VALIDATION OF TRDS EXTERNAL DOSE ESTIMATES BY EPR MEASUREMENTS SUPPORTED BY ASSESSMENTS OF STRONTIUM-90 CONCENTRATION AND MONTE CARLO SIMULATIONS OF ELECTRON TRANSPORT IN DENTAL TISSUES

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ABSTRACT

Validation of the new estimates of external dose is considered to be a critical factor in the continuing credibility of the Techa River Dosimetry System (TRDS-2000) results and the companion epidemiologic studies they support. Luminescence measurements on quartz extracted from bricks in old buildings in Metlino have confirmed calculations of dose (Jacob et al. 2003; Taranenko et al. 2003). Some measurements of fluorescence in situ hybridization on lymphocytes taken from former residents of Metlino also tend to confirm doses calculated with the use of TRDS-2000 (see Degteva et al. 2004), but these measurements were handicapped by the limited number of cells scored. Electron paramagnetic resonance (EPR) appears to be the method of choice for validating TRDS-based estimates. EPR measurements are difficult, however, as the levels of dose are low compared to the sensitivity of the method, and EPR measurements are also influenced by background dose and internal dose from the incorporation of ^{90}Sr in tooth enamel and dentin.

The purposes of this document are (1) description of the results of external dose estimates derived from use of the TRDS-2000 and recent improvements of the Techa River Dosimetry System; (2) description of available results of individual-dose estimates based on EPR data for the Techa River residents; (3) extraction of the component of external dose from the results of EPR measurements; (4) comparative analysis of external doses derived from EPR data with the TRDS-based estimates; and (5) discussion of the perspective of future investigations on external dose verification for the Urals region.

Work continues on improving the TRDS by implementation of the new two-compartment Techa River Model, extending the calculation to include doses received beyond the prior cutoff of 1960, and by including doses previously neglected as being in the "background region." These improvements do not result in significant changes for residents of Metlino, which is the closest village to the site of releases. However, for persons living at >150 km from the site of release, the results are significantly higher.

The results considered in this paper include EPR measurements on 27 teeth from 13 adult residents in Metlino; 218 teeth from residents in "background" areas; and 265 teeth from 179 donors living further downstream on the Tech River. Also considered are the results of radiochemical measurements of ^{90}Sr in teeth from 152 permanent residents of the middle and lower regions of the Techa River. Results of Monte Carlo calculations for dose-rate coefficients (*DCR*s) for various source and target tissues in teeth at each position of the denture are presented; these values of *DCR* allow for the subtraction of dose due to incorporated ^{90}Sr, which can be the major source of dose to teeth of residents on the middle to lower regions of the Techa.

The conclusions are that individual doses can be validated for residents of Metlino and nearby villages. However, for residents on the middle to lower regions only average doses can be considered due to the low values of total dose and the significant contribution of ^{90}Sr to the EPR-based doses. The results of these average values fully disprove the allegations of Mokrov (2003; 2004) that the TRDS-2000-based external doses for residents of Metlino and Muslyumovo are too low by a factor of 3–5. The EPR-based doses for permanent residents of Muslyumovo are nearly an order of magnitude lower than those postulated by Mokrov.

1. INTRODUCTION

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 effluent into the rather small Techa River. This resulted in chronic external and internal exposure of about 30,000 residents of riverside communities. The major intake of ^{90}Sr by inhabitants of the area occurred in 1950–1951. An average of about 3,000 kBq of ^{90}Sr was ingested with river water by each resident of the upper- and mid-Techa region. The "Extended Techa River Cohort" (ETRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM). A special database was established for the follow-up of the exposed population. This database contains the roster of exposed persons, their residence histories, and the results of medical and dosimetric examinations. The long-term dosimetric study represents a unique database on the contents of ^{90}Sr in humans, including measurements of the radionuclide in bones, teeth and whole body for more than 15,000 exposed persons for a period of more than 45 years.

Russian and United States scientists have been involved in collaborative research programs under the sponsorship of the U.S.–Russian Joint Coordinating Committee on Radiation Effects Research (JCCRER) since 1995. JCCRER Project 1.1 is a comprehensive program to develop improvements in the dosimetry system for the population exposed as a result of the releases from the MPA (Degteva et al. 2000a). As a result of the recent completion of the first phase of Project 1.1 (1996–2000), many improvements had been made in the derivation and implementation of the Techa River Dosimetry System-2000 (TRDS-2000*); these improvements resulted in major changes in doses calculated for members of the ETRC. For example, the external doses were re-evaluated on the basis of more complete examination of the existing data and on more realistic (rather than radiation-protection) assumptions, and the currently estimated doses from external exposure decreased by as much as a factor of ten compared to earlier estimates (Vorobiova et al. 1999; Degteva et al. 2000a,b). And finally, the uncertainty in both the internal and external doses was evaluated for the first time (Napier et al. 2000; Shagina et al. 2000).

Validation of the new estimates of external dose is considered to be a critical factor in the continuing credibility of the TRDS-2000 results and the companion epidemiologic studies they support. Recent successes in the measurement of doses by thermoluminescence (TL) of natural materials and by electron paramagnetic resonance (EPR) of tooth enamel have demonstrated that these measurements can be applied to the Techa River situation. The validation task of the current project is planned as the combined analysis of the entire pool of measured samples. This combined analysis and the supportive modeling necessary for the interpretation of the EPR results is used for the purpose of validation of estimates of external dose and further evaluation of associated uncertainties.

Preliminary EPR studies of the Techa River population (Romanyukha et al. 1996a,b; Tolstykh et al. 2000) have shown that there are three sources of the absorbed dose: external

* The TRDS-2000 is a codified database processor that is used to calculate the doses for members of the ETRC.

exposure, internal exposure (mainly due to ^{90}Sr), and background radiation including all other sources of exposure, except that arising from the Techa River. Thus, EPR measurements by themselves are not sufficient for determining the dose to important organs, and additional knowledge of the contribution from radionuclides incorporated into teeth, as well as of background exposure, is necessary. Therefore, EPR measurements must be supported by an evaluation of the background dose, extensive modeling, and determination of the concentration and distribution of ^{90}Sr in tooth tissues.

The purposes of this document are the following:

- description of the results of external dose estimates derived from use of the TRDS-2000 and recent improvements of the Techa River Dosimetry System;
- description of available results of individual-dose estimates based on EPR data for the Techa River residents;
- extraction of the component of external dose from the results of EPR measurements;
- comparative analysis of external doses derived from EPR data with the TRDS-based estimates; and
- discussion of the perspective of future investigations on external dose verification for the Urals region.

Efforts described here have included work in close cooperation with other institutions (mainly, Institute of Radiation Protection of GSF-Center for Environment and Health, Munich, Germany, and the Institute for Metal Physics, Ekaterinburg, Russia) to study and analyze the results of several groups of methods and data sets:

- EPR spectroscopy (including intercomparison among different laboratories involved and different techniques used for sample preparation, EPR measurement, and spectrum analysis);
- measurements of ^{90}Sr concentration in tooth tissues (enamel, dentin);
- modeling of strontium metabolism in teeth (necessary to reconstruct the complete time pattern of ^{90}Sr retention in tooth tissues since the onset of intake); and
- Monte Carlo modeling of electron and photon transport and distribution of absorbed dose throughout human tissues (including development of geometric models of teeth).

The results presented in this document are firmly based on the findings described in our previous Milestone Reports (Shishkina et al. 2001a; 2003a; Vorobiova et al. 1999; 2003), Unscheduled Reports (Anspaugh et al. 2001; Shishkina et al. 2002) and peer-review papers by Anspaugh et al. (2003); Degteva et al. (2000b; 2004); Shishkina et al. (2003b); and Tolstykh et al. (2000; 2003), which are briefly summarized in the next chapter.

2. BACKGROUND

EPR analysis of teeth basically measures defects in hydroxyapatite that have been induced by ionizing radiation. The method has proven quite useful as a biological indicator of exposure. However, there are drawbacks to the method. One of the more severe is that a tooth must be available for analysis. Due to ethical considerations, subjects must not be asked to donate teeth for this purpose, but available teeth are limited to those that have been extracted for reasons of dental health. The collection of teeth extracted on the basis of medical indications from residents of the Urals for the purposes of providing validation using the EPR method was started in 1992. The majority of samples were received from dentists of rural clinics in the Chelyabinsk Oblast (Kunashaksky, Sosnovsky and Krasnoarmejsky Raions) and Kurgan Oblast (Dalmatovsky and Katajsky Raions). It must be noted that the majority of the members of the Extended Techa River Cohort (ETRC) now live in these five raions and the "unexposed" part of the population is considered as a comparison group for epidemiologic studies (Degteva et al. 1997). A special database named "TOOTH" had been established at the URCRM in 2001 (Shishkina et al. 2001a). This database (linked to the corresponding tooth-tissue bank) contains data on all tooth samples collected and individual data on all tooth donors. The current (June 2004) status of this database is described in the following section.

2.1. THE CURRENT STATUS OF DATABASE "TOOTH"

The current status of database "TOOTH" and the associated tooth-tissue bank is shown in Table 1. As can be seen 3,757 samples from 2,370 donors are available in total. Only 816 samples (22%) have been measured by the EPR method, and the remaining teeth are available for further investigations.

Teeth were collected both from exposed and background donors. The URCRM roster for the exposed population includes data for the following cohorts:

- Extended Techa River Cohort" (ETRC);
- Techa River Offspring Cohort (TROC);
- East Urals Radioactive Trace (EURT) residents; and

Table 1. Status of database "TOOTH" on June 2004.

Characteristics	Number of teeth (donors)		
	Exposed teeth (donors)	Background teeth (donors)	Total
All teeth	1,142 (670)	2,615 (1,700)	3,757 (2,370)
Teeth with EPR measurements	438 (306)	378 (366)	816 (672)
Teeth suitable for EPR measurements	488 (306)	1,289 (962)	1,777 (1,268)

- The so-called Urals Liquidators (UL), persons who did not live in contaminated areas but who could be exposed as a result of work on these territories.

For our study it was necessary to identify “pure background;” therefore, matching of donors to members of each of these cohorts was performed. The result of such matching has shown that ETRC members represent 70% of the exposed donors; TROC and EURT members each represent about 10%, and the remaining 9% are liquidators. The total number of teeth from members of the ETRC is 853 samples from 499 donors.

2.2. BACKGROUND EPR DOSE ESTIMATES FOR THE URALS REGION

The background-EPR signal arises from the radioactive contents of the immediate environment (external gamma rays from the uranium and thorium series and ^{40}K in soil or building materials, etc.), gamma and beta rays from ^{40}K in the body, cosmic rays, and medical exposure (Nilsson et al. 2001). The rate of accumulation of enamel dose from background radiation can vary for donors living in different geographical regions due to variation of natural radiation background (Romanyukha et al. 1999).

The background level of EPR signals in enamel for this study was investigated using 218 teeth (63 molars, 128 premolars, and 27 incisors) selected from the database “TOOTH.” These samples were obtained from donors who lived in presumably uncontaminated rural areas of the Urals region. The details of this study were described in our Report for Milestone 7, Part 1 (Shishkina et al. 2003a).

The ultraviolet (UV) component of sunlight also induces an additional contribution to the EPR signal for enamel of the outer sides of anterior teeth (Ivannikov et al. 1997; Nilsson et al. 2001). Therefore, posterior teeth or samples prepared from the inner part of anterior teeth are preferable for dose reconstruction. Unfortunately, it was impossible to measure separately the inner and outer fractions of enamel for a portion of the incisor teeth in this study, due to the small mass of enamel. For such cases, the EPR signal of the total incisor enamel was measured, and an appropriate background value (including the UV contribution) was estimated.

According to (Shishkina et al. 2003a), no statistically significant age dependence of background levels was found in the investigated age range (from 45 to 95 years) of Urals donors born in 1910–1953. The expected age dependence is probably masked by significant variability in individual values. The average-background level for posterior teeth (molars and premolars) was estimated to be equal to 90±24 mGy; for anterior teeth, average values were obtained for incisors as 121±48 mGy for the inner portion of enamel, 234±79 mGy for the outer portion of anterior teeth, and 154±55 mGy for the total enamel (inner and outer portions combined). It must be noted that the background estimates given before by Shishkina et al. (2003a) included a value for the so called intrinsic signal (or system bias) equal to 60 mGy. The estimates given in this report are free of this bias.

2.3. AVAILABLE DATA ON ^{90}SR CONCENTRATION IN DENTAL TISSUES

Measurements of ^{90}Sr in permanent teeth extracted from Urals residents were performed in 1959−1964. The teeth under investigation were extracted for medical/dental indications and then were examined using the radiochemical method described by Gussev et al. (1959). Later (in 1998) these results were matched with the roster of exposed persons. This data set includes measurements for 535 teeth obtained from 452 persons who lived on the contaminated territories of the Urals region.

For radiochemical measurements teeth from different positions were collected. As a rule, the samples were of posterior teeth, i.e., molars and premolars. The ^{90}Sr concentration in teeth was estimated in units of ^{90}Sr activity per mass of calcium [Bq (g Ca)$^{-1}$]. The calcium content in a tooth sample was calculated as 37% of the ashed weight, which is an average value for human ashed bone (ICRP 1995). The resulting value for the average concentration of calcium in teeth was 235 mg Ca per g of tissue. Because the enamel is only about 17−30% of the total dental mass (and for carious teeth part of the enamel has been lost), radiochemical data could be interpreted with a satisfactory level of accuracy as the average concentration of ^{90}Sr in dentin of molars and premolars.

It was found by Tolstykh et al. (2003) that the strontium concentration is not age dependent for adults (age 25 y and more) and is equal to about 16 Bq (g Ca)$^{-1}$. It is known that mineral metabolism in adult teeth is determined by secondary and reparative (irritation) dentin formation, and the constant ^{90}Sr contents for adult teeth over a long age period indicates that these processes occur at a relatively constant rate. The concentration of ^{90}Sr in dentin of persons younger than 15 y at the time of intake was about eight times higher than for those who were adults, and maximum values were registered for infants who were born in 1949−1950. The difference between infants and adults is about one order of magnitude. It is known that permanent dentition for all teeth (except wisdom teeth) is completed by the age of 12−15 y; therefore, for ages younger than 12−15 y the retention of ^{90}Sr in different dental tissues depends upon the state of maturation for the individual tooth. This phenomenon explains the elevated levels and the high variability in the concentration of ^{90}Sr in teeth for persons younger than 12−15 y.

Special investigations of ^{90}Sr distribution in dental tissues of Techa River residents were performed in 1958−1970 with use of autoradiography (Ivanov 1959; Saurov et al. 1972) and later using photostimulable phosphor imaging (Romanyukha et al. 2002). Results demonstrate two different patterns of ^{90}Sr deposition: (1) for persons who were adult in the period of intake, ^{90}Sr is concentrated mainly in the dentin on the surface of the pulp channel and in the root and (2) for persons with tooth formation during the period of intake, ^{90}Sr is concentrated mainly in the enamel and dentin layer near the junction of enamel and dentin. It seems obvious that tooth samples with ^{90}Sr incorporated in enamel are useless for the purpose of assessment of external dose, and teeth for which the period of enamel calcification overlapped with the period of ^{90}Sr intake must be excluded from any study of external dose. As shown by Tolstykh et al. (2003), very high levels of dose are absorbed in enamel of such teeth, and doses in tooth enamel do not correlate with doses absorbed in other organs and tissues (because path lengths of beta particles in dental tissues are short).

