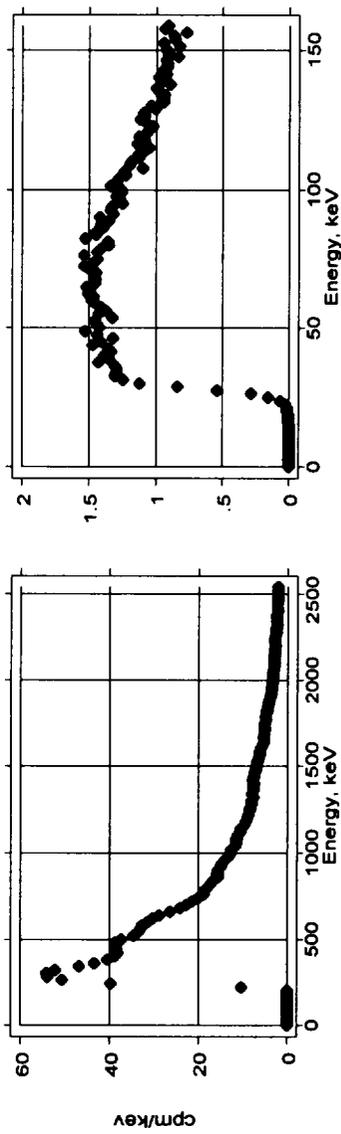


Fig.4. Background spectrum in empty room.

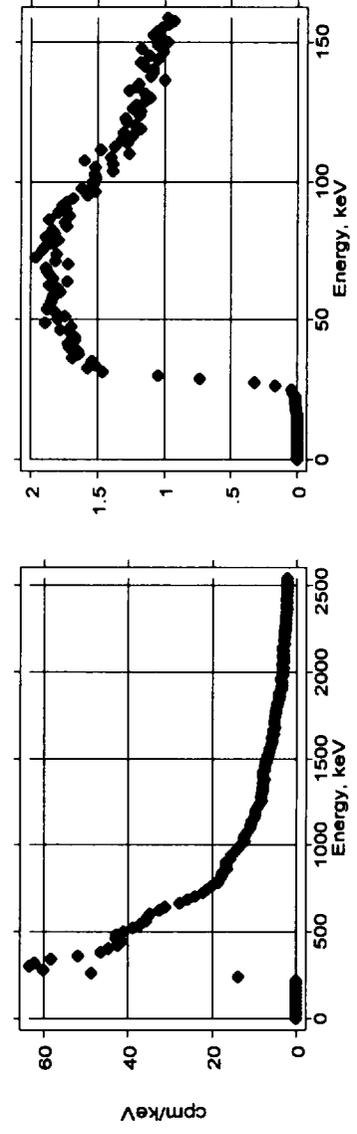


Fig.5. Background spectrum with phantom F3.

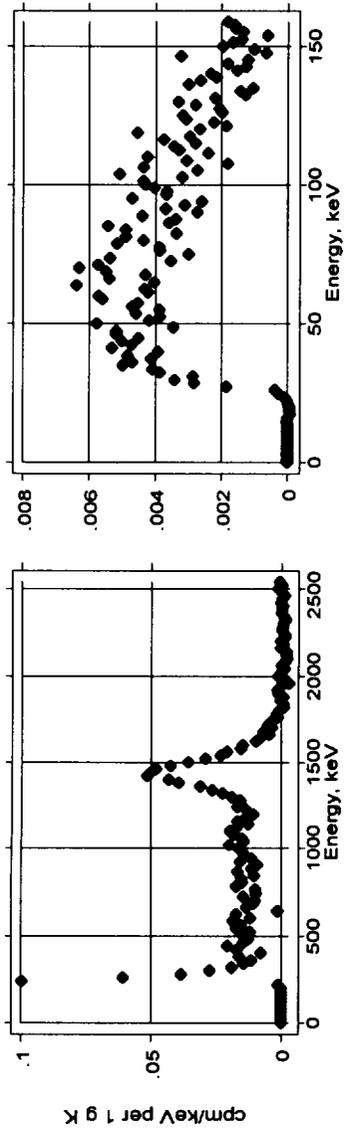


Fig.6. Spectrum of K-40 in phantom F3.

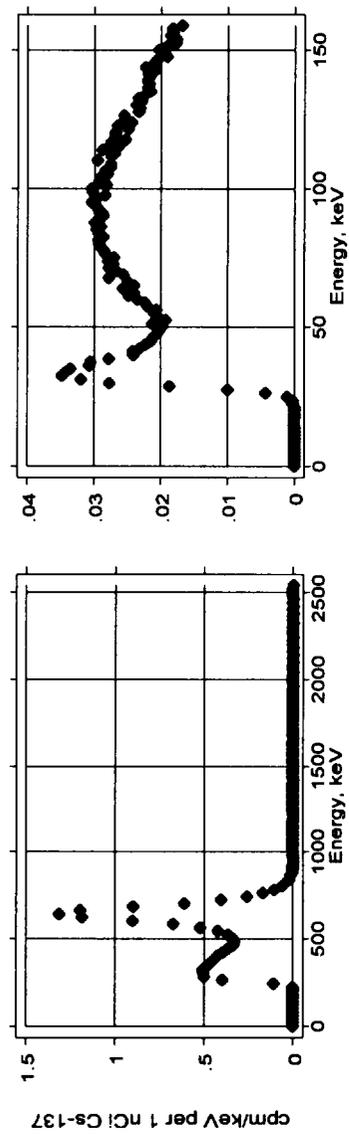


Fig.7. Spectrum of Cs-137 in phantom F3.

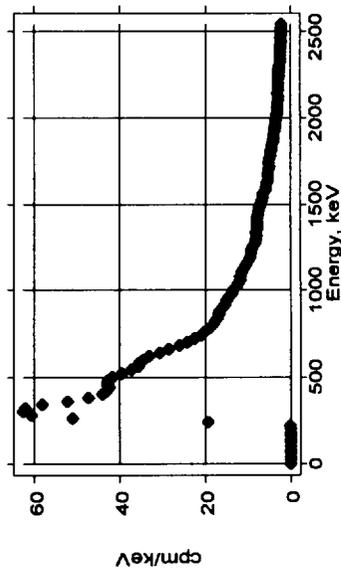
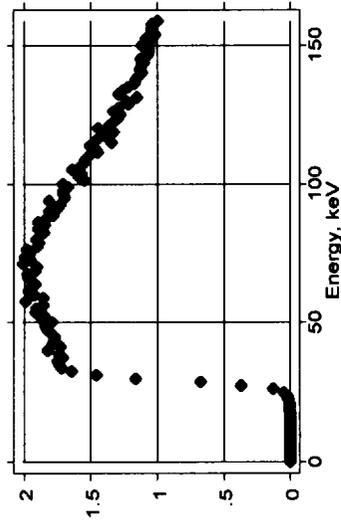


Fig.8. Background spectrum with phantom F4.

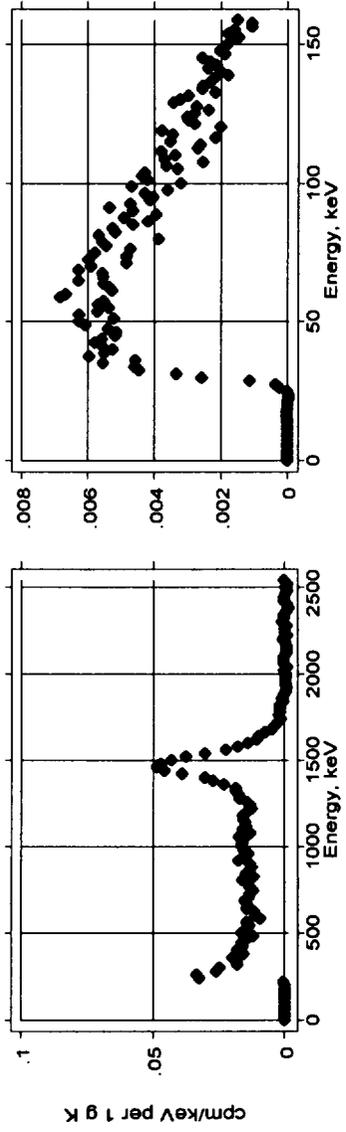


Fig.9. Spectrum of K-40 in phantom F4.

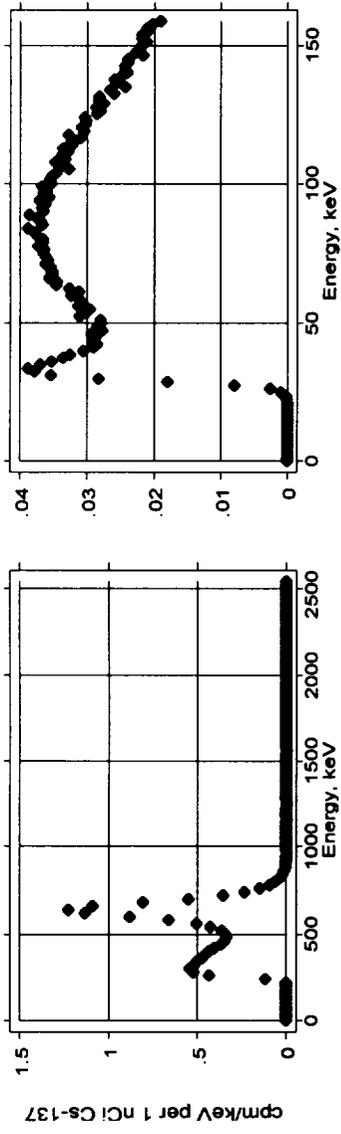


Fig.10. Spectrum of Cs-137 in phantom F4.

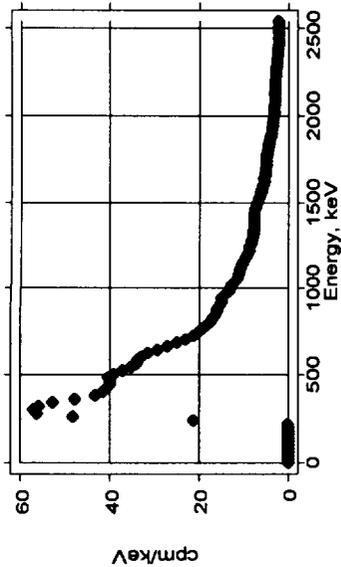
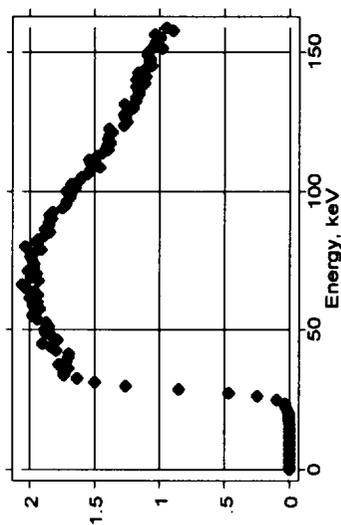


Fig.11. Background spectrum with phantom F5.

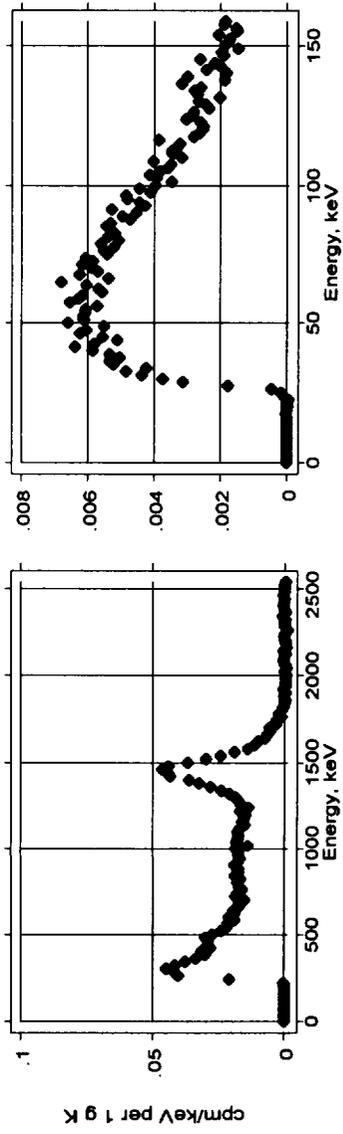


Fig.12. Spectrum of K-40 in phantom F5.

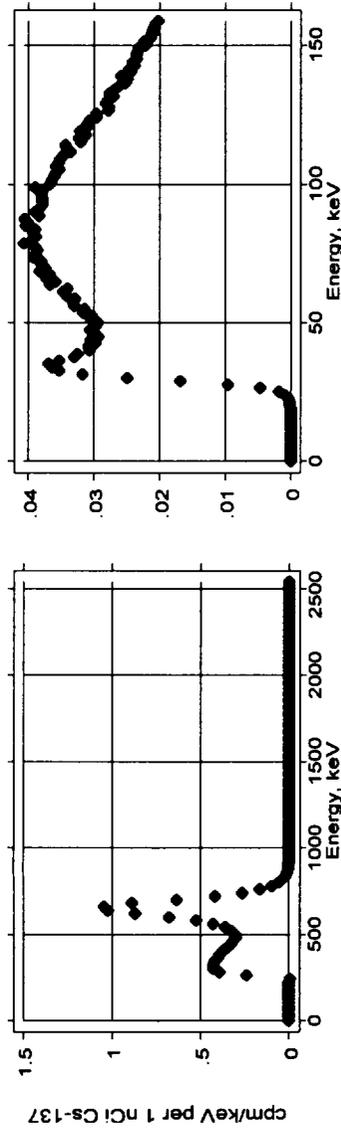


Fig.13. Spectrum of Cs-137 in phantom F5.

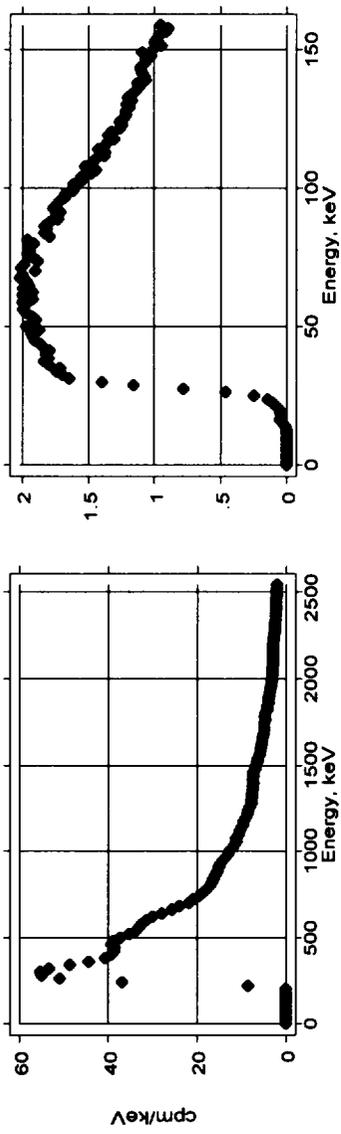


Fig.14. Background spectrum with phantom F6.

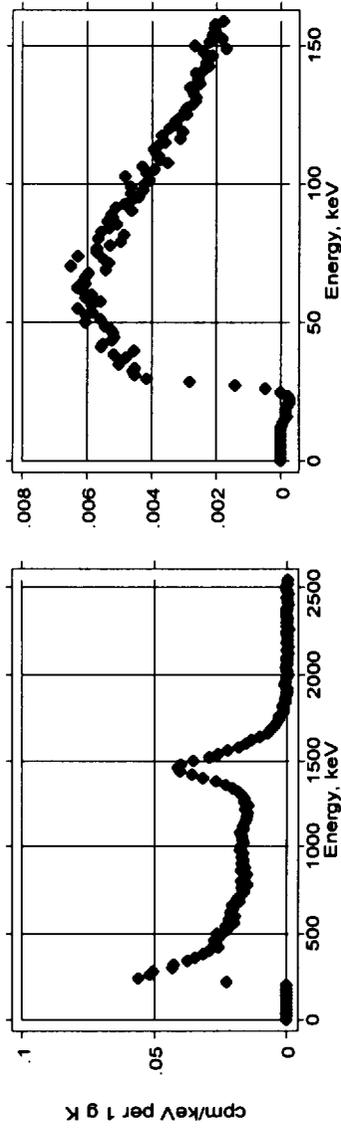


Fig.15. Spectrum of K-40 in phantom F6.

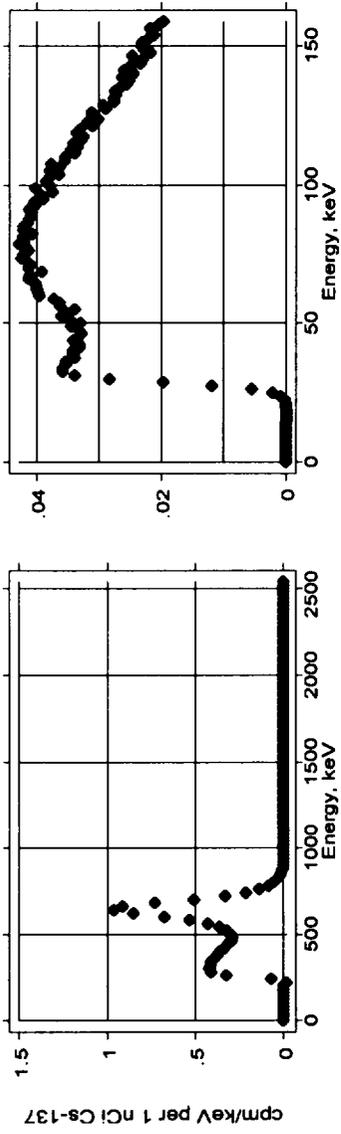


Fig. 16. Spectrum of Cs-137 in phantom F6.

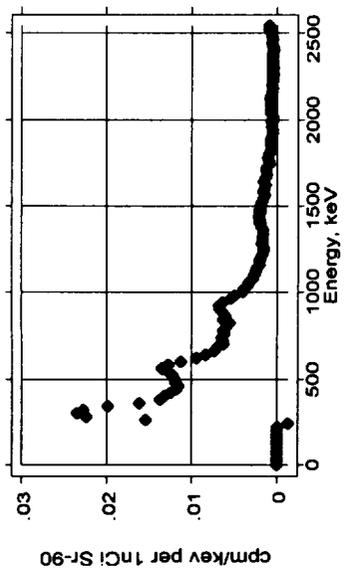
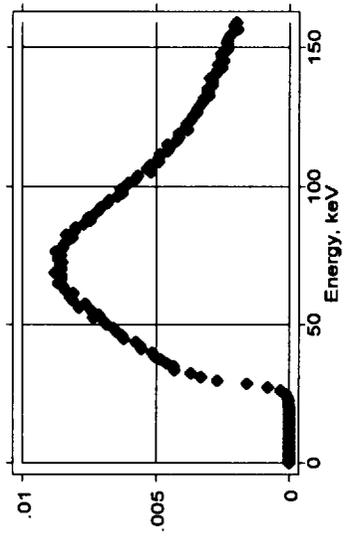


Fig.17. Spectrum of phantom FST 06T.

**INDIVIDUAL-BODY-BURDEN HISTORIES AND RESULTING INTERNAL  
ORGAN DOSES EVALUATED ON THE BASIS OF THE TECHA RIVER  
DOSIMETRY SYSTEM APPROACH**

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**Final Report for Milestone 3**

**US-Russian Joint Coordinating Committee on Radiation Effects Research  
Project 1.1  
“Development of an Improved Dose Reconstruction System for the General  
Population Affected by the Operation of the Mayak Production Association”**

**October 1998**

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## 1. INTRODUCTION

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. This plant began operation in 1948, and during its early days there were technological failures that resulted in the release of large amounts of radioactive waste into the rather small Techa River. The residents of the riverside communities were exposed to chronic external and internal irradiation. Extensive monitoring efforts for the environment and the population at this site began in 1951. The "Techa River Cohort" (TRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM), and an increase in both leukemia and solid tumors with radiation dose has been noted (Kossenko et al. 1997). This finding suggests that, with continuing improvements in the quality of the follow-up and dosimetry, study of the TRC has the potential to provide quantitative estimates of the risks of stochastic health effects produced by chronic low-dose-rate radiation exposure in the general population. Study of this population affords a unique opportunity to address the question of the existence of a dose-rate-reduction factor for the induction of stochastic effects in an unselected general population. A definitive answer to this question would have relevance to the regulation of radiation exposure throughout the world.

The purpose of the US–Russia Joint Coordinating Committee on Radiation Effects Research (JCCRER) Project 1.1 is to define and implement a protocol for improvements in the dose-reconstruction system (known as the Techa River Dosimetry System or TRDS) for the TRC, which numbers about 30,000 people (Degteva et al. 1996c). The current dose-reconstruction system is grounded firmly on whole-body counts for half of the members of the cohort (for the evaluation of internal dose, which was mainly due to incorporated  $^{90}\text{Sr}$ ) and on direct measurements of external gamma-exposure rates. This project is concerned with a comprehensive program to develop improvements in the existing dosimetry system for the TRC members by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results.

A set of conceptual models that defines the relationships, pathways, and parameters that form the basis of the dose-reconstruction efforts has been described in the Final Report of a one-year pilot study (Degteva et al. 1996a). The hierarchy of information required for calculating doses to people who lived along the Techa River has also been described. One of the important tasks formulated within the framework of Project 1.1 is the analysis of available historical data on source terms of the releases and on environmental contamination.

The purposes of this document are the following:

- To present available data on human-body contamination by long-lived radionuclides and to assess of the applicability of such data for the reconstruction of dose from the incorporation of radionuclides;

- To describe the improvements, which include new data and models, in the TRDS modules used for internal dose calculations; and
- To analyze the distribution of calculated doses in different organs due to the ingestion of radionuclides as a result of the Techa River contamination.

## 2. ANALYSIS OF DATA ON INDIVIDUAL-BODY-BURDEN HISTORIES

### 2.1. DATA BASE ON HUMAN-BODY CONTAMINATION

Extensive monitoring efforts for the Techa Riverside residents began in 1951. These efforts included a large-scale program for measuring human-body contamination by long-lived radionuclides. For the follow-up of the exposed population a special data base was established in the Urals Research Center for Radiation Medicine (Degteva et al. 1996b). This data base contains the roster of exposed persons, their residence histories, and the results of medical and dosimetric examinations. Over half of the members of the TRC have had individual measurements of  $^{90}\text{Sr}$  burden. This data base provides an objective basis for environmental dose reconstruction and for the assessment of the health effects on a general population exposed to chronic radiation.

