ISSUES IN THE VALIDATION OF EXTERNAL DOSE: BACKGROUND AND INTERNAL DOSE COMPONENTS OF CUMULATIVE DOSE ESTIMATED USING THE ELECTRON PARAMAGNETIC RESONANCE (EPR) METHOD

E.A. Shishkina, V.A. Shved, M.O. Degteva, E.I. Tolstykh, D.V. Ivanov, S.N. Bayankin, A. Wieser, H.Y. Göksu, N.A. El-Faramawy, N. Semiochkina, P. Jacob, L.R. Anspaugh and B.A. Napier

Urals Research Center for Radiation Medicine
Chelyabinsk, Russian Federation

Institute of Metal Physics,
Ekaterinburg, Russian Federation

GSF-Forschungszentrum für Umwelt und Gesundheit, Institut für Strahlenschutz,
Neuherberg, Germany

University of Utah
Salt Lake City, Utah, USA

Pacific Northwest National Laboratory
Richland, Washington, USA

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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 TRDS-2000 results and the companion epidemiological 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. There are three sources of absorbed dose: external 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. A number of issues related to determining external dose from EPR measurements are evaluated in this report.

Methodological aspects (such as the problem of impurities in the teeth, chemical treatment of samples, method reliability and limitations, and uncertainty analysis) for the current status of EPR dosimetry were investigated. The magnitude of uncertainty is found to depend on the quality of the spectrum and on the mass of sample. The contributions to the total uncertainty by different factors were analyzed for EPR-tooth-dosimetry systems used at the IMP and the GSF laboratories, and errors in reproducibility were evaluated. Overall, it is possible to assume that the total uncertainty for analysis of data from one laboratory is ±120 mGy and for the combined analysis of IMP and GSF data as ±140 mGy. A new spectrum-deconvolution procedure was prepared that is highly appropriate for the low dose range.

Teeth that were received from non-exposed donors from the Urals rural population (background teeth) were evaluated for average-background-EPR dose for anterior and posterior teeth of persons within an age range similar to the range for members of the ETRC. Two approaches were used (mathematical and physical averaging) for both anterior and posterior teeth. An age dependence was not found for teeth from donors with a range of age from 45 to 95 years (1910–1953 birth years); the average dose estimated is 150±24 mGy for posterior teeth, 182±48 mGy for the lingual portion of anterior teeth, 294±79 mGy for the buccal portion of anterior teeth, and 214±55 mGy for total incisors (lingual and buccal portions combined).

An important element of the current investigation of tooth internal dose was determination of the concentration of ^{90}Sr in the tooth tissues using a new method of contact-TL beta counting. For application of this method to detection of ^{90}Sr in tooth tissues, special approaches for methods of calibration and correction for sample mass were developed. An approach is also described for the calibration of the photostimulated luminescence (PSL) method that can be used to map ^{90}Sr distribution through tooth tissues.

Finally, suggestions are made for future EPR measurements: Laboratories should be used that have new equipment and have access to improvements in all steps of the overall technology, including methods of signal extraction. The TL technique for ^{90}Sr determination in teeth is a powerful new analytical technique that will also provide new insights.

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. 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 the 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. 2000). 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[1]); 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. 2000). 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 epidemiological 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 will be 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:

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

external 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 (Shved and Shishkina 2000; Shishkina et al. 2001; Shishkina et al. 2002).

Plans include work in close cooperation with other institutions 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);
- Determination of ^{90}Sr concentration in tooth tissues (enamel, dentin) by low-level beta counting, radiochemistry, TL-contact beta detection, or other methods;
- 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).

Preliminary results of background-dose evaluation were presented in the Milestone 2 report (Shishkina et al. 2001). In this report it was indicated that, in the process of background-dose assessment, signals due to impurities were found in some EPR spectra. Moreover, the intercalibration between laboratories at the German Institute of Radiation Protection -Center for Environment and Health, Munich, (GSF) and the Institute for Metal Physics, Ekaterinburg (IMP), demonstrated poor agreement for the reconstruction of low doses (<400 mGy). This poor agreement was explained by methodological limitations or by absence of criteria for dealing with signals due to impurities. Therefore, the problem of background-dose estimation required additional investigation.

The internal dose component can be estimated using Monte Carlo modeling of electron transport (described in Shishkina et al. 2002), biokinetic modeling of ^{90}Sr metabolism in tooth tissues (Tolstykh et al. 2000; Shishkina et al. 2002), and determination of current ^{90}Sr concentration in tooth tissues. The estimation of low radionuclide concentrations in small sample volumes by beta counting (using a non-destructive method) is not precise. At GSF, a new method of contact-TL beta detection was developed (Göksu et al. 2002). For application of this method to detection of ^{90}Sr in tooth tissues, special approaches to calibration and corrections for sample mass had to be developed.

Thus, additional work was necessary to allow interpretation of EPR measurements for validation of external dose. Therefore, this report reflects the intensive efforts toward solving these additional problems that arose in the process of our investigations.

The current investigation was devoted to evaluation of all dose-forming factors for tooth enamel. In other words, in the framework of EPR-dose assessment the following tasks were specially investigated:

- evaluation of EPR-dosimetry results for the non-exposed population of the Urals (background-level estimation) and
- development of methods for the estimation of ^{90}Sr concentration in tooth tissues.

Accurate knowledge of background-radiation dose is a critical element of EPR studies of exposed populations. In this document, the accumulated experimental data of EPR measurements for members of the Urals non-exposed population have been analyzed relative to (1) experimental methodology and uncertainty in the EPR method and (2) estimation of background dose.

The current investigation also was devoted to the question of determination of ^{90}Sr concentration in tooth samples, including methodological aspects of contact-TL beta detection and use of the photostimulated luminescence (PSL), of Fuji plate, method.

2. BACKGROUND

EPR measurements provide an estimate of the total absorbed dose in tooth tissues during the lifetime of the human subject. As described in our Milestone 2 Report (Shishkina et al. 2001), background dose is one of three components contributing to the total EPR dose in enamel.

The EPR spectrum of non-irradiated enamel consists of a signal from stable CO_2^- radicals, which occur for reasons other than exposure in radiation incidents. The background-EPR signal results 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 ultraviolet component of sunlight induces an additional contribution to the EPR signal, especially for incisors (Ivannikov et al. 1997; Nilsson et al. 2001). In addition, enamel has a native signal not related to irradiation (Ivannikov et al. 1997). The concentration of stable radicals in enamel increases with age (Ivannikov et al. 2000). The accumulation rate of enamel dose from background radiation varies for donors living in different geographical regions due to variation of natural radiation background (Romanyukha et al. 1999).