2.4. REQUIREMENTS FOR EPR MEASUREMENTS TO SUPPORT A VALIDATION STUDY OF TRDS-BASED EXTERNAL DOSES ON THE TECHA RIVER

As discussed above, in general a measured EPR dose, D_T, (which results from a conversion of a signal to a dose based upon some suitable calibration) is made up of three parts:

$$D_T = D_{ext} + D_{\mathrm{int}} + D_{back} \ , \tag{1}$$

where

D_{ext} = dose due to external exposure resulting from the Techa River contamination;

D_{int} = dose due to internal exposure resulting from radionuclides ingested mainly with water from the Techa River; and

D_{back}= dose due to background sources of radiation (discussed in Section 2.2).

For the purpose of our validation study we seek to compare the derived value of D_{ext} with estimates of external dose in the enamel that have been calculated on the basis of the TRDS code. The value of D_{int} is the dose in the enamel that is due to the presence of $^{90}Sr/^{90}Y$ and ^{89}Sr in dental tissues and perhaps other radionuclides in the underlying soft tissues. Parameter D_{back} consists of "background" radiation.

There are several groups of requirements necessary for the success of a validation study of external doses calculated with the use of the TRDS. These requirements (discussed in Shishkina et al. 2001a; 2002; 2003a,b and Anspaugh et al. 2001) are aimed at reducing the uncertainties associated with each step in the evaluation of individual external doses performed on the basis of EPR measurements.

Thus, selection of EPR measurements for any study on validation must be based on this rule: Teeth used for validation must not contain a significant amount of ^{90}Sr in enamel. As discussed above, high levels of ^{90}Sr concentration in enamel and therefore ultrahigh absorbed EPR doses are observed, if the intake of ^{90}Sr occurs during the period of tooth calcification and crown formation. Maximal releases of radionuclides into the Techa River started from March 1950. Therefore, for those individuals who lived on the Techa River since 1949 and earlier, only teeth with crowns completed before 1950 can be used for validation. If a person started to live in the Techa region after 1950, tooth samples from this donor should be used only if teeth from this person would have completed crowns at the date of arrival.

The development of dentition and periods of crown formation were discussed in Tolstykh et al. (2003) and Shishkina et al. (2001a). Table 2 presents the criteria for selection of teeth for

Table 2. Permanent tooth development and criteria of selection for EPR measurements for the validation study based on the tooth donor's age.

Position in denture	Tooth name	Age at crown completion, y	Range of birth years for permanent residents	Minimal permissible age, years
1	Central incisor	4–5	<1946	5
2	Lateral incisor	4–5	<1946	5
3	Canine	6–7	<1944	7
4	First premolar	6.0–8.1	<1943	8
5	Second premolar	6.7–9.3	<1942	9
6	First molar	3.1–4.9	<1946	5
7	Second molar	7.3–10.2	<1941	10
8	Third molar	12.0–17.1	<1934	17

EPR measurement for the validation study according to year of birth of the donor and the position of the investigated tooth.

An additional rule has been introduced for incisors in order to minimize uncertainties in the background contribution to the enamel dose. If possible (and available) only EPR measurements performed on the inner fraction of enamel are considered. If measurements on the inner fraction are not available, then data obtained on the total enamel (inner plus outer fractions) are considered. And, in cases when the inner fraction of enamel has been lost (there are four such cases), EPR measurements obtained on the outer fraction of enamel are considered.

Preliminary assessments of expected values of different contributions to the total enamel dose (Table 3) for adult residents of Metlino (upper Techa region) and Muslyumovo (middle Techa region) were presented at the Techa River Dosimetry Review Workshop held on December 8–10, 2003, at the State Research Centre Institute of Biophysics, Moscow, by E. Shishkina and M. Vorobiova. Background levels were taken from an EPR study of non-exposed donors (described above in Section 2.2). The contributions of betas from $^{90}Sr/^{90}Y$

Table 3. Expected average values of different contributions to total enamel dose for adult permanent residents of Metlino and Muslyumovo.

Settlement	Tooth position	Background dose, mGy	Dose from $^{90}Sr/^{90}Y$ in dentin, mGy	External dose, mGy	Total enamel dose, mGy
Metlino	1	150	50	660	860
	6	90	30		780
Muslyumovo	1	150	70	15	235
	6	90	40		145

incorporated in dentin were evaluated using Monte Carlo simulations and average values of ^{90}Sr concentration in mature teeth (as described by Tolstykh et al. 2000). The contributions of external exposure given in Table 3 were calculated with the use of the TRDS-2000 approach. Assessments are given in Table 3 for central incisors (Position 1) and first molars (Position 6) having maximally different levels of background and beta contributions (other types of teeth have intermediate levels).

It must be noted that the contributions to D_{int} due to ^{90}Sr in enamel, ^{137}Cs in soft tissues and the contributions of all short-lived radionuclides (including ^{89}Sr) were not considered in this preliminary analysis, because such contributions were expected to be relatively small.

As can be seen from Table 3, the contribution of external exposure for Metlino residents is expected as 77–85% of the total-enamel dose, and the component from $^{90}Sr/^{90}Y$ is relatively low (contributing only 4–6%). Also, average levels of the total enamel dose are high enough to be reliably measured by the EPR method. Just the opposite pattern is expected for Muslyumovo where the contribution of external exposure is evaluated as about 6–10%, and the contribution of $^{90}Sr/^{90}Y$ betas is about 28–30% of the total enamel dose. Also, the average levels of the total enamel dose are expected to be low (comparable with the levels of background sources of radiation).

The data from Table 3 are very important, because they have allowed us to determine a strategy for the validation of external dose. These data indicate clearly that for Metlino validation of external doses using the EPR method is expected to be feasible and possibly could even be done on an individual basis. So, teeth from donors who lived in Metlino or nearby downstream villages represent the best opportunity for validation of the TRDS-2000 external doses. The validation task appears to be problematic for residents on the middle and lower Techa region due to high contributions of background and internal exposure and also due to low levels of the total enamel dose. This task could be performed only if the following requirements would be met: (1) the accuracy of EPR measurements is good enough to allow reliable estimates at background levels of dose; (2) the investigated numbers of exposed and background subjects are large enough to provide a statistically significant difference between sample-average values; and (3) the contribution of internal exposure from all sources is evaluated on an individual basis for all tooth samples included in the analysis.

Following this strategy, we focus in this report on data obtained on teeth from Metlino donors, give some preliminary evaluation of the results obtained for the pooled sample of teeth from subjects from the middle and lower Techa region, and outline the issues for the "intermediate" group of subjects, including those residing in settlements located between Metlino and Muslyumovo.

3. CALCULATIONS OF ENAMEL DOSE DUE TO EXTERNAL EXPOSURE ON THE BASIS OF THE TRDS APPROACH

The Techa River Dosimetry System (TRDS) is being developed in the framework of the current project to determine individual-organ doses for the members of the ETRC. The TRDS is a modular database processor; that is, depending on the input data for an individual, various elements of several databases (or modules) are combined to provide the dosimetric outputs requested by the user. These modules were created in 2000 (the so-called TRDS-2000) and can be further updated or improved. It is planned to incorporate the achievements of the period after year 2000 into a new version of the dosimetry system (TRDS-2006) that will be used for individual-dose calculation. One of such achievements is the development of a new two-compartment Techa River Model described in our Milestone 5 Report (Vorobiova et al. 2003). The revised model provides improved results for radionuclide concentration in bottom sediments in 1950–1951 (that can be used for the calculation of external dose rates on the riverbank). Thus, in the validation study we use two versions of external dose calculations: The first version is based on the one-compartment Techa River Model (Model 1 had been used in TRDS-2000); and the second one is based on the new two-compartment model (Model 2 is planned for use in TRDS-2006).

3.1. TRDS-2000 EXTERNAL DOSE ESTIMATES

The method being used for the TRDS-2000-based estimates of external dose is relatively simple and can be written as a single equation:

$$D_{o,Age}^{L} = \sum_{y=1}^{n} A_{o,Age}\, P_{Riv,y}^{L} \left(T_{1,Age} + k_{Out/Riv}^{L} \cdot T_{2,Age} + k_{In/Out} \cdot k_{Out/Riv}^{L} \cdot T_{3,Age} \right), \qquad (2)$$

where:

$D_{o,Age}^{L}$ = cumulative absorbed dose from external exposure (Gy) in organ *o* of a particular individual who lived in location *L* on the Techa River (function of age);

L = river-location (village) identifier;

y = year of environmental exposure;

n = the endpoint of external exposure for a particular individual;

$A_{o,Age}$ = conversion factor from absorbed dose in air to absorbed dose in organ o (function of age, related to y);

$P_{Riv,y}^{L}$ = dose rate in air (Gy year-1) near river shoreline at location L in summer time of year y;

$k_{Out/Riv}^{L}$ = average ratio of dose rate in air outdoors within residence area to dose rate near river shoreline at location L;

$k_{In/Out}$ = ratio of dose rate in air indoors to dose rate outdoors;

$T_{1,Age}$ = time spent on river bank (relative to whole year) (function of age, related to y);

$T_{2,Age}$ = time spent outdoors (relative to whole year) (function of age, related to y); and

$T_{3,Age}$ = time spent indoors (relative to whole year) (function of age, related to y).

The data used for the derivation of parameters in eqn (2) are described in detail by Vorobiova et al. (1999) and Degteva et al. (2000a,b). To evaluate external doses near the shoreline, the results of exposure-rate measurements (available since 1951) were used. To reconstruct external dose rates in 1949–1951, the model (Model 1, Vorobiova and Degteva 1999) describing radionuclide transport along the river and the accumulation of radionuclides by bottom sediments was used. Dose rates in air were calculated on the basis of modeled radionuclide concentrations in bottom sediments and conversion coefficients obtained by Monte Carlo simulations of air kerma for contaminated soil (Eckerman and Ryman 1993) with a dose-reduction factor for river shorelines.

3.2. CHANGES IN TRDS-BASED EXTERNAL DOSE ESTIMATES DUE TO IMPLEMENTATION OF THE TWO-COMPARTMENT TECHA RIVER MODEL

As discussed above, a simple model describing the free flowing Techa River as a one-compartment system was used in TRDS-2000 to derive estimates of gamma-exposure rates near the river shoreline in 1950 and 1951. Recently, a two-compartment model has been developed that provides a better description of the total radionuclide concentration in river water for the middle and lower reaches of the Techa River (Vorobiova et al. 2003). The two compartments consist of the free flowing Techa River divided according to different characteristics of the riverbed and its floodplain. That part of the river from the release point to the village of Muslyumovo is swampy with a poorly marked winding bed overgrown with plants; the bed deposits consist of turf-silt or clay. Downstream of Muslyumovo the river has a well-marked bed consisting of layers of sand and slime and in some places clay.

The dose rates in air near the river shoreline in 1950–1951 were re-calculated on the basis of modeled radionuclide concentrations in bottom sediments obtained by use of the two-compartment model as described above. The results of these calculations are presented in Fig. 1.

As can be seen, for 1950 the two-compartment model gives higher estimates of external dose rate for all distances except for the section 100–120 km from the site of release. The increase in estimates is significant for distances more than 150 km. As for 1951, both models give similar estimates for the upper Techa region, but the new model gives slightly lower values for the middle Techa and slightly higher values for the lower Techa.

Therefore, the implication of the two-compartment river model will result in increased estimates of external doses for the majority of the ETRC members. This increase will be taken into account in the next version of dosimetric system.

3.3. EVALUATION OF EXTERNAL DOSE FOR THE PERIOD AFTER 1960

As discussed in our previous reports, there were some countermeasures taken in the 1950s to minimize the exposure of the residents continuing to live near the Techa River in non-relocated settlements. The use of river water was banned in 1953; the floodplain area within

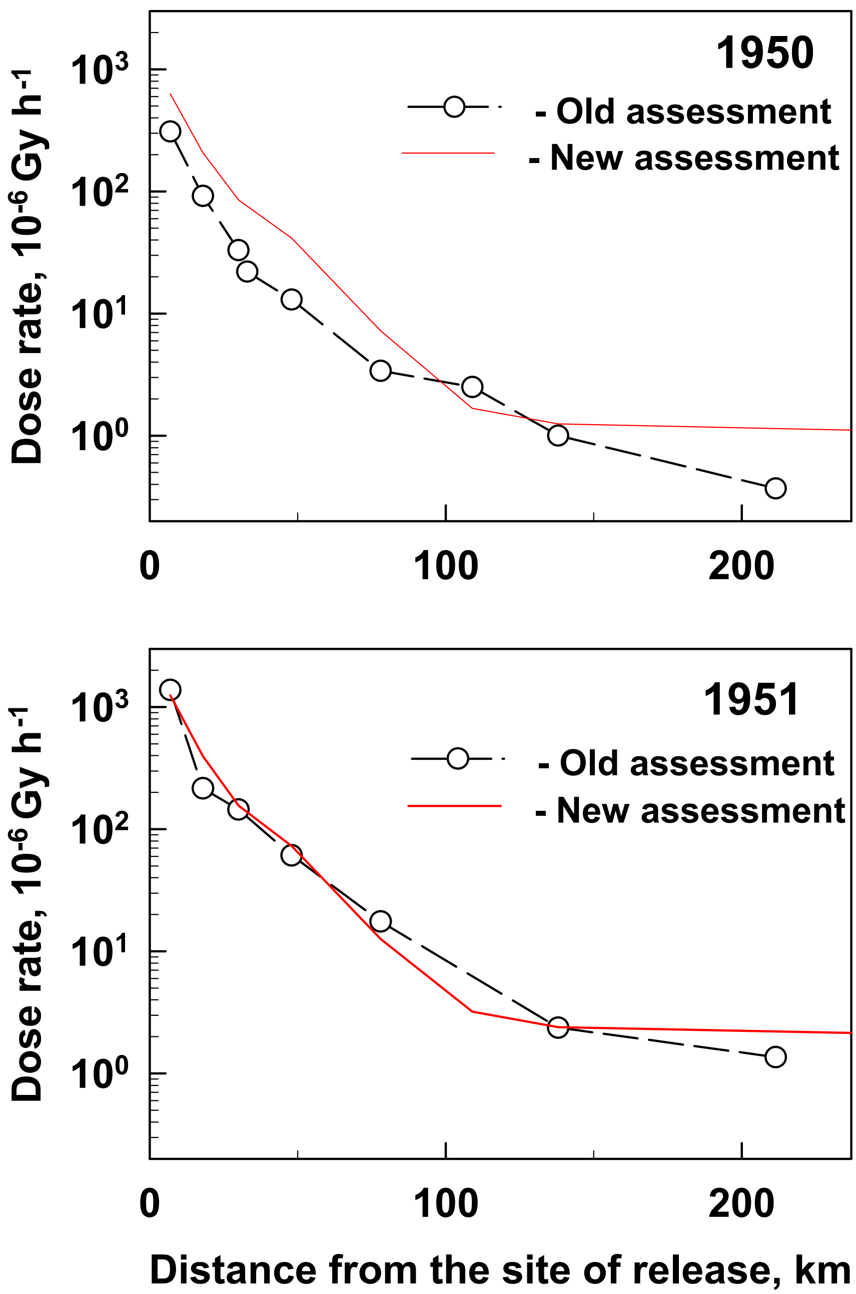

Fig. 1. Old and new assessments of absorbed dose rate in air near the Techa River shoreline in 1950 and 1951.

residence areas was fenced in 1956; persons from households closer to the river in Muslyumovo, Brodokalmak, Russkaya Techa and Nizhnepetropavlovskoye were relocated in 1958–1960 to areas far from the river (but within the residence areas of the same settlements). These countermeasures were the reasons for the assumption implemented in TRDS-2000 that external exposure of the residents stopped in 1960.