The data base on human-body contamination includes findings obtained by different methods that reflect the evolution of experimental dosimetry since the early 1950s. The first analyses involved radiometric measurements of bioassay and autopsy samples. From 1951 to 1960 ashed samples were measured using a gas-flow counter calibrated with a  $^{90}\text{Sr}$ - $^{90}\text{Y}$  standard (Gussev et al. 1959). The average  $^{90}\text{Sr}$  concentration in the skeleton was calculated on the basis of measurements of several samples of trabecular and compact bone. Since 1958 a radiochemical method (based on the coprecipitation of  $^{90}\text{Sr}$  with nitrates) has been used (Gussev et al. 1959). Fig. 1a presents the findings for subjects measured using both techniques; the indicated agreement assures consistency and continuity of results.

The autopsy program continued up to 1993, and more than 7500 analyses were performed on bone samples from 5400 autopsies of Urals residents. Unfortunately, these data were not accurately matched with the roster of exposed subjects until 1995, when the creation of a computerized autopsy registry permitted the selection of 809 measurements for 232 member of the TRC.

*Post mortem* measurements were not a sufficient basis for the enrollment of exposed persons in dosimetric and epidemiologic investigations, and since 1959 *in vivo* measurements of surface-beta activity on front teeth have been performed. This measurement was made possible with the development of detectors that could be placed in the mouth of the person to be measured (Keirim-Marcus et al. 1961). The technique was very simple and the measuring device was cheap and portable, but the findings showed that only the narrow range of age cohorts born in 1945-1955 had detectable amounts of  $^{90}\text{Sr}$  in front teeth (Kozheurov 1994).

Since 1974, residents of the Techa Riverside have also been examined for their  $^{90}\text{Sr}$ - and  $^{137}\text{Cs}$ -body burdens with a specially developed whole-body counter (WBC). The measurements of  $^{90}\text{Sr}$  were achieved by measuring with a phoswich detector the bremsstrahlung of  $^{90}\text{Y}$  (daughter of  $^{90}\text{Sr}$  with a half-life of 64 h) beta rays; for this purpose scanning-bed geometry enclosed in a special shielding room was used (Kozheurov 1994). Analyses of  $^{137}\text{Cs}$  were accomplished at the same time with the same detector by the measurement of the 662-keV photons from the decay of  $^{137\text{m}}\text{Ba}$  (daughter of  $^{137}\text{Cs}$  with a half-life of 2.6 min). The autopsy program for members of the TRC was reduced after a sufficient number of subjects were measured both with the WBC and on the basis of the analysis of *post mortem* samples, and the consistency between these two methods was confirmed (Fig. 1b). The use of the WBC has resulted in 31,800 individual measurements on 15,250 members of the TRC.

## 2.2. ASSESSMENT OF DATA AVAILABLE

Any retrospective dosimetry study is specific with respect to the data available, and assessment of such data is an important step in determining the optimum strategy of dose reconstruction. The complicated exposure situation that occurred on the Techa Riverside, coupled with the extensive migration (evacuation and relocation) of exposed persons, leads to the conclusion that dose cannot accurately be reconstructed on the basis of theoretical source terms and models of radionuclide transfer through ecological and agricultural systems. However, the availability of direct measurements of  $^{90}\text{Sr}$ -body burdens on half of the members of the cohort provides an unusually well-defined basis for the reconstruction of internal dose for the entire cohort (Degteva et al. 1998).

### 2.2.1. Strontium-90

Fig. 2 presents the time dependence of  $^{90}\text{Sr}$  content in the skeleton of adult residents of the upper- and mid-Techa region. Average levels have decreased more than ten fold during 40 years but “river  $^{90}\text{Sr}$ ” is more than two orders of magnitude higher than global fallout levels (Boecker et al. 1991). Fig. 2 illustrates that there is reliable information on  $^{90}\text{Sr}$ -body burdens for the Techa Riverside residents beginning from the first years after the onset of contamination.

Maximum and average values of  $^{90}\text{Sr}$ -body contents for the different villages are shown in Fig. 3, as a function of the distance from the site of release to the village. The slope of maximum-body-burden levels is the same as that for the concentration of  $^{90}\text{Sr}$  in river water during the period of massive releases. This observation appears to confirm that for these people almost all of the  $^{90}\text{Sr}$  was ingested with river water during that period. The average level has “zero-slope” for distances less than 80 km. This resulted from the implementation of countermeasures. Residents of the upper Techa were prohibited to use river water and relocations were started in 1951, whereas these same measures were extended only after 1956 to the lower parts of the river. The substantial scatter of average values reflects differences in the provision of water supply from non-contaminated sources (wells or tributaries) for different villages during the period of massive releases. Fig. 3 demonstrates that  $^{90}\text{Sr}$  intake has been influenced strongly by

social factors for many of the Techa River residents; therefore, it is impossible to assess the real intake pattern on the basis of environmental data only.

The results of multitude measurements made with the WBC and with the tooth detector show a clear relationship between both the  $^{90}\text{Sr}$ -body burden and  $^{90}\text{Sr}$  content in tooth enamel with the year of birth (Fig. 4). However, the age dependencies are quite different for teeth and skeleton and the two kinds of measurements have been used for different purposes. Tooth data were utilized for  $^{90}\text{Sr}$ -intake reconstruction (Kozheurov and Degteva 1994), while the WBC data provided the basis for the elaboration of an age-dependent model of strontium retention (Degteva and Kozheurov 1994).

### 2.2.2. Cesium-137

The variation with time of  $^{137}\text{Cs}$ -body burdens for the Techa River residents is shown in Fig. 5 and is compared to the variation of body burdens from global fallout (Moiseev 1985). For the period before 1986 "river  $^{137}\text{Cs}$ " is about twice the global level, but there is a peak after the Chernobyl accident. This peak reflects mainly the ingestion of foodstuffs contaminated as a result of the Chernobyl accident. Due to the short biological half-life (roughly 100 days for adults) of  $^{137}\text{Cs}$ , the large amounts of this radionuclide ingested in 1950-1951 had already been eliminated when measurements with the WBC were begun.

### 2.2.3. Plutonium-239,240

There was no special program for the measurement of  $^{239,240}\text{Pu}$  in members of the TRC cohort; only seven subjects from this cohort were measured in Branch 1 of the Moscow Biophysics Institute (Suslova et al. 1996) within the framework of a study of Chelyabinsk Region residents (the study involved about 7000 samples from 680 autopsies). Plutonium-239,240 body burdens decreased sharply with distance of place of residence from the MPA. Levels of  $^{239,240}\text{Pu}$  were about twice those of global fallout for the Techa River residents and the population of settlements located at the same distances from the Mayak, but not on the river. This indicates that  $^{239,240}\text{Pu}$  intake occurred due to airborne releases, and not from contamination of the river.

## 2.3. RECONSTRUCTION OF STRONTIUM-90 INTAKE

Rates of ingestion of  $^{90}\text{Sr}$  by inhabitants of Muslyumovo, a reference settlement on the Techa River, were reconstructed on the basis of beta-ray measurements of teeth. The formation of the permanent front tooth enamel occurs within a short age interval, and the metabolism in the enamel is extremely slow. Therefore, a sharp peak of  $^{90}\text{Sr}$  uptake in childhood (the right curve in Fig. 4) reflects accurately the intake dynamics in those persons whose enamel formation occurred during the period of high  $^{90}\text{Sr}$  contamination. The ratio of intake in children to that of adults was determined by analyzing the contributions of different dietary components to the total diet. The detailed intake reconstruction for all age classes of the population for the relevant years is described elsewhere (Kozheurov and Degteva 1994). The principle of computation was to express

the average values of the observations for different age cohorts in terms of a comparatively simple model that contained unknown dietary contents of  $^{90}\text{Sr}$  for each year and unknown age-dependent uptake factors, and to determine these unknown parameters from a least squares fit of the model to the data.

To reconstruct  $^{90}\text{Sr}$  intake for other settlements it was assumed that the ratios of average intakes equal the ratios of the mean age-standardized  $^{90}\text{Sr}$  contents in the skeleton and in the teeth for the relevant settlements. This assumption, which was made both for existing and demolished villages, is a simplification of the real situation. For the upper Techa residents relocated in 1955-1956 additional  $^{90}\text{Sr}$  intake due to the 1957 Kyshtym accident is evaluated as 150 kBq\* (Balonov et al. 1995). The people who were not relocated from the Techa Riverside received approximately an additional 40 kBq of  $^{90}\text{Sr}$ . Thus, the real pattern of intake was different for relocated and non-relocated people, but this difference is minor in comparison with the 3000 kBq of  $^{90}\text{Sr}$  ingested during 1950-1952.

#### 2.4. VALIDATION OF THE BIOKINETIC MODEL FOR STRONTIUM

An age-dependent biokinetic model for  $^{90}\text{Sr}$  was described in detail elsewhere (Degteva and Kozheurov 1994). Fig. 4 shows model calculations of  $^{90}\text{Sr}$  retention in the skeleton for different ages in comparison with the WBC measurements. As seen, the model describes the distinct maximum of long term  $^{90}\text{Sr}$  retention that is observed in those persons who were teenagers during the period of major intake. In addition, model calculations for adults were compared with measured levels over 40 years of observation. Fig. 6a demonstrates mean levels for the upper- and mid-Techa region where the measurements were started in 1951, and Fig. 6b shows the measurements for the lower reaches that were started after 1956. Two model curves outline the corridor of values for age cohorts included in the measurements. The consistency of model calculations and measured values is obvious. Fig. 7 illustrates the possibility of the reconstruction of individual-body-burden histories on the basis of reference-model curves and individual-WBC measurements.

### 3. TECHA RIVER DOSIMETRY SYSTEM (TRDS) APPROACH TO ORGAN-DOSE EVALUATION

#### 3.1. DESCRIPTION OF TRDS-96

The first version of the Techa River Dosimetry System (TRDS-96) was developed at URCRM under Contract with the Federal Department of the Russian Ministry of Health (Contract No 94427, 1994-1996). The main goal of TRDS development was to reconstruct external and internal radiation doses for 34,000 individuals born in 1949 and earlier and who lived (or continue to live) in the Techa Riverside settlements. The

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\* This estimate is considered to be very conservative. It likely is a substantial overestimate of the actual intake.

purpose of the dose reconstruction is to support companion epidemiologic studies of radiogenic leukemia and solid cancers.

TRDS-96 consisted of the three following parts:

1. An environmental module that contained:
  - 1.1. Age-dependent mean-annual-intake levels of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  for all Techa Riverside settlements from 1950 through 1960 and for East Urals Radioactive Trace (EURT)<sup>†</sup> settlements (except for three evacuated early after the accident) from 1957 through 1960.
  - 1.2. Mean-annual doses of external exposure for two-age categories of residents for all Techa Riverside settlements from 1950 through 1956 and for EURT settlements (except for three evacuated early after the accident) from 1957 through 1960.
2. A metabolic module that contained the results of age-dependent model calculations of doses in red bone marrow, bone surfaces, upper and lower parts of the large intestine, and other soft tissues per unit intake of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ .
3. An individual-data module that contained the following information for 33,400 members of the MAIN Registry extracted from the data base MAN (Techa River Cohort): unique code to identify the individual, year of birth, year of entry to the catchment area, year of migration from the catchment area, vital status, year of vital status determination, residence history in contaminated area (terms and places of residence in the Techa Riverside and EURT settlements from 1949 up to the year of migration).

A special code was developed to use the data of the three modules indicated above to calculate organ doses for each individual according to the age and residence history for every calendar year beginning from 1950 up to the date of the most recent determination of vital status. This version of the TRDS has already been used to calculate such doses and, in conjunction with the additional data in the TRDS, to evaluate radiation-risk coefficients. Fig. 8 illustrates examples of input-data sets for two members of the TRC.

For further studies on the TRC, plans have been made to update or improve the information within each of the modules, and it will be possible to include other radionuclides and models without changes to the general system. The work aimed at the improvements of epidemiologic data for the TRC (Module 3) has already begun under the NCI-RERF-URCRM Project and under JCCRER Project 1.2. This current report presents the improvements in the data and models used for evaluation of internal dose.

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<sup>†</sup> The EURT refers to the fallout from the 1957 accidental explosion of a waste-storage tank. This incident is commonly referred to as the Kyshtym accident.

### 3.2. IMPROVEMENTS IN INTERNAL DOSE RECONSTRUCTION

The following two tasks have been accomplished in order to improve the TRDS modules used for the assessment of internal dose:

- The rates of ingestion of  $^{89}\text{Sr}$  and  $^{137}\text{Cs}$  were verified, and rates of ingestion for  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{103,106}\text{Ru}$  and  $^{141,144}\text{Ce}$  were newly derived on the basis of results of modeling the flow of contaminants through the Techa River (Vorobiova et al. 1997).
- Data on age-dependent metabolism and dosimetry for the short-lived radionuclides listed above have been included in TRDS Module 2.

Age-dependent mean-annual-intake levels for  $^{137}\text{Cs}$  and short-lived radionuclides were calculated on the basis of the following assumptions. As most of the ingestion of radionuclides occurred with the consumption of river water in 1950-1952, intakes of  $^{137}\text{Cs}$  and short-lived radionuclides were derived from estimates of age-dependent intakes of  $^{90}\text{Sr}$  scaled in terms of radionuclide composition of the river water. The ratios of radionuclide concentrations to  $^{90}\text{Sr}$  as functions of calendar year and distance downstream from the site of release were calculated using the Techa River Model described in Vorobiova et al. (1997).

For  $^{90}\text{Sr}$  (and short-lived  $^{89}\text{Sr}$ ) the biokinetic model developed on the basis of Techa River data was used and dose coefficients to target tissues were taken from Spiers et al. (1978). Dose coefficients were calculated using the DosAge computer code developed at URCRM (Degteva et al. 1992).

For  $^{137}\text{Cs}$  and other radionuclides the age-dependent biokinetic models from ICRP Publication 67 (ICRP 1993) were utilized. As this publication contains data only for six age groups, dose coefficients on a year-by-year basis for these radionuclides were calculated for us by Dr. V. Berkovsky (Radiation Protection Institute, Kiev, Ukraine) using the special software IDSS (Berkovsky 1992).

Now, the TRDS Environmental Module contains age-dependent mean-annual-intake levels of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{103,106}\text{Ru}$ ,  $^{137}\text{Cs}$ , and  $^{141,144}\text{Ce}$  for all Techa Riverside settlements from 1950 through 1960 and the Metabolic Module contains the results of age-dependent model calculations of dose in red bone marrow, bone surfaces, walls of the upper and lower parts of the large intestine, wall of the small intestine, stomach wall, ovaries, testes and uterus per unit intake of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{103,106}\text{Ru}$ ,  $^{137}\text{Cs}$ , and  $^{141,144}\text{Ce}$ .

The TRDS code is designed so that data included in its modules can be updated and so that additional sources of exposure can be included. It is planned to include in the TRDS the intake of short-lived radionuclides due to the residence of some TRC members in the area contaminated as a result of the Kyshtym accident (EURT territory); this will be done following the completion of work to verify ingestion levels of radionuclides for persons living on the EURT. This work is ongoing now in the Environmental

Department of the URCRM under Contract with the Russian Ministry on Emergency Situations (EMERCOM). After completion of this work, all pathways of internal exposure due to major radiation accidents that occurred in the fifties in the Urals (Techa River and EURT) will be included in the TRDS. Future improvements of the current version of this dosimetric system can be envisaged with the inclusion of the relatively minor amounts of radionuclide ingestion that continued after 1960 due to Techa River Valley and EURT contamination. Also, it would be interesting to evaluate additional doses due to routine airborne releases ( $^{131}\text{I}$  and  $^{239,240}\text{Pu}$ ) from the MPA and due to the Lake Karachay trace<sup>†</sup> of 1967 ( $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ). Preliminary analysis has shown that the contamination of 1967 is superimposed predominantly on the area from which the population had been already evacuated as a result of the Kyshtym accident. As for the routine airborne releases, exposure extended in all directions from the MPA, including the upper- and mid-Techa regions. Therefore, the resulting doses may have similar values for the Techa River residents and for the residents of nearby settlements that are used as comparison groups in epidemiologic studies. However, this additional source of exposure for the members of the epidemiologic studies could be an important confounder in the analysis; thus, the dose received by these persons from airborne emissions from the MPA should be evaluated in the near future.

#### 4. ANALYSIS OF THE ORGAN DOSES EVALUATED

##### 4.1. DOSES IN RED BONE MARROW AND BONE SURFACES

As described above, doses for members of the TRC are calculated on the basis of age- and location-specific mean-annual-intake levels of radionuclides, age-dependent biokinetic models for radionuclides, and individual-residence histories for each subject. Fig. 9 presents dose distributions in red bone marrow (RBM) among the TRC members. Accumulated organ doses were calculated for each individual for every calendar year from the first date of residence near the contaminated river up to the date of vital status determination. The left columns in Fig. 9 illustrate doses for 30,500 subjects who came to live on the Techa River before 1960. Some cohort members were lost to follow-up early due to death or migration to other regions, and 99.9% of people who came to the Techa after 1952 received doses less than 0.1 Gy. The right hand columns in Fig. 9 illustrate doses for 14,500 subjects who lived near the river during the period of massive releases and were under observation for more than 30 years. In both cases more than half of the people have RBM doses between 0.1 and 0.5 Gy, but the proportion of subjects with high doses is higher in the subcohort of "permanent residents." Absorbed doses in cells on bone surfaces (BS) have the same distributions as do the RBM doses, but the values are about two times higher.

The doses described above were calculated on the basis of site-specific intake levels and individual-residence histories of each subject (TRDS approach). For subjects

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<sup>†</sup> Lake Karachay received waste waters from the MPA. In 1967 this lake dried out, and the contaminated sediments were suspended and re-deposited to such an extent that a "trace" or fallout pattern was formed.

measured by the WBC it is possible to evaluate individual internal doses by assuming doses are proportional to  $^{90}\text{Sr}$ -body burdens within the same age cohort. Such evaluations were made for a subcohort of 6270 "permanent residents" with repeated WBC measurements normalized to a fixed date by taking into account age- and sex-dependent elimination rates of  $^{90}\text{Sr}$  from the body (Tolstykh et al. 1997). Individual doses for these subjects were calculated on the basis of the above assumptions by multiplying individual  $^{90}\text{Sr}$ -body burdens by the ratio of age-mean dose to age-mean  $^{90}\text{Sr}$ -body burden (this principle is illustrated in Fig. 7). It is interesting to compare dose distributions obtained using the two different approaches. Fig. 10 presents RBM-dose distributions calculated with the TRDS ("residence history") and "body burden" approaches. As seen, the "body burden" approach gives a more asymmetrical curve with a long tail. This reflects individual variability in intake and biokinetic parameters that could not be described by location- and age-specific models. Nevertheless, the mean and median values are close for both approaches. The regression equation for "body burden" relative to "residence history" doses is:  $y = 0.056 + 0.995x$  with correlation coefficient equal 0.25. Therefore, the "residence history" approach gives a good evaluation of the "central tendencies" of dose received by the cohort, but does not show individual variations in dose. This restricts the applicability of site-specific dose assessments. Such assessments may be used for risk evaluation of stochastic effects among large population groups, but are of less value in studies on dose dependencies for deterministic effects in small groups of individuals.

#### 4.2. DOSES IN OTHER TISSUES

The method of TRDS dose calculation in other tissues was the same as that for RBM. Absorbed doses were calculated for the following tissues: walls of the upper and lower parts of the large intestine (ULI and LLI), wall of the small intestine (SI), stomach wall (ST), ovaries, testes and uterus. Fig. 11 demonstrates the distributions of doses in ULI and LLI among the members of the TRC. The comparison of "old" and "new" values shows that the inclusion of short-lived radionuclides results in a threefold increase in dose for the upper Techa River residents: Maximal values become 0.98 Gy instead of 0.38 Gy for the LLI and 0.34 Gy instead of 0.12 Gy for the ULI. The old TRDS approach gave an average value of absorbed dose for all tissues except RBM, BS, ULI and LLI walls, because only  $^{137}\text{Cs}$ , which has uniform distribution throughout the body, was taken into account. The levels of internal dose for other soft tissues were lower than 0.047 Gy for all members of the TRC and lower than 0.02 Gy for 95% of people. The new TRDS approach allows the separate consideration of other parts of the gastrointestinal tract (small intestine and stomach), which could have elevated exposure due to the ingestion of radionuclides with low gastrointestinal absorption (such as  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{144}\text{Ce}$ , etc.). The new calculation shows that the dose to the small intestine of the upper Techa River residents increases by 50% and reaches a value of 0.07 Gy. As for the stomach, testes, ovaries and uterus, the inclusion of short-lived radionuclides results in an increase of less than 10% in absorbed dose for the upper Techa River residents. The distributions of dose for these tissues are very similar: The maximum level is 0.05 Gy and about 90% of people have doses lower than 0.02 Gy. This means that the primary

radionuclide contributing dose to these tissues (as well as for other tissues not considered) is  $^{137}\text{Cs}$ .

## 5. SUMMARY AND CONCLUSION

Methods are described for reconstructing internal doses from chronic environmental exposure of the population living on the banks of the Techa River, which was contaminated by fission products in the early fifties. Extensive monitoring efforts that started in 1951 for this cohort of about 30,000 people have provided an objective basis for analyzing different approaches to individual-dose reconstruction. The data on  $^{90}\text{Sr}$  in humans from the Techa River Cohort could be used for validation and improvement of biokinetic models of bone-seeking radionuclides to evaluate individual doses for risk-assessment purposes. Improvements in the Techa River Dosimetry System have been accomplished that provide more complete information on levels of internal exposure, especially for the residents of the upper Techa River region who received maximal doses.

## ACKNOWLEDGEMENTS

This work has been funded by the US Department of Energy's Office of International Health Studies, the Federal Department of the Ministry of Health of the Russian Federation, the US Environmental Protection Agency, and the US National Aeronautics and Space Administration. We acknowledge the invaluable assistance of Vladimir Berkovsky (Radiation Protection Institute, Kiev), who has provided us with the results of dose-coefficient calculations performed using his original software IDSS.