The assessment of the background-dose level specific to the Urals region is very important in estimation of the external dose for the Techa River population exposed as a result of Mayak activities.

The assessment of background dose is a very difficult task in spite of numerous EPR investigations carried out in recent years. The properties of tooth enamel as a detector of ionizing radiation have been insufficiently studied. In addition, the uncertainty of the EPR method in the low dose range must be defined more exactly for further analysis of EPR results.

It should be noted that enamel is a biological mineral and individual variations of physical, chemical and biological properties influence the individual measurements of EPR dose relative to the average dose for a population.

Also, among the existing EPR results, inexplicably large differences in EPR doses have been revealed for different teeth belonging to the same donors. The potential for

difference between EPR doses in anterior and posterior teeth was described in the literature and is ascribed to ultra-violet (UV) radiation. According to Ivannikov et al. (1997) and Nilsson et al (2001), the UV component of solar light has a significant contribution to the total-background dose as measured by EPR. Therefore, it has been recommended that anterior teeth be excluded from EPR investigations, or that only enamel from the inner side of anterior teeth be used. But the collection of teeth from members of the Techa River population has not been easy, and each tooth sample is of great value. An aim of the assessment of enamel-background-EPR dose to the inner side of anterior teeth is the determination of whether and how anterior teeth from exposed donors may be used in EPR investigations.

An additional complication is that our "TOOTH" database (Shishkina et al. 2001) also includes the results of early EPR measurements on the entire enamel of incisors. Thus, another goal of the current investigation of enamel dosimetric properties for anterior teeth is to understand whether and how we can use the early EPR results for anterior teeth for the validation of external doses.

Statistical investigations of background dose in teeth from different positions in the denture have been carried out for the Techa River population. The difference between EPR doses in enamel from anterior and posterior teeth has a logical explanation, but the differences among EPR doses for posterior teeth extracted from the same donor are not explainable. According to (Ivannikov et al. 1997), the levels of background dose in the premolars and molars from donors living in Kaluga Oblast are not significantly different. Therefore, we either have a phenomenon for the Techa River population in which the dosimetric properties of different teeth in the denture differ, or we have to test and improve the methods of EPR measurements.

Preliminary investigations of background dose for donors from the Urals region have revealed some additional complications in this estimation. EPR spectra of enamel from some portion of Ural donors contain an interfering parasitic signal (Shishkina et al. 2001, Shishkina et al. 2002). A possible reason for the appearance of such a signal is impurities in the enamel. Therefore, we have an additional problem concerning this interfering signal. It is necessary to determine a method for selecting, and excluding from analysis, EPR spectra that contain this interfering signal. This fact influences the calculation of quantities of teeth necessary for EPR measurements.

For testing methodical aspects, the collaboration of scientists from the GSF and the IMP using similar EPR methods (Shishkina et al. 2001) was very useful. Based on data in the last intercalibration study (Wieser et al. 2000a, b), the reliability of results in the low dose range (<400 mGy) can be estimated as about ±180%.

3. MATERIALS USED IN EPR STUDY

EPR measurements were carried out on extracted permanent teeth from residents of the Urals. The teeth under investigation were extracted on the basis of medical indications. 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

epidemiological studies. The birth years of the donors were in the range from 1910 to 1955, which in general corresponds to the age of the exposed population living on the Techa River.

The condition of the crown and enamel were checked. Teeth given for EPR measurements were selected on the basis of no x-ray exposures from medical inspection. About one third of the teeth had visual defects of the crowns. Widespread defects were cracks, holes, and chips of enamel, abrasion of enamel, and sometimes abrasion of part of the tooth. Some of the teeth had a permanent filling. About one third of selected teeth had caries (by visual observation), which had resulted in destruction of tooth, absence of enamel, and fragmented dentin. It should be noted that, after the process of chemical treatment (the sample preparation procedure), the quantity of enamel decreased, and in some cases the mass of samples was insufficient for measurement by the EPR method.

For estimation of background dose two approaches were used. The first was mathematical averaging of individual measurements (for 1st molars). The second was physical averaging (by use of mixtures of enamel from several donors). Such physically averaged measurements were done for 2nd premolars and different fractions of incisor enamel (lingual, buccal and whole enamel).

3.1. Description of tooth groups under investigation

Teeth with different positions in the denture were investigated for the assessment of background: 1st lower molars, 2nd premolars, and incisors. EPR measurements on enamel samples were performed for the assessment of individual-background dose and for the dose of mixtures of enamel samples from different donors. Each enamel sample prepared from a tooth by chemical treatment was given a preliminary EPR measurement to allow rejection of samples with impurities in enamel, insufficient mass, or apparent high doses. The occurrence of high doses, not characteristic of background doses, may be explained as arising from donors who had been exposed to non-background sources. However, for some reason, these persons had been mistakenly identified as non-exposed to dental x-rays had not been identified as members of the ETRC.

3.1.1. First lower molars

Eighty-four first lower molars (sixth tooth position in the denture) were given to scientists at the IMP for EPR measurements between the years 2000 and 2002. During the period 2000–2001, samples were prepared from 69 of these teeth. Thirty tooth-enamel samples from 1st lower molars were measured at the IMP and five at the GSF. Additionally, samples of enamel from 34 teeth were given to the GSF for measurement, and then 28 of these received later measurements at the IMP.

In 2002, 15 teeth were sawed into equal parts and two samples of enamel from each tooth were prepared. One half of the samples were treated using the IMP method, and the other half were treated using the GSF procedure. These 30 half-tooth samples were then measured by the laboratory that prepared them, either IMP or GSF.

Thus, the two laboratories combined planned to provide 127 measurements of 99 samples prepared from 84 first lower molars. However, five teeth from the 84 were rejected from EPR investigation for reasons of mass limitation and impurities in enamel (because more than one sample had been prepared for some of these teeth, the total

number of samples rejected was nine). Summary statistics for the investigated samples of first lower molars are shown in Table 1.

3.1.2. Second premolars

One hundred forty two upper and lower second premolars (fifth tooth position in the denture) were selected for EPR measurement from non-exposed donors. The range of birth years was from 1918 to 1953. Eighty six of these teeth did not have visible defects of the crown. Fourteen of the teeth had a permanent filling.