However, there are some data supporting that external exposure of residents continued after 1960. First, the ^{137}Cs-body burdens measured by whole-body counter (WBC) for those who lived on the Techa River in the 1970s showed for some persons periodic peaks (up to 37 kBq). This clearly indicated that these persons had contact with the contaminated river. In 1974–1980 the Techa Riverside residents filled out special questionnaires (at the time they were being measured with the WBC) concerning their contacts with the contaminated river. According to these "interview data," 46% of Muslyumovo residents continued to use the Techa River for their domestic needs in that period of time: 19% used river water for drinking; 39% bathed; 14% laundered; 8% fished and 12% mowed grass on meadows.

Another indication of such contacts was described by Kravtsova et al. (1994). This was a time-and-motion study conducted in Muslyumovo by special observers who monitoring times spent by residents near the river shoreline. This study demonstrated that some of the Muslyumovo residents used the river during the 1990s for bathing, fishing, and other domestic needs. As discussed in Vorobiova et al. (1999), the periods of time spent on the river shoreline evaluated by Kravtsova et al. for the 1990s are comparable with the results from Saurov (1992) received in the 1950s and used in the TRDS-2000. Therefore, it is apparent that external exposure of the residents on the Techa River continued after 1960, and it is necessary to evaluate the contribution of this exposure to the total external dose.

Fig. 2 shows the decrease of dose rate in air measured near the Techa River shoreline in Muslyumovo. As can be seen, an exponential decrease with a slope close to the decay constant of ^{137}Cs is observed (0.02349 and 0.02310 y^{-1}, respectively); this looks reasonable, and such information can be used in calculations of external doses for residents in all Techa River settlements.

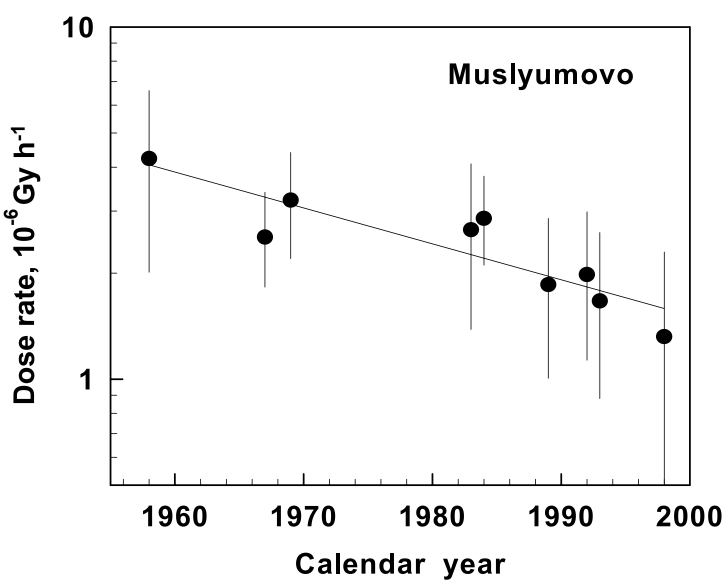

Fig. 2. Decrease of dose rate in air with time near the Techa River shoreline in Muslyumovo.

In addition, small levels of dose rate due to ^{137}Cs in residence areas after 1960 were not taken into account in the TRDS-2000 (because no appropriate estimates existed for these low levels of exposure). According to a recent publication by Golikov et al. (2004), the measurements of ^{137}Cs-dose rates made in 1998–1999 by field spectrometry showed 1.5×10^{-6} Gy h^{-1} for flood plain and 15×10^{-9} Gy h^{-1} for residence areas in Muslyumovo. Respective values for Brodokalmak were 0.4×10^{-6} and 8×10^{-9} Gy h^{-1}. It must be noted that prolonged exposure during 30–40 years at such levels could result in the accumulation of several mGy of external exposure, which would be comparable with the TRDS-2000-based estimates for the first ten years after the start of radioactive releases (1950–1960) for residents on the lower Techa. Therefore, this factor could also contribute to the total external dose and should be evaluated and taken into account in the next version of the dosimetric system.

3.4. REVISED VALUES OF EXTERNAL DOSES FOR DIFFERENT LOCATIONS ALONG THE TECHA RIVER

Revised estimates of external doses absorbed in tooth enamel of the adult permanent residents of settlements along the Techa River have been calculated for the period 1950–1960 with use of external dose rates derived from the two-compartment river model. Results for these revised estimates are shown in Table 4 in comparison with the TRDS-2000-based estimates. In addition, one more change has been included in the revised estimates for this period. According to the assumption made in the TRDS-2000, if the excess dose rates in houses and/or residence areas did not exceed the background-radiation level (which was observed for some settlements in the middle and lower Techa), these very low dose rates were not taken into account in the calculations of external doses. Now, all these small contributions are included in the revised estimates of external doses.

As can be seen from Table 4, the new estimates are not significantly different from the old ones for the upper Techa settlements, where the levels of external dose were relatively high. As for settlements located at more than 125 km from the site of release, the revised estimates are 2–3 times larger. This increase is caused mainly by the implementation of the new two-compartment river model for the calculation of doses in 1950–1951.

The last column in Table 4 shows the total dose accumulated in tooth enamel with the assumption of continuous residence on the Techa River during 1950–2000. As can be seen, if the residents continued to be in contact with the contaminated river during forty years (1960–2000) they received a value of dose comparable with that received during the first ten years after contamination. It must be noted that such contacts were not monitored in the period 1960–1990, and it is not possible to conclude which estimate (from Column 4 or Column 5) is more appropriate. The twofold difference between these two estimates of external dose stresses once more the importance of the application of instrumental methods of retrospective dosimetry (such as EPR and FISH) for the validation of individual external doses on the Techa River.

4. MATERIALS AND METHODS USED IN THE EPR STUDY

All EPR measurements described in this report were carried out on extracted permanent teeth from members of the ETRC; such teeth were selected on the basis of information from the

Table 4. Comparison of TRDS-based estimates of external dose absorbed in tooth enamel of permanent adult residents of different settlements along the Techa River.

Settlement	Distance from site of release, km	TRDS-2000 external dose, mGy	Revised external dose absorbed before 1960, mGy	Total external dose for the period 1950-2000, mGy
Metlino	7	663	694	-
Techa Brod	18	623	710	-
Asanovo and Nazarovo	33	227	236	-
M. Taskino	41	128	145	-
Gerasimovka	43	216	238	-
GRP	45	79	93	-
Nadyrov Most	48	54	65	-
Nadyrovo	50	137	148	-
Ibragimovo	54	125	140	-
Isaevo	60	67	77	-
Podssobnoe hoz.	65	37	44	-
Muslyumovo	78	14	18	41
Kurmanovo	88	10	14	-
Karpino	96	11	17	-
Zamanikha	100	9	12	-
Vetrodujka	105	7	11	-
Brodokalmak	109	6	9	23
Osolodka	125	4	8	-
Panovo	128	4	7	-
Cherepanovo	137	4	8	-
Russkaya Techa	138	3	6	15
Baklanovo	141	4	7	-
Nizhnepetropavlovskoye	148	3	6	17
Beloyarka-2	155	5	10	-
Lobanovo	163	4	9	22
Anchugovo	170	3	8	19
Verkhnyaya Techa	176	3	8	19
Skilyagino	180	2	7	17
Bugaevo	186	2	7	17
Dubasovo	200	2	7	-
Bisserovo	202	3	8	20
Shutikhinskoye	203	3	8	20
Progress	207	3	8	-
Pershinskoye	212	3	8	20
Klyuchevskoye	223	3	8	20
Ganino and Markovo	230	3	8	-
Zatechenskoye	237	3	8	20

database "TOOTH." Birth years of the donors were in the range from 1912 to 1945. Crown (enamel) formation was completed before the start of exposure for all samples included in the current analysis (the requirement discussed in Section 2.4).

4.1. DESCRIPTION OF TOOTH GROUPS UNDER INVESTIGATION

Data on the total number of teeth and EPR measurements for each settlement along the Techa River are presented in Table 5. As can be seen, 192 donors have been investigated in total. From one to five teeth were measured per donor (292 teeth in total). The number of EPR measurements (349) is larger than the number of teeth because some teeth have repeated EPR measurements performed in different laboratories.

It must be noted that the number of investigated teeth from exposed donors is about three times lower than indicated previously by Anspaugh et al. (2001) as the number of teeth required for analysis in order to validate reliably estimates of external dose for the members of the ETRC. Nevertheless, validation of calculations of external dose, even at this stage of development, is considered essential, as the allegation has been made that current calculated estimates of external dose are too low by a factor of three to five (Mokrov 2002; 2003; 2004).

4.2. EPR-DOSE RECONSTRUCTION

The majority of EPR measurements have been performed at the Institute of Radiation Protection, GSF-National Research Center for Environment and Health (GSF), Munich, Germany; and at the Institute of Metal Physics (IMP), Ekaterinburg, Russia. The EPR techniques used in the laboratories have been described in detail elsewhere (Wieser et al. 2000a,b; Shishkina et al. 2003a). The EPR spectrometers and procedures used for enamel-dose reconstruction are shown in Table 6. As can be seen, similar procedures of sample preparation (chemical treatment in concentrated alkali solution), spectrum processing (deconvolution using a set of Gaussian functions), and dose evaluation (universal calibration curve) have been used in both laboratories. Both laboratories participated in several international intercomparisons (Romanyukha et al. 2000; Wieser et al. 2000a,b). The intercomparisons have demonstrated that contemporary EPR-dosimetry systems can reconstruct doses within ±21% for levels higher than 390 mGy, and ±82 mGy for doses below 390 mGy, with a probability of 0.90.

In order to confirm that the data received can be analyzed together, the accuracy of the GSF and the IMP EPR-dosimetry systems was specially checked using 51 samples obtained from Urals donors (Ivanov et al. 2001; Shishkina et al. 2001a). A correlation coefficient of 0.99±0.01 (p <0.001) was obtained between the results of the two laboratories. The slope of the regression line was very close to unity, which indicated that the IMP and the GSF calibrations for radiation sensitivity of the samples agree within 1%. The results also demonstrated the absence of a systematic bias between EPR doses measured by the two laboratories, and thus provided the basis for combined analysis of the data.

Table 5. Total number of teeth and EPR measurements for each settlement along the Techa River (status on June 2004).

Settlement	Distance from site of release, km	Number of donors	Number of teeth	Number of EPR measurements
Metlino	7	13	27	40
Techa Brod	18	-	-	-
Asanovo and Nazarovo	33	13	17	18
M. Taskino	41	6	8	10
Gerasimovka	43	15	30	46
GRP	45	10	16	22
Nadyrov Most	48	5	7	12
Nadyrovo	50	9	23	28
Ibragimovo	54	6	12	14
Isaevo	60	17	30	32
Podssobnoe hoz.	65	7	12	12
Muslyumovo	78	30	39	39
Kurmanovo	88	26	30	32
Karpino	96	1	1	1
Zamanikha	100	3	3	5
Vetrodujka	105	-	-	-
Brodokalmak	109	10	11	12
Osolodka	125	1	1	1
Panovo	128	-	-	-
Cherepanovo	137	1	3	3
Russkaya Techa	138	4	4	4
Baklanovo	141	-	-	-
Nizhnepetropavlovskoye	148	2	3	4
Beloyarka-2	155	1	1	1
Lobanovo	163	2	2	2
Anchugovo	170	-	-	-
Verkhnyaya Techa	176	-	-	-
Skilyagino	180	-	-	-
Bugaevo	186	1	1	1
Dubasovo	200	1	2	2
Bisserovo	202	-	-	-
Shutikhinskoye	203	-	-	-
Progress	207	-	-	-
Pershinskoye	212	2	3	3
Klyuchevskoye	223	3	3	4
Ganino and Markovo	230	-	-	-
Zatechenskoye	237	3	3	3
Total		**192**	**292**	**349**

Table 6. Summary of methods used for EPR-dose reconstruction (a detailed description is given in Wieser et al. (2000a) and Shishkina et al. (2003b).

Laboratory	Equipment	Sample-preparation method	Signal-evaluation method	Dose-calibration method
GSF, Germany	Bruker, ECS106	Chemical treatment with NaOH (grain size 0.1–0.6 mm)	Deconvolution using a set of Gaussian functions	Universal calibration curve obtained with permanent molars from German donors
IMP, Russia	GDR, ERS231	Chemical treatment with KOH (grain size 0.1–0.6 mm)	Deconvolution using a set of Gaussian functions	Universal calibration curve obtained with permanent molars from Urals donors

4.3. CRITERION FOR THE ESTIMATION OF EPR-MEASUREMENT RELIABILITY

Information on more than 800 EPR measurements on tooth enamel is currently available in the URCRM database. Teeth were available from various positions and were of different health conditions; consequently, the quality of measured samples varied and this fact affected the reliability of estimated doses. In addition, samples were measured in different laboratories using different procedures and equipment. Thus, the quality of data available is not identical and the relative reliability of the measured results should be considered in the analysis of the whole data set.

In the course of the current investigation, a criterion for the reliability of EPR measurement results has been established. At present each EPR measurement is characterized by a certain value, called the "criterion of measurement," which is evaluated as a sum of points. A detailed description of this criterion is presented in Appendix 1.

The reliability criterion is based on factors affecting EPR-dose-estimation quality, such as (1) tooth position; (2) sample weight; (3) estimation of spectrum quality with "signal-to-noise" ratio; (4) laboratory that performed the measurements; and (5) coefficient of variation. A number of points is assigned to the EPR measurement for each factor and then points of all factors are added together. The number of points for each criterion depends on the measured dose. The total number of points represents the criterion of measurement reliability. The total of all five factors summing to the criterion of measurement reliability could vary from 0 to 16.5.

This criterion is useful for the evaluation of EPR data. Also, this criterion is used when EPR measurements for different tooth samples (when available from one person) are averaged to receive the most reliable estimate of enamel dose (Section 4.4).

4.4. STATISTICAL METHODS USED FOR EVALUATION OF ENAMEL DOSE

4.4.1. Statement of the problem

Measurements, X_j, of the value A received during different conditions in compliance with the model of measurements can be described by the following:

$$X_j = A + \xi_j, \quad j = 1,2...N . \tag{3}$$

The inaccuracy of the measurements, ξ_j, is characterized by an average value of zero (there is no constant bias) and by the dispersion σ_j^2, generally speaking, for various values of j. The various measurements are assumed to be independent.

Expert evaluation of the quality of each separate measurement with subscript j is the whole positive number *"c"*. These values vary from *0* up to *C,* which are assigned to the measurement in compliance with the rule – the greater value of an expert criterion is answered by the less qualitative measurement (i.e., more reliable measurements are assigned a smaller value of points).

It is necessary to receive an estimation $\hat{A}$ of the measured value A; this is done by taking into account the accuracy of the measurements and expert criteria (described in Section 4.3 and Appendix 1).

4.4.2. The weighted least squares method

Let's attribute to each measurement weight, W_j, which takes into account the ***comparative value*** of the measurement, information on the accuracy of the measurement, and an expert estimation of the quality of this measurement.