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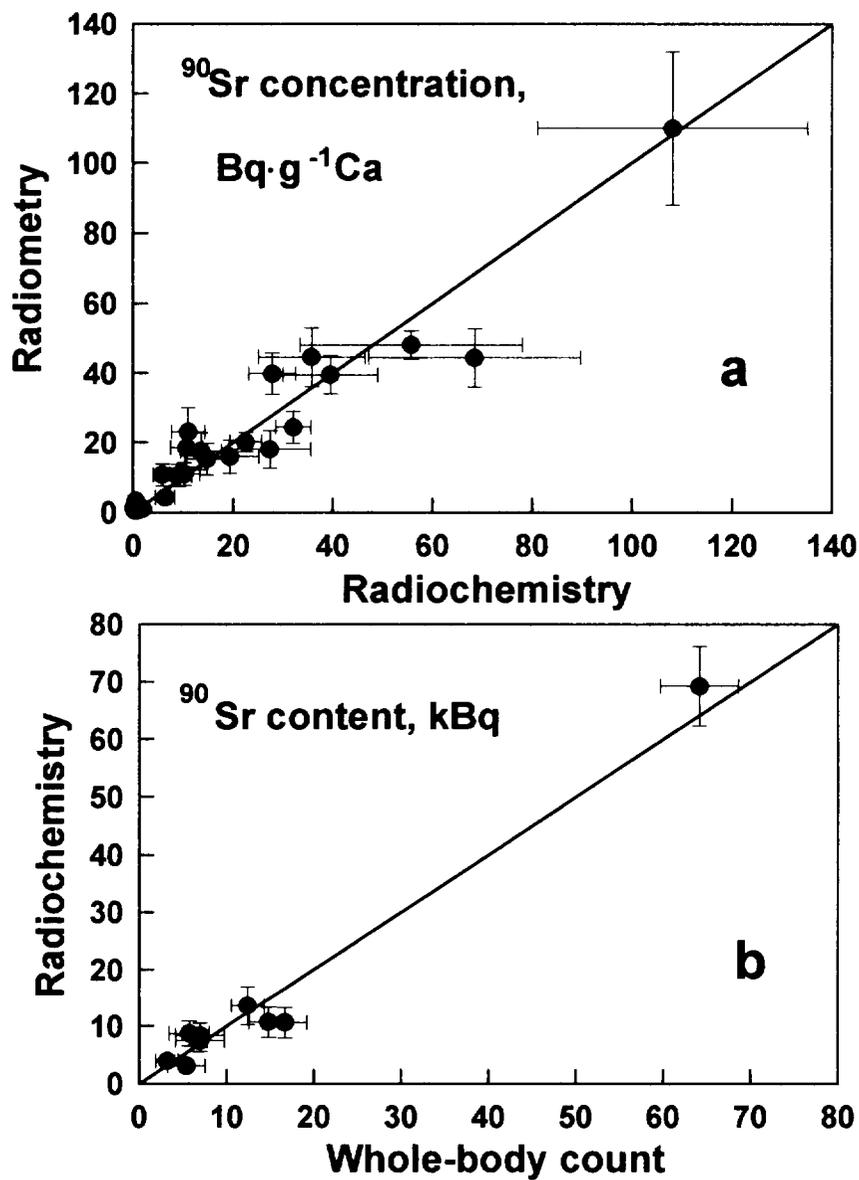


Fig. 1. The consistency of methods used for <sup>90</sup>Sr evaluation in humans: (a) radiometric versus radiochemical measurements of autopsy samples and (b) radiochemical measurements of autopsy samples versus whole-body counting.

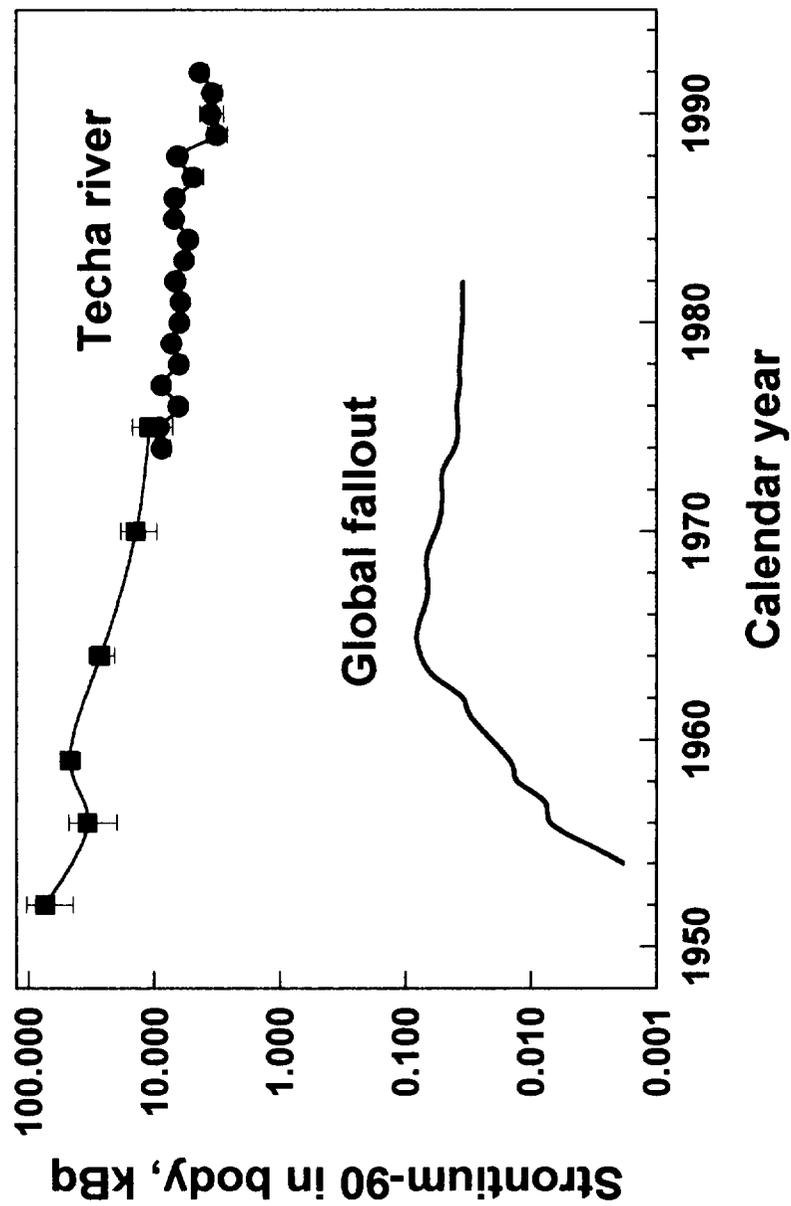


Fig. 2.  $^{90}\text{Sr}$ -body burdens for adult residents of the upper- and mid-Techa region in comparison with global fallout levels: ■ - autopsy data, ● - whole-body count.

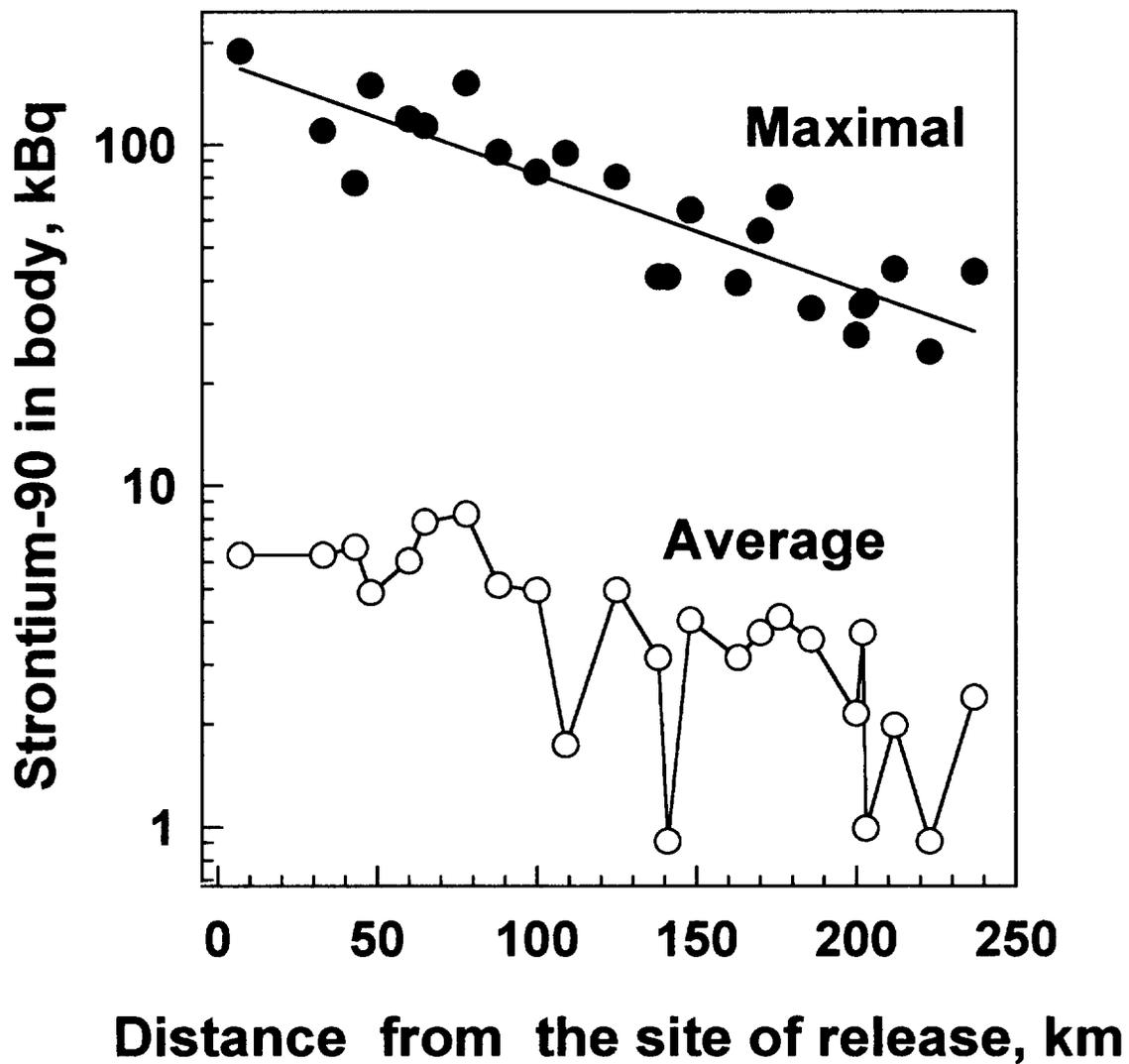


Fig. 3. Maximal and average values of  $^{90}\text{Sr}$ -body burden in different settlements on the Techa River as a function of distance along the river from the site of release.

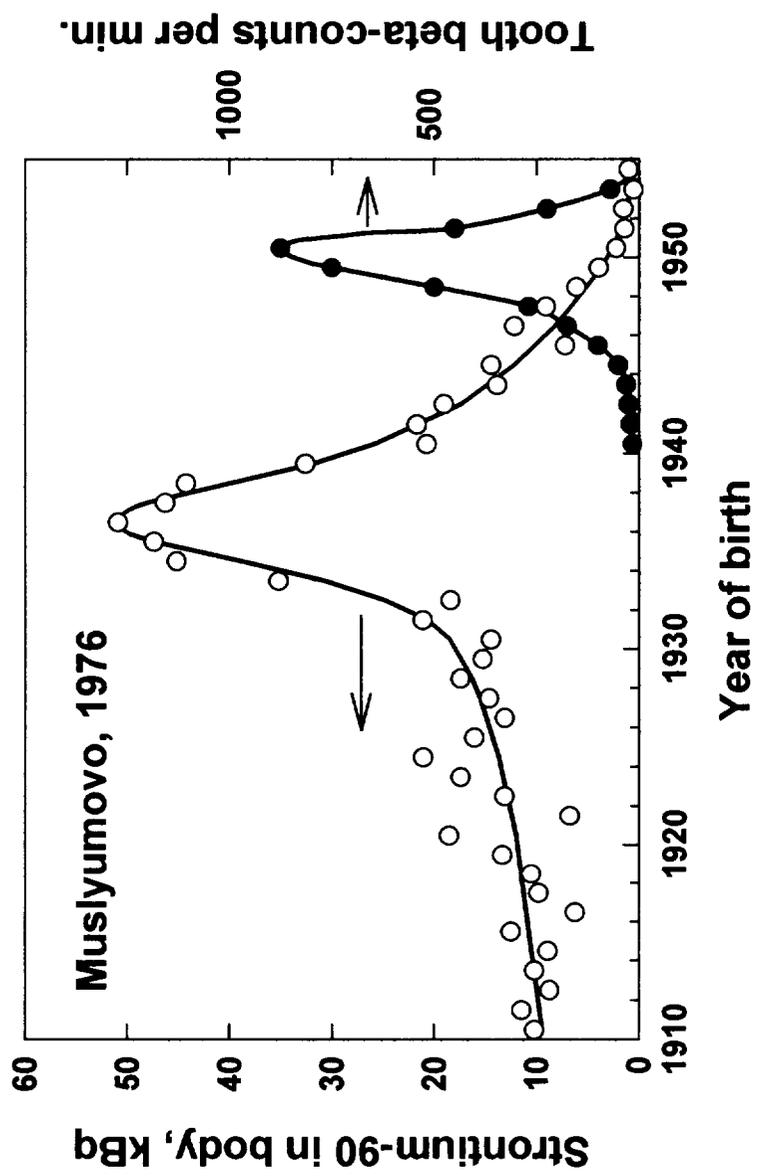


Fig. 4. Average values of WBC measurements (open circles) and tooth beta-count rates (closed circles) for different age cohorts of Muslyumovo residents in 1976. Left curve: model calculation on the basis of mean  $^{90}\text{Sr}$ -intake levels. Right curve: cubic spline fit to beta-count rates of teeth.

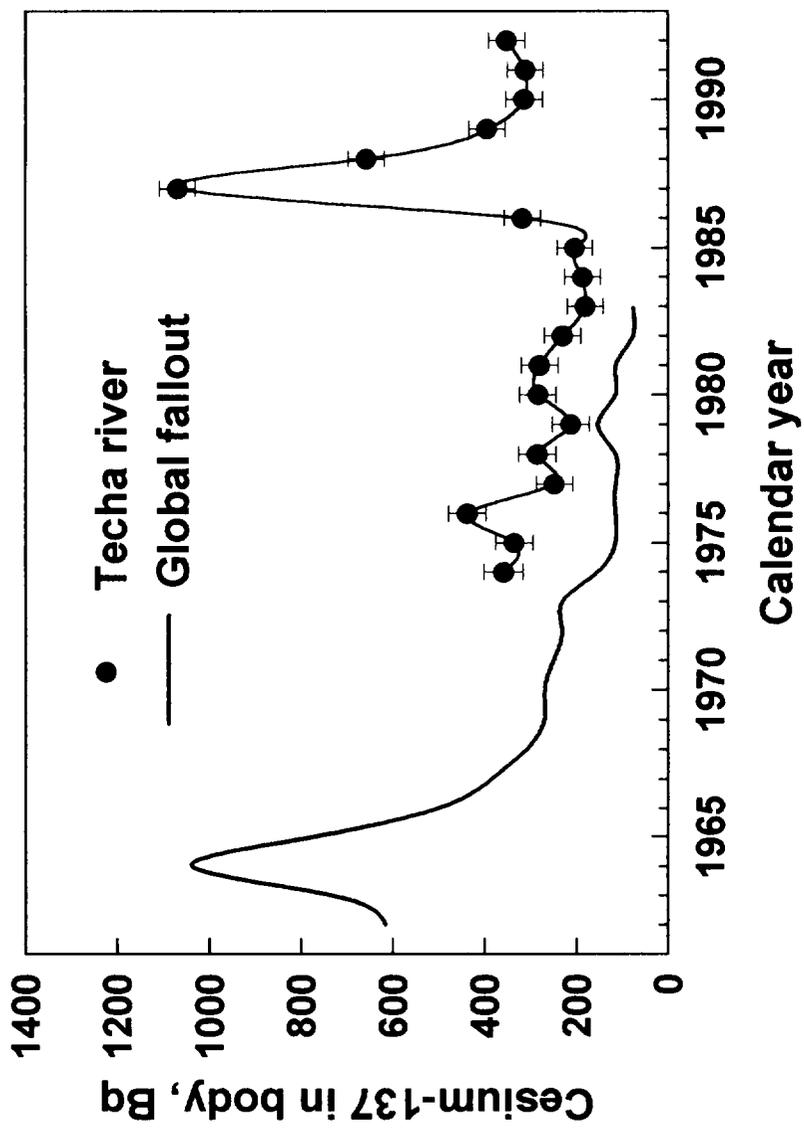


Fig. 5. <sup>137</sup>Cs-body burdens for adult residents of the upper and mid-Techa region in comparison with global fallout levels.

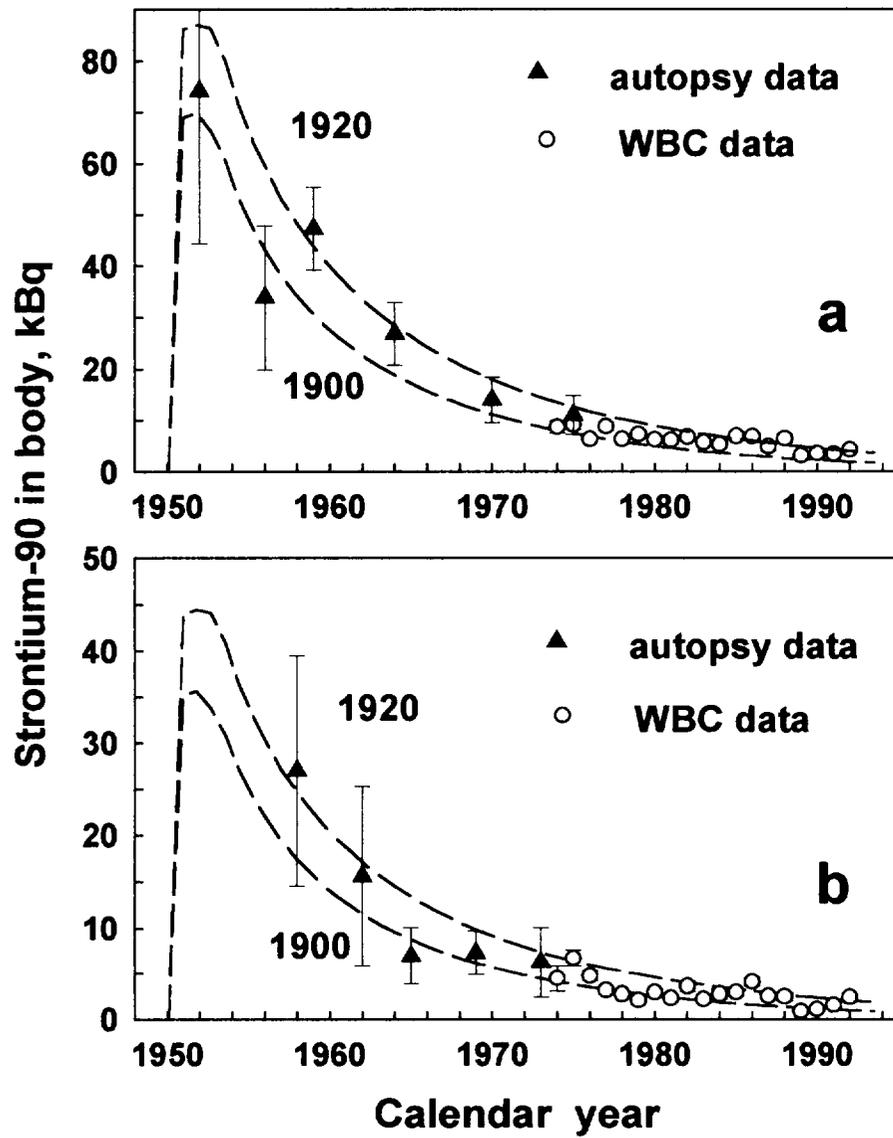


Fig. 6. Mean levels of  $^{90}\text{Sr}$  in adult humans for the Techa Riverside settlements (a) up to 80 km from the site of release and (b) for distances of 90-150 km. The model curves outline the corridor of values between age cohorts born in 1900 and 1920.