After preliminary EPR measurements, four teeth were excluded from further investigations, because their estimated doses were higher than 2 Gy. Six teeth had impurities in the enamel, and because of parasitic signals, their dose could not be evaluated. Four teeth had insufficient mass of enamel. Therefore, 14 teeth (14 enamel samples) from 142 teeth (142 enamel samples) were rejected from analysis (~10%).

EPR measurements were provided using mixes of enamel from different teeth. The mixes were prepared for teeth of donors subdivided into five age groups. Enamel samples for the first and second groups were prepared at the GSF (donor date of birth ≤1925 and 1926–1935), and the other samples were prepared at the IMP. Each mixture was subdivided into a number of portions. In total, 137 portions of mixtures prepared using 128 teeth were measured.

3.1.3. Incisors

For the EPR investigations, 27 upper and lower incisors (first and second teeth in the denture) were selected. The range of birth years was from 1912 to 1955. All incisors were cut into lingual and buccal portions of enamel; therefore, 54 samples of enamel were prepared. All samples were prepared at the IMP. EPR measurements of incisors were provided using mixtures of lingual or buccal enamel from different teeth. The mixes were prepared for teeth of donors subdivided into four age groups, and an additional fifth group averaged for the entire range of ages. Each mixture was subdivided into a number of portions. In total, 21 portions of mixtures prepared using 27 teeth were measured. After initial measurement, the mixtures of lingual and buccal enamel in each age group were mixed together, and measured again in order to evaluate background dose for the entire enamel of incisors (16 portions of entire enamel were derived).

Table 1. Summary statistics for tooth samples of first lower molars from non-exposed donors investigated at the IMP and the GSF.

Laboratory	Number of samples prepared by chemical treatment	Number of samples rejected due to mass limitation and impurities in enamel	Total number of measured samples
IMP	79	7	66
GSF	20	2	52
Total	99	9	118

3.1.4. Total statistics

A total of 253 teeth were used for the investigation by EPR of background dose. From the 253 teeth, 295 tooth samples were prepared by chemical treatment. Of the total number of teeth, 19 teeth (23 samples) were rejected from EPR investigations (Table 2).

3.2. Sample loss in the process of investigation

From the discussion above, it is clear that some of the enamel samples under investigation were lost for the following reasons:

1) Inadequate mass of enamel

This factor is to be expected, because the available teeth were extracted for medical reasons and can be anticipated to be of bad quality. The minimum useful sample mass can depend on EPR equipment. About 4% of molars were rejected from investigation by equipment in use at the IMP and the GSF. The number of samples of insufficient mass also depends on tooth position. For incisors, the mass limitation is more acute, because the tooth must be divided into inner and outer parts. On the basis of data from the IMP, about 15% of the samples of incisor enamel cannot be investigated on an individual basis.

2) Presence of impurities in the teeth

The presence of impurities in the tooth enamel is independent of method. This question will be discussed below. However, the influence of impurities on sample-collection strategy is very significant. For molars, for example, about 4.6% of samples must be rejected from investigation because of the presence of impurities. The impurity problem is important predominantly for determination of low dose levels.

3) Anomalously high dose levels

For 2nd premolars from donors registered as non-exposed, doses of greater than 2 Gy were detected for 3% of teeth. Such high doses can be explained by registration errors by the dentists or as a tooth from a donor who should have been a member of the ETRC (that

Table 2. Final statistics of enamel samples prepared by chemical treatment and used for preliminary measurement by the EPR technique.

Laboratory	Number of samples prepared by chemical treatment for preliminary EPR measurement[a]	Number of samples with low mass and impurities	Number of measured teeth[c]
IMP	217	21	221
GSF	78	2	72
Total	295	23[b]	234[d]

[a] The quantity of teeth is not equal to quantity of tooth samples;
[b] One sample was not measured in both labs due to low mass;
[c] Individual and mixed measurements;
[d] Fifty-nine teeth were measured at both the IMP and the GSF.

is, an exposed person). These errors cannot be corrected, and they show the influence of the human element on sampling. It is possible to verify the data for the exposed cohort, but for the non-exposed population it is a very difficult task. The probability of such identification errors must be taken into account in the analysis and sampling strategy for the evaluation of background dose.

For the above reasons, losses of samples must be considered in the planning of investigations by EPR dosimetry, and the initial number of samples must be higher than the number of expected measurements by about 14–25%.

4. METHODOLOGICAL ASPECTS OF THE EPR STUDY

EPR measurements were provided at the IMP and the GSF laboratories. The procedures for sample preparation, EPR measurements, spectrum analysis, and calibration for the GSF and the IMP are described in detail in Appendix 1. Methodological aspects (such as the problem of impurities in the teeth, chemical treatment of samples, method reliability and limitations, and uncertainty analysis) for the current status of EPR dosimetry were investigated.

4.1. The problem of impurities in teeth

The first methodological task of EPR measurements was the investigation of the problem of impurities in the enamel that cause anomalous spectra. This requires the development of criteria for separation of "bad" spectra, which should not be analyzed. Only after this step is completed can we provide precise selection of samples for EPR-dose assessment and for individual measurements and analysis of background signal.

4.1.1. Description of signals due to impurities

The problem of parasitic signals in EPR spectra caused by paramagnetic impurities in teeth is genuine for cases of low radiation-signal intensity (for doses below about 300–400 mGy). Special attention to this problem has to be paid in EPR studies of background doses for persons in the Urals, i.e., for persons who never lived in contaminated areas of the Techa Riverside or on the East Urals Radioactive Trace and who had no occupational exposure (Shishkina et al. 2001). It has been suggested that some of these parasitic signals are introduced by metal impurities in enamel (Shishkina et al. 2001). The cause of such impurities could be metal dentures and fillings, which some people (especially old persons) could have had in their mouths for many years. According to the literature (Kopeikin et al. 1978), such metals as gold, silver, copper, iron, etc., were widely utilized in the practice of dental prosthetics. Therefore, metal dentures and fillings could be a cause for the presence of metal ions in saliva, which could cause ion exchange between the saliva and hydroxyapatite crystallites. The crystal structure of apatite ($Ca_{10}(PO_4)_6X_2$; where 'X' could be OH, F, Cl, Br or I) is a good host for impurity ions (Ikeya 1993). For example, Ca^{2+} can be replaced by univalent cations like Na^{+}, K^{+}, bivalent cations like Mg^{2+}, Sr^{2+}, Fe^{2+}, trivalent cations like Al^{3+}, Y^{3+}, and so on.

Basically, two different types of impurity signals were observed in EPR spectra of background-enamel samples.