The numbers W_j have the following properties:

$$W_j > 0, \tag{4}$$

$$\sum W_j = 1, \text{ and} \tag{5}$$

$$W_j = k \cdot f(c_j) \frac{1}{\sigma_j^2}. \tag{6}$$

Function $f(c)$ in eqn (6) is monotonically decreasing, and when *"c"* is equal to zero takes on a value of 1. The constant, k, may be obtained for the given function $f(c)$ from the condition in eqn (7):

$$k = \frac{1}{\sum f(c_j)\frac{1}{\sigma_j^2}}. \qquad (7)$$

Function $f(c)$ accumulates a priori information on the comparative relative value of various measurements. Different non-experimental reasons can be used for its construction.

The estimation of $\hat{A}$ in this situation can be found with the use of the weighted least squares method with weights W_j:

$$\sum (X_j - \hat{A})^2 \cdot W_j \quad \Rightarrow \quad \min; \qquad (8)$$

the result is

$$\hat{A} = \sum W_j \cdot X_j . \qquad (9)$$

We note that this estimation is unbiased $M(\hat{A}) = A$ and has dispersion equal to

$$D(\hat{A}) = \sum W_j^2 \cdot \sigma_j^2 = \frac{\sum_{j=1}^{N} \frac{f^2(c_j)}{\sigma_j^2}}{\left[\sum_{j=1}^{N} \frac{f(c_j)}{\sigma_j^2}\right]^2} = \sigma^2(\hat{A}). \qquad (10)$$

4.4.3. Construction of the function *f*(*c*)

As discussed above, the criterion of reliability is defined by five factors and the sum of the points for each factor can vary from 0 up to 16.5. The analysis of the available EPR measurements and a priori estimations of the quality of these measurements allows the choice of a ratio:

$$f(c) = \frac{1}{\left(1 + (\alpha \cdot c)^2\right)}, \qquad (11)$$

as a function $f(c)$.

Thus, the factor α that determines the rate of decrease and position of a point in excess of the function, is derived from the condition that the importance of measurements, which value of criterion "c" is in a range from 0 up to 4 points, would not be less than 0.5 (Fig. 3). The result is $\alpha = 0.25$.

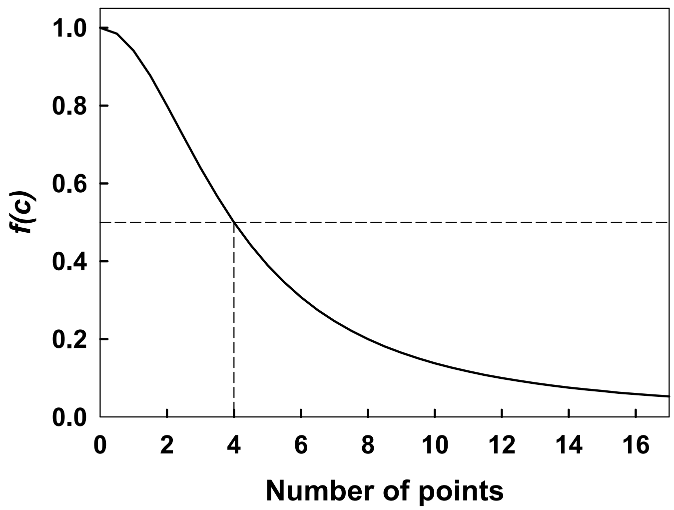

Fig. 3. Function f(c), where "c" is the criterion of EPR measurement reliability.

4.4.4. Variation of the estimation

The total number (M) of donors surveyed was 192; thus, the j^{th} donor has N_i EPR measurements, $X_i^j, i = 1,2,...N_j$. The total number of measurements is 349. We shall denote the i^{th} weight of measurement for the j^{th} donor as w_i^j, and the standard deviation as $\sigma(\hat{A}_j)$.

Then, we consider the standardized residual, which is determined as

$$r_i^j = \frac{w_i^j\left(X_i^j - \hat{A}_j\right)}{\sigma(\hat{A}_j)}, i = 1,2,..., N_j, \quad j = 1,2,..., M \,. \tag{12}$$

The standardized residuals have the distribution given in Fig. 4 and are characterized with an average of 0.0 and standard deviation equal to 1.52.

The frequency analysis of this distribution shows that it is compactly concentrated around zero: All observations are between -11.6 and 13.3; thus, 95 % of all observed residuals are in an interval from-1.65 till 1.71, and 98 % in the interval from -6.15 up to 6 (Table 7).

So with reliability $1-\alpha$ (α is negligible quantity), not less then $(1-\alpha)$ ×100 % of the measurements will be within the limits:

$$\hat{A}_j - \frac{r_{1-\alpha} \cdot \sigma(A_j)}{w} \le X \le \hat{A}_j + \frac{r_{1-\alpha} \cdot \sigma(A_j)}{w}, \tag{13}$$

where

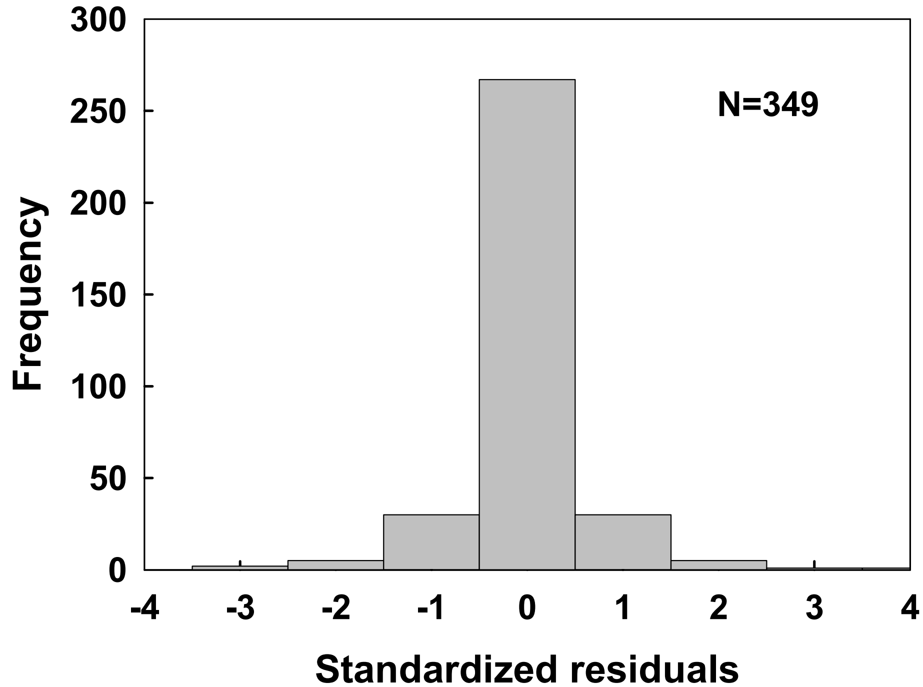

Fig. 4. Frequency histogram of standardized residuals.

Table 7. Frequency analysis of the distribution of standardized residuals.

Percentile	Reciprocal
0.1	-11.628
0.5	-10.427
1.0	-6.150
2.5	-1.649
97.5	1.708
99.0	5.596
99.5	12.038
99.9	13.268

$r_{1-\alpha}$ = α - reciprocal distributions ($P(|r| < r_{1-\alpha}) = 1 - \alpha$) and

w =weight of measurement.

For example, it is possible for $r_{1-\alpha}$ to take the value of 1.7 for $\alpha = 0.05$; for $\alpha = 0.1$ the corresponding value of $r_{1-\alpha}$ =1.1.

The method described in this section has been applied to the donors who have more than one EPR measurement in order to receive the most reliable estimate of average enamel dose. It must be noted that the majority of such donors are residents of the upper Techa region (the most important group in the validation study).

5. ESTIMATION OF STRONTIUM CONTRIBUTION TO ENAMEL DOSE

As discussed above, evaluation of the dose component in enamel contributed by $^{90}Sr/^{90}Y$ and ^{89}Sr distributed in dental tissues is a very important task for the success of the validation study (because strontium concentration can vary from tooth to tooth). Work has begun recently to perform parallel measurements of ^{90}Sr concentrations in all tooth samples in order to evaluate carefully and to subtract the internal dose component from the total enamel dose measured by the EPR method. The thermoluminescent (TL)-contact detection method based on thin layer α-Al_2O_3:C detectors is used (Göksu et al. 2002; Veronese et al. 2004; Shishkina et al. 2004). This method is not destructive and it has high sensitivity. Nevertheless, only a few results of measurements are currently available from this method. A correct interpretation of TL data (as well as of other information derived from measurements of exposed people) requires careful assessment of background levels before the start of routine measurements. Also, it must be noted that in order to receive reliable results, the detectors must be exposed during about 30–40 days, and only 6–10 samples can be measured simultaneously. Therefore, it is possible to anticipate that the results of individual-^{90}Sr assessments for the majority of teeth that have been measured by EPR method would be available not earlier than 1–2 years.

Nevertheless, as noted above, validation of calculations of external dose, even at this stage of development, is considered essential in order to resist the allegation that the use of the TRDS-2000 significantly underestimates actual values of external dose (Mokrov 2002). The following two arguments provide support for this validation study:

1. The levels of total enamel dose for residents of Metlino are high enough to be measured reliably by the EPR method, and the contribution of $^{90}Sr/^{90}Y$ betas is estimated as 4–6% of the total dose (Table 3). Therefore, even if the contribution of internal exposure is not subtracted from the total dose, EPR-based estimates can be considered as upper limits of actual external doses, and can be compared with the TRDS-based assessments for residents of Metlino.
2. A preliminary evaluation can be performed also for EPR measurements on a pooled sample of teeth from permanent residents, who lived in the middle and lower Techa regions during the major period of massive releases, 1950–1953. Average levels of ^{90}Sr concentration determined on the basis of radiochemical measurements carried out in 1959–1964 for a similar group of subjects can be assigned to this pooled sample. Thus, an approximate estimate of average dose due to betas can be made and then subtracted from the total enamel dose. Also, it should be noted that all EPR measurements for this group of subjects have been performed on posterior teeth (this avoids the issues inherent to anterior teeth, such as larger uncertainties of EPR measurements due to small mass of enamel and higher levels of background signal). As discussed above (Section 2.3), the majority of radiochemical measurements were also carried out on posterior teeth.

This section includes the analysis of radiochemical measurements for the group of permanent residents of the middle and lower Techa regions, and the evaluation of average enamel doses due to strontium incorporated in tooth dentin.

5.1. ANALYSIS OF DATA ON ^{90}SR CONCENTRATION IN DENTAL TISSUES

Radiochemical data are available for 152 permanent residents of the middle and lower Techa regions who were born during or before 1946 (the range of birth years that should be used for EPR analysis according to information in Table 2). The dependence of average ^{90}Sr concentration on donor's age at the beginning of intake (in 1950) is shown in Fig. 5. As can be seen, a significant decrease of ^{90}Sr concentration in dentin with age in 1950 is observed, and this decrease levels off only after the age of 17–19 y. As discussed by Tolstykh et al. (2003), the dentin (root dentin in particular) has a longer period of maturation than does enamel, and this phenomenon can explain the age dependence observed. The constant ^{90}Sr contents after the age of 19 y indicates that the process of mineral metabolism in adult teeth occurs at a relatively constant rate.

The age dependence of average ^{90}Sr concentration in dentin presented in Fig. 5 is described by the function:

$$Y = Y_0 + a\exp(-bX)\,, \tag{14}$$

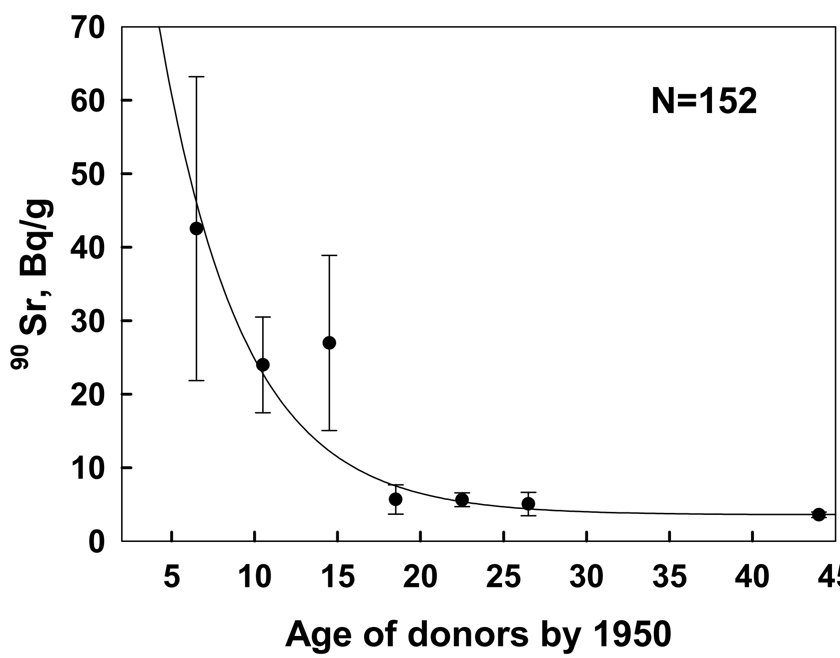

Fig. 5. Age-dependent average concentration of ^{90}Sr in tooth samples from permanent residents of the mid and lower Techa regions. Results were obtained by radiochemical analysis. Bars reflect standard errors.

where

Y = ^{90}Sr concentration in dentin, Bq/g;

X = age of donor in 1950; and

Y_0 = 3.6, a = 154, and b = 0.20 – fitted values of parameters.

As can be seen, this function (shown by the curve in Fig. 5) satisfactorily describes experimental data and thus can be used for evaluation of enamel dose from $^{90}Sr/^{90}Y$ incorporated in tooth dentin.

Nevertheless, as mentioned by Tolstykh et al. (2003), exact tooth positions are not available for the majority of the samples (it is known only that molars and premolars predominantly were measured). The results obtained for age group 5–10 y can include some samples with uncompleted crown (Positions 4, 5, 7 and 8). The large uncertainty inherent to this age group also indicates such a possibility. Therefore, in the current analysis we consider only donors older than 10 y at the beginning of intake (birth years less 1940). This warrants that the enamel of all posterior teeth (except wisdom teeth) has been completed.

As discussed by Tolstykh et al. (2003), the metabolism of strontium and calcium in enamel after completion is extremely slow due to the high level of mineralization of enamel and the absence of living cells. It was shown by Ivanov and Kulish (1959), Saurov et al. (1972) and Romanyukha et al. (2002) that, for persons who were adult during the period of intake, ^{90}Sr is concentrated in dentin mainly on the surface of the pulp channel and in the root. Nevertheless, such processes as the non-organic ion turnover at the enamel surface and ion transport with enamel fluid occur in mature enamel and can cause the incorporation of some amounts of ^{90}Sr into the enamel of such teeth. Limited data sets on ^{90}Sr incorporation in the enamel of mature teeth are available. Kleschenko et al. (1994) reported about 0–0.2 Bq of ^{90}Sr per gram of enamel on the basis of low-level beta counting measurements of four samples from the residents of the lower Techa region. According to Shishkina et al. (2001b) the enamel-to-dentin ratio of ^{90}Sr concentration after a single intravenous intake in adult dogs is between 0.1 and 0.3. The first preliminary data obtained by the TL-contact detection method for teeth from residents on the upper Techa region gave similar values for enamel-to-dentin ratio (0.2–0.4). The contribution to enamel dose due to low-level strontium concentration in the enamel itself cannot be evaluated until appropriate TL data become available. Thus, the estimates of enamel dose due to strontium incorporation in dentin presented in this section should be evaluated as an underestimation of the actual dose due to beta exposure.