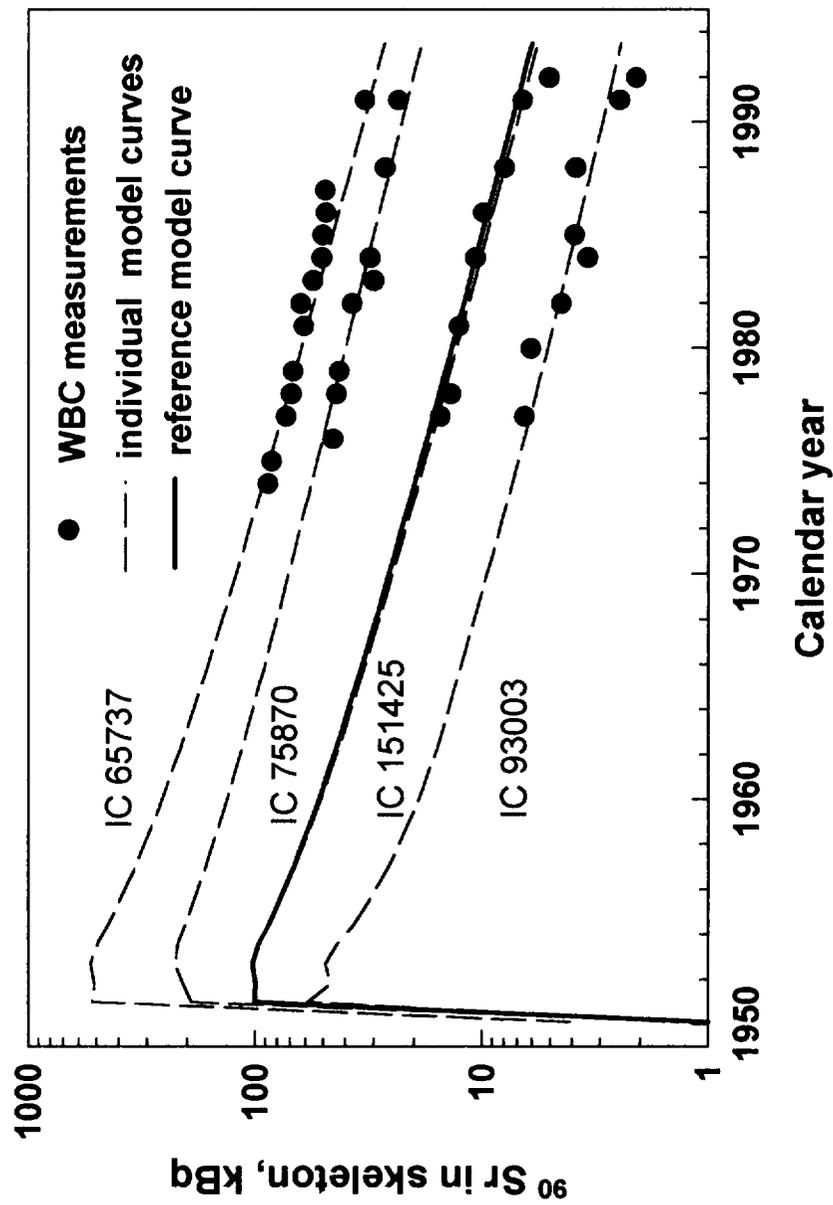


Fig. 7. Examples of reconstruction of  $^{90}\text{Sr}$  in skeleton on the basis of WBC data. The solid line represents a reference-model curve describing average  $^{90}\text{Sr}$  content from the onset of intake for the residents of the upper- and mid-Techa region. Dashed lines represent individual curves fitted to WBC measurements for four persons with corresponding Identification Codes (IC).

1.

Identification code	Date of birth	Vital status
9546	17.04.1938	Medical check-up in 1996

Residence history

Settlement	Period	Notes
Kasli	1938-1950	-
Metlino	1950-1954	Techa (7 km)
Kazhakup'	1954-1959	EURT (2Ci/km <sup>2</sup> )
Petrovsky	1959-1996	-

2.

Identification code	Date of birth	Vital status
65737	04.06.1928	Died in 1995

Residence history

Settlement	Period	Notes
Ibragimovo	1928-1953	Techa (54 km)
Muslyumovo	1953-1995	Techa (78 km)

Fig. 8. Two examples of individual-data sets for dose reconstruction.

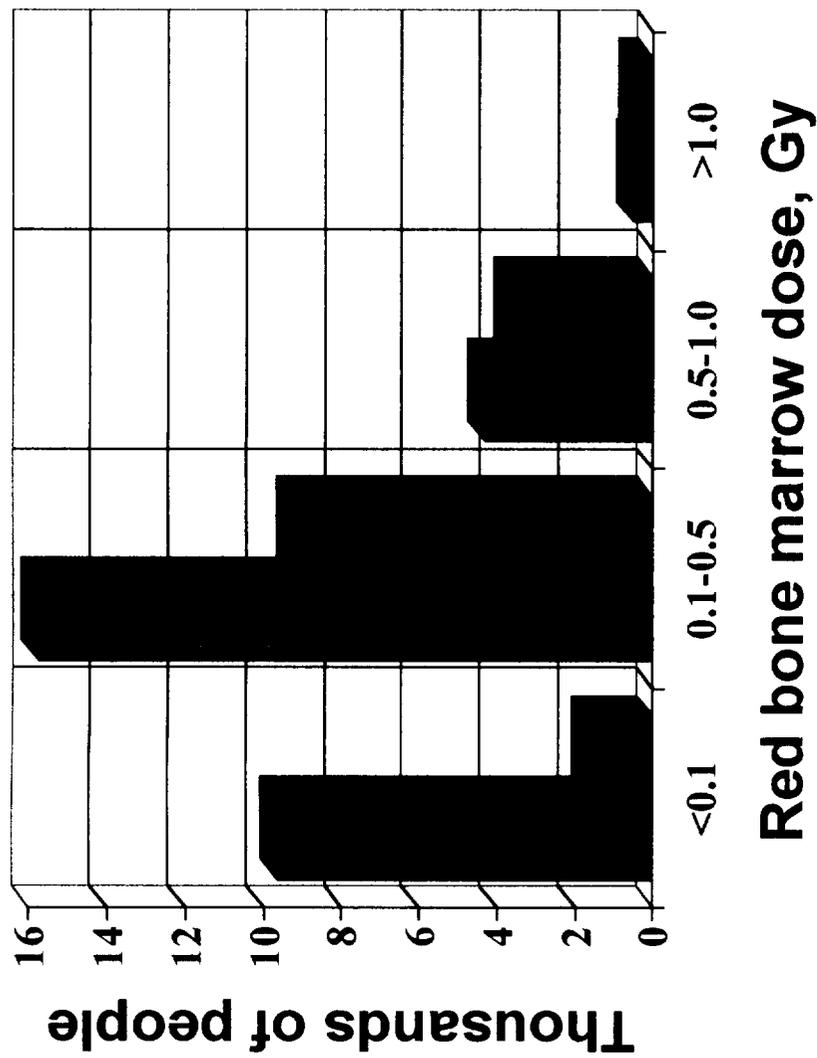
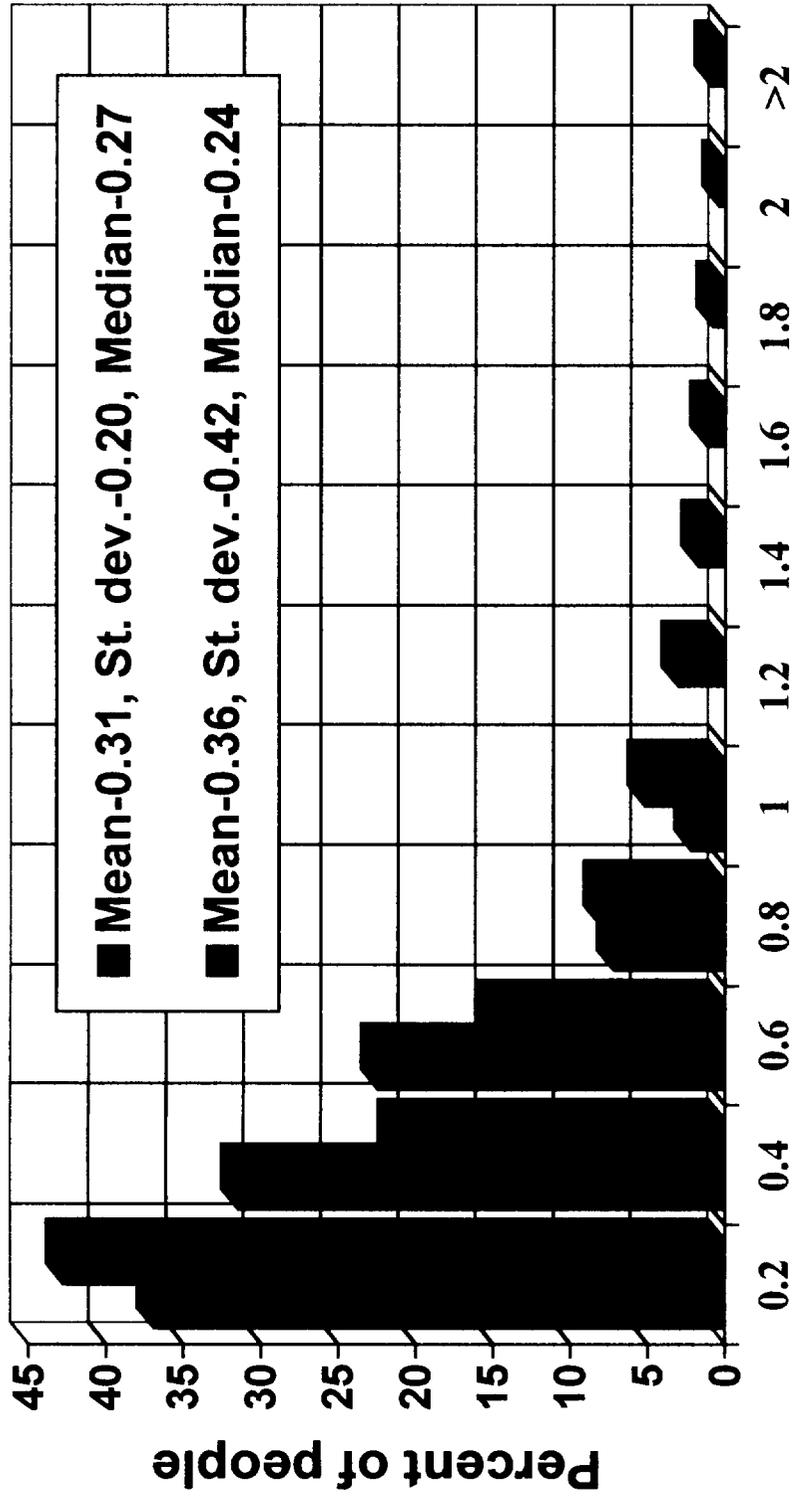


Fig. 9. Distribution of dose in red bone marrow (RBM) for 30,500 subjects who lived on the Techa River before 1960 (left, or blue columns) and for 14,500 permanent residents (right, or maroon, columns).



### Red bone marrow dose, Gy

Fig. 10. Distributions of dose to the red bone marrow (RBM) for the subcohort of permanent residents with WBC measurements. Left (or blue) columns: "residence history" approach. Right (or maroon) columns: "body burden" approach.

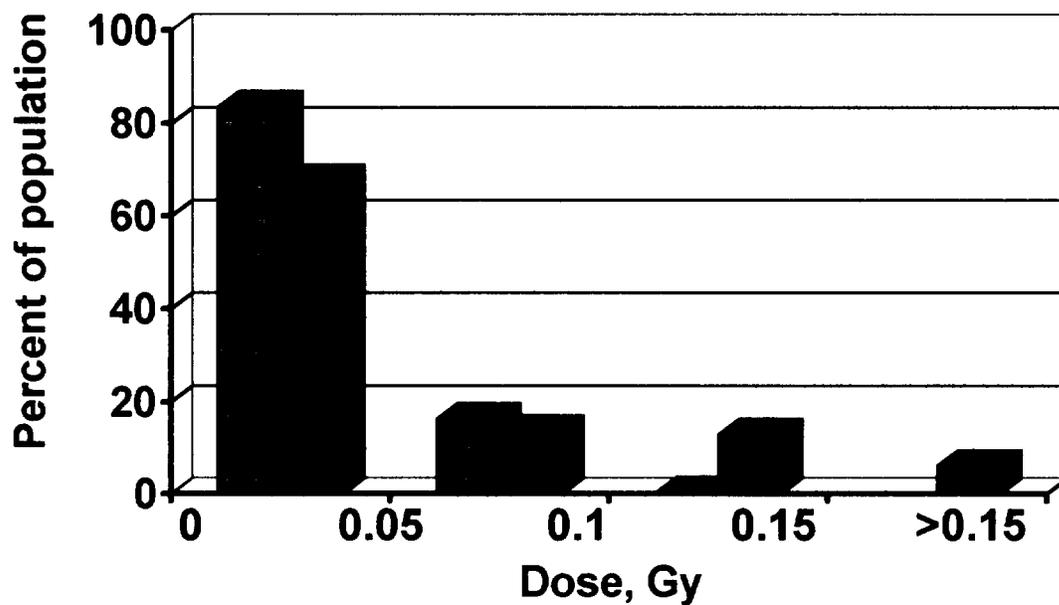
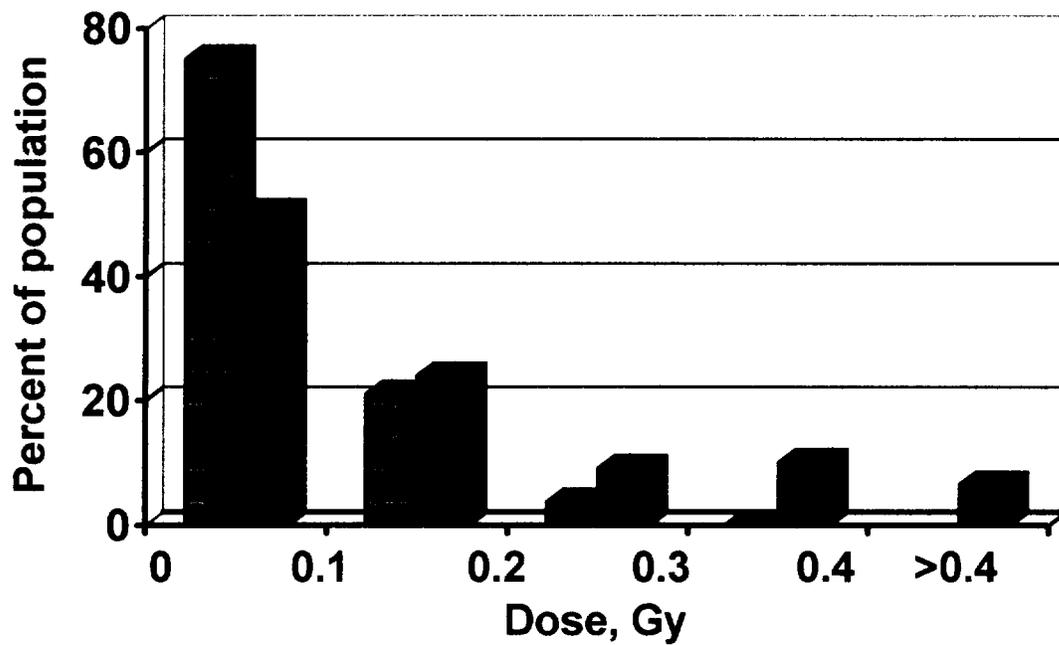


Fig. 11. Lower large intestine (LLI, upper panel) and upper large intestine (ULI, lower panel) dose distributions for the Techa River Cohort due to ingestion of all radionuclides (right, or maroon, columns) and due to ingestion of only  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$  (left, or blue, columns).

***Retrospective Dosimetry Using Electron Paramagnetic Resonance (EPR)  
and Thermoluminescence (TL) Techniques in Contaminated Areas of the  
Former Soviet Union.***

***Final Report of work performed at the Center for Applied Dosimetry,  
University of Utah under  
DOE Cooperative Agreement  
# DE-FC03-97SF21354***

***September 24, 1998***

***Submitted by Edwin H. Haskell***

***Retrospective Dosimetry Using Electron Paramagnetic Resonance (EPR) and Thermoluminescence (TL) Techniques in Contaminated Areas of the Former Soviet Union. Report on work performed at the Center for Applied Dosimetry, University of Utah.***

**Summary:**

Details of TL analyses appear in the March, 1998 progress report which together with a collaborative publication (Bougrov et. al, 1998) represents the scope of the TL effort. The remainder of this report addresses concerns which surfaced following work at the University of Utah laboratory by Dr. Alex Romanyukha of Ekaterinburg, Russia suggesting that carious teeth might show altered EPR dosimetric properties. The preliminary report of Dr. Romanyukha is included as Appendix I in this report. Measurements were made of additional Russian teeth by Mr. Robert Hayes. Those results revealed no systematic differences in the dose estimates of carious Russian teeth prepared with chemical preparation versus those analyzed with mechanical treatment only. These results were obtained with additive dose irradiations and native signal subtraction. In July of this year Dr. Sergey Sholom visited the University of Utah under NCI support for a three week period and completed analysis of carious and noncarious portions of Ukrainian teeth prepared by mechanical means. His results (Appendix II) revealed no adverse dosimetric effects of the carious portions versus the healthy portions of the teeth analyzed when mechanical preparation was employed. There was an increase in the dosimetric component of the EPR signal, but no significant differences in the sensitivity of the teeth or in the estimate of doses applied to the teeth before analysis. Because of higher random uncertainties associated with aggressive KOH preparation we recommend that analyses be done with mechanical separation followed by moderate KOH or NaOH treatment to removed impurities if indicated by the initial EPR spectrum.

During the visit of Dr. Alexander Romanyukha to the University of Utah earlier this year carious teeth from the Urals region of Russia were prepared and measured by EPR spectrometry. Questions arising from the analysis of the teeth were reported to EH-63 in January and are delimited in Appendix I. Subsequent to that visit we have completed full analysis of the original samples and have characterized additional samples in an attempt to clarify the origin of the effects observed. Those effects which were found to be reproducible are described in a paper which was submitted to the journal Applied Radiation and Isotopes. A manuscript of this paper is included as Appendix II and the main findings are summarized here.

Higher scatter in major EPR parameters (native signal intensities, native signal widths, dosimetric signal sensitivities and induction of an interfering signal near the measurement location of the dosimetric signal) was observed in carious samples aggressively treated with KOH (saturated solution, long duration, elevated temperature, hypersonic bath). The native signal intensities were greatly reduced with this treatment. The change in sensitivity of the dosimetric signal resulted in systematic errors in dose estimation when a standard calibration curve was used. The use of the additive dose method minimized errors associated with sensitivity changes but other uncertainties remained elevated.

We have continued this investigation using 11 additional teeth collected from the same region. These teeth were prepared solely by mechanical crushing followed by heavy liquid separation of the dentine and enamel. The excessive scatter in EPR parameters seen with chemical treatment was eliminated with this method of preparation. From this it was concluded that excessive KOH treatment can both randomly and systematically perturb the EPR spectral characteristics of tooth enamel. A preliminary copy of the report on this study is given as Appendix III and is intended to be included in the Ph.D. thesis of Mr. Robert Hayes.

A preliminary investigation of mechanically prepared healthy, precarious and carious portions of Ukrainian teeth was conducted by Dr. Sergey Sholom during his visit to the University of Utah in July of this year (Appendix IV) under NCI support. The results revealed no systematic changes in sensitivity for any portions of the teeth. An increase in the native signal with increasing carious content was observed, but no systematic effect was seen on the estimation of doses applied prior to analysis. Moderate treatment in KOH was found to reduce uncertainties in one tooth which exhibited an anomalous EPR signal with mechanical separation only. In summary, only the intensity of the native signal was seen to be effected and any modifications in native signal shape appeared to be insignificant. Additionally, KOH treatment when not used in an excessive manner was found to be useful for removing impurity signals which would have otherwise interfered with the dose reconstruction.

In a more detailed followup of the questions raised in the initial report (Appendix I), we will review each of the points made with a comment reflecting current status. Note that all of the more substantial findings and conclusions are detailed in the appendices.

(prior report comments numbered and given in plain text. Follow up comments based on subsequent research and analysis are printed in italics)

- 1) KOH has a more rapid and pronounced effect on "cleaning up" the enamel spectrum than does NaOH. This spectral cleaning is critical to spectral reproducibility at low doses.

*The results of Dr. Sholom indicated that mechanically prepared samples may be analyzed with reasonable accuracy at doses below 1 Gy. Moderate KOH treatment improves measurement on samples with impurities.*

- 2) KOH treatment can reduce and even eliminate the native signal. Removal of the native signal has long been thought to be one means of improving measurement accuracy.

*With significant reduction of the native signal there is also a tendency for reduction of the sensitivity of the dosimetric signal. Analysis of chemically treated versus mechanically treated Urals teeth revealed no systematic deviation in dose estimate for teeth of the same age suggesting that dose estimation is not effected by this sensitivity reduction (when measured using the additive dose method).*

- 3) Aggressive KOH treatment induces a signal at radiation measurement locations equivalent to approximately 300 mGy over a 70 hour period.

*This effect was found to be reproducible to within 30 to 40% when healthy American teeth were examined. It is possible that the signal may be due to the effects of the elevated temperature of the hypersonic bath treatment, the KOH or NaOH itself, or to an impurity present in the solutions. The issue has not yet been resolved.*

- 4) 70 hour KOH treatment causes an increase in sensitivity to radiation of approximately 15%.

*This was found to be an approximation only for the healthier teeth. Further analysis demonstrated that for certain massively carious teeth, the treatment may significantly decrease sensitivity.*

5) NaOH treatment induces a signal at the radiogenic signal's location equivalent to approximately 20mGy per hour of treatment.

*See comments above.*

6) The above effects are exaggerated when teeth with caries are treated.

*A higher scatter is found in the spectral characteristics of carious teeth treated with KOH. Sensitivity changes may differ for healthy versus carious teeth treated in KOH. The induction of a confounding signal has not been definitively quantified in healthy versus carious teeth.*

7) The native signal is broadened in teeth with caries.

*This effect appears to be an artifact of the KOH/hypersonic bath treatment.*

8) This effect extends beyond the region of visible disease into apparently healthy portions of the tooth.

*This has been demonstrated to be negligible when aggressive KOH treatment is not employed.*

9) Teeth with caries have a lower sensitivity to added radiation. One tooth examined had its sensitivity reduced by a factor of 6.

*This effect again appears to be due to the KOH treatment. No effect of caries on sensitivity was seen with mechanical treatment.*

10) Approximately 10% of the Urals teeth examined were unsuitable for analysis due to high caries content.

*Analysis of mechanically treated carious Urals samples and mechanically treated carious Ukrainian samples indicates that it may not be necessary to exclude such teeth.*

### **Previous and Present Conclusions and Recommendations**

1) The dosimetric effects of teeth with caries can be large. The effects extend beyond the visibly diseased regions.

*We have now found that the dominant effect of caries is on the intensity of the native signal when the KOH treatment is not employed. The shape of the native signal increases with proximity to a carious lesion but the effect of this on dosimetry seems to be minimal. Significant effects at low doses have not yet been ruled out.*

2) At the present time, measurements of teeth with extensive caries should not be used for dosimetry purposes. Teeth with caries from the exposed Mayak and Techa river populations should be archived until methods are developed for the analysis of such samples.

*Dosimetry using a calibration curve should not be attempted on these samples. Dosimetry using mechanical treatment and native signal subtraction should not result in significant systematic errors.*

- 3) Effects of tooth preservative methods (70% ETOH, 10% formaldehyde, etc) should be evaluated

*This conclusion should be expanded to include effects of hypersonic bath treatments alone.*

- 4) The extent to which the pre-carious portion of a tooth exhibits altered dosimetry properties should be examined. It may be possible to develop spectral screening procedures to identify pre-carious alterations by identifying spectral broadening (observed in this study) during analysis. This would be useful for pre-caries teeth with little or no visual indication of disease which may slip by a preliminary screening process.