The first type of signal is of broad shape and high intensity. A typical example is illustrated in Fig. 1. This signal is very broad and has very large amplitude in comparison with typical EPR spectra of background-tooth enamel. This kind of signal can vary considerably in position, width and intensity (only part of the impurity signal is seen in Fig. 1 due to its great width). When such signals are observed, dosimetry on the basis of radiation-induced radicals is not possible. Fortunately, such cases are rarely observed. In our experience, they were seen in 2.5% of spectra for background teeth.

To illustrate that the possible cause of impurity Type 1 could be the micro-insertion of metal into a tooth, the EPR spectrum of enamel obtained from a tooth that initially had a yellow metal filling is shown in Fig. 2 (the filling itself was removed in the process of sample preparation, but enamel grains were likely to have retained parts of the filling). This impurity signal is not as broad as in Fig. 1, but it is much wider and has much higher intensity than a typical background radiation-induced signal of tooth enamel.

In spite of the absence of a filling in the sample shown in Fig. 1, it could have received metal micro-insertions due to rubbing against its antagonist (in the process of mastication), if this antagonist had a filling or was part of a denture. Such a possibility cannot be excluded, because the presence of metal fillings and dentures is known only for the tooth samples collected for analysis.

Spectra due to the second type of impurities were observed more frequently. The second type of spectrum contains many additional signals with different positions. Additional lines can interfere with the dosimetric signal and can cause a high uncertainty for dose reconstruction. The second type of signal (Fig. 3) has a smaller intensity than that of Type 1 The nature of this type of impurity is not yet clear.

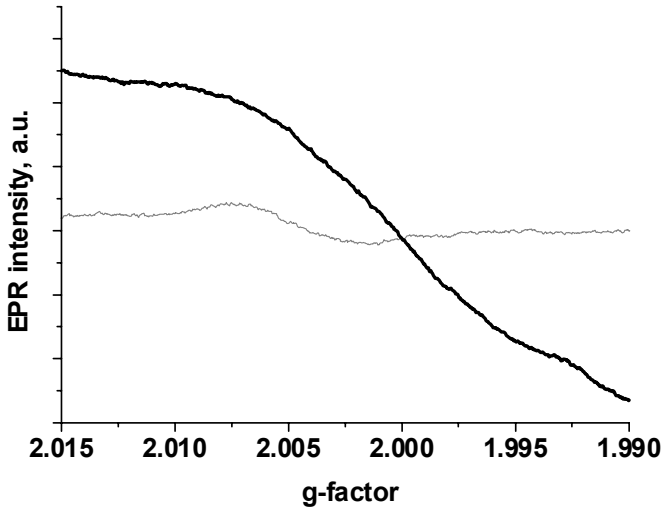

Fig. 1. Example of spectrum influenced by an impurity of Type 1. The heavy line is the spectrum from a tooth (Tooth Code, TC 777) with an impurity; the light line is a typical spectrum of an enamel sample (TC 690) exhibiting a normal background spectrum.

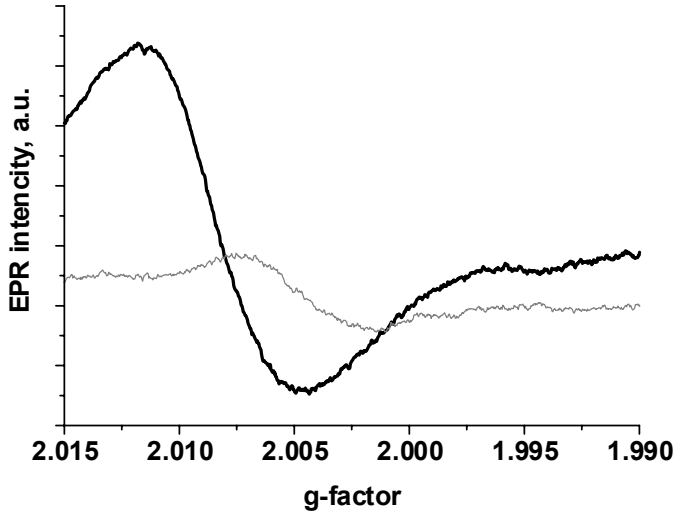

Fig. 2. Comparison of the enamel spectrum of tooth (TC 1245), which had a metal filling (heavy line), and a typical background spectrum of tooth enamel (light line).

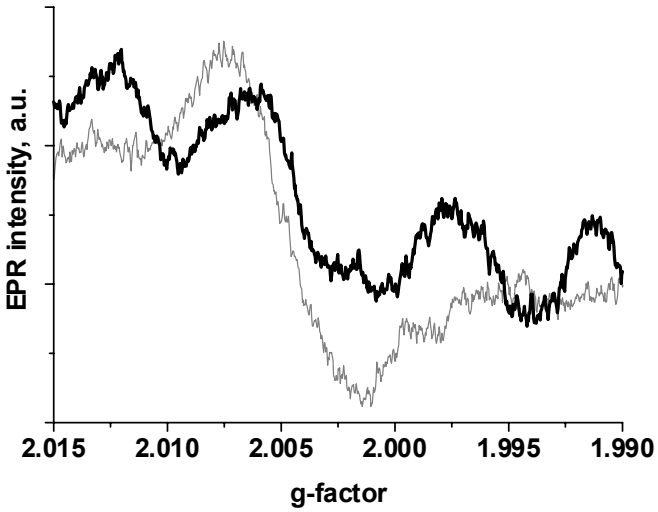

Fig. 3. Comparison of a spectrum with impurities of unknown nature (TC 988, heavy line) and a typical spectrum (light line).

In the literature, such cases are seldom mentioned and almost never scrutinized (Skvortsov et al. 1995), but such spectra can be observed in illustrations in some publications [see, for example, fig. 1 in Egersdörfer et al. (1996); fig. 1 in Rossi et al. (2000)]. At the IMP, samples of tooth enamel have been measured since 1995 by several scientists, and periodically each of the investigators has faced this challenge. However, when higher level doses such as those for the Techa River Cohort were investigated, the influence of impurities has been negligible, because of the very high levels of radiation-

induced signal in those samples. Only for the evaluation of background (or very low) dose is this problem acute.

Because the question of the introduction of impurities through the sample-preparation procedure at the IMP was raised by specialists at the GSF, we compared the sample-preparation procedures used at the IMP and the GSF in an effort to evaluate this possibility.