5.2. MONTE CARLO SIMULATION OF DOSE DISTRIBUTION IN DENTAL TISSUES

Enamel and dentin dose-rate coefficients were calculated for two different tooth models: a simple tooth model (Shved and Shishkina 2000) and an accurate model (Shishkina et al. 2002). In this paper we use dose-rate coefficients calculated for the simple model by Monte Carlo simulation of electron transport using the CASCADE-5 code (Lappa and Burmistrov 1994). The coupled electron-photon transport physics in the CASCADE-5 code takes into account in a rather accurate way the diffusion and slowing down of all radiations in the electron-photon cascade established in the heterogeneous and axis symmetric media in the energy range 10^{-2}–10^{9} MeV. This code uses Moliere theory and includes unique non-analogue methods for both linear transport characteristics and fluctuations. The constants of interaction between electron and substance were calculated using data on user-defined chemical composition of the cylindrical zone media and their density. The following approximations were used with consideration that teeth at various positions in the denture have different shapes:

1. The CASCADE-5 program is limited to cylindrical geometry. In this case we can describe the plate geometry only as a prism with a circular base. The crown of anterior teeth (incisors and canines) was described as a system of parallel plates: enamel-dentin-enamel (Fig. 6). The crown of posterior teeth (molars and premolars) was modeled as a system of cylinders inserted into each other (Fig. 6). The model dimensions of each tooth in the denture were developed on the basis of odontometric investigations of teeth from members of the Urals population (Shved and Shishkina 2001).
2. The density of tooth tissues was considered as a constant; and
3. The distribution of ^{90}Sr/^{90}Y in the sources within tooth tissues was assumed uniform.

The photon and electron thresholds were stipulated as 10^{-2} MeV. The constants of interaction between electron and substance were calculated using data on the chemical composition of tooth tissues (Derise and Ritchey 1974; Bazhanov 1987; Driessens and Verbeeck 1991) and their density (Manly et al. 1939).

Based upon the described assumptions and results of odontometric measurements of average teeth sizes for the Urals residents, the dose-rate coefficients (*DRC*s) for different source and target tooth tissues have been calculated by the Monte Carlo method. Enamel and dentin dose-rate coefficients were computed for each tooth of the denture. The unit of the dose-rate coefficients is Gy s^{-1} per Bq g^{-1}. The results of calculations are presented in Appendix 2. As can be seen, the values of *DRC* for "enamel→enamel" are 3–9 times higher than for "crown dentin→enamel." This means that small concentrations of ^{90}Sr in enamel can produce absorbed

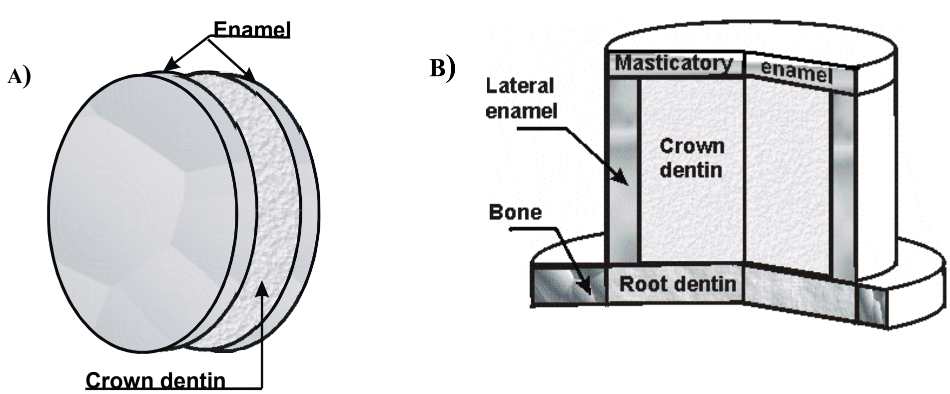

Fig. 6. Geometrical approximations of tooth geometry: (a) for anterior teeth; (b) for posterior teeth.

doses comparable with those produced by much higher concentrations of ^{90}Sr incorporated in the dentin. This should be taken into account in future investigations, when TL data on estimates of ^{90}Sr concentration in enamel for each investigated tooth will be available.

DRC values for crown and root dentin as source tissues and enamel as target tissue are used in the calculations of enamel dose due to beta exposure described in the next section.

5.3. CALCULATION OF ENAMEL DOSE DUE TO BETA EXPOSURE FROM STRONTIUM RADIONUCLIDES INCORPORATED IN DENTIN

Enamel doses are calculated as the product of dose-rate coefficients for respective types of teeth and the numbers of radioactive disintegrations per gram of dentin from 1950 through the year of EPR measurement. For the calculation of the number of disintegrations it was assumed that all ^{90}Sr was ingested in 1950 and after that the strontium content in dentin decreased with an effective rate of 4.25% per year (Tolstykh et al. 2000). The absolute values of ^{90}Sr retention in dentin were fitted to the radiochemical data of 1960–1964 (given in Fig. 5).

In order to calculate ^{90}Sr concentration for specific tooth position eqn (14) has been re-written by replacing 'donor age' with 'tooth age.' Donor age at the beginning of calcification in the crowns of posterior teeth varies from 0 (for the first molars) to 7.5 years (for the third molars) with the average value equal to 2.9 year (see table 2 in Tolstykh et al. 2003). Because eqn (14) describes the average for all posterior teeth, it is assumed that the conversion from 'donor age' to 'tooth age' can be achieved simply by moving the curve to the left by 2.9 y. Such modification allows us to evaluate more precisely the ^{90}Sr concentrations for each tooth sample used in the EPR study by taking into account the age of each tooth.

For the calculation of enamel dose due to beta exposure contributions from both $^{90}Sr/^{90}Y$ and short-lived ^{89}Sr are taken into account. It must be noted that some simplifications were assumed in these preliminary calculations of dose due to short-lived ^{89}Sr. In particular, it was assumed that the amounts of intake of ^{89}Sr and ^{90}Sr were equal (according to the source term used in TRDS-2000, the ratio of ^{89}Sr-to-^{90}Sr is equal to 0.9) and the *DRC* values for ^{89}Sr were taken as equal for all posterior teeth (0.01095 nGy s^{-1} per Bq g^{-1} for crown dentin and 0.0002 nGy s^{-1} per Bq g^{-1} for root dentin).

For the calculation of the number of disintegrations it was assumed that all ^{90}Sr and ^{89}Sr was ingested in 1950. After ingestion the ^{90}Sr concentration in dentin decreased with an effective rate of 4.25% per year (the rate of biological elimination plus radioactive decay) for a living tooth. After tooth extraction ^{90}Sr concentration decreased due to radioactive decay only (2.4 % per year). The rate of decrease of ^{89}Sr activity is assumed to be 50.2% per year (only radioactive decay) both for the living tooth and for a tooth after extraction.

The number of disintegrations, *N*, in the time period $(T_2–T_1)$ is calculated using eqn (15):

$$N = \int_{T1}^{T2} N_0 \cdot \exp(-\lambda T)dt \,, \qquad (15)$$

where N_0 is the radionuclide concentration in dentin at time T_1 and λ is the rate of decrease.

The method of internal beta-dose calculation described in this section has been applied to all tooth samples from the middle and lower Techa regions measured by the EPR method. The comparison of the calculated results with EPR-based enamel doses is presented in Section 6.3. It is stressed again that this approach must be considered as preliminary and can be applied only to *pooled samples* of EPR measurements for permanent residents, who lived in the middle and lower Techa regions. Average levels of ^{90}Sr concentration in dentin determined on the basis of radiochemical measurements for a *similar group of subjects* have been assigned to this pooled sample. As discussed above, careful evaluation of the internal dose component *on a individual basis* (including all sources of internal exposure for each tooth sample available for EPR analysis) is a very important task for the success of the validation study. This issue will be discussed further in Section 7.2.

6. VALIDATION OF EXTERNAL DOSE ESTIMATES USING EPR DATA

6.1. ANALYSIS OF EPR DATA

It follows from data in Table 5 that there are not enough EPR measurements at this time to validate the TRDS-2000 estimates of external doses on village-by-village basis, because the number of donors is small for several settlements and sometimes is zero. Nevertheless, even at this stage of development, EPR data are available for the entire range of distances from the site of releases, and thus a preliminary evaluation of the dependence of average EPR dose for permanent residents ($N = 140$) on distance from the release site can be given (Table 8).

As seen from Table 8, the EPR data were grouped in six clusters by distance from the release site. Some decrease in the average EPR dose is observed for the upper Techa region (<60 km from the site of release). This decrease is not monotonic, because the third cluster (45–50 km) gives the same value of median and two times higher value of mean in comparison with the first cluster (7 km). This relatively small cluster includes two persons (both lived in the same household) with very high levels of enamel dose (6.1 Gy and 2.4 Gy). This special case is discussed in detail below (Section 7.5). Standard errors of the mean are large for teeth from the upper Techa region (0.1–0.6 Gy), which can be explained by the large individual variability of EPR doses and the small numbers of donors in the first three clusters. As discussed in Degteva et al. (1997), it is very difficult to collect samples from the residents of the upper Techa region, because they were evacuated from their initial places of residence (and some of them were evacuated twice, because their new residence sites were contaminated as a result of the Kyshtym accident in 1957).

The last three clusters have very close values both for means and medians, which could be interpreted as the absence of a distance dependence beyond 60 km, or very small (statistically insignificant) decrease in average EPR dose for the middle and lower Techa regions. Standard errors of the mean are smaller than for the upper Techa region (0.03–0.07 Gy).

Table 8. Evaluation of the dependence of EPR-based enamel dose for permanent residents on distance from the site of release (background subtracted).

Cluster/ Locations	Distance, km	Number of donors	Mean EPR dose±SEM, mGy	Median EPR dose, mGy	Maximum EPR dose, mGy
Metlino	7	10	680±200	516	2327
Asanovo, M.Taskino, Gerasimovka	30-43	19	310±90	202	1233
GRP, Nadyrov Most, Nadyrovo[a]	45-50	11[a] 9	1030±540 330±100	395 159	6094 830
Ibragimovo, Isaevo, Podssobnoe	54-65	25	250±70	128	1272
Muslyumovo, Kurmanovo	78-88	46	250±60	156	2550
Lower Techa	96-237	30	200±30	158	720

[a] There are two persons (both lived in the same household) with very high levels of enamel dose (6.1 and 2.4 Gy) in this cluster. The evaluation has been done for the whole sample ($N = 11$) and also for the sample without these two persons ($N = 9$).

As discussed above, in the current section of this report we have included a detailed analysis of data from the first cluster (Metlino) and a preliminary evaluation of pooled data from the last two clusters (middle and lower Techa regions). Issues of validation for persons who resided at distances 30–65 km from the site of releases is discussed below in Section 7.6.

6.2. VALIDATION OF TRDS-2000-BASED EXTERNAL DOSES FOR RESIDENTS OF METLINO

The results of EPR measurements (performed on teeth with crown completed at the beginning of exposure) for the members of the ETRC who resided in Metlino are presented in Table 9. The total-enamel dose for each tooth sample measured at the IMP and the GSF (without subtraction of background dose) is given in this table. As can be seen, from one to five teeth were measured per donor. The scattering of EPR results for different teeth from the same donor may be explained by uncertainty of the EPR method as well as by differences in background levels for teeth at different positions in the denture.

In order to receive the most reliable estimate of enamel dose for each person due to his/her residence on the Techa River, EPR measurements for different tooth samples (when available from one person) were averaged after the subtraction of background EPR levels for the respective types of tooth samples.

Table 9. Results of EPR investigation for Metlino residents having mature teeth in the period of exposure.

Individual Code (IC)	Birth year	Type of sample[a]	Total EPR dose[b], Gy	
			GSF	IMP
470	1932	Molar	0.47±0.10	0.38±0.08
		Premolar	0.31±0.08	0.37±0.08
		Incisor #1 (inner)	0.53±0.11	-
		Incisor #2 (inner)	-	0.24±0.08
		Incisor #3 (total)	-	0.24±0.08
1154	1935	Incisor (total)	1.10±0.23	-
4235	1938	Premolar	0.07±0.08	0.19±0.10
		Incisor #1 (inner)	-	0.32±0.08
		Incisor #2 (inner)	-	0.16±0.10
4531	1936	Molar	0.51±0.10	0.56±0.13
4759	1931	Molar	-	0.91±0.19
6903	1937	Incisor #1 (inner)	0.76±0.16	-
		Incisor #2 (total)	-	0.67±0.14
7411	1937	Incisor (total)	-	0.72±0.15
8092	1930	Molar	0.10±0.08	0.07±0.08
8451	1933	Premolar	0.37±0.08	0.40±0.15
12460	1942	Incisor #1 (total)	0.52±0.11	-
		Incisor #2 (total)	0.56±0.12	-
		Incisor #3 (total)	0.92±0.19	-
18255	1937	Molar	2.68±0.55	2.20±0.45
19287	1935	Premolar	0.32±0.08	0.26±0.08
		Molar	0.41±0.08	0.18±0.09
20436	1928	Molar	-	0.14±0.08

[a]Ciphers indicate the numbers of teeth from the donor, if several samples of the same type have been measured.
[b]Including background dose

A comparison of measured EPR results for persons having mature teeth at the period of exposure ($N = 13$) with enamel doses calculated using the TRDS-2000 approach specific for these persons' village-residence histories is shown in Fig. 7 and Table 10. The slope of the linear fit to the data is equal to 0.91, and the Spearman rank-correlation coefficient is reasonable ($r_s = 0.66$, $p < 0.05$). However, the relationship of the two data sets does not appear as good as the statistics imply; this requires discussion.

The estimates of external dose calculated with use of the TRDS-2000 are determined mainly by input data on village-residence history. For the data on Metlino, there are three visual clusters in Fig. 7: (1) two points close to zero, which represent persons exposed less than one year; (2) one point having an intermediate position (a person exposed during three years); and (3) ten points with similar values of TRDS-2000 doses (0.65–0.7 Gy) belonging to permanent residents (exposed during 6–7 years). As discussed above, the TRDS-2000 estimates are based on village-average parameters; that is why these estimates for permanent residents have such a

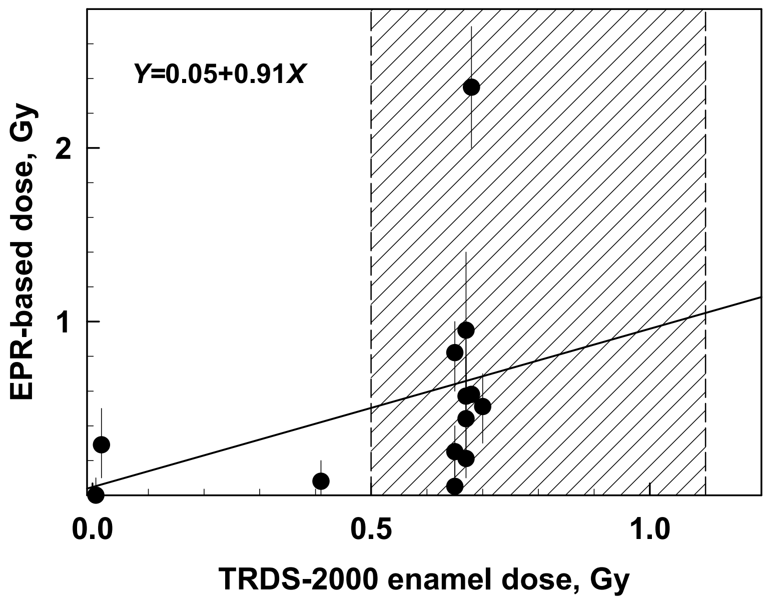

Fig. 7. Correlation between individual enamel doses evaluated on the basis of EPR measurements of persons with mature teeth at the period of exposure and individualized external enamel doses calculated with use of the TRDS-2000 approach.