*We have demonstrated that this is not necessary if a standard native signal subtraction method is employed without excessive KOH/hypersonic bath treatments.*

- 5) Systematic uncertainties due to sensitivity change and increase in radiation signal size may be introduced by the use of KOH and NaOH treatment. This effect should be examined further to determine the magnitude of the effect in a number of healthy and diseased teeth, and an optimal cleaning regime with minimal artifact introduction should be developed.

*Our preliminary study on healthy, precarious and carious portions of teeth indicates that mechanical preparation avoids or minimizes these effects. A detailed study such as that suggested above would be of great value to the dosimetric community since the majority of scientists use some variant of the alkali treatment in a hypersonic bath. We have since opted to use only mechanical crushing of a tooth crown followed by heavy liquid separation of the enamel from the dentine. Until the effects of NaOH, KOH and hypersonic bath treatment can be qualitatively delineated including the effects of temperature, time and concentration, the uncertainties of dose estimates obtained using chemical treatment will be elevated. If chemical treatment must be used we recommend short duration, moderate concentration (2N or less) and low temperature hypersonic treatment.*

- 6) Given that EPR dosimetry on tooth enamel is in a rapid state of development, non destructive methods should be used whenever possible so that samples can be reanalyzed as the technology advances. Development of a procedure allowing nondestructive measurements with correction for tooth to tooth sensitivity differences was recently proposed.

*The nondestructive analysis procedure which does not rely on a standard calibration curve is detailed in a paper submitted to the journal Applied Radiation and Isotopes and is included as Appendix IV. This method has been successfully carried out and demonstrated as shown in Appendix II.*

- 7) The use of non-destructive methods with a standard EPR sensitivity curve can produce large errors in teeth with caries. Non destructive methods must take into account individual sensitivities, alternatively, the uncertainties associated with those measurements should be more realistic estimated.

*It now appears that caries themselves are not the source for sensitivity variations but that preparation effects and possibly geographical factors are more important.*

- 8) All existing methods of spectral analysis should be examined and those most suitable for handling native signal variations should be compared for speed and accuracy of analysis.

*Dose estimates obtained with Dr. Sholom's samples were done using native signal subtraction of healthy Ukrainian teeth. The effects on dose estimation using other forms of analysis (differential microwave power method or signal deconvolution) is not known.*

## Appendix I

Effects of Caries and Sample Preparation Procedures on Accuracy of EPR Dose Measurements: Preliminary Results  
Report of visit by Dr. Alex Romanyukha to the Center for Applied Dosimetry of the University of Utah  
October 1997 to February 1998  
A. Romanyukha, E. Haskell, R. Hayes and G. Kenner

## Appendix II

EPR-dosimetry with carious teeth  
Preliminary report  
of visit by Dr. Sergey Sholom  
to the Center for Applied Dosimetry of the University of Utah  
July 11 - August 1, 1998.

## Appendix III

Variations in Tooth Enamel EPR Spectra  
Alexander A. Romanyukha, Robert B Hayes, Edwin H Haskell and Gerry H Kenner.  
Submitted to Radiation Protection Dosimetry

## Appendix IV

Verification of a New Method for Virtually Nondestructive Additive Dose EPR Dosimetry  
Robert B Hayes, Edwin H Haskell, Gerry H Kenner, Jeffrey K Barrus and Alexander A. Romanyukha. Manuscript in preparation.

## Appendix V

Preliminary report on the Development of a Virtually Non-Destructive Additive Dose Technique for EPR Dosimetry  
Edwin H. Haskell, Robert B. Hayes, Alexander A. Romanyukha, and Gerry H. Kenner.  
Submitted to Applied Radiation and Isotopes

## Appendix VI

Results of EPR measurements on teeth from the Techa River Control Population

## Appendix VII

Inventory of additional unmeasured teeth.

## Appendix VIII

Publications supported in full or in part by this contract.

## Appendix 1.

### Effects of Caries and Sample Preparation Procedures on Accuracy of EPR Dose Measurements: Preliminary Results

A. Romanyuhka, E. Haskell, R. Hayes and G. Kenner

In October, 1997, Dr. Alex Romanyuhka began a working visit to the University of Utah with the aim of 1) evaluating and gaining proficiency in the routine EPR procedure in us at the Utah laboratory and 2) measuring background dose in several dozen teeth obtained from potential background regions (Chelyabinsk and Kurgun Oblasts) in the South Urals between 50 and 100km from the Techa River. During this visit we discovered variations in the EPR measurement of radiation doses in background teeth from the Urals region of Russia beyond those expected from our previous experience. Closer examination revealed differences in the shape of the "native" signal of the Urals teeth relative to that of the standard background spectrum in use at the University of Utah for subtraction of the native signal during spectral analysis. It appeared that small variations in the shape of the native signals were causing errors in the dose estimations of up to several hundred mGy. We were also using a sensitivity standard appropriate for healthy U.S. teeth, but assumed that the accepted sensitivity variations between teeth of 10 to 15% would not be a major source of error .

We had used KOH with ultrasonic treatment for preparation of the Urals teeth (in a manner similar to that used by the Ukrainian Research Center for Radiation Medicine in Kiev), while our native background standard had been prepared using NaOH with ultrasonic treatment (the usual procedure at the University of Utah). It was impossible, therefore, to immediately identify the source of our errors as being the teeth, their less than healthy state, the sample preparation treatment, the sensitivity standard or some combination thereof. Although our immediate problem was largely rectified by development and application of a new background signal and measurement of individual sensitivities for each tooth examined, we felt that the variations we had observed required closer examination.

We broadened our research program from routine dose estimation of the 36 background teeth to an examination of variations in the sensitivities of the teeth, the magnitude of the spectral differences of the native signals, the effects on dose estimation and the sources of the spectral and sensitivity variations. We should point out that the Inco Copurnicus EPR program of the European Union had attempted to address the question of variation due to sample preparation and had not uncovered the types of problems which we were now observing. This was likely due to several factors; 1) we were not analyzing entirely healthy teeth, 2) analyses were now being made on a single spectrometer making direct spectral comparisons feasible, and 3) greater precision was possible using instrumentation and methodology for removal of sample anisotropies and dynamic instrumental variations. The latter improved reproducibilities by an order of magnitude allowing detailed examination of small spectral variations.

Our research program was designed to address the sample preparation methods currently in use by major laboratories: No treatment, Obninsk; KOH with ultrasound at 60°C, Kiev; NaOH with ultrasound at 60°C, Utah. It was assumed that any effects induced by NaOH treatments with ultrasound at approximately 40°C as done at GSF and Ekaterinburg would be revealed by treatment at the higher temperature of 60°C used here.

#### Research Program:

- 1) Comparison of changes (spectral and sensitivity) induced by chemical treatment. Methods were compared by treating separate portions of the same tooth in NaOH and KOH for varying periods of time. Results were compared against untreated portions.
- 2) Comparison of spectral parameters and sensitivities of healthy versus diseased (caries) teeth.
- 3) Relative effects of chemical treatments on healthy versus diseased teeth.

## Summary of Results:

- KOH has a more rapid and pronounced effect on "cleaning up" the enamel spectrum than does NaOH. This spectral cleaning is critical to spectral reproducibility at low doses.
- KOH treatment can reduce and even eliminate the native signal. Removal of the native signal has long been thought to be one means of improving measurement accuracy.
- KOH treatment induces a signal at radiation measurement locations equivalent to approximately 300mGy over a 70 hour period.
- 70 hour KOH treatment causes an increase in sensitivity to radiation of approximately 15%.
- NaOH treatment induces a signal at the radiogenic signal's location equivalent to approximately 20mGy per hour of treatment.
- The above effects are exaggerated when teeth with caries are treated.
- The native signal is broadened in teeth with caries.
- This effect extends beyond the region of visible disease into apparently healthy portions of the tooth.
- Teeth with caries have a lower sensitivity to added radiation. One tooth examined had its sensitivity reduced by a factor of 6.
- Approximately 10% of the Urals teeth examined were unsuitable for analysis due to high caries content.

## Conclusions and recommendations

1. The dosimetric effects of teeth with caries can be large. The effects extend beyond the visibly diseased regions.
2. At the present time, measurements of teeth with extensive caries should not be used for dosimetry purposes. Teeth with caries from the exposed Mayak and Techa river populations should be archived until methods are developed for the analysis of such samples.
3. Effects of tooth preservative methods (70% ETOH, 10% formaldehyde, etc) should be evaluated
4. The extent to which the pre-carious portion of a tooth exhibits altered dosimetry properties should be examined. It may be possible to develop spectral screening procedures to identify pre-carious alterations by identifying spectral broadening (observed in this study) during analysis. This would be useful for pre-carious teeth with little or no visual indication of disease which may slip by a preliminary screening process.
5. Systematic uncertainties due to sensitivity change and increase in radiation signal size may be introduced by the use of KOH and NaOH treatment. This effect should be examined further to determine the magnitude of the effect in a number of healthy and diseased teeth, and an optimal cleaning regime with minimal artifact introduction should be developed.
6. Given that EPR dosimetry on tooth enamel is in a rapid state of development, non destructive methods should be used whenever possible so that samples can be reanalyzed as the technology advances. Development of a procedure allowing nondestructive measurements with correction for tooth to tooth sensitivity differences was recently proposed to EH-63. (see attachment).
7. The use of non-destructive methods with a standard EPR sensitivity curve can produce large errors in teeth with caries. Non destructive methods must take into account individual sensitivities, alternatively, the uncertainties associated with those measurements should be more realistic estimated.
8. All existing methods of spectral analysis should be examined and those most suitable for handling native signal variations should be compared for speed and accuracy of analysis.

## Appendix II

### *EPR-DOSIMETRY WITH CARIOUS TEETH*

#### *Preliminary report*

of Sergey Sholom's visit  
to the Center for Applied Dosimetry of the University of Utah

July 11 - August 1, 1998.

#### DESIGN OF EXPERIMENT

Four carious and two healthy control teeth were chosen for this study. Three of the carious teeth were irradiated before any treatment with doses of 250, 600 and 1000 mGy respectively. These doses were considered as an analog of an "unknown" accidental dose. For the first step enamel was carefully separated from dentine with a stomatological drill and divided into three aliquots: carious, non-carious and intermediate. Several pictures were taken at this time for each tooth and each aliquot. A representative specimen is shown in Fig. 1. Pictures **a** and **b** correspond to different projections of the whole tooth, while picture **c** shows carious (upper four) and non-carious (bottom four) pieces of enamel from this tooth. White areas of enamel can be seen in these pictures which correspond to the carious-affected parts of the tooth.

Aliquots of 100-mg each were then taken from each healthy tooth and from the carious, non-carious and intermediate portions of the carious teeth. These aliquots were used for recording initial EPR spectra for studying the native and small-size dosimetric signals.

Aliquots of 20-40-mg from each tooth were used for determining radiation sensitivity after irradiation with a dose of 10 Gy using a Co-60 irradiator at the Center for Applied Dosimetry.

Prior to EPR measurements of the dosed samples, the samples were annealed at 95° C for 2 hours (Sholom et al., 1998).

Spectral acquisition was done on a Bruker Model ESP 300E spectrometer. Measurement parameters were the same as those used for routine EPR-dosimetry with teeth at the Center for Applied Dosimetry. Each sample was measured three times with shaking between each measurement. A constant rotation goniometer was used during acquisition (Haskell et al., 1997). Average values of the three measurements were used for the data plots.

#### RESULTS AND DISCUSSION

The intensities of native signals measured for the different aliquots of carious teeth are shown in Fig.2. For comparison, the same signal intensity that corresponds to healthy teeth is also shown. It is possible to see that the native signal decreases from a maximum value which is observed in carious aliquots to a minimum in non-carious aliquots. The reason of this dependence is the higher organic component in the carious portions. Only minor differences were seen in the shape of the native signals for the different aliquots.

The sensitivities of the above aliquots were measured after a 10 Gy gamma dose (Fig. 3). There is only a small difference (within 10%) between the sensitivities of the different aliquots of the carious teeth. These results reveal no significant problems with the use of carious teeth for EPR dosimetry.

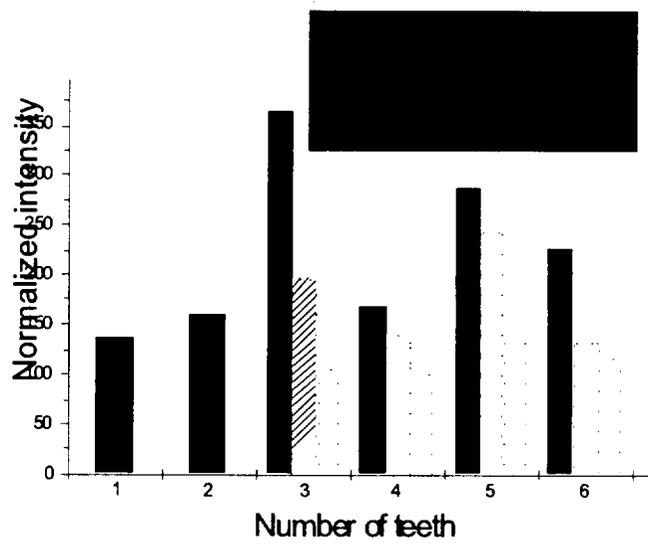


Fig. 2. Native signal intensity in the healthy teeth (number 1 and 2) and different aliquots of carious teeth (number 3-6).

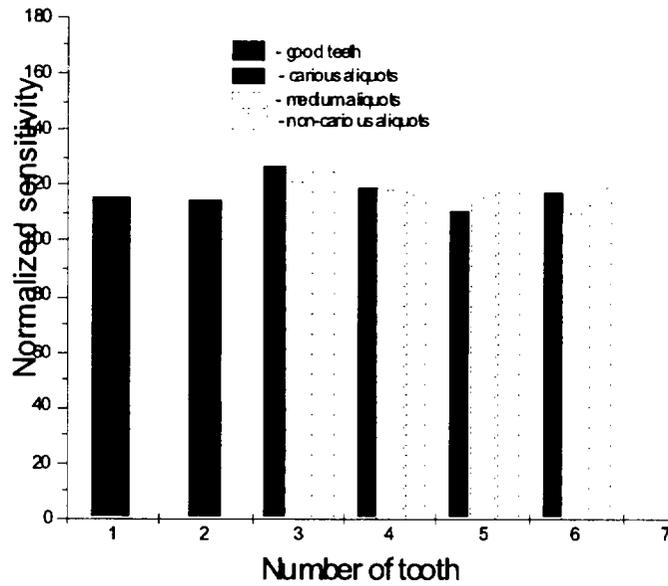


Fig. 3 Radiation sensitivity of the healthy teeth (number 1 and 2) and different aliquots of carious teeth (number 3-6).

This was verified by the results of dose reconstruction for carious teeth number 3-5 (number 6 didn't have an initial dose). These results are shown in Table 1, where the values of nominal (applied) doses for each sample are also indicated. It is possible to see from the data of this table that in general we have very good correlation between both values even for carious aliquots. There are two exceptions: one for a nominal dose of 250mGy, and one for 1000 mGy, both using the medium quality aliquots. The first is explained by the presence of an some impurity that produces an additional signal overlapped with the dosimetric signal of enamel. Applying chemical treatment with KOH to that sample, we successfully removed this signal and obtained new values for the reconstructed dose of approximately 287 mGy, in better agreement with the applied dose. The reason for the discrepancy with the 1000 mGy medium quality aliquot remains unclear. A possible explanation is that native signal standard used in this study does not match the native signal component for this aliquot after irradiation. Statistical fluctuation due to the small number of samples is another possibility.

### CONCLUSION

This study supports the use of carious teeth for purposes of EPR dosimetry. Lowest uncertainties, however, were obtained with the non-carious aliquots (less than 30 mGy for reconstructed doses). The only significant effect of carries seen in this study is an increase in native signal intensity. Such an increase should have no systematic effect on dose assessment.

### Reference

- Haskell E. H., Hayes R. B. and Kenner G. H. (1997) Improved accuracy of EPR dosimetry using a constant rotation goniometer. *Radiat. Measur.* **27**, 325-329.
- Sholom S. V., Haskell E. H., Hayes R. B., Chumak V. V. and Kenner G. H. (1998a) Influence of crushing and additive irradiation procedures on EPR dosimetry of tooth enamel. *Radiat. Measur.* **29**, 105-111.

Table 1. Doses reconstructed by using of different aliquots of teeth.

Number of tooth	Characteristic of aliquot	Reconstructed value, mGy	Error, $\pm\sigma$ , mGy	Nominal value, mGy
3	Carious	261	44	250
	Medium	411	25	250
	Non-carious	260	25	250
4	Carious	535	48	600
	Medium	546	137	600
	Non-carious	591	51	600
5	Carious	939	79	1000
	Medium	858	111	1000
	Non-carious	974	54	1000

### Appendix III

#### VARIATIONS IN TOOTH ENAMEL EPR SPECTRA

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#### Abstract

The presence of stable radiation induced radicals in the mineral component of tooth enamel allows use of this material as a biological dosimeter. Estimation of the dose absorbed in tooth enamel can be done by EPR. Generally, for the purpose of dose reconstruction, the EPR spectrum of tooth enamel is interpreted in terms of two main components. The first is a broad background signal often called the native signal centered at a g-value of 2.005. The origin of this signal is not precisely known. The second main component in the tooth enamel spectrum ( $g_{\perp}=2.0018$ ,  $g_{\parallel}=1.9985$ ) is purely radiation induced and can be used for retrospective dosimetry. Internal structure of the native signal and variations of its amplitude and linewidth were investigated for the samples prepared from modern teeth obtained from different geographic locations (USA and Russia). Possible reasons for the variations observed are discussed as are the potential effects of the variations on the reliability of dose estimation.

## INTRODUCTION

It is well known that tooth enamel can be used as an *in vivo* dosimeter for retrospective Electron Paramagnetic Resonance (EPR) dosimetry<sup>(1)</sup>. This is done by taking EPR measurements of the stable radiation induced radicals in tooth enamel. Tooth enamel is composed of hydroxyapatite crystallites bound by an organic matrix. Chemically, tooth enamel is about 97% carbonated hydroxyapatite ( $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ ) 2% water and 1% organic component (mostly protein)<sup>(2)</sup>. There are also considerable amounts of various impurities in tooth enamel, most important are 0.67% Na, 0.35% Mg, 0.35% Cl, 0.04% K<sup>(2)</sup>. During the mineralization process of tooth enamel, carbonate ions are incorporated into the hydroxyapatite. Structurally tooth enamel consist of hydroxyapatite needle crystallites with lengths approximately 500-600 nm dispersed in an aqueous-organic gel. A typical EPR spectrum of irradiated tooth enamel is depicted in Fig. 1. The term "typical" is used because tooth enamel is a human tissue and there are variations in its chemical composition depending on certain individual peculiarities like a diet, environmental pollution, dental diseases etc. To investigate these dependancies, modern teeth originating from Russia (Urals) having dental diseases were compared with healthy wisdom teeth from the USA (Utah).

## EXPERIMENTAL

There were two portions of teeth investigated in the framework of the present study. The first 17 teeth were collected in the Southern Urals area of Russia. This region of Russia was the site of serious radiation accidents caused by the first Soviet industrial nuclear facility Mayak<sup>(3,4)</sup>. However, the villages where the teeth were collected for the present study are located outside of the main radioactively polluted area. Therefore, this Southern Urals population group can be considered as a background for Mayak nuclear workers and the Techa riverside residents.

All collected teeth were extracted because of different dental diseases by dentists at the dental clinic of the village of Kunashak, Chelaybinsk region. Generally, there were two types of diseased teeth. The first type included carious teeth and the second type included those extracted because of periodontal disease. Periodontal disease is not a disease of the teeth themselves but rather of the gums. The teeth in this category had more or less sound tooth enamel. An additional number of teeth (four) were collected in Salt Lake City, Utah. They were healthy deciduous (wisdom) teeth extracted according to standard dental practices in the USA.

The procedure of sample preparation for EPR measurements consisted of the following steps:

- 1) Cutting off the root from the crown;
- 2) Crushing the crown into 3-4 pieces with a porcelain mortar and pestle;
- 3) Double ultrasonic treatment with supersaturated KOH aqua solution at 60°C. The first treatment was for 600 min. and the second for 500 min. For the second treatment, fresh KOH solution was used. Between the first and second treatments, the sample was washed with distilled water 4 times by filling, shaking and decanting.
- 4) Final washing of the samples with distilled water 5 times at 1/2 hour each in the ultrasonic bath with fresh water replacement being done each time.
- 5) Drying in a desiccator for 1 hour at 60 °C.
- 6) Final sample crushing in a mortar with pestle followed by sample sieving to grain sizes of 250-850  $\mu\text{m}$ .

The spectrometer parameters used for the study were, 0.5 mT modulation amplitude, 100 kHz modulation frequency, 25 mW microwave power, 10.0 mT scan width, 30 accumulations per spectrum, 1024 field point resolution, 41 ms time constant and conversion time. To average the anisotropy of the EPR tooth enamel spectrum a special goniometer connected with a stepper motor was used<sup>(5,6)</sup>. Subtraction of the empty EPR tube signal from the tooth enamel spectrum was made to eliminate contributions of the EPR signals from the empty tube and the microwave cavity (see for details<sup>(5,6)</sup>). Thus spectral acquisition was done in three steps: scan empty EPR tube signal with 15 sweeps; scan sample with 30 sweeps; scan empty EPR tube again with 15 sweeps. Finally, the sum of the 2 empty EPR tube scans (15+15 sweeps) was subtracted from the sample spectrum (30 sweeps).

Calibration of the EPR radiation response was made by irradiating the tooth enamel samples with 1 Gy from a  $^{60}\text{Co}$  source<sup>(7)</sup>. To estimate the experimental error (scattering) in the reconstructed dose, EPR spectra were taken 2 to 3 times for every sample before and after irradiation.