4.1.2. Comparison of sample-preparation procedures at the GSF and the IMP

Two main differences in the procedures used for sample preparation at the GSF and the IMP are (1) the use of an additional treatment (Titriplex III and ethanol washing) at the GSF; (2) the use of KOH at the IMP versus NaOH at the GSF.

Theoretically, washing with Titriplex III can remove metal ions located on the surface of enamel grains. Preliminary attempts at washing samples with Type 2 impurities in Titriplex III after initial preparation indicated a slight decrease of signals due to impurities, but washing samples with Type 1 impurities in Titriplex III had no effect. However, this is a very preliminary conclusion.

The use of KOH instead of NaOH speeds up the process of dentin removal by etching. Also, it has an effect on decreasing the non-radiation EPR signal due to organic radicals (Romanyukha et al. 2000).

A special study was performed in order to compare the qualities of sample preparation. For this research, 15 samples of first lower molar teeth (6^{th} lower position) were selected. Donors of these teeth belonged to the control cohort, i.e., background levels of dose were expected for these samples. Teeth were separated into two parts. The first part of each tooth was prepared at the IMP and the second part of each tooth was prepared at the GSF. All samples then were measured at the GSF. Comparing amplitudes of background organic radicals for the two halves of teeth, one can see that the amplitude for the IMP-prepared samples was lower than that for the GSF-prepared samples (Fig. 4). On average for the IMP-prepared samples, the amplitude is lower by 56 a.u., which corresponds to 244 mGy in accumulated dose units. Therefore, the IMP-sample-preparation procedure demonstrates better removal of the organic components from tooth enamel.

However, visual comparison of recorded spectra (Fig. 5) confirms a large quantity of signals due to impurities in the IMP-prepared samples. Possibly, treatment in the stronger alkali KOH used at the IMP, besides removing the organic component of enamel, also activates sites of paramagnetic impurity in tooth enamel. Unfortunately, the result is a worsening in quality of the spectrum. This fact can be very important, if visual assessments in the spectra analyses have played a significant part in the total uncertainty of measurement results.

Thus, difficulties exist with unbiased analysis of spectra that include signals due to the presence of impurities. It is evident that criteria are necessary for useful sample preparation and for the estimation of spectrum quality. Such criteria should allow the impartial selection of unanalyzed spectra. It is also necessary to evaluate the reliability of dose estimation on the basis of low-level radiation signals.

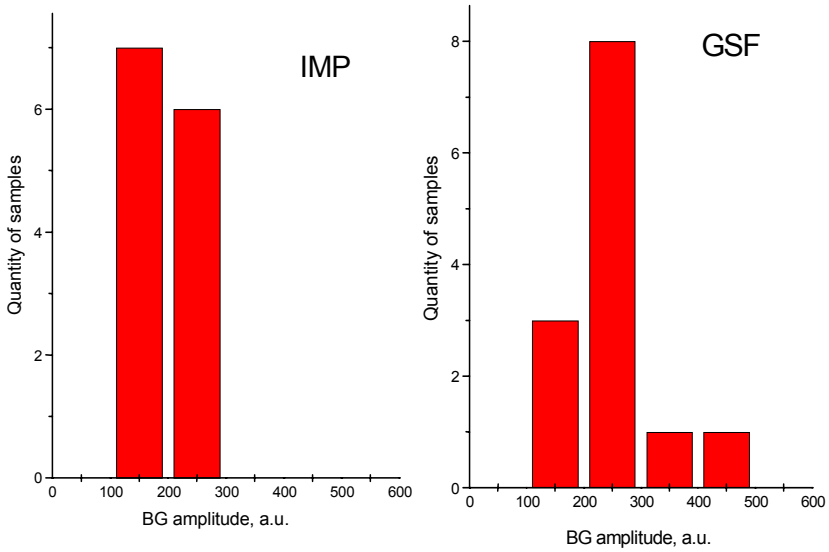

Fig. 4. Comparison of background EPR-signal distributions for IMP-prepared samples (left) and for GSF-prepared samples (right).

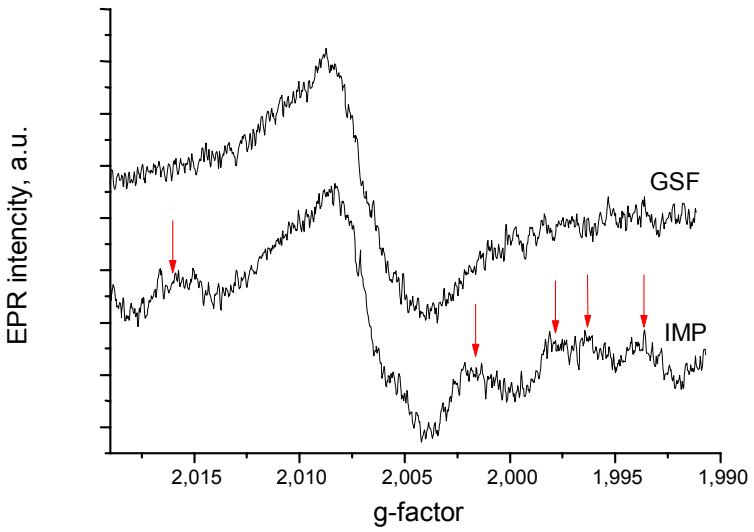

Fig. 5. Examples of EPR spectra of samples of tooth enamel. Lower line—spectrum of IMP-prepared sample, upper line—spectrum of GSF-prepared sample (TC 2162). The arrows show peaks that are believed to be due to impurities.

4.1.3. Criteria for the estimation of spectrum quality

It is very important to establish the appropriateness of a spectrum for analysis. The following criterion for rejecting spectra was suggested: *If the spectrum contains parasitic signals lying partly or fully in the range of g-factors 1.995–2.006, and the amplitudes of those signals are equal to or greater than the amplitude of the dosimetric signal, then dose reconstruction from the spectrum of this sample is not possible*.

However, in most cases it is difficult to estimate the amplitudes of the signals due to impurities. This estimation also depends on the experience of the operator analyzing the spectra. Different operators can give different estimates of quality for individual spectra. In other words, it is a visual criterion.

Hence, it was necessary to develop an unbiased criterion. A preliminary criterion with this aim was formulated using estimates of the amplitude of the signal-to-noise ratio (*SNR*). Noise intensity was determined as the difference between maximum and minimum points at the wings of the spectrum. Therefore, if the *SNR* is less than 1.0, the spectrum has poor quality and dose reconstructed on its basis is not suitable for individual dosimetry. This preliminary criterion has been included in software used at the GSF and the IMP for spectra deconvolution.