Table10. Comparison of EPR-based dose estimates for Metlino residents with mature teeth at the period of exposure (N = 13) and enamel doses evaluated for the same sample of persons using the TRDS-2000 approach.

Statistical parameter	EPR-based dose, Gy	TRDS-2000 dose, Gy	Test for difference (*p*-values)[a]
Median	0.44	0.67	0.35
Mean ± SEM	0.55 ± 0.17	0.55 ± 0.07	0.99
90% CI for mean	0.27 – 0.83	0.43 – 0.67	
Spearman rank correlation: 0.66 (p <0.05)[b]			

[a] The Wilcoxon signed rank test is used for medians, and the *t* test for independent samples is used for means.

[b] Statistically significant

narrow range. Nevertheless, the 90% confidence intervals evaluated using Monte Carlo simulations (shown in Fig. 7 as the shaded zone) are relatively large (0.5–1.1 Gy). The large uncertainty reflects the current non-consideration of the precise location of an individual within Metlino. Work is now underway to improve TRDS-2000 estimates by assigning individuals the location of their historic residences. This will result in splitting of the third cluster and will allow better evaluation of the correlation among individual-dose assessments. At the present time, it can be stated that the highest point (associated with a person who lived close to the river) will move significantly to the right, and the minimum point (associated with a person who lived far from the river) in the third cluster will move to the left.

It must be noted that no age dependency of EPR estimates has been found for the Metlino donors under study (the range of birth years is 1928–1942 corresponding ages at the beginning of exposure of 8–22 years).

In spite of all these uncertainties, the sample-average values derived from TRDS-2000 calculations and from EPR measurements (Table 10) coincide well with each other (0.55 ± 0.07 Gy and 0.55 ± 0.17 Gy, respectively); this allows us to conclude that TRDS-2000 external dose assessments for the upper Techa region are consistent with the data of EPR assay.

6.3. COMPARISON OF EPR DATA WITH CALCULATIONS OF EXTERNAL DOSES FOR MUSLYUMOVO AND VILLAGES FURTHER DOWNSTREAM

Fifty-eight donors (ETRC members) who had teeth with completed enamel in the period of exposure and permanently resided in Muslyumovo and villages further downstream have been investigated using the EPR method. As opposed to the situation for residents of Metlino, a strong age dependence is observed for this group of subjects (Fig. 8).

This age dependence is fitted by the same function as for radiochemical data [eqn. (14)] with the following values of parameters: $Y_0 = 93$, $a = 1647$, and $b = 0.23$). As can be seen, the

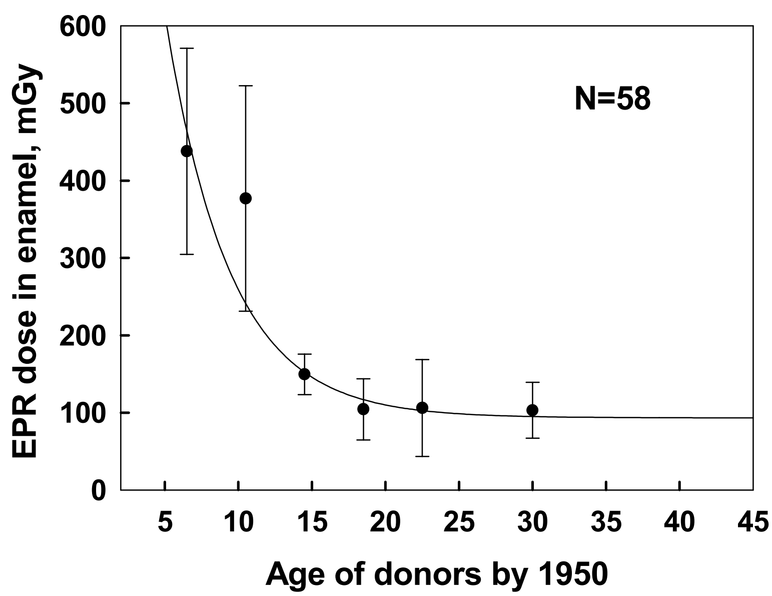

Fig. 8. Age-dependent average EPR-based dose in tooth samples for permanent residents of the mid- and lower Techa regions. Bars reflect standard errors.

slopes of the curves (values of parameter *b*) are similar for the EPR and radiochemical data (0.23±0.10 and 0.20±0.04 respectively). Also, after the age of 19 y, the value of enamel dose becomes constant in the same manner as does the concentration of ^{90}Sr in dentin (Fig. 5). Nevertheless, the age dependences for EPR-based doses and internal beta doses from strontium incorporated in dentin (evaluated for similar tooth samples using dose-rate coefficients from Appendix 2 and average values of ^{90}Sr concentration from Fig. 5) are not parallel, because the ratios of parameters a/Y_0 are not equal (18 for EPR and 43 for radiochemical data); this needs some explanation.

It is known that after eruption (which follows crown completion) the macro- and micro-structure of permanent teeth continues to change with age. According to Samusev et al. (2002) just after eruption the pulp cavity is relatively large, then the volume of the pulp cavity decreases due to formation of secondary dentin. This process has a maximum speed at ages of 13–19 y (during this period the pulp cavity decreases to half of its initial size). Also, the growth and calcification of roots for the majority of permanent teeth occur at ages of 10–16 y. So, the concentration of ^{90}Sr incorporated in crown and root dentin during the period of adolescence could be substantially non-uniform. Thus, separate measurements of ^{90}Sr concentrations in crown and root dentin are necessary for the correct evaluation of internal beta dose in enamel of teeth obtained from persons who were adolescents in the period of intake (it should be remembered that the values of *DRC* for “crown dentin→enamel” and “root dentin→enamel” are significantly different). The simplifying assumption on uniform ^{90}Sr distribution though all dentin (both crown and root) made above in order to use radiochemical data therefore can be applied for the evaluation of internal beta dose in enamel of teeth obtained from persons older than 19 y. This restriction reduces the number of samples from the mid and lower Techa regions to 21 persons.

As can be seen from Fig. 9, the distribution of individual values in this sample of adult persons is not normal: the average difference between EPR-based doses and calculated values of internal beta doses is equal to 39±28 mGy, and the median is equal to 7 mGy.

A comparison of EPR results with enamel doses calculated using the TRDS-2000 approach specific for these persons’ village-residence histories is shown in Table 11. The average external dose for this group of subjects calculated according to the TRDS-2000 approach (dose rates in 1950–1951 based on the one-compartment river model and assumed end of exposure in 1960) is evaluated as 10.2±1.0 mGy. This is not significantly lower than the mean EPR-based dose. The comparison of medians using the non-parametric Wilcoxon signed rank test also does not show a statistically significant difference between TRDS-2000-based and EPR-based doses for this sample of subjects..

If we perform similar calculations for this sample of subjects but using the values of dose rates in 1950–1951 derived from the two-compartment river model, we receive an average value of external dose (assumed end of exposure in 1960) equal to 13±1 mGy (Table 12). In addition, taking into account the contribution of low-level exposure after 1960 for those who continue contact with contaminated river and flood plain (“contacting persons”), we receive an average value equal to 24±3 mGy (Table 12). As can be seen from this Table, there are no statistically

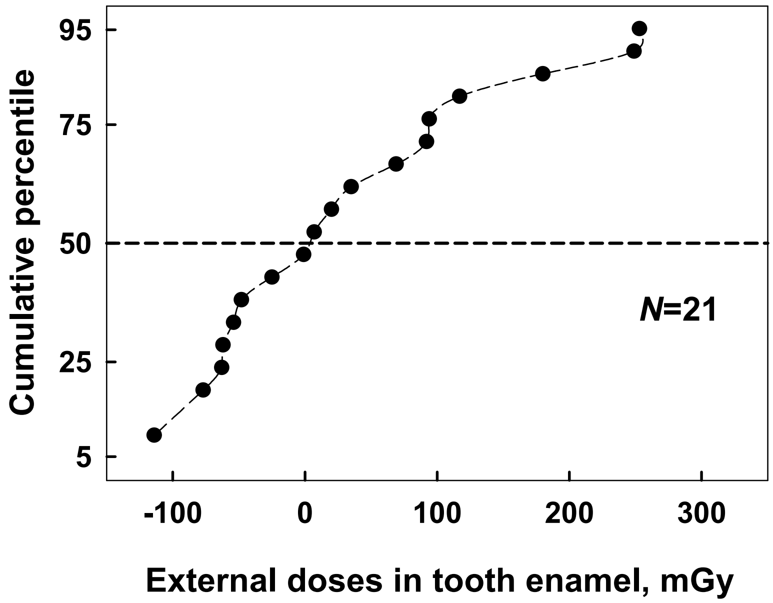

Fig. 9. Probability distribution of EPR-based external doses for permanent residents of the mid- and lower Techa regions.

Table 11. Comparison of EPR-based dose estimates for residents on the mid and lower Techa regions with mature teeth at the period of exposure and enamel doses evaluated for the same sample of persons using the TRDS-2000 approach. Twenty-one donors are considered.

Statistical parameter	EPR-based dose, mGy	TRDS-2000 enamel dose, mGy	Testing for difference (*p*-values)[a]
Median	7	10.4	0.64
Mean ± SEM	39±28	10.2±1.0	0.32
90% CI for mean	-7–85	8.5–11.9	

[a] The Wilcoxon signed rank test is used for medians, and the *t* test for paired samples is used for means

confirmed differences between the revised TRDS-based and EPR-based doses for this sample of subjects.

Thus, the comparative analysis described above gives us the absence of statistically confirmed differences between EPR-based dose and enamel dose evaluated using the TRDS approach (both original and revised). This is explained by the small doses and the small number of tooth samples that can be considered under the assumption of uniform distribution of ^{90}Sr in dentin. These findings should be considered as preliminary and will be discussed below (Section 7.4).

Table 12. Comparison of EPR-based dose estimates for residents on the mid- and lower-Techa regions with mature teeth at the period of exposure and enamel doses evaluated for the same sample of persons using the revised TRDS approach (21 donors are considered).

Statistical parameter	EPR-based dose, mGy	Revised dose for non-contacting persons, mGy	Revised dose for contacting persons, mGy	Testing for difference between EPR and revised doses (*p*-values)[a]
Median	7	14	14	0.72 for non-contacting and 0.99 for contacting persons
Mean ± SEM	39±28	13±1	24±3	0.38 for non-contacting and 0.60 for contacting persons
90% CI for mean	-7–85	12–15	19–29	

[a] Wilcoxon signed rank test is used for medians and *t* test for paired samples is used for means.

7. DISCUSSION

This milestone report completes the series of tasks within Work Package 3, "Validation of TRDS external doses by combining EPR results with modeling," which was planned within the framework of the current phase of JCCRER Project 1.1. Data presented in the report include:

1. Revision of TRDS-2000-based estimates of external dose by implementation of the revised Techa River Model, and the evaluation of doses for those who permanently lived near the Techa River after 1960;
2. Statistical analysis of EPR measurements available for the members of the ETRC;
3. Estimation of the contribution of radiostrontiums to enamel dose; and
4. Validation of TRDS external dose estimates by combining EPR results with modeling.

7.1. ANALYSIS OF THE REVISED TRDS EXTERNAL DOSES

Two versions of TRDS-based external dose estimates have been considered in this report:

- The first version is based on TRDS-2000 modules calculated using the one-compartment Techa River Model for dose rates in 1950–1951; and
- The second version (named also 'revised external dose estimates') is calculated using the revised two-compartment Techa River Model described in our Milestone 5 Report (Vorobiova et al. 2003) and which we plan to include in TRDS-2006.

As described in Vorobiova et al. (2003), the same source term (as used before in TRDS-2000) was used for fitting the parameters of the two-compartment model. This model has provided better assessments for radionuclide concentrations in bottom sediments in 1950–1951 that are used for the calculation of external gamma-dose rates on the riverbank. The implementation of the two-compartment river model results in a small increase of external doses for residents on the upper and middle Techa regions. As for the settlements located at more than 125 km from the site of release, revised estimates become 2–3 times larger, because the second compartment in the revised river model results in a flatter gradient of radionuclide concentration in bottom sediments with downstream distance.

A recent publication by Golikov et al. (2004) describes current measurements of ^{137}Cs-dose rates made by field spectrometry on floodplain soils and within residence areas in Muslyumovo and Brodokalmak. These results are another reason to revise estimates of external dose for non-evacuated residents in the middle and lower Techa regions. These data have allowed us to quantify low-level (comparable with natural background) rates of "extra" external exposure chronically affecting the population during a long time period. As shown in Section 3.4, such chronic low-level external exposure during forty years (1960–2000) results in values of dose comparable with those received during the first years after contamination.

According to "interview data," 46% of questioned Muslyumovo residents continued to use the Techa River water and floodplain areas for their domestic needs even after they had been prohibited to do this. However, it is impossible to reconstruct exactly the frequency and duration of the contacts with the contaminated river and floodplain in 1960–1990. Therefore, the range of variation of external dose between two endpoints of behavioral regimes ("absence of contacts" and "contacts continuing exactly as before the prohibition") must be included in estimates of uncertainty.

Thus, the revision of TRDS-based estimates of external dose does not result in significant changes for the residents of evacuated settlements in the upper reaches of the river, where the levels of external dose were relatively high. However, implementation of the two-compartment river model and especially the re-evaluation of doses obtained by those who permanently lived on the middle and lower Techa River after 1960 result in a significant increase in external dose assessments for the residents of this region.

7.2. ANALYSIS OF AVAILABLE EPR DATA

The data presented in this report have shown that at the current time EPR measurements performed on 292 teeth from 192 members of the ETRC are available for the purpose of validating TRDS-based estimates of external dose. This is three times lower than the number of teeth indicated previously by Anspaugh et al. (2001) as required for a full-scale validation study. Nevertheless, the analysis of available data, even at this stage of development, is considered essential because:

- The TRDS-based external doses for the most contaminated village (Metlino, located in the upper Techa at 7 km downstream from the release site) are in good agreement with the EPR data described above. The TRDS-based estimates of dose are also in agreement with the results obtained by two additional independent methods (luminescence measurements of quartz in bricks and fluorescence in situ hybridization measurements of chromosome translocations in lymphocytes) as described by Jacob et al. (2003) and Degteva et al. (2004); and

- The detailed analysis of EPR data for the pooled sample of teeth from residents of the middle and lower Techa regions provide a method for careful evaluation of the contribution of all sources to the total enamel dose. This allows for a more clear understanding of the tasks that should be done at the next step of the validation study.

These two points are discussed below in detail.