For doses below 200-300 mGy, the native signal obscures the dosimetric signal preventing its direct measurement. There are three main approaches to solving this problem: 1) subtraction of the native signal using a standard spectrum constructed from specially prepared non-irradiated tooth enamel samples or dentin<sup>(8-10)</sup>, 2) computer simulation of the tooth enamel

spectrum<sup>(11-13)</sup> and 3) selective microwave saturation of the native component<sup>(14)</sup>. In the present study the first approach was used. Details of the method applied can be found in<sup>(5,6)</sup>.

## RESULTS AND DISCUSSIONS

The tooth enamel samples prepared from American teeth showed very similar EPR spectra without marked differences from sample to sample. However, the samples prepared from Russian teeth demonstrated considerable variation of the EPR spectral shape. Some typical examples of the EPR spectra observed are given in Fig. 2. All presented spectra are normalized by sample mass. It is easy to see from Fig. 2 that the native signal for the Russian teeth is much weaker than for the American teeth. For some samples like R14 the native signal is practically absent. Fig. 3 demonstrates variation of the radiation response of the samples investigated. Again for the American teeth the native signal has a higher intensity than the Russian samples. However, resolution of the dosimetric signal for the Russian teeth is much better due to the weak native signal.

A first approximation of the EPR spectral variation of the tooth enamel (Fig. 1) can be made on the basis of its two main components. The first is the native signal which is a broad background component centered at  $g=2.005$  and having a linewidth of 0.8-1.0 mT. The origin of this signal is not precisely known but is generally attributed to the organic matrix of the tooth enamel. The EPR parameters of the native signal do not strongly depend on the absorbed dose<sup>(15)</sup>. The second main component in the tooth enamel spectrum is purely radiation induced. This signal has axial g-factor anisotropy lineshape ( $g_{\parallel}=2.0018$ ,  $g_{\perp}=1.997$ ) with linewidth 0.4 mT. In 1987, Callens et al.<sup>(16)</sup> demonstrated that this dosimetric signal can be attributed mainly to  $\text{CO}_2^-$  radicals located at the crystal surface. They also demonstrated that there are also contributions from  $\text{CO}_2^-$  radicals localized in the  $\text{PO}_4^-$  and  $\text{OH}^-$  sites of hydroxyapatite lattice and some other carbonate radicals including  $\text{CO}_3^{3-}$ ,  $\text{CO}_3^-$  and  $\text{CO}^-$ .

Three parameters were used for the quantitative estimations in the framework of the present paper. For the native signal the peak-to-peak linewidth and peak-to-peak amplitude normalized on the sample mass. For the dosimetric signal, we used the radiation sensitivity, e.g. the EPR response per mg of material per mGy of absorbed dose. The resulting variations in the chosen parameters are shown in Fig. 4.

From the data given in Fig. 4, the average amplitude of the native signal for the Russian teeth is almost a factor of 3 lower than this parameter for American teeth. In fact, for samples 13, 14, 25, 28, 37 and 52 the average native signal was about 6 times lower. One possible reason for this effect is a weak organic matrix of the Russian diseased teeth which would be more easily removed by the applied caustic chemical treatment relative to that of the healthy American teeth. On the other hand, the observed variation in the native signal intensity for the Russian teeth is likely due to variations in the extent and type of dental disease.

The variation of the native signal linewidth (Fig. 4) suggests the presence of at least two spectral components of the native signal. One being a symmetric singlet with a linewidth of 0.75 mT and another being a singlet shifted by  $\Delta g=0.001$  having a linewidth of about 1.5 mT. Such parameters are in reasonable agreement with the parameters from the model suggested by Jonas<sup>(12)</sup>:  $\Delta H_1=0.8$  mT,  $g_1=2.002$ ,  $\Delta H_2=1.8$  mT,  $g_2=2.007$ . The first spectral component of the native signal can be virtually eliminated via chemical treatment (ultrasonic bath + 60°C + KOH) and probably originates from the organic component of the tooth enamel. This assessment is in agreement with studies showing that in deorganified bone and dentin the narrow component of the native signal can almost entirely be removed<sup>(17,18)</sup>. The narrow component of the native signal dominates the EPR spectrum. The broad component likely has another origin.

The variations found in the shape and structure of the native signal make it difficult to construct a native signal standard for reconstruction of low doses. Thus, it limits the application of this approach which, is being used by many investigators<sup>(19,20)</sup>. Similarly, the relative variation of the sample sensitivities (Fig. 4A vs. Fig. 4B) would argue strongly for the need to do individual sample radiation sensitivity calibrations. Calibration curves (average radiation sensitivity) are also used by many investigators<sup>(19)</sup>.

The radiation sensitivity of the Russian teeth varies much more than the American teeth and is generally lower (Fig. 4A&B). The radiation sensitivity of teeth is determined by carbonate concentration in hydroxyapatite<sup>(21)</sup>. It is known<sup>(2)</sup> that during the development of caries, the carbonate concentration is dynamic. Therefore the low radiation sensitivity of the caries teeth is not unexpected. However, for the teeth extracted because of periodontal disease the cause is not so clear. It could be that the carbonate concentration in Russian teeth is generally lower than that in American teeth. Another

possibility is that for Russian teeth, the carbonate content decreases with time or age relative to the American teeth which were extracted shortly after eruption (the Russian teeth were kept in-vivo for many decades).

The question now arises of what value these effects are concerning dose reconstruction? The answer is not obvious. If, for example, the radiation dose was obtained before the carbonate concentration changes occurred due to the disease, then the radiation sensitivity of the healthy teeth should be used. *Vice versa* if the radiation dose was obtained already after the caries process began in the tooth enamel, then the current radiation sensitivity should be used. This seems intuitive but should be independently verified.

Another important conclusion concerning the role of the native signal can be made on the basis of our results. The  $\text{CO}_2^-$  anisotropic signal in tooth enamel is known to be a biomarker of the dose absorbed. We have found that the native signal is likely a biomarker of the pathological processes that have occurred in tooth enamel. Its intensity and linewidth may describe obscured caries processes in tooth enamel and may help to make appropriate choices for the radiation sensitivity to be used in dose reconstruction.

#### ACKNOWLEDGEMENTS

The authors are grateful to Dr. Marina Degteva for organization of the sample collection at the Southern Urals and Dr. David Smoot of the Salt Lake City FHP (Family Health Plan) Hospital who furnished the U.S. wisdom teeth used in this study. This work was supported by DOE contract DE-FC03-97SF21354.

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## Figure legends

**Fig. 1.** Typical EPR spectrum of tooth enamel irradiated with 3 Gy. Dosimetric signal is shown by a thick line. R is a measure of the dose absorbed. The Native signal is shown by a thin line.

**Fig. 2.** EPR spectra of different tooth enamel non-irradiated samples. All spectra are normalized by mass. From the top, spectra of samples R14, R22, A3 are plotted, respectively.

**Fig. 3.** EPR spectra of different tooth enamel samples irradiated with a 1 Gy dose. All spectra are normalized by mass. From the top, spectra of samples R14, R22, A3 are plotted, respectively.

**Fig. 4.** EPR variations in the spectral parameters of tooth enamel. Figure 4A shows the values for the Russian samples. Figure 4B shows the comparable values for the American samples.

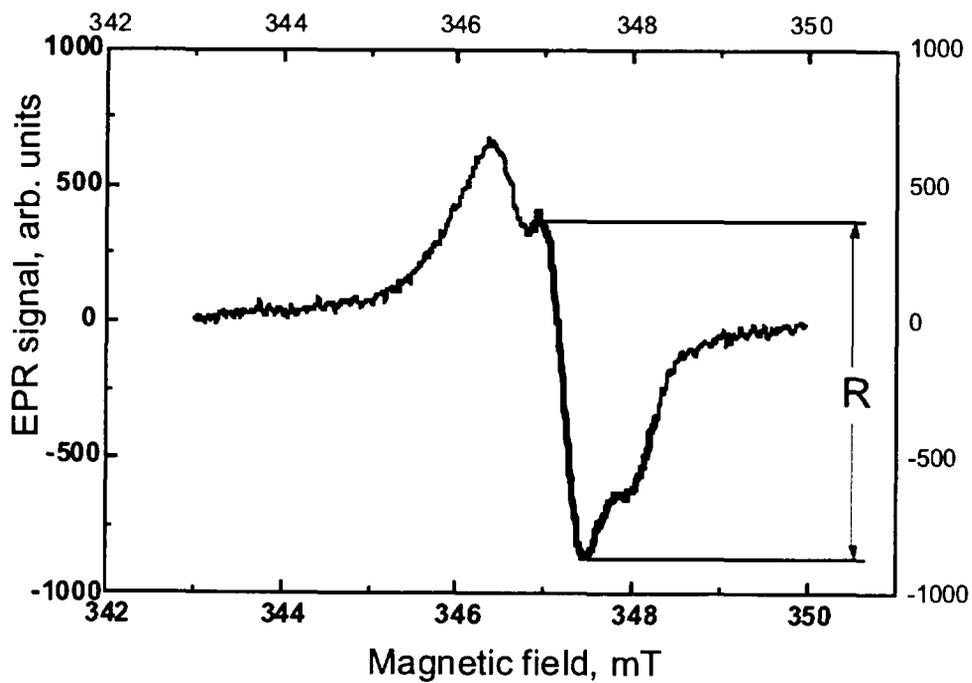


Fig. 1. Typical EPR spectrum of tooth enamel irradiated with 3 Gy. Dosimetric signal is shown by a thick line. R is a measure of the dose absorbed. The Native signal is shown by a thin line.

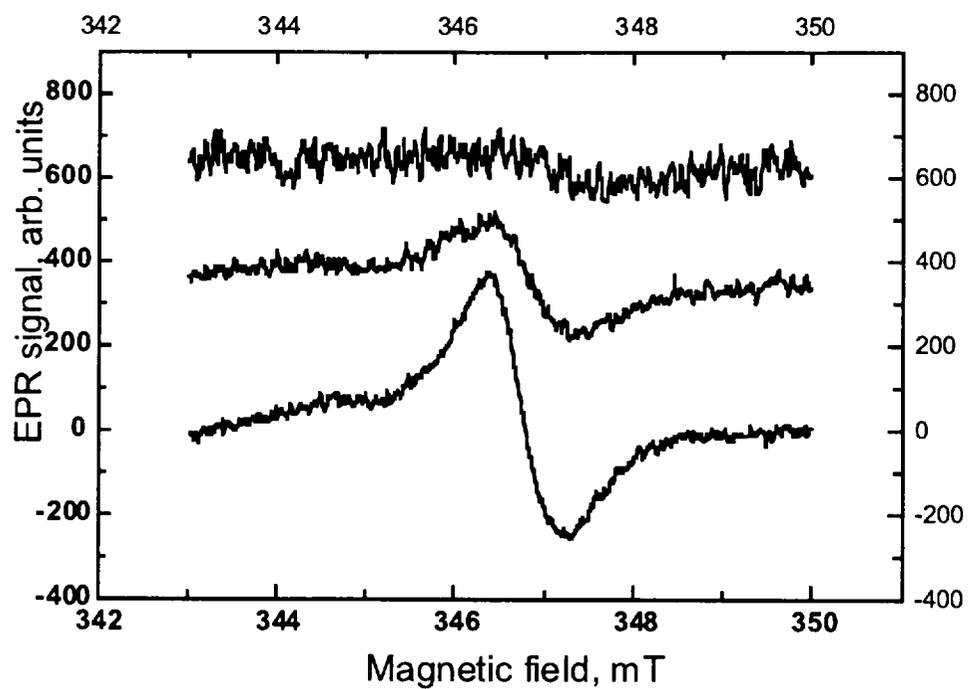


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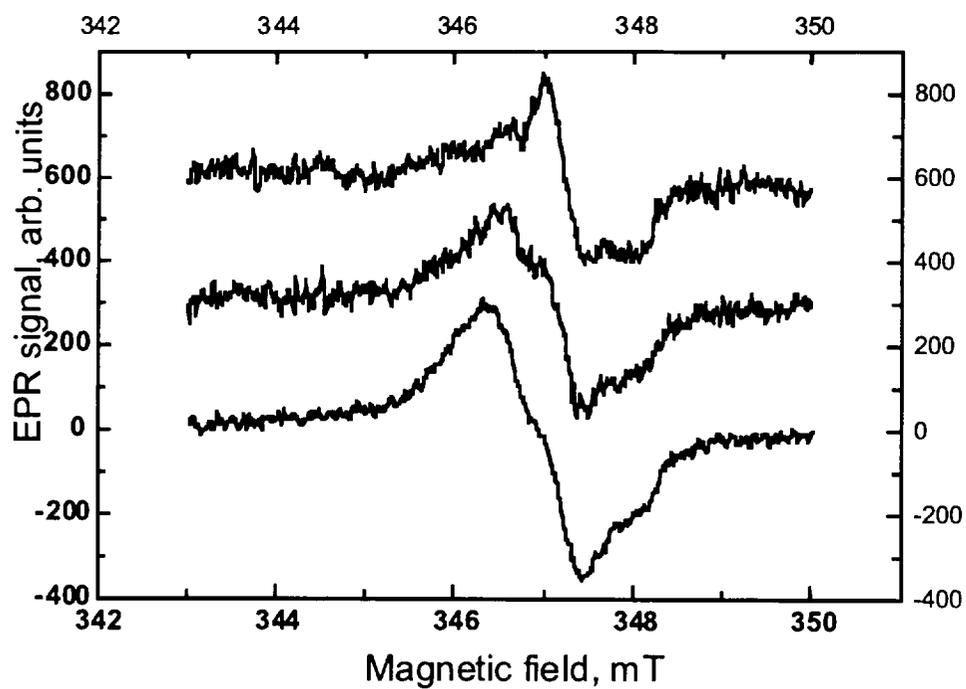
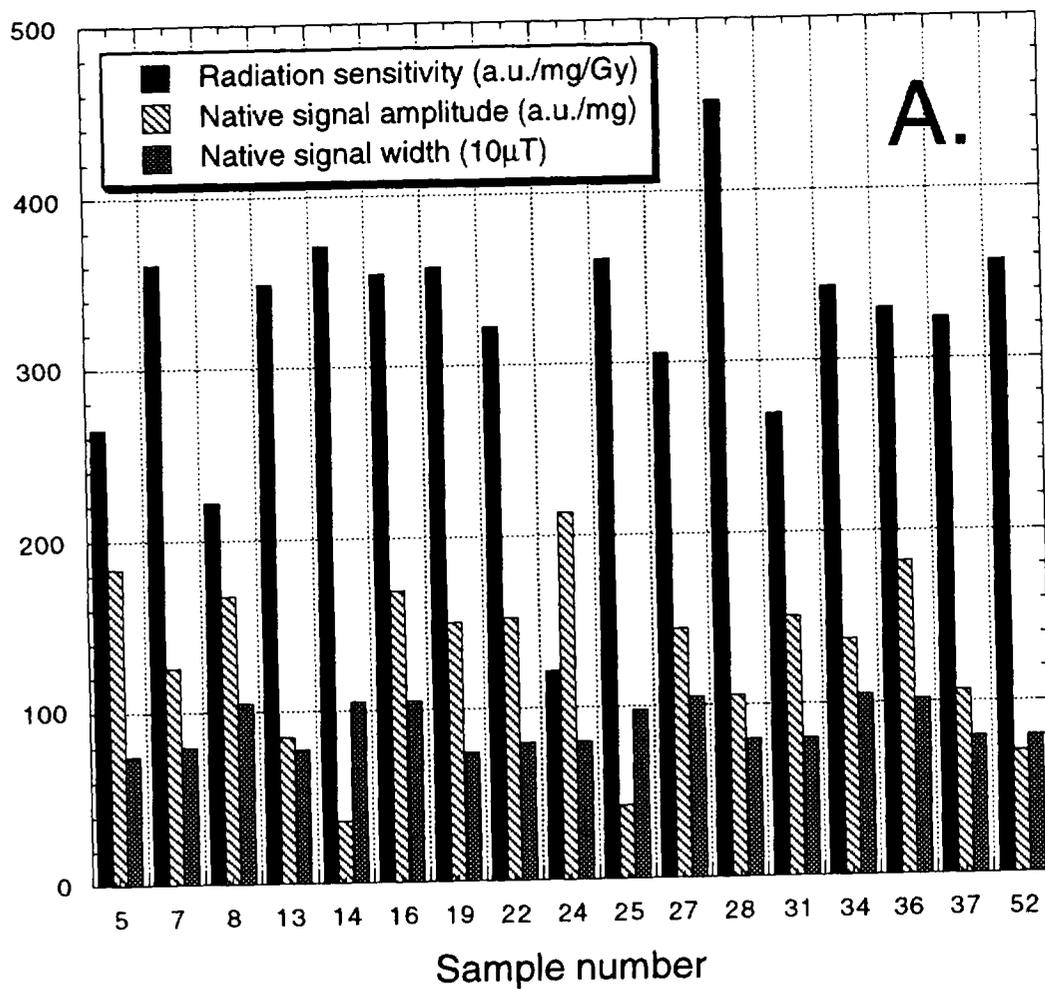


Fig. 3. EPR spectra of different tooth enamel samples irradiated with a 1 Gy dose. All spectra are normalized by mass. From the top, spectra of samples R14, R22, A3 are plotted, respectively.

Fig. 4A.



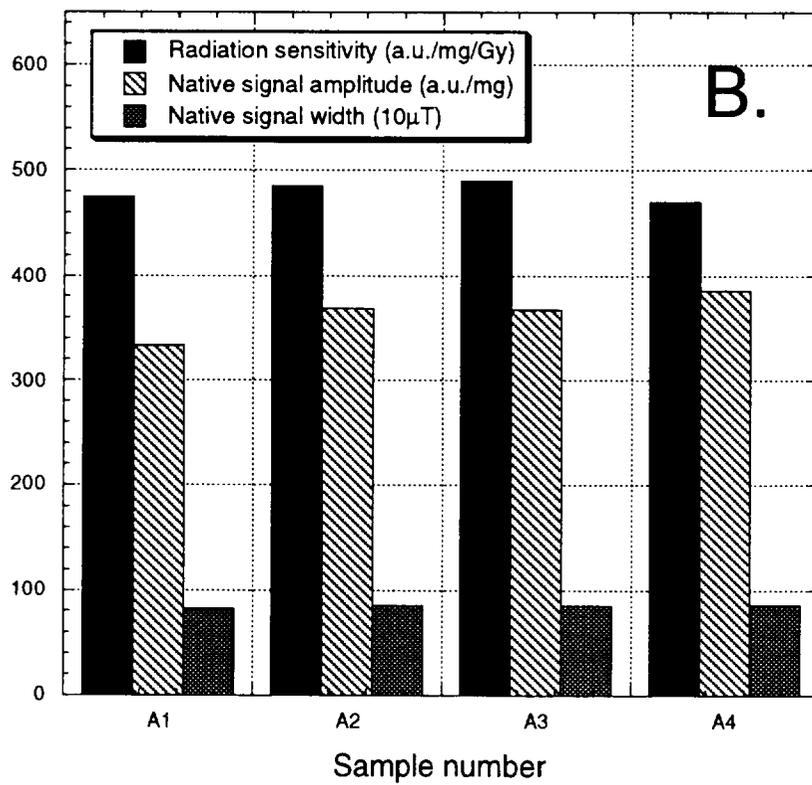


Fig. 4B.

Appendix IV

Verification of a New Method for Virtually Nondestructive Additive Dose EPR  
Dosimetry

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**Abstract-** By using techniques for high measurement reproducibility and accuracy, we have demonstrated the feasibility for a pseudo-nondestructive additive dose EPR technique that does not require irradiating a large fraction of the dosimeter material while simultaneously allowing for individual sample sensitivity calibration. This is shown to be attainable by irradiating only a small fraction (10-20%) of the sample for a representative sensitivity calibration. Corrections had to be made for the cavity response characteristics when using variable mass samples. Likewise, methods were employed to correct for empty tube signals, sample anisotropy and frequency drift. Results showed the method used here will reconstruct doses to within 10% of those reconstructed using standard additive dose methods.

### *Introduction*

Electron paramagnetic resonance (EPR) is a nondestructive method of testing materials whose basic physics of operation correspond to those of nuclear magnetic resonance (NMR). Rather than detecting nuclear spins as in NMR, EPR is tuned to detect unpaired electrons such as those created in crystalline materials upon exposure to ionizing radiation. Here, the radiation induced EPR signal will have an intensity proportional to the total dose acquired. EPR dosimetry techniques commonly use the additive dose method (Chumack et al. 1996A) to calibrate individual sample sensitivity to irradiation. In doing so, the sample is reirradiated many times over to determine the relative signal increase as a function of applied dose. From this, one is able to back extrapolate from the original signal intensity to reconstruct the initial dose.

When additive irradiations are applied, the method is no longer nondestructive as is NMR. For this reason, many investigators have opted to use only calibration curves which are simply average sample sensitivities for a given population. It is known however (Pass et al. 1997) that sensitivity (EPR signal intensity per unit dose) can vary by as much as 30% for a given population group. It has also been shown that sensitivity can vary from tooth to tooth from a single individual (Iwasaki et al. 1995). It has even been shown that for some massively carious teeth, sensitivity can be low by as much as a factor of 5 or more than the average (Romanyukha et al. 1998). It is for these reasons that additive irradiations are commonly employed so as to avoid errors incurred through assuming an "average" sensitivity. Again, the price that must be paid for using additive dose methods is that the sample must be destructively evaluated. This has many detrimental implications. Some of these are that later investigations cannot reevaluate the samples using future improvements in technology, samples cannot be examined for differential dose effects subsequent to irradiations (Shimano et al. 1989 Nishiwaki et al. 1990), interlaboratory intercomparisons and or cross checks cannot be done etc.

It was for the above reasons that Haskell et al. (1998?) suggested a non-destructive additive dose method where only a small fraction of the original sample is irradiated to a single large dose to determine sample sensitivity. In this paper we demonstrate the application of this method and cover many of the pertinent factors that must be considered and accounted for in order to attain sufficient levels of accuracy and precision.