All measured samples were analyzed using this criterion. The distribution of noise amplitude for spectra of samples prepared at the GSF and the IMP is the same. No correlation is observed between the laboratories in sample preparation and quantity of impurities in the spectra.

No dependence was found between the *SNR* and sample mass or donor age for available samples. Preliminary analysis indicates no correlation between the *SNR* and presence of caries as recorded in the database "TOOTH" (Shishkina et al. 2001).

The signal-to-noise criterion appears to be too rigid, because the radiation signal can have a clear peak in spite of the fact that the noise amplitude (at the wings of spectrum) can be higher. In this case, the distance between the unknown and radiation signals allows separation of the radiation peak. Therefore, in a number of cases, the visual criterion can be very useful. We compared these two criteria as applied to a statistical investigation of background-dose estimation for 1st molars.

Application of two criteria for estimation of unusable spectra in the analysis of individual distributions of background doses for first lower molars

This research includes results obtained for 50 first lower molars (6th lower position). All samples were prepared and measured at the IMP. The quality of spectra was estimated using the following two criteria:

(1) Signal-to-noise ratio (*SNR*) and

(2) Visual criterion. The quality of each one of three recorded spectra was visually estimated as appropriate or not for analysis and the frequency of selection of good spectra was calculated.

In comparing results of sample separation, one can see that the distribution of background doses is lognormal. However, the signal-to-noise criterion rejects spectra with signals lower than the noise amplitude at the wings of the spectra, even if they are

clearly seen. These signals are usually lower than 100–150 mGy. So, both the median and the mean of the sample set shift to higher dose values (Table 3). This fact can result in overestimation of average dose. Therefore, in the following we will use estimation with the criterion of visual selection. Moreover, visual separation is not so stringent and the number of appropriate samples using this criterion is more than that using the signal-to-noise criterion.

As can be seen from Table 3, the use of the *SNR* criterion rejects about 44% of the spectra. The visual criterion is less strict and the rejection of samples is only 18%. The use of both the *SNR* and visual determination of signal-peak presence provides a sample rejection of about 50%. At the same time, the shift in median of the dose distribution due to signal-to-noise separation is about 60 mGy toward higher doses.

Therefore, all our estimates in the current stage of investigation of background doses have used the visual criterion. In this case, about 21–23% of teeth have been rejected from analysis (taking into account insufficient mass of some enamel samples). However, this is not a final result, because the problem of defining an objective and reliable criterion for spectrum selection (and corresponding sample selection) remains under investigation.

4.2. Uncertainty analysis for EPR measurements

The uncertainty of absorbed dose in enamel is due to uncertainties in calibration, enamel response to ionizing radiation, intrinsic signal, and sensitivity of the EPR spectrometer. Analysis of the results of international intercomparisons demonstrated that an EPR-dosimetry system, using a spectrum-deconvolution procedure and a universal calibration curve, could reconstruct doses within ±25 % above 400 mGy, and ±100 mGy below 400 mGy with the probability of 0.95 (Wieser et al. 2000a,b).

Comparison of the results obtained for teeth donated by persons exposed on the Techa River and the residents of the background areas of the Urals demonstrated the consistency of data, and the absence of systematic shift between the measurements, from these two laboratories. Nevertheless, the 95% confidence interval of the deviation between the individual IMP and GSF results in the low dose range is equal to ±180 mGy.

The sources of such divergences for assessment of dose by EPR measurement is puzzling, because similar methods are used at the IMP and the GSF. However, two different spectrometers were used, and some differences exist in the sample-preparation methods. The spectrum analyses and calibrations were essentially the same. As shown

Table 3. Statistical information for spectra selection using different criteria.

Parameter	Without using any criteria	*SNR* criterion	Visual criterion	Both criteria
Number of samples	50	28	41	25
Mean, mGy	149	178	147	174
Median, mGy	125	183	120	174
Std. deviation, mGy	71	77	73	80

above, the differences in the chemical treatments can cause large uncertainty only because of operator arbitrariness during spectrum analysis (for the case of the influence of impurities on spectrum shape). The calibration cannot result in discrepancies, because the slopes of calibration curves for the IMP and the GSF are practically equivalent (Fig. A2 in Appendix 1). The source of irradiation error is not higher than 3%; variation of enamel sensitivity to radiation in the investigated population is 8%; and the dose accumulation in enamel powder differs from the unbroken lattice enamel by not more than 2%. Therefore, the calibration uncertainty is about 9%.

Possible sources of uncertainties are in the spectrum-processing algorithm, equipment characteristics, and the stability of signal recording reflected in the reproducibility of measurement results. Because the reproducibility of results can be regarded as characteristic of the reliability of the method, this characteristic was tested based on statistical investigations of measurements of background doses. The differences both between various operators' dose assessments and between various equipment used by the same operator were also tested.

4.2.1. Uncertainty due to operator arbitrariness

For comparison of uncertainty introduced by the operator, spectra of 32 first lower molars (after selection of good spectra) from non-exposed donors were used. All enamel samples were prepared at the IMP. Two different operators (one from the GSF and another from the IMP) analyzed the same spectra recorded using GSF equipment. Fig. 6 shows the results of comparison of doses obtained by two operators.

The equation of the regression line for the data in Fig. 6 is:

$$D_{IMP} = 70(\pm 15) + 0.51(\pm 0.07) * D_{GSF} \qquad r = 0.64 \tag{1}$$

The 95% confidence interval of the deviation between the individual EPR results (obtained by two operators using GSF equipment) in the low dose range is ±140 mGy.

In Fig. 7, the data are shown as boxes representing statistical values for the results obtained by the GSF and the IMP operators.

It is obvious that these two distributions are not equal. In spite of the very similar mean and median values, the range of data obtained by the GSF operator is wider than that obtained by the IMP operator. This fact is reflected by the low correlation coefficient (Fig. 6) and the wide confidence interval. Thus, the uncertainty of common analysis of individual measurements obtained by IMP and GSF operators in the lower dose range (<400 mGy) is not less than ±140 mGy.

4.2.2. Instrumental uncertainty

Twenty-three samples of 1st lower molars were recorded and analyzed at the IMP and at the GSF by one operator. Therefore, it was possible to compare results obtained by one operator using different spectrometers (Fig. 8).