7.3. COMPARISON OF TRDS-BASED EXTERNAL DOSES FOR METLINO WITH THE RESULTS OF DIFFERENT ASSAYS

Some work on validation of the TRDS-based estimates of external dose in Metlino had already been completed with use of the technique of luminescent dosimetry of quartz found in bricks of old buildings located on the banks of the Techa River combined with Monte Carlo photon-transport modeling (Bougrov et al. 1998; Jacob et al. 2003; Taranenko et al. 2003). It was found that the TRDS-2000-based value of absorbed dose in air accumulated at the river shoreline in Metlino (the closest settlement to the site of radioactive releases) is in good agreement with the estimate of dose reconstructed independently on the basis of luminescence measurements (Jacob et al. 2003). The important conclusion made in this study is, "There is no indication for a considerable underestimation of the absorbed dose in air accumulated in Metlino at the shore of the Techa River in the period 1949–1956 [that is about the corresponding value used in the TRDS-2000]."

Nevertheless, a major source of uncertainty of the TRDS-2000-based estimates of individual external dose is associated with assumptions about the geographical variation of dose within residence areas and assumptions about behavior of the cohort members in terms of the amount of time spent near the river (Napier et al. 2001). The last factor can not be evaluated by retrospective dosimetry methods using objects with fixed in situ position (like the investigated bricks). That is why the experimental techniques for individual-dose reconstruction are of great importance for our validation study.

As shown above, EPR-based measurement of dose in human teeth is a valuable experimental technique for the study of retrospective dosimetry of individuals exposed on the upper reaches of the Techa River. The sample-average value (± SEM) of enamel dose derived from EPR measurements of Metlino residents having mature teeth at the period of exposure ($N = 13$) is equal to 0.55±0.17 Gy, which coincides well with the value of 0.55±0.07 Gy predicted by use of TRDS-2000 for these persons and their village-residence histories. Results also show a statistically significant correlation between the two estimates of external dose. Individual EPR-based estimates have a wider scattering than do doses calculated using TRDS-2000. This point needs further investigation aimed at reducing the uncertainty in EPR measurements, verification of background levels, and careful evaluation and subtraction of the contribution of $^{90}Sr/^{90}Y$ from the total enamel dose measured by the EPR method.

Another approach for individual-dose reconstruction is fluorescence in situ hybridization (FISH) of lymphocytes. A FISH-based analysis of stable translocations in members of the Techa River population was reported by Bauchinger et al. (1998). Unfortunately, individual doses were presented in this report for only a few persons, because the number of cells scored for the majority of cases was not sufficient for the routine statistical technique (based on the Poisson distribution) used to derive absorbed dose and its confidence limits. A new statistical approach (based on Monte Carlo simulation with the use of the binomial distribution) was suggested for the interpretation of results of available FISH assays, when zero or only a few translocations are observed (Anspaugh et al. 2000). This approach has allowed evaluation of individual doses on the basis of FISH data for persons previously investigated by Bauchinger et al. (1998), but for whom results were not reported.

The re-analysis of FISH-data for residents of Metlino is described in our recent paper (Degteva et al. 2004). FISH measurements were made for 31 residents of Metlino and for 39 individuals believed to be unexposed (Bauchinger et al. 1998). The EPR- and FISH-based estimates agreed well for permanent residents of Metlino; 0.67±0.21 Gy and 0.48±0.18 Gy (mean±standard error of the mean), respectively. The FISH-based dose for all investigated ETRC members, 0.38±0.10 Gy, compared well to the TRDS-2000 prediction of external dose, 0.31±0.03 Gy, to red bone marrow for these persons.

The comparable estimates obtained with the use of TRDS-2000 and derived independently from the results of three experimental techniques using different objects for study (EPR on teeth, FISH on lymphocytes and luminescence on bricks) have confirmed the quality of the resultant dosimetric data on external exposure as reconstructed for residents of Metlino in the upper Techa River. However, for the validation of dose estimates for *specific individuals*, an improvement that takes into account the location of the houses (where individuals lived during the period of exposure) will be implemented in the next version of the dosimetry system.

7.4. ANALYSIS OF THE RESULTS OBTAINED FOR MUSLYUMOVO AND VILLAGES FURTHER DOWNSTREAM

Results obtained for the pooled sample of teeth from residents of the middle and lower Techa regions should be considered as preliminary, because the contribution of external exposure to the total enamel dose measured by EPR is relatively small; therefore, external dose estimates derived from EPR data depend strongly on the precision of the evaluation of background and internal dose components. The contribution of internal exposure to total-enamel dose was evaluated on the basis of a similar (but not the same) sample of subjects, and also not all components of internal exposure were taken into account. Therefore, the EPR-based estimates, obtained by subtracting from the total enamel dose (measured by EPR) the components contributed by background exposure and beta particles of radionuclides (^{90}Sr/^{90}Y and ^{89}Sr) incorporated in dentin, should be considered as some surrogate of external dose for this group of subjects. Nevertheless, the analysis presented here is very important for future work, because it includes for the first time the evaluation of the age dependency of ^{90}Sr concentration in dentin of teeth with completed crowns obtained for those who lived in the middle and lower Techa regions.

As shown in Section 5.1, a significant decrease of ^{90}Sr concentration in dentin is observed with donor's age up to 17–19 y. As discussed by Tolstykh et al. (2003), dentin (root dentin in particular) has a longer period of maturation than does enamel, and this phenomenon can explain the age dependence observed. Intensive formation of secondary crown dentin as well as growth and calcification of roots occur in permanent teeth during the period of adolescence. These processes result in significant unevenness of distribution and a large dispersion of values of ^{90}Sr concentration in dentin, if the radionuclide intake occurs during this age period. Therefore, a more precise determination of ^{90}Sr distribution through dental tissues (compared to group-average values provided by historical radiochemical data) is necessary for the evaluation of enamel dose in this age group. It is anticipated that such data will be provided by the contact-TL method applied separately for crown and root dentin samples from each tooth under study.

According to the data presented in Section 6.3, EPR-based doses for residents on the middle and lower Techa regions have a similar age dependence as ^{90}Sr concentration in dentin. Nevertheless, a stable systematic difference (which could be attributed to the contribution of external exposure) is observed only for persons older than 19 y. Statistical analysis of this subset of data indicates the absence of a statistically significant differences between EPR-based and TRDS-based (both original and revised) estimates of external dose in enamel.

In spite of the absence of statistically significant differences, some tendency is observed for mean EPR-based dose to be higher than the value calculated for the same sample of residents of the middle and lower Techa regions. This difference (about 15–30 mGy) can be due to the following sources of internal exposure, which are not included in the current dose estimates:

- Enamel exposure from beta particles emitted by ^{90}Sr/^{90}Y and ^{89}Sr incorporated in the enamel itself; and
- Enamel exposure from betas and photons emitted by ^{137}Cs/^{137m}Ba incorporated in soft tissues.

As shown in Section 5.2, values of dose-rate coefficients for “enamel→enamel” are 3–9 times higher than for “crown dentin→enamel.” The first results for tooth samples from residents on the Techa River obtained with thin layer contact-TL detectors show that for mature teeth the ratio of ^{90}Sr concentrations in enamel-to-dentin is equal to 2.5–5. This means that ^{90}Sr in enamel of mature teeth can produce absorbed doses comparable with those produced by ^{90}Sr incorporated in the dentin. Therefore, it is very important to evaluate this source of internal exposure, when appropriate results of TL measurements will be available.

A preliminary evaluation of enamel dose due to ^{137}Cs/^{137m}Ba uniformly distributed in soft tissues has been performed by Monte Carlo simulation of electron and photon transport with use of the MCNP code (Briesmeister 2000) in the simple anthropomorphic phantom BOAMB (this phantom was described by Gualdrini and Casalini 1998). The results of this simulation show that enamel dose due to ^{137}Cs/^{137m}Ba is about 1.3 times higher than the average dose for the whole body of the phantom. Applying this coefficient to internal dose estimates for the Muslyumovo residents, we can conclude that enamel doses due to ^{137}Cs/^{137m}Ba can reach 15–30 mGy. Such a contribution is considered to be significant and therefore should be taken into account in future analysis.

Summarizing everything discussed above we conclude the following: The total enamel dose (including background) for residents of the middle and lower Techa regions is formed mainly due to background sources and the exposure of radionuclides incorporated in dental (^{90}Sr and ^{89}Sr) and soft (^{137}Cs) tissues. The contribution of external exposure is evaluated as about 10–20% of the total enamel dose. Taking into account the low levels of total enamel dose (200–400 mGy in average), we conclude that the validation of dose estimates for *specific individuals* is not possible for this region. Nevertheless, we do believe that group-average-dose estimates derived from EPR data can provide the basis for validation, if the following requirements are met: (1) the accuracy of EPR measurements is good enough to allow reliable estimates at background levels of dose; (2) the investigated groups of exposed and background subjects are large enough to provide a statistically significant difference between sample-average values; and (3) the contribution of internal exposure from all sources is evaluated on an individual basis for all tooth samples included in the analysis. Therefore, the reduction of the uncertainties associated with each step in individual external dose evaluation performed on the basis of EPR measurements is a crucial point during the next stage of the validation study.

7.5. COMPARISON OF EPR DATA FOR METLINO AND MUSLYUMOVO WITH THE RECENT ALLEGATIONS BY MOKROV

In spite of the limitations described above validation of our calculations of external dose, even at the current stage of development, is considered essential, as the allegation has been made that estimates of dose based upon TRDS-2000 significantly underestimate actual external doses (Mokrov 2002; 2003; 2004). Mokrov gave the following assessments recently:

- External doses for residents in Metlino for the period 1949–1951 were 0.9–6.6 Sv, depending upon the locations of the resident's house (Mokrov 2004); and
- External doses for the Muslyumovo population can vary from 0.5 to 2.0 Sv depending on the time of stay at the contaminated floodplain (Mokrov 2003).

Available EPR data for the permanent residents of Metlino and Muslyumovo do not confirm these allegations by Mokrov, because after subtraction of the background component, the average enamel doses (± standard error of mean) derived from the EPR measurements are equal to

- 0.67 ± 0.21 Gy for *adult permanent residents of Metlino,* and
- 0.14 ± 0.04 Gy for *adult permanent residents of Muslyumovo.*

These values are significantly lower than Mokrov's estimates. Also, if we subtract beta-exposure due to strontium radionuclides incorporated in dentin (as discussed above, this contribution is significant for residents of Muslyumovo), the result is an EPR-based estimate of external dose as *0.07 ± 0.04 Gy* for the population of Muslyumovo. The last estimate is an order of magnitude lower than the minimum assessment given by Mokrov.

It also must be noted that Mokrov does not mention the organ to which his estimates apply. If we suppose that his values can be interpreted as 'average dose in soft tissues,' the discrepancy with EPR-based results becomes even larger, because absorbed dose from environmental radiation fields is about 1.6 times higher for enamel than for the majority of soft tissues. Therefore, available EPR data for Metlino and Muslyumovo residents fully disprove the recent allegations by Mokrov.

7.6. ISSUES IN THE VALIDATION OF EXTERNAL DOSES FOR RESIDENTS OF SETTLEMENTS LOCATED BETWEEN METLINO AND MUSLYUMOVO AND AVENUES FOR FURTHER INVESTIGATIONS

Validation of external doses for residents in the settlements located between Metlino and Muslyumovo is an active area of investigation. Twelve small evacuated villages located at distances 18–65 km from the release site are included in this group. According to the TRDS estimates (Table 4) a sharp (16-fold) decrease of external dose is observed with downstream distance for this part of the Techa River. Some decrease in the average EPR dose is also observed for this region, but it is not so significant (Table 8) due to the contribution of internal exposure. The contribution of internal exposure to the total enamel dose for persons in this region increases from 10% (as for Metlino) to 40–60% (as for Muslyumovo). Therefore, it is not possible to neglect this factor of exposure (as had been done in the evaluation of data for residents of Metlino). On the other hand, there are not sufficient historical measurements of ^{90}Sr in teeth for residents in these settlements, so we should not use the approach that has been applied to the pooled sample 'Muslyumovo and lower Techa.' Therefore, the results of contact TL-dosimetry, which would allow the evaluation of ^{90}Sr concentration on an individual basis, are especially important for correct interpretation of EPR-based estimates of dose for residents in this region.

The number of tooth samples collected from residents for each of these small villages is small, and the quality of samples varies significantly. Thus, it is very important to apply the criterion for the estimation of EPR measurement reliability for statistical inference based on available data sets (Sections 4.3 and 4.4).

Another issue (important for all aspects of dose reconstruction for residents on the Techa River) is the variability of individual external doses inside settlements. The TRDS-2000 estimates are based on village-average parameters and do not consider the precise location of an individual (the position of the household where a person lived during the period of exposure). Because dose rates in air varied considerably within residence areas, the knowledge of precise location can reduce significantly the uncertainty of external dose assessments. Work is now underway to reconstruct household position on an individual basis (for those ETRC members with such data available) and to include this factor in the next version of the dosimetry system.

In consideration of the configurations of residence areas and the peculiarities of river bed and flood-lands, there is a peculiar feature that is specific to Nadyrovo Village (located at 50 km from the release site). The Techa River has a 'blind creek' (or appendix) located within the residence area of Nadyrovo (a similar situation is not observed in any of the other villages). It is possible that the incorporation of radionuclides in the bottom sediments (and resulting dose rates in air) for this 'blind creek' is not described by the model of the free-flowing Techa River. Such a specific situation might have resulted in higher levels of dose rates on the banks of this 'blind creek.' Unfortunately, there are no historical measurements of radionuclide concentrations and exposure rates in the period of radioactive releases (1949–1956) for this site, and the specific situation in Nadyrovo was not taken into account in the evaluations made with the use of TRDS-2000.

The set of EPR data for the residents of Nadyrovo (Table 8) shows very high levels of enamel dose (6.1 Gy and 2.5 Gy) for two persons (full sisters) identified as having lived in the same household just on the bank of this 'blind creek.' The EPR doses for the remaining residents of this village are significantly lower. The interpretation of this finding has two aspects. The first aspect is the necessity to develop a model describing the radioactive contamination of stagnant reservoirs (similar to that in Nadyrovo) and to evaluate possible external doses for those who lived on such sites. The second aspect of the problem is connected with further improvement of EPR methodology discussed below.

The enamel dose for the first sister 6.1±1.3 Gy is derived from the EPR measurements of two incisor teeth and has high uncertainty. The enamel dose for the second sister 2.4±0.5 Gy was derived as a weighted average of the EPR measurements of five posterior teeth and one incisor. The results for posterior teeth varied in the range 0.7–3.1 Gy and the dose for the incisor was 6.2 Gy (comparable with the dose of the first sister). These findings address the uncertainties of the EPR method and in particular the uncertainty of measurements of incisor teeth.

It must be noted that unrealistically high doses are sometimes measured by the EPR method in tooth enamel of incisors obtained from background donors. It has been identified that one major cause is natural UV-light exposure to the front part of incisors. In the current practice of EPR dosimetry it is recommended to use only the back part of incisors for dose reconstruction. This recommendation is sometimes not followed, because there is frequently only small masses of posterior enamel. Another problem with incisors is that radiation sensitivity of its enamel may differ from that for molar teeth. Therefore, more work is required to focus on development of a methodology for EPR measurements of small masses and also establishing a calibration curve for enamel from incisors.

8. CONCLUSIONS

This milestone completes the series of tasks within Work Package 3, "Validation of TRDS external doses by combining EPR results with modeling," planned in the frame of the current phase of JCCRER Project 1.1. Analysis of the findings obtained leads to the following conclusions:

1. Work on continued revision of TRDS-2000-based estimates of external dose has been performed by implementation of a new Techa River Model, and doses for those who lived permanently near the Techa River after 1960 have been evaluated. The revision of TRDS-based external dose estimates does not result in significant changes for the residents of evacuated settlements in the upper reaches of the river, where the levels of the external dose were relatively high. However, the implementation of the new two-compartment river model and especially the evaluation of doses obtained by those who permanently lived in the middle and lower regions of the Techa River and continued to

use the river for domestic needs after 1960 result in a significant increase in the external dose assessments for the residents of this region.