One of the more important effects that had to be corrected for in measuring small masses was that of the empty EPR tube signal (Fig. 1). This signal becomes a dominant error contribution for sufficiently small sample masses and/or sufficiently small doses. As shown in Figure 1, the magnitude of these signals is strongly dependant on microwave power although, as pointed out by Haskell et al. (1997?), these signals are also proportional to the modulation amplitude. At 25 mW power and 0.5 mT modulation, these signals are typically equivalent to a few hundred mGy in absolute peak to peak for a 100 mg sample although they are not generally located directly at the dosimetric signal measurement positions (Fig. 1).

Another very important effect that had to be corrected for was that of the cavity response. It has often been reported in the literature that the EPR response for the dosimetric signal in tooth enamel is linear as a function of sample mass, even to values exceeding 200 mg (Iwasaki et al. 1990, Ikeya et al. 1993). We have found that for a quick inspection, this indeed appears to be true but that a more thorough investigation displays definitive departures from linearity which must be accounted for in order to use any linear sample mass normalization procedure (Fig. 2).

### MATERIALS AND METHODS

Eleven samples were taken from the surrounding regions of the Techa river. These samples were selected as a control cohort for epidemiological studies of the Techa river residents who were exposed to massive radionuclide releases in the early part of the cold war (Romanyukha et al. 1996). The teeth first had their roots removed using a diamond tipped cut off saw. The remaining crown was then crushed in a ceramic mortar and pestle to grain sizes of 250-600  $\mu\text{m}$  diameter. The enamel granules were then separated by heavy liquid separation using bromoform (s.g. = 2.9  $\text{g}/\text{cm}^3$ ). This resulted in a high purity enamel granule material. The samples were then rinsed in acetone (x3) followed by methanol (x3) and finally distilled water (x3). After this, the samples were allowed to sit in any ambient temperature desiccator for 1 month to allow relaxation of any mechanically generated radicals and evaporation of any adsorbed organic solvents.

All irradiations were done using a Co-60 volume irradiator (US Nuclear) having a dose rate of 7.0 Gy/hr. Because a volume irradiator was used, samples were irradiated in an aluminum cylinder for electronic equilibrium. Although an aluminum cap was placed on the cylinder, it did not extend all the way down to the sample surface. Subsequent to all irradiations, samples were annealed according to Sholom et al. (1998). Samples were not scanned until a day after each annealing to allow an equilibrium content of adsorbed water to be reached and so insure constant dielectric properties during EPR scanning.

The spectrometer used was an x-band Bruker ESP 300E fitted with a TE<sub>102</sub> resonant cavity. The constant rotation goniometer of Haskell et al. (1997?) was used for all spectral acquisitions with the modified instrumentation of Hayes et al. (1998). The EPR parameters employed were 0.5 mT modulation, 25 mW power, 41 ms time constant, 10 mT sweep width, 1024 field point resolution, 30 accumulations per spectrum at 41 seconds per accumulation. All dose reconstructions were done using the method of Haskell et al. (1998?) which account for empty tube signals using an in cavity Mn<sup>++</sup> standard (Haskell et al. 1997?) and subtraction of the native signal similar to the method of Skvortzov et al (1995). All dose estimates were done using the automation routine of Haskell et al. (1998?) where only a 10 mGy purely random uncertainty is introduced by the program.

All eleven samples were scanned three times prior to any irradiation. The original average sample mass for the group of samples was 141±31 mg. A small modification was implemented on the spectral acquisition algorithm implemented by Haskell et al. (1998?). This was to scan the empty tube spectrum once every three sample scans. This was done because the dynamic character of the empty tube signals are negligible over the time period used. All samples were vigorously shaken between each scan.

After all samples had their initial three scans completed, 20 mg was measured out from each sample and given a 10 Gy hydroxyapatite dose. Subsequent to annealing, these aliquots were then scanned three times each. These three measurements in addition to the three unirradiated sample scans made up a two point dose response as described by Franklin (1986) and Hayes et al. (1997). Because only 20 mg (within 10-20% of the total sample mass) had to be irradiated in order to calibrate the sample sensitivity, this method can be considered a virtually non-destructive method.

The remaining large fraction of the sample was additively irradiated in accordance with the recommendations of Chumack et al. (1996B). Here we used only the first of the three undosed sample scans (normalized by sample mass according to Fig 2) as the zero dose value for these dose responses. Five additional irradiations were given at additive dose values of 1.6 times the original dose estimate obtained via the virtually non-destructive method. Because both approaches (the standard additive dose method and the non-destructive method) used 6 spectra total taken under identical conditions (not including sample mass and dose), a direct comparison between the two methods was made possible.

The calibration of the cavity response function (Fig. 2) was done with an enamel sample having a dose of 550 Gy and a grain size range of 250-600 μm. Here, the grain sizes were identical to those used in the experiment so that a direct correlation to the experimental values could be made.

## Results

The effect of signal intensity on sample mass is shown in Fig. 2. Here, it can be seen that the EPR signal response appears to be linearly dependant on the sample mass up to 175 mg (solid line regression on open squares up to 175 mg) as reported by Iwasaki et al. (1990). A closer look reveals some slight nonlinear dependencies which are modeled with a fourth order polynomial (dashed curve regression on open squares). The absolute deviation from linearity is small over the apparent linear range although this constitutes a very large relative error at the small mass values. This large relative error at small masses is also shown in Fig. 2 by the solid curve fit to the filled diamonds. The filled diamonds represent the relative EPR signal intensity per mg as a function of sample mass and also has a fourth order polynomial fit applied to the data. The fit demonstrates that near zero mass, the response is 30% lower than at the maximum response occurring near 84 mg. Likewise, at the 200 mg mass value, the response demonstrates an 13% decrease relative to the maximum. The overall deviation from the fit is only 1.6%.

A comparison of the reconstructed doses using the destructive and non-destructive methods is given in Fig. 3. The one to one correspondence line is given as the dotted line whereas the solid line is the unweighted linear least squares fit to the data. The curve fit is

of the form  $Y = mX + b$  and resulted in values of  $m = 1.10 \pm 0.12$  and  $b = -0.3 \pm 16$  mGy with a correlation coefficient of 0.95.

The average statistical uncertainty in the dose estimates from all of the dose reconstructions was  $55 \pm 22$  mGy for the nondestructive method and  $15 \pm 8$  mGy for the destructive method. The larger errors from the nondestructive method are not solely due to the lower mass used in those measurements. Although the same number of measurements were made for both methods, the nondestructive method had measurements only done at the zero and highest dose administered. This is the optimum dose distribution attainable for the condition where errors are dominated by only relative uncertainties ( $n_l/n_h = 1$ , Bluszcz 1988). The optimum distribution found for the case where constant errors are the dominant uncertainty is given by Franklin (1986) as  $n_l/n_h = Y_h/Y_l$ . Here  $n_l$  is the number of measurements at the zero laboratory dose point,  $n_h$  is the number of measurements at the high dose value,  $Y_l$  is the signal intensity at the zero laboratory dose point and  $Y_h$  is the signal intensity at the high dose point. The optimum dose distribution as pointed out by Bluszcz (1988) is the sum in quadrature of the two distributions as given in equation (2), here appropriate weighting consideration must be given to the relative error magnitude  $\delta$  in percent and the constant error component  $\sigma$ .

$$\frac{n_l}{n_h} = \sqrt{\frac{10^{-4} \times \delta^2 + \sigma^2 / Y_h^2}{10^{-4} \times \delta^2 + \sigma^2 / Y_l^2}} \quad (2)$$

Using the actual measured values of  $\delta = 1.5$ ,  $\sigma = 10$ ,  $Y_h = 3600$  and  $Y_l = 60$  in equation 2 gives an optimum distribution of  $n_l/n_h = 3$ . Effectively, the optimum distribution of the six point should have been 5 measurements at the zero and one at the 10 Gy dose point. This resulted in the fact that the y-intercept uncertainty (in dose equivalent) of the nondestructive method was on average  $22 \pm 9$  mGy whereas for the destructive method this value was  $7 \pm 3$  mGy. An additional source of uncertainty in the 20 mg samples used in the nondestructive method is the relatively steeper slope shown in Fig 2 where mass normalization introduces a larger error for the smaller sample masses.

Sample sensitivities were found to be systematically lower for the nondestructively evaluated samples. This can be explained by the irradiation configuration. That the top of the samples were exposed to air inside the cylinder, the effects from this would be more significant for the small mass than for the larger mass samples as a relative effect. This assumes electron density is lower at the air enamel interface than the enamel aluminum interface.

### Discussion

The correlation of the two methods shown in Fig. 3 demonstrates that the two methods are equivalent to within detection accuracy. This because the  $10 \pm 12\%$  overestimate of dose estimate (derived from the unweighted least squares fit) using the nondestructive method is not statistically distinguishable from zero. Furthermore, the intercept from the fit also demonstrates that no systematic errors are present within detection capabilities.

It is clear however that both methods provide very good precision at levels acceptable for epidemiological purposes. The precision attained in the methods is due in large part to the implementation of sophisticated spectral manipulations and measurement procedures. One of the more important spectral manipulations is that of the empty EPR tube signal (Fig 1). The other is the suppression of sample anisotropy through use of a constant rotation goniometer (Hayes et al. 1998, Haskell et al. 1997?).

The empty tube signal is actually a superposition of the empty resonator signals (via oxidation on the cavity lining, atmospheric oxygen etc.) and those of the empty tube and in-cavity standard with its associated fixtures (Hayes et al. 1998). The spectra of the empty tube has visually discernable contributions to the total sample spectrum at microwave powers at or above 10 mW as shown in Fig. 1A. By periodically scanning the empty sample tube, these spectral components (Fig. 1B) can be subtracted from the sample spectrum as described by Haskell et al. (1998?). The results of subtracting the empty tube signals (Fig.

1B) from the sample spectra taken at multiple powers (Fig. 1A) is shown in Fig. 1C. A quick review of the literature shows that when investigators have published EPR spectra of tooth enamel samples at higher microwave powers, the effects shown in Fig. 1A are typical (Copeland et al. 1993, Hoshi et al. 1985, Rodas Duran et al. 1985, Skvortzov et al. 1996, Stepanyenko et al. 1996). It is important to remember that the effects from the empty tube spectra are not only important in small mass measurements but perhaps even more important for measurements of samples with small doses. Here, the compounded effects from both small doses and small masses can occur.

Another very important effect to be considered in measuring small sample masses is that of sample anisotropy. It was shown by Hayes et al. (1998) that for a single enamel piece, the peak to peak intensity of the dosimetric signal can vary by more than 50%. This is a striking effect in light of the fact that Momborquette and Weil (199?) demonstrated that  $10^6$  to  $10^8$  particles are required in EPR for a powder distribution to be made. Hayes et al. (1998) demonstrated that useful dosimetric information can still be obtained from samples exhibiting the worst possible conditions of anisotropy. This required using a constant rotation goniometer and a microwave frequency stabilization method such as the relative g-factor alignment method using the signal from an in cavity  $Mn^{++}$  standard.

The resonator cavity response function (Fig. 2) can also be a significant factor to be considered for small mass samples. In fact, for small mass samples, the Q factor of the cavity is higher than when larger mass samples are scanned (similar to the Q factor increase found when scanning an empty tube spectrum (Haskell et al. 1997?)). This effect can be countered if the position of the small mass sample is not sufficiently near the maximum response position in the resonant cavity. The EPR response as a function of sample mass shown in Fig. 2 appears entirely linear up to 175 mg when a linear function is fitted to the data. By plotting the EPR response per unit mass as a function of sample mass (filled diamonds in Fig. 2), clear deviations from linearity are displayed. This is because the absolute deviation at the small masses is quite small but the relative deviation approaches as much as 30% when the mass goes to zero. Any procedure that would then utilize mass normalization based on a linear function with zero intercept would then be subject to some value of relative error incurred in the small mass sample.

### Conclusion

Despite some problems with the irradiation configuration used, non-destructive EPR dosimetry has been validated as a viable method for retrospective dosimetry. It was found that for this to be realized, accounting for the resonant cavity response function and empty EPR tube signals was critical. In order to fully resolve the cavity response function, a very high signal to noise should be realized at the low mass values. Of less importance but still necessary was the suppression of sample anisotropy using a constant rotation goniometer in conjunction with a g-factor stabilization method as described by Hayes et al. (1998?).

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## Figure Captions

Figure 1. Microwave power dependencies of the empty tube signal and native signal in tooth enamel. Figure 1A shows the sample spectrum from a tooth enamel sample scanned at multiple powers. Figure 1B shows the empty tube spectra taken sequentially with the sample spectra of Figure 1A. Figure 1C demonstrates the resultant spectra constructed when the empty tube spectra (Figure 1B) are subtracted from their corresponding sample spectra (Figure 1A) according to the method of Haskell et al. (1998?). The signals in Figures 1A and 1B at ca. 342.3 mT and 351.2 mT are from the in cavity Mn<sup>++</sup> standard with an internal cavity placement similar to that of Haskell et al. (1998?).

Figure 2. EPR signal response characteristics as a function of sample mass. The open squares demonstrate the straightforward application done by Iwasaki et al. (199?) in which the EPR signal intensity is plotted as a function of sample mass. The solid line is a least squares fit to the data up to 175 mg. The dashed line is a fourth order

polynomial fit to all of the points (open squares). The filled diamonds represent the EPR signal intensity per unit mass plotted as a function of sample mass with a fourth order polynomial fit as well (solid line curve).

Figure 3. Correlation of the nondestructively determined dose estimates to the destructively determined doses. The solid line is the unweighted least squares fit to the data, the dashed line is the one to one correspondence line.

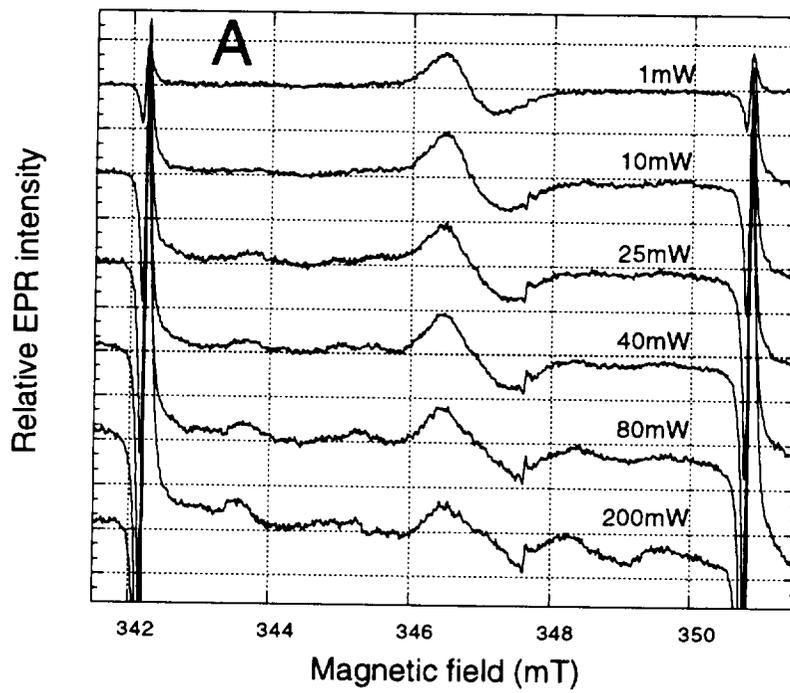


Figure 1A

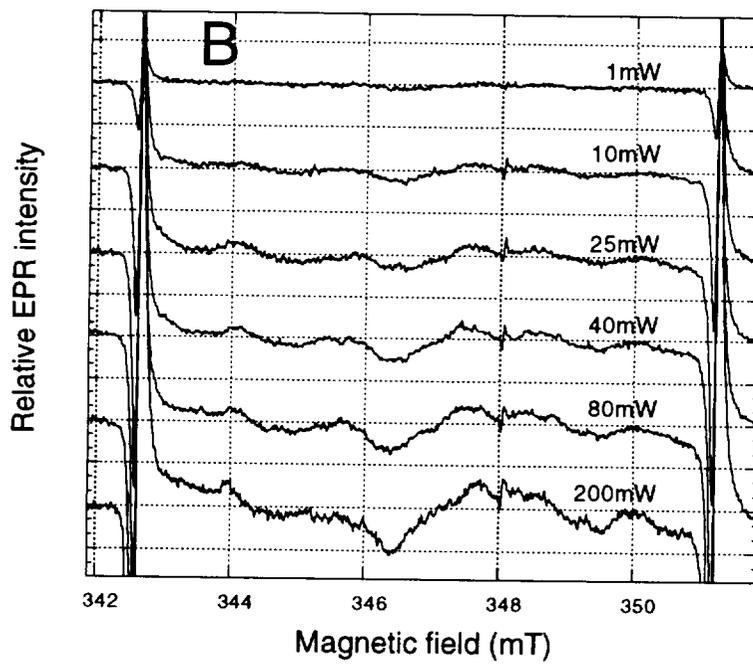


Figure 1B

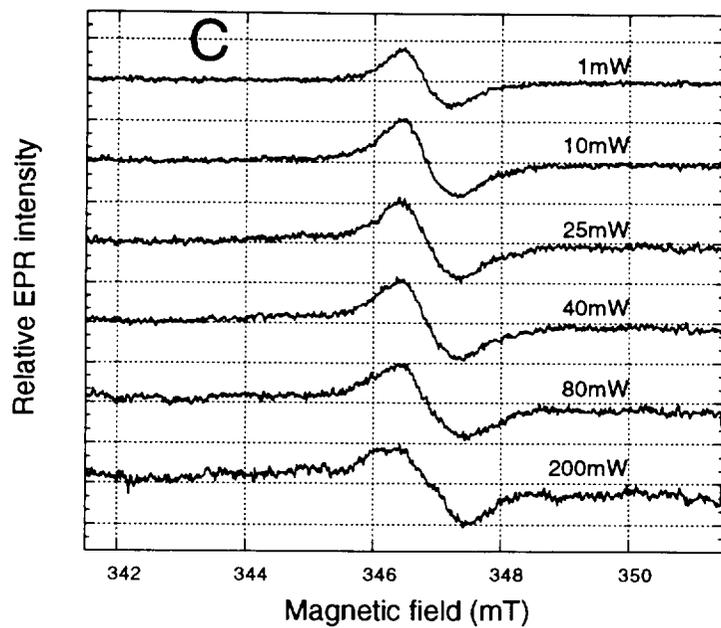


Figure 1C

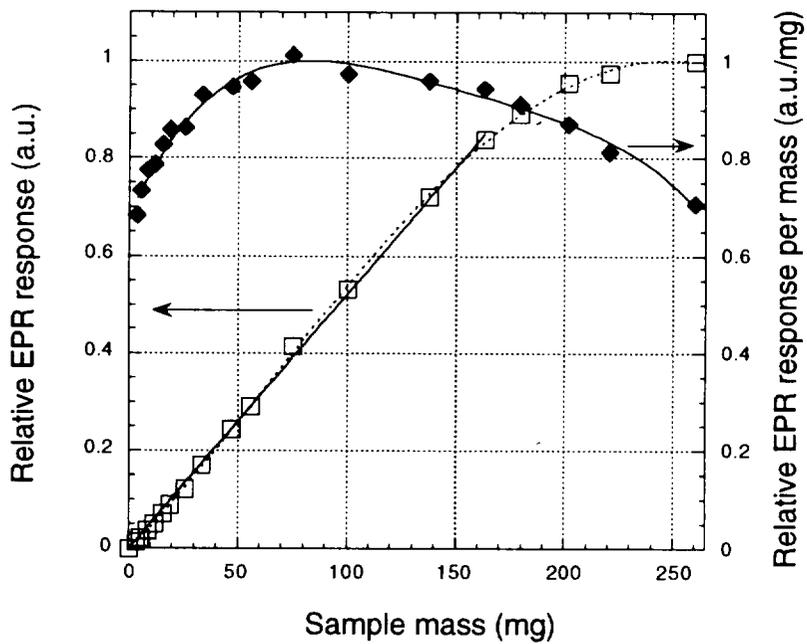


Figure 2

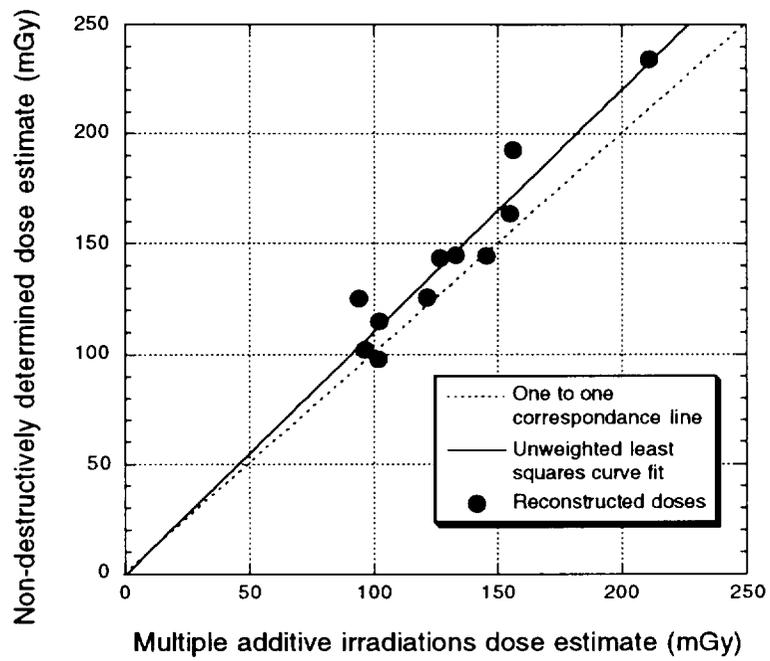


Figure 3

Appendix V

Preliminary report on the development of a virtually non-destructive additive dose technique for EPR dosimetry

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*Abstract-We propose a new approach to the additive dose method in EPR dosimetry studies for tooth enamel specimens. We outline a specialized routine whereby the sample may be left for the most part unirradiated while only a small aliquot of the sample will be additively irradiated to relatively large doses. The routine is done in such a way as to not significantly compromise either precision or accuracy of the dose reconstruction. It is also demonstrated that the overall throughput of the dose reconstruction is not appreciably compromised. With this potential ability, the utility of an international dose/sensitivity standard for EPR dosimetry of teeth is considered.*

## **Introduction**

When EPR dosimetry with tooth enamel was initially reported, the methodology only allowed doses down to 1 Gy to be reconstructed (Brady et al. 1968). Subsequent investigators then pushed this below the 1 Gy level (Pass and Aldrich 1985, Ikeya et al. 1986) and it wasn't until the 1990's that the present levels of detection were empirically demonstrated (Romanyukha et al. 1994; Egersdörfer et al. 1996, Romanyukha et al. 1996A). A recent international intercomparison demonstrated  $\pm 100$  mGy accuracy at the 100 mGy dose level (Chumak et al. 1996). A follow up intercomparison demonstrated that 20% accuracy at the 200 mGy level could be realized (Haskell et al. 1997A) if subtle effects in the dose reconstruction process were accounted for (Haskell et al. 1997B). The latter intercomparison showed that using nonstandard instrumentation, precision could be attained at the 25 mGy level (Haskell et al. 1997C). For these reasons, it can be expected that future advancements will continue to improve the technology.