The equation of the regression line in Fig. 8 is:

$$D_{IMP} = 40(\pm 20) + 0.6(\pm 0.13) * D_{GSF} \qquad r = 0.51 \tag{2}$$

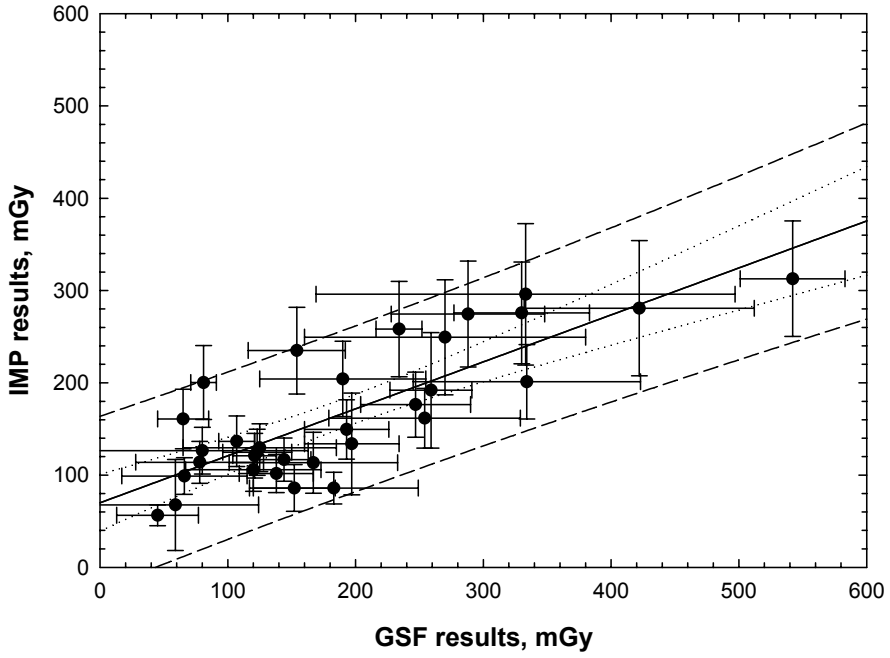

Fig. 6. Comparison of results obtained by IMP and GSF operators analyzing the same 32 spectra. The solid line is the line of linear regression. The dotted lines are the 95% confidence intervals of regression. The dashed lines are the 95% prediction interval of regression corresponding to the 95% confidence interval of the deviations between the individual EPR results.

The 95% confidence interval of the deviation between the individual-EPR results (obtained by the same operator using IMP and GSF equipment) in the low dose range is ±120 mGy, which is slightly less than in the case of processing by different operators.

In Fig. 9, the data are shown as boxes representing statistical values for the GSF and the IMP results obtained by the same operator.

It is obvious that these two distributions are more similar than in the case of processing the same samples by different operators. The difference between maximal and minimal values in each group of measurements is not so large as in the case shown in Fig. 7. In spite of the similar mean and median values, the range of data scattering obtained using the GSF equipment is somewhat wider than that obtained using the IMP equipment. This may be because the GSF spectrometer is more sensitive. Thus, the uncertainty of common analysis of individual measurements obtained by one operator using equipment at IMP and GSF is not less than ±120 mGy.

It should be noted that this comparison does not reflect the full influence of the equipment on the measurement results, because the spectrum processing cannot be excluding as an uncertainty factor. Moreover, it is clear that the difference in equipment can't be detected based on these data, because even in the case where the same operator

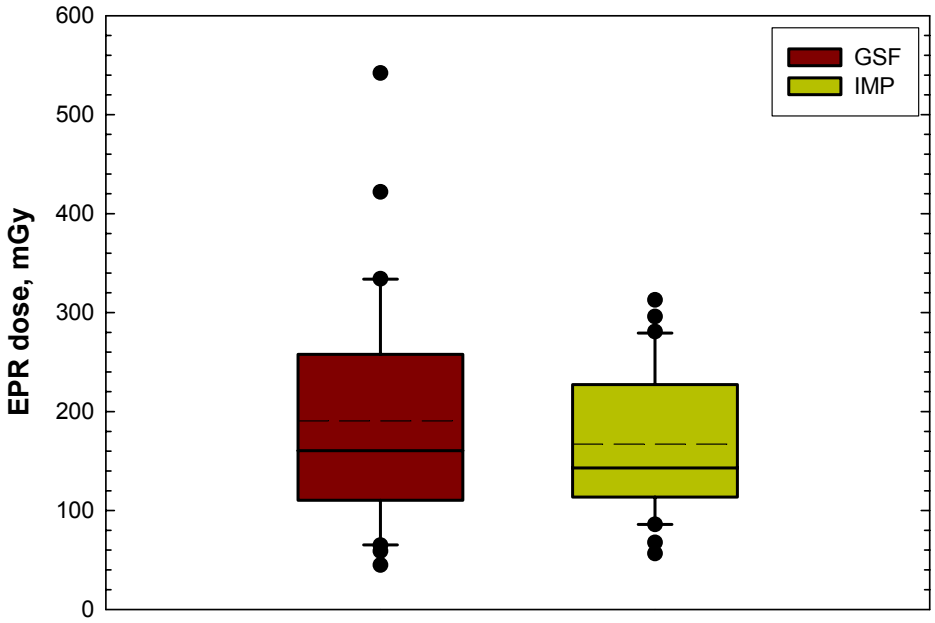

Fig. 7. Statistical values for the distributions of EPR results obtained by IMP and GSF operators using the same spectra. The boundary of the box closest to zero indicates the 25th percentile, solid lines within the boxes mark the median, dashed lines within the boxes mark the mean value, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers (error bars) above and below the box indicate the 95th and 5th percentiles. In addition, outlying points are indicated.

used different equipment, the correlation between the two sets of results is very approximate.

Thus, the current data indicate that for low dose levels a major uncertainty is due to spectrum simulation and processing. Moreover, for reconstruction of background doses, the person operating the equipment introduces the most uncertainty. Therefore, it is necessary to improve the processing of spectra based on improvements in spectrum simulation, so that such an automated procedure can minimize the arbitrariness of the operator.

This conclusion demonstrated the necessity of improvements in spectrum analysis and promoted development of a new algorithm for spectrum processing that minimized the arbitrariness of the operator in the individual dose assessments (which is very important for Techa River dosimetry). However, because the average and median values in the statistical estimations of background doses are similar (the difference is not significant) for the IMP and the GSF, the current results can be used as characteristic of background dose for molars.