2. The number of ETRC members investigated with the EPR method at this date (292 teeth from 192 ETRC members) is three times lower than the number of teeth indicated previously as required for a full-scale validation study. Nevertheless, the analysis of available data, even at this stage of development, is considered essential because it is possible to evaluate dosimetric results for two important groups of subjects: the residents of Metlino (settlement closest to the site of release) and data for a pooled sample of teeth obtained from the residents of Muslyumovo and further down the Techa River.
3. The comparable estimates obtained for Metlino in the upper Techa River with the use of TRDS-2000 and derived independently from the results of three experimental techniques using different objects for study (EPR on teeth, FISH on lymphocytes and luminescence on bricks) have confirmed the quality of the resultant dosimetric data on external exposure. However, for the validation of dose estimates for *specific individuals*, an improvement that takes into account the location of the houses (where individuals lived during the period of exposure) will be implemented in the next version of the dosimetry system.
4. The total enamel dose (including background) for the residents of the middle and lower Techa regions is formed mainly due to background sources and the exposure of radionuclides incorporated in dental ($^{90}Sr/^{90}Y$ and ^{89}Sr) and soft (^{137}Cs) tissues. The contribution of external exposure is evaluated as about 10–20% of the total enamel dose. Because of the low levels of total enamel dose (200–400 mGy on average), the validation of dose estimates for *specific individuals* is not possible for this region. Nevertheless, we do believe that group-average-dose estimates derived from EPR data can provide the basis for validation, if the following requirements are met: (1) the accuracy of EPR measurements is good enough to allow reliable estimates at background levels of dose; (2) the investigated groups of exposed and background subjects are large enough to provide a statistically significant difference between sample-average values; and (3) the contribution of internal exposure from all sources is evaluated on an individual basis for all tooth samples included in the analysis. Therefore, the reduction of the uncertainties associated with each step in individual external dose evaluation performed on the basis of EPR measurements is a crucial point for the next stage of our validation study.
5. Available EPR measurements obtained for permanent residents of Metlino and Muslyumovo are entirely consistent with the TRDS-2000-based estimates of external dose. On the other hand, the available EPR measurements are significantly lower (up to an order of magnitude) than the assessments of external dose given recently by Mokrov (2003; 2004). Therefore, the EPR data fully disprove the allegations by Mokrov.
6. Validation of external doses for the settlements located between Metlino and Muslyumovo is an active area of investigation. To perform this task more work is required and should be focused on development of a methodology for EPR measurements of small masses and also establishing a calibration curve for enamel from incisors. In addition, the results of contact TL-dosimetry (expected in the near future) that allow the

evaluation of ^{90}Sr concentration on an individual basis are especially important for the correct interpretation of EPR data for this group.

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APPENDIX 1

ASSESSMENT CRITERION FOR THE RELIABILITY OF EPR-MEASUREMENT RESULTS

Information on more than 800 EPR tooth enamel measurements is available in the URCRM database. Teeth for measurements were sampled from various positions and were of different health conditions; consequently, the quality of measured samples varied and this fact affected the reliability of dose estimates. In addition, samples were measured in different laboratories with the use of different procedures and equipment. Thus, the quality of data available is not identical, and the relative reliability of measured results should be considered in the course of analysis of the whole data set.

Most samples were measured at the IMP or the GSF and the comparability and quality of the results of these two laboratories were studied extensively. According to Shishkina et al. (2001a) there was no systematic shift detected between the EPR measured results of the IMP and the GSF. The intercalibration between these two laboratories demonstrated high correlation between measured data within the range of higher doses. According to a joint investigation of teeth of unexposed donors (low dose range) the statistical characteristics of background doses also coincided. But individual variations demonstrated a large scattering (Shishkina et al. 2001a), which could be caused by the large error of individual-dose measurements below 300 mGy. Moreover, the reliability of results within the low dose range is affected by metal impurities in hydroxyapatite that causes corruption of the EPR spectrum. So, a criterion was developed and tested for spectrum-quality assessment (Shishkina et al. 2003a). Further analysis of the effect of the procedure applied at the IMP and the GSF allowed distinguishing three measured dose groups with significantly different grades of reliability:

- <450 mGy. Dose estimates with high uncertainty, which depend on the spectrum quality. A normal distribution of EPR-amplitude probability is typical for this dose group. The distribution width of each spectrum depends on the uncertainty in spectrum deconvolution and should be estimated individually in each case.
- 450–700 mGy. A truncated normal distribution of EPR-amplitude probability is typical for this dose range. The distribution width of each spectrum, which depends on the uncertainty in spectrum deconvolution, is to be estimated individually. The contribution of uncertainty in deconvolution to the total uncertainty is significantly high within this dose range.
- >700 mGy. The uncertainty in deconvolution is insignificant. The main contribution to the total uncertainty within this dose range is the uncertainty of reproducibility due to signal anisotropy.

The general criterion of reliability is based on factors affecting EPR-dose-estimation quality such as:

1) tooth position;
2) sample mass;
3) signal-to-noise ratio;
4) laboratory that performed the measurements; and
5) coefficient of variation.

A number of points is assigned to each factor, and then the points of all factors are summed. The number of points for each criterion depends on the value of the measured dose. The total number of points represents the criterion of the measurement reliability.

The principal features of the point-assignment procedure for each criterion are described below.

1) Factor "Tooth position"

From 0 up to 4 points could be assigned for this factor. Enamel of premolars and molars is mostly suitable for EPR measurements. Incisor enamel suffers under additional "dose" due to the ultra-violet component of sunlight. That is why it is preferable to measure the lingual portion of incisor enamel. In case the inner portion is not available, it is possible to measure the buccal portion of enamel. Unfortunately, the crown of more than half of the incisors sampled is scuffed, and the amount of enamel is small. In such cases the whole incisor enamel is measured. In addition, before 2001 incisor enamel was measured in total, without partitioning into lingual and buccal portions. The measurement quality of "incisor enamel in total" is the lowest, because it is not possible to determine whether the lingual or the buccal portion was larger in the measured sample.

In such a way the number of points assigned depends on the tooth position, which could be molar and premolar enamel, incisor enamel, enamel of the buccal portion of incisors and enamel of the lingual portion of incisors (Table A1). If the tooth position and respectively the measured tissue are not known, an incisor was presumed and 4 points were assigned to the factor "tissue."

2) Factor "Sample mass"

The reliability of EPR measurements depends on the sample mass. The lower the weight the higher the measurement uncertainty. Enamel weight plays an especially critical role in EPR-measurement reliability at low EPR doses (below 450 mGy). Table A2 represents the algorithm of the assessment of the sample-weight effect on the reliability of EPR-measurement results.

Table A1. Reliability criterion of EPR measurements. Sample tissue as the factor.

Sample tissue	Number of points
Premolar and molar enamel	0
Enamel of lingual portion of incisors	0.5
Enamel of the buccal portion of incisors	3
Incisors enamel in total	4

Table A2. Reliability criterion of EPR measurements. Enamel-sample weight as the factor.

Sample weight, mg	Number of points	
	EPR dose ≥450 mGy	EPR dose <450 mGy
Mass≥70	0	0
40≤Mass<70	0.5	1
25≤Mass<40	1	2
15≤Mass<25	2	3
Mass<15	3	4
Mass unknown	0.5	0.5

If the sample weight was not recorded in the laboratory and this value is not available in the database, the value 0.5 was assigned to the factor. It is necessary to explain why the number of points assigned in this case was not large. There were 114 enamel samples with unknown weight in total. The greatest part (about 70%) of these samples were molar and premolar samples. For such samples a large amount of enamel is typical, i.e., the weight of these samples was practically always above 40–70 mg, which means that the number of points should not exceed 0.5. There are about 30% of incisors with unknown weight. Enamel of these incisors was not divided into buccal and lingual portions before measurement. Because of this fact the weight of tooth samples was within 40–70 mg range or more. Furthermore, 60% of the measurements demonstrated EPR doses above 450 mGy, so the number of points varies on the average from 0 to 0.5.

3) Factor "Signal-to-noise ratio"

As mentioned above, the ratio "signal/noise" (S/N) is known for measurements performed at the IMP laboratory. The point-assignment procedure for measurements at the IMP laboratory is represented in Table A3. The number of points varies from 0 to 4 according to the ratio of "signal-to-noise" and the EPR dose.

The factor S/N is not known for earlier measurements at the IMP (carried out before 1998, because spectra of those measurements are now not available) as well as for the measurements in any other laboratory.

The number of points assigned to the sample in case that the factor S/N is unknown depends on the laboratory and the EPR-measured dose of this sample (Table A4). At high doses (above 700 mGy) the magnitude of the radiation signal is much higher than that from noise and is practically always well distinguishable. That is why for high doses in every laboratory the noise effect on the dose estimation quality is taken as zero. At doses below 700 mGy the number of points assigned to a laboratory depends on the type of equipment used (spectrometer response) and on the sample-preparation procedure (with chemical processing of enamel or without).

Table A3. Reliability criterion of EPR measurements.
Signal-to-noise (S/N) ratio as the factor.

Signal/noise	Number of points	
	EPR dose ≥450 mGy	EPR dose <450 mGy
S/N≥1.2	0	0
1.2>S/N≥0.8	0.5	1
0.8>S/N≥0.5	1	2
0.5>S/N≥0.2	2	3
0.2>S/N	3	4

Table A4. Reliability criterion of EPR measurements.
Assignment of points if the factor S/N is unknown.

Laboratory	Number of points		
	EPR dose of sample		
	≥700	450–700	≤450
IMP	0	0.5	1
GSF	0	0.5	1
MRNC	0	1	2
ICP	0	1	2
NIST	0	0.5	1

4) Factor "EPR laboratory"

Several points are assigned to each laboratory depending on the results of international and internal intercalibrations. The assignment of points is indicated in Table A5. The factor zero was taken for IMP and GSF because the measured results of these two laboratories were repeatedly compared with each other and many parallel measurements were carried out testing many more samples than in international intercalibrations. The high factor for NIST measurements is caused by the fact that this institution did not participate in any intercalibrations, and it is not possible to assess the systematic error of its data.

5) Factor "Coefficient of variation"

Each EPR measurement has its own standard deviation (STD). The ratio of standard deviation-to-EPR dose was chosen as the fifth factor of reliability. The point-assignment procedure is the same for all laboratories and is shown in (Table A6). The results of point assignments showed that this factor varies from 0 to 2.5.

Table A5. Reliability criterion of EPR measurements. EPR laboratory as the factor.

Laboratory	Number of points
IMP	0
GSF	0
MRNC	0.5
ICP	1.5
NIST	2.5

Table A6. Reliability criterion of EPR measurements. "Coefficient of variation" as the factor.

Dose, mGy	STD, mGy	Number of points
≤170	≤57	0
≤170	57<STD≤114	1
≤170	114<STD≤170	2
≥170	Any	STD/Dose

The sum of all five factors for the criterion of measurement reliability could vary from 0 to 16.5. This characteristic of EPR results allows for the common analysis of all EPR measurements of tooth enamel carried out in different laboratories.

In total the reliability criterion was calculated for 788 enamel measurements and entered into the URCRM database.

APPENDIX 2

DOSE-RATE COEFFICIENTS OBTAINED BY MONTE CARLO SIMULATION

Table A7. Dose-rate coefficients for $^{90}Sr/^{90}Y$ in anterior teeth, (nGy s^{-1}) per (Bq g^{-1}).

Target-tissues	Enamel		Dentin	
Sources of irradiation	Dentin	Enamel	Dentin	Enamel
		Upper jaw		
1st incisor	0.017	0.076	0.063	0.014
2nd incisor	0.017	0.077	0.063	0.016
Canine	0.016	0.084	0.066	0.012
		Lower jaw		
1st incisor	0.017	0.071	0.060	0.014
2nd incisor	0.018	0.071	0.062	0.014
Canine	0.018	0.073	0.067	0.011

Table A8. Dose-rate coefficients for $^{90}Sr/^{90}Y$ in premolars, (nGy s^{-1}) per (Bq g^{-1}).

Source of irradiation	Target tissues			
	Dentin	Whole enamel	Enamel of lateral surface	Enamel of masticatory surface
		1st upper premolar		
Enamel	0.0107	0.0446	0.0415	0.0491
Crown dentin	0.0681	0.0146	0.0157	0.0130
Root dentin	0.0047	0.0012	0.0020	0.0000
Alveolar bone	0.0011	0.0037	0.0060	0.0004
		2nd upper premolar		
Enamel	0.0105	0.0444	0.0430	0.0467
Crown dentin	0.0682	0.0148	0.0156	0.0136
Root dentin	0.0046	0.0015	0.0025	0.0000
Alveolar bone	0.0010	0.0038	0.0060	0.0004
		1st low premolar		
Enamel	0.0116	0.0450	0.0439	0.0472
Crown dentin	0.0669	0.0145	0.0153	0.0127
Root dentin	0.0028	0.0013	0.0019	0.0000
Alveolar bone	0.0012	0.0040	0.0058	0.0004
		2nd low premolar		
Enamel	0.0125	0.0469	0.0455	0.0497
Crown dentin	0.0663	0.0134	0.0144	0.0116
Root dentin	0.0029	0.0013	0.0020	0.0000
Alveolar bone	0.0011	0.0039	0.0056	0.0004

Table A9. Dose-rate coefficients for $^{90}Sr/^{90}Y$ in molars, (nGy s^{-1}) per (Bq g^{-1}).

Sources of irradiation	Target tissues			
	Dentin	Whole enamel	Enamel of lateral surface	Enamel of masticatory surface
	1st upper molar			
Enamel	0.0144	0.0645	0.0613	0.0678
Crown dentin	0.0680	0.0071	0.0084	0.0057
Root dentin	0.0037	0.0015	0.0029	0.0000
Alveolar bone	0.0002	0.0022	0.0039	0.0003
	2nd upper molar			
Enamel	0.0103	0.0492	0.0485	0.0503
Crown dentin	0.0702	0.0134	0.0140	0.0125
Root dentin	0.0046	0.0015	0.0025	0.0000
Alveolar bone	0.0006	0.0032	0.0051	0.0003
	3rd upper molar			
Enamel	0.0100	0.0482	0.0473	0.0496
Crown dentin	0.0704	0.0142	0.0149	0.0132
Root dentin	0.0027	0.0013	0.0020	0.0000
Alveolar bone	0.0007	0.0033	0.0052	0.0003
	1st low molar			
Enamel	0.0149	0.0632	0.0615	0.0650
Crown dentin	0.0668	0.0070	0.0080	0.0059
Root dentin	0.0037	0.0017	0.0032	0.0000
Alveolar bone	0.0002	0.0023	0.0041	0.0004
	2nd low molar			
Enamel	0.0104	0.0489	0.0478	0.0509
Crown dentin	0.0705	0.0133	0.0143	0.0116
Root dentin	0.0045	0.0015	0.0024	0.0000
Alveolar bone	0.0006	0.0032	0.0051	0.0003
	3rd low molar			
Enamel	0.0105	0.0487	0.0473	0.0507
Crown dentin	0.0697	0.0136	0.0142	0.0127
Root dentin	0.0029	0.0013	0.0021	0.0000
Alveolar bone	0.0007	0.0034	0.0053	0.0003