Generally, EPR dose reconstruction is done using the additive dose method where the sample is measured before, and subsequent to, multiple irradiations to generate the dose response curve (from which the reconstructed dose is derived). After the dose response has been measured, however, the sample has a significant radiation dose in excess of the reconstructed dose and is useless for further repeated analysis of the initial dose received. Because of this, implementation of EPR dose reconstruction techniques that do not use the standard additive dose method would be advisable so that independent measurements or future technological advancements could be applied to the original samples.

*One approach to addressing this issue is to use a calibration curve (average radiation sensitivity in EPR counts per mg per mGy) to replace the dose response curve created with the additive dose method. It is known, however, that sample sensitivity differs between different teeth of a single as well as different individuals (Iwasaki et al. 1995). Furthermore, for a few select countries, it has been found that average sensitivities vary and that sensitivity also has a dependency on the relative health of a tooth (Alexander Romanyukha manuscript in preparation). Indeed, for tooth enamel damaged by caries, we have found that sensitivity can vary by more than a factor of five (Alexander Romanyukha manuscript in preparation). This of concern as most teeth used in large scale accident dose reconstruction will be obtained as the result of periodontal disease or massive caries. For these reasons, individual sample sensitivity calibrations for dose reconstruction are warranted.*

*We propose a methodology whereby calibration of sample sensitivity can be carried out virtually non-destructively by additively irradiating only a small aliquot (20-30 mg) of the sample to a dose in the range of  $\geq 10$  Gy. The calibration curve generated from this sample could then be applied to subsequent nondestructive measurements of the remainder of the sample. Since sample sensitivity is a function of instrumental conditions and parameters, the calibration curve would only be appropriate to the parameters used for the collection of the curve and on the machine from which the data was initially generated.*

*To circumvent this problem, we also propose the development and distribution of a standard sensitivity sample. The standard would include both dosed and undosed portions*

*of a homogenized batch of natural or artificial enamel. Each calibration curve generated for a new tooth sample would then be reported in terms of sensitivity relative to that of the standard samples measured under identical instrumental conditions. This relative sensitivity value would then remain with the measured and unmeasured portions of the tooth and could be applied to the tooth by any laboratory using any parameters for future nondestructive measurements.*

*We also demonstrate that this method can be done in such a way that sample throughput, accuracy or precision (to within 5%) are not compromised.*

## **Materials and Methods**

Samples were obtained from the region surrounding the Techa river of the former Soviet Union. These teeth were selected as part of a background study monitoring the health effects from massive accidental radionuclide releases in the Techa river from 1949-1957 (Romanyukha et al. 1996B).

*All samples were irradiated according to the method prescribed by Hayes et al. (1997) whereby only one additive irradiation is applied and linearity is assumed. All samples had two or three spectra taken prior to irradiation followed by three spectra taken subsequent to irradiation. All maximum doses were kept below 20 Gy to insure saturation effects would be negligible (Rink and Schwarcz 1994, Grün 1996). Sample shaking was implemented prior to each spectral acquisition.*

All samples were scanned using a constant rotation goniometer (Haskell et al. 1997C). Spectral manipulations including native and empty EPR tube subtractions were done using the Mn<sup>++</sup> line method (Haskell et al. 1998A, Haskell et al. 1998B). Irradiations and annealings were done according to Sholom et al. (1998). The spectrometer parameters used for the study were, 0.5 mT modulation amplitude, 100 kHz modulation frequency, 25 mW microwave power, 10.0 mT scan width, 1024 point field resolution, 30 sweep accumulations per spectrum, 41 ms time constant and conversion time. The receiver gain was 10<sup>5</sup> for all spectra and the goniometer rotation rate was 0.3°/sec. The spectrometer used was a Bruker ESP Model 300E with a standard TE<sub>102</sub> cavity.

*Two groups of samples were prepared consisting of multiple teeth with each tooth being kept separate in each set. All samples were scanned prior to irradiation and had masses ranging from 40 to 200 mg.*

### **Group 1.**

This group consisted of 34 samples which were prepared by an initial crushing followed by a subsequent chemical purification with KOH. We used a much stronger treatment than that used in previous studies (Romanyukha et al. 1994). This included using supersaturated KOH at 60° C. After the 0 Gy scans were taken of this group, subportion aliquots were irradiated to 1 Gy and scanned. These aliquots had masses ranging from 25% to 100% of the original sample mass.

### **Group 2.**

This group of samples were similarly prepared but without the KOH treatment (the differences in sample preparation effects are expected to be reported elsewhere in conjunction with a more broad study but will not effect the results of this study). The grain sizes used were 250-850 µm obtained by sample sieving. The initial sample masses ranged from 90 mg to 110 mg. The additive irradiation given was 10 Gy to subportion aliquots of 25 mg. The value of 10 Gy was calculated to be the dose that would give ≤5% variation in reproducibility.

## Results

### Group 1

The 1 Gy reproducibility obtained in this group is shown in Figure 1 as a function of the sample mass. The dashed line in the figure is an unweighted least squares fit to the data. No correlation between reproducibility and sample mass was found at the 0 Gy dose level for this set of samples as shown in Figure 2. The correlation of this data (Figures 1 and 2) is shown in Figure 3 (here, the effects of sample mass on precision are not taken into account). The reproducibility at the 1 Gy level tended to be worse on average than that at the 0 Gy level by 17 mGy. The diagonal line in Figure 3 is the 1 to 1 correspondence line.

### Group 2

It was found for this group that at the 0 Gy level using 100 mg mass, the reproducibility was equivalent to  $21 \pm 11$  mGy. For the 25 mg samples at the 10 Gy level, the reproducibility was  $4\% \pm 1\%$  ( $400 \pm 100$  mGy). The reproducibility attained for the 25 mg, 10 Gy irradiated samples can be seen qualitatively in Figure 4 where a typical dose response for group 2 is shown. The results of all measurements from group 2 are given in Table 1.

## Discussion

### Group 1

It is not surprising that measurement reproducibility decreases with decreasing mass (Figure 1). What is illuminating is that the deviation from average appears to become very strong when the sample mass passes below about 75 mg. On average however, the reproducibility stays to within 10% over the entire mass range evaluated (ca. 50 to 200 mg). At the zero dose level (Figure 2), the precision is more or less constant and can be assumed to be a measure of the overall random noise in the entire measurement process ( $19 \pm 16$  mGy in this case).

Because there is a correlation between the reproducibility at zero added dose and the 1 Gy dose, we can infer that precision in EPR dosimetry is strongly sample dependent. The lower reproducibility found at the 1 Gy dose relative to that at the 0 Gy dose (Figure 3), can be attributed to machine instabilities. We estimate the daily stability of our spectrometer to be in the range of 1% absolutely for any given EPR intensity measured.

### Group 2

Group 1 results contrast with those for group 2 for samples measured with 25 mg at the 10 Gy level as given in Table 1. Here we find that it is possible to measure the sensitivity of a sample to within  $\pm 5\%$  using the following methods; 1) constant rotation goniometer (Haskell et al. 1997C), 2) native and empty EPR tube signal background subtraction using the  $Mn^{++}$  line method (Haskell et al. 1998A, Haskell et al. 1998B) and 3) high dose irradiation of a small sample fraction. Average sensitivity can be assumed if the aliquot used for the sensitivity calibration is randomly selected from a granulated sample.

### General

For the laboratory doing the initial dose reconstruction and sensitivity measurement, the amount of time required for a given dose reconstruction will not be significantly altered. For those laboratories doing subsequent analysis of the same samples, throughput is expected to increase due to the fact that the sample sensitivity will already be known and so additive irradiations will not be necessary.

The implementation of the pseudo-nondestructive method we suggest here opens some new possibilities for those doing dose reconstruction. An international dose standard could be constructed by taking a highly homogenized sample of 250-800  $\mu\text{m}$  tooth enamel grains and giving it a precise dose or doses in the range of 1 to 20 Gy at a primary source standard (NPL, NIST etc.). This would then be followed by distribution of adequate portions of the sample to the dose reconstruction community.

If such an international dose standard were measured under identical conditions as the sample aliquots used for sensitivity assessments, sample sensitivity values could be recorded and thereby reported in units relative to the international dose standard. With such an approach, another measurement technique or laboratory could nondestructively measure the dose to the previously analyzed sample, without having to measure irradiated aliquots of those same samples. Because the sensitivity would have already been measured and reported relative to the international dose standard, the other laboratory or method could use the relative sensitivity previously calibrated for their particular machine, cavity, method etc. For example, the selective saturation method (Ignatiev et al. 1995) will have a different inherent sensitivity than that of the native signal subtraction method (Skvortzov et al. 1995) even if done on the same spectrometer. This method would be applicable to all the of the current methodologies currently available to carry out a dose reconstruction (Jonas 1995, Chumak et al. 1996, Galtsev et al. 1996, Ignatiev et al. 1995, Skvortzov et al. 1995, Schabl 1996, Callens et al. 1996).

An important point to be remembered is that due to the fact that sample mass normalization is an integral part of this approach, all sample measurements (by all laboratories) would have to be done where a linear EPR response vs. sample mass relationship is guaranteed (Iwasaki et al. 1990, we recommend 200 mg or less for x-band). This is not expected to be a problem because the relative gain in potential accuracy or precision above this linear range will generally be negligible.

## Conclusion

This preliminary study supports the feasibility of small sample sensitivity measurement. The remainder of an unirradiated sample could then be stored for future evaluation or sent to other laboratories for quality assurance, method intercomparison, new technology testing etc.

The approach to EPR dose reconstruction which we are endorsing only addresses the issue of nondestructive sample evaluation with individual sample calibration. Other problems relating to error inducing factors have not here been addressed. Examples of such problems are impurity signals (Haskell et al. 1997B), sample anisotropy (Aoba et al. 1985), the irradiation energy spectrum or the source and/or type of irradiation (Wieser et al. 1996).

An international tooth enamel reference standard has been proposed which would allow relative reporting of measured sensitivities. Relative values could then be reported using all published tooth enamel dose reconstruction methods. This would expedite intercomparisons, quality checking or future measurements of a given sample since nondestructive testing could be applied to all but a small portion of the original sample.

**Acknowledgements** - The authors are grateful to Dr. Marina Degteva for organization of the sample collection at the Southern Urals. This work was supported by DOE contract DE-FC03-97SF21354.

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*Table 1. Group 2 measurement results. Values reported are the 1 standard deviation values measured from three separate spectra of each sample at each dose.*

Sample name	Sample mass (mg)	0 Gy Reproducibility (mGy)	10 Gy Reproducibility (mGy)	10 Gy Reproducibility (%)
R 67	112	36		
R 67	24		310	3.1
R 97	111	12		
R 97	25		470	4.7
R 105	83	21		
R 105	23		230	2.3
R 120	106	15		
R 120	23		460	4.6

## Figure Captions

- Figure 1. EPR measurement reproducibility at the 1 Gy dose level as a function of sample mass for the dosimetric signal in the group 1 samples. This plot is of the scatter about the mean from multiple measurements and is being reported at the 1 standard deviation level. The solid line through the data is the unweighted least squares fit to all of the data points. The resulting values of the curve fit  $Y=mX+b$  were  $b=5.3\pm 1.1\%$  and  $m=-0.019\pm 0.012\%/mg$ .
- Figure 2. EPR measurement reproducibility at the 0 Gy dose level as a function of sample mass for the dosimetric signal in the group 1 samples. Most of the samples were from individuals over the age of 40 and so the dosimetric signals measured were on average about 150 mGy. This plot is of the scatter about the mean of these measurements to the 1 standard deviation level.
- Figure 3. Correlation of reproducibility at the 1 Gy level to that at the 0 Gy level. The dashed line running diagonally through the figure is the one to one correspondence line.
- Figure 4. Typical dose response carried out using the proposed psuedo-nondestructive EPR technique for the group 2 samples. The ordinate axis is measured in EPR peak to peak counts per mg of material.

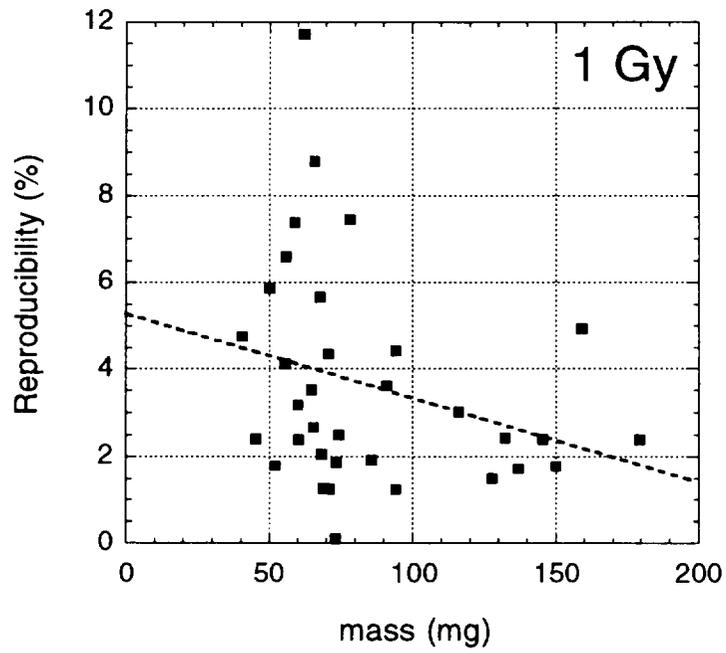


Figure 1

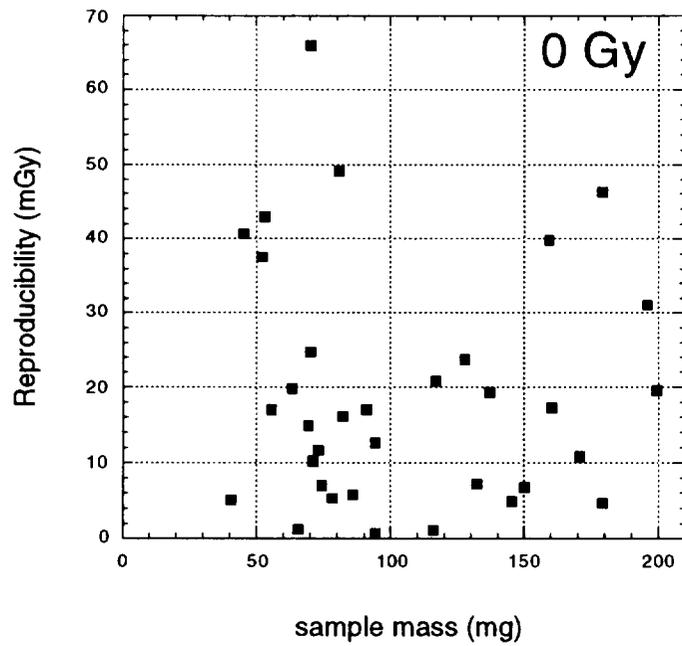


Figure 2

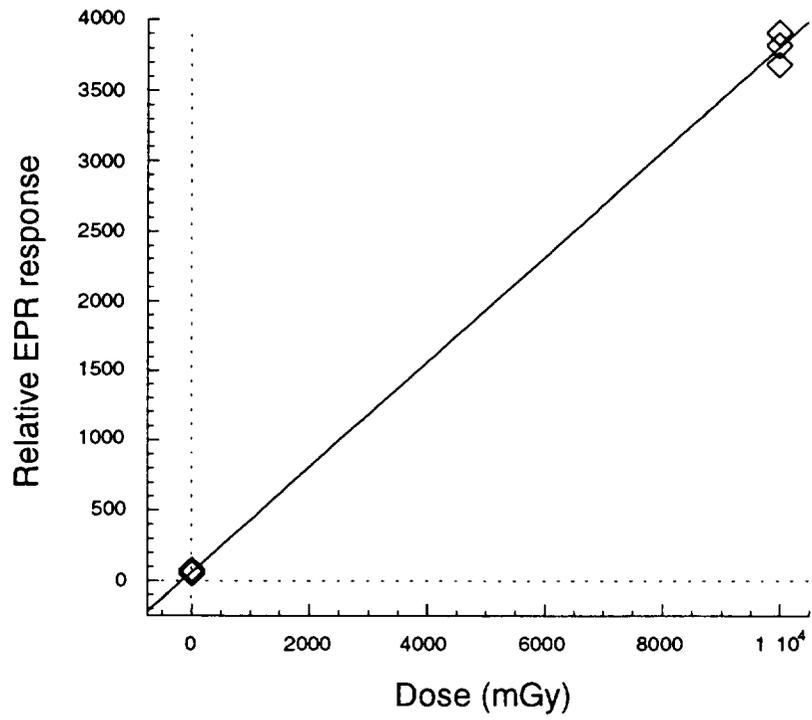


Figure 3

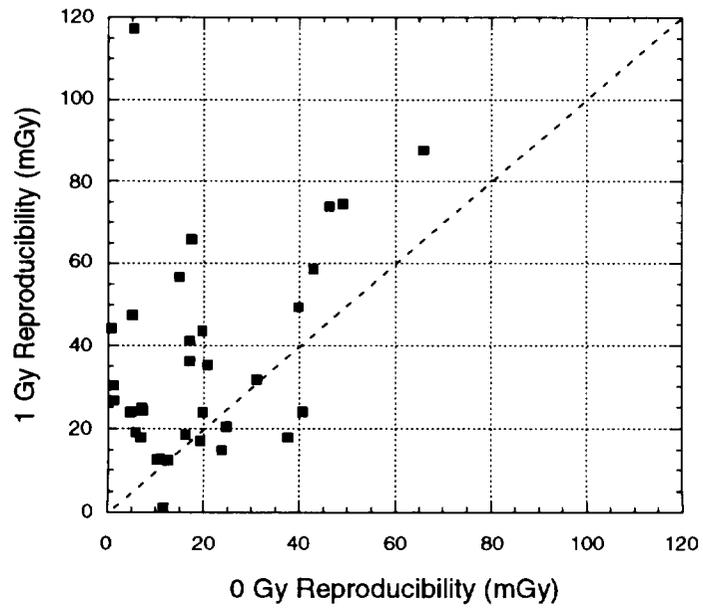


Figure 4

Appendix VI

Results of EPR measurements on teeth from the Techa River Control Population

Once evaluated	Dose (mGy)	
Techa#5	85	
Techa#7	96	
Techa8_2	315	
Techa#13	26	
Techa14	104	
Techa16_1	298	
Techa18_1	157	
Techa#19	110	
Techa22_1	133	
Techa24_1	407	
Techa25_1	143	
Techa27_1	170	
Techa#28	126	
Techa31_1	122	
Techa#34	155	
Techa#36	202	
Techa#37	102	
Techa41_1	94	
Techa52	79	
Techa67	17	
Techa97	20	
Techa105	140	
Techa111	6	
Techa120	22	
Techa69	106	
Twice evaluated		s.d. (mGy)
Techa#50	85	23
Techa59_0Gy	113.5	9
1		
Techa64_1	149	20
Techa78_1	156	16
Techa79_1	202.5	155
Techa#48	124.5	6
Techa#81	36.5	1
Techa#92	262.5	11
Techa101_1	91	14
Techa#113	61	8
Techa#117	199	3
Mechanical separation		
Techa100	145	

Techa119	135
Techa11	99
Techa43	139
Techa55	109
Techa57	174
Techa70	110
Techa72	159
Techa86	123
Techa97	100
Techa103	354
Techa100y	105

Appendix VII

Inventory of additional unmeasured teeth.

Sample	Year	Position	Position code=(Upper or Lower)/(Right or Left)/(position #)
R12	1932	UL4	
R38	1935	LR1	
R18	1931	LR4	
R58	1930	LR4	
R45	1932	LL6	
R52	1927	UL4	
R1	1924	LR3	
R53	1926	UL3	
R56	1938	LR5	
R40	1934	LR2	
R7	1953	LL8	
R28	1939	LL7	
R15	1941	LR4	
R60	1940	UR2	
R10	1943	UL1	
R42	1927	LR4	
R23	1921	UR2	
R23	1921	UR1	
R23	1921	UL1	
R17	1923	LR3	
R8	1949	LL3	
R8	1949	LL2	
R46	1948	UR6	
R30	1932	UR2	
R54	1930	LL1	
R49	1947	LR1	
R26	1937	UR4	
R39	1939	LR4	
R35	1934	UL3	
R6	1929	UL3	
R6	1929	LR2	
R51	1926	UL1	
R29	1937	UL4	
R29	1937	UL3	
R47	1938	UL6	
R9	1936	UL1	
R21	1929	UR2	
R44	1948	LL6	

## Appendix VIII

Publications supported in full or in part by this contract.

An Assessment of the Levenberg-Marquardt Fitting Algorithm on Saturating Exponential Data Sets. R.B. Hayes, E.H. Haskell, and G.H. Kenner. In Press. Ancient TL

A Technique for Increasing Reproducibility in EPR Dosimetry of Tooth Enamel. R. Hayes, E. Haskell, A.A. Romanyukha and G. Kenner. In Press. Meas. Sci. Tech.

An EPR Dosimetry Method for Rapid Scanning of Deciduous Teeth Following a Radiation Accident. E. Haskell, R. Hayes, and G. Kenner. Health Physics. In press. Health Physics.

Variations in Tooth Enamel EPR Spectra Alexander A. Romanyukha, Robert B Hayes, Edwin H Haskell and Gerry H Kenner. Submitted to Radiation Protection Dosimetry.

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