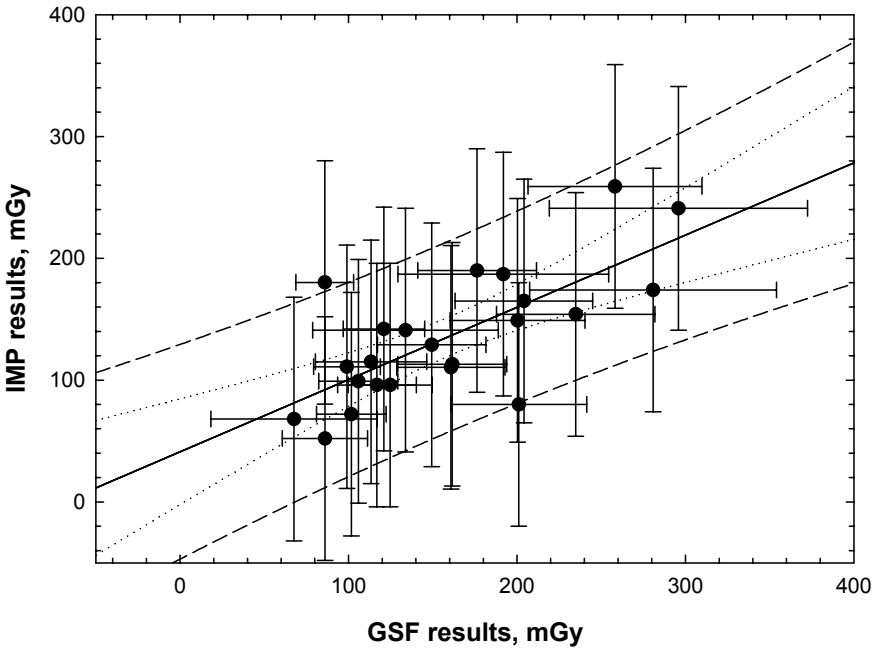

Fig. 8. Comparison of results obtained by one operator using different equipment. The number of samples is 23. The solid line is the line of linear regression. The dotted lines are the 95% confidence intervals of regression. The dashed lines are the 95% prediction intervals corresponding to the 95% confidence interval of the deviation between the individual-EPR results.

4.2.3. Reproducibility of the method for estimation of low doses

The reproducibility of EPR-measurement results was tested using two groups of 2nd premolars, separated according to the donor's birth year:

(1) 1925 and earlier (17 teeth) and

(2) from 1926 to 1935 inclusive (38 teeth).

All enamel samples from these groups were prepared at the GSF. All donors lived in non-contaminated areas. The age separation was intended to minimize the scatter in individual doses in each sample bracket.

After initial measurements to exclude samples with poor quality enamel, all sample powders in each group were thoroughly mixed. Then all enamel was separated into samples with weights optimal for measurement (100±5 mg). Samples in each group were numbered. Group 1 numbered 20 samples, and Group 2 numbered 50 samples.

Analysis of the measurements was based on the assumption that the same dose should have been measured in each portion of the mixture from each group. This is an approximation for two reasons: (1) even in one tooth the energy deposition in the enamel is non-uniform and (2) the main factor influencing the reproducibility of measurement results is anisotropy of the EPR signal. Under this assumption, the scatter of measured

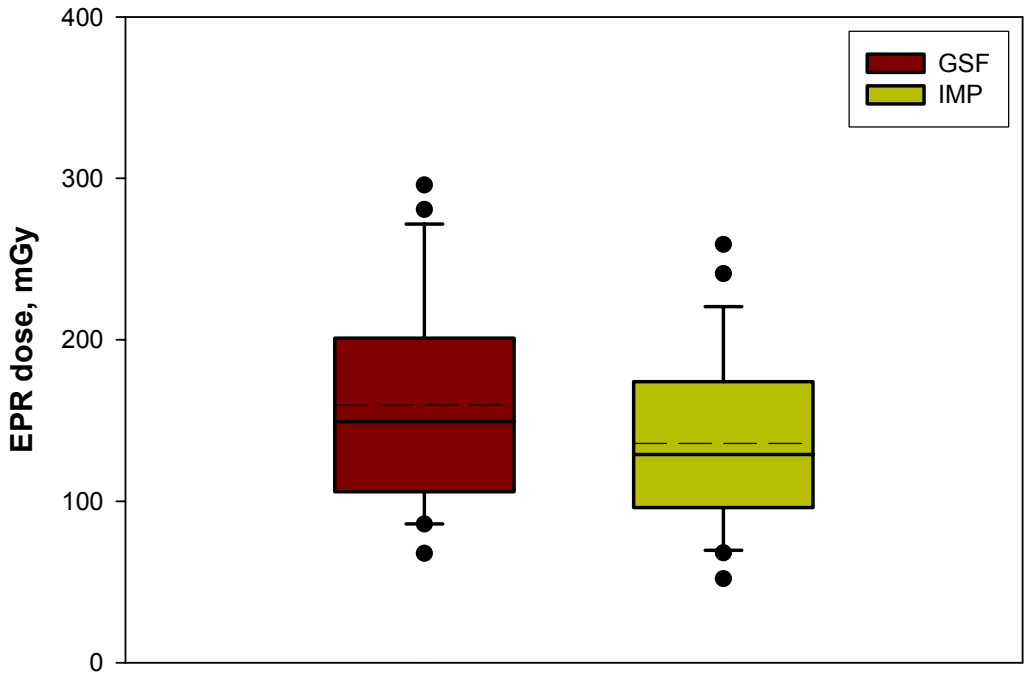

Fig. 9. Statistical values for distributions of EPR results obtained by the use of IMP and GSF equipment and evaluated by one operator. The boundary of the box closest to zero indicates the 25th percentile, solid lines within the boxes mark the median, dashed lines within the boxes mark the mean value, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers (error bars) above and below the box indicate the 95th and 5th percentiles. In addition, outlying points are indicated.

dose results in the different portions of the mixtures should reflect the reproducibility of experimental evaluation of the level of background dose.

Because the same operator using the same spectrometer provided all measurements, the results of the reproducibility evaluation can be attributed to the uncertainty of the reproducibility of the EPR method used for these measurements.

The dose distribution is shown in Fig. 10a for the mixture for Group 1 and in Fig. 10b for the mixture of Group 2. The dose distribution for each case can be approximated by a normal distribution, which validates the one-dose assumption. The statistical parameters for both distributions are shown in Table 4.

Table 4. Descriptive statistics for distributions of doses for Groups 1 and 2.

	Mean	Valid no.	Minimum	Maximum	Std. deviation	Range
Group 1	115	20	57	205	37	148
Group 2	94	50	38	208	33	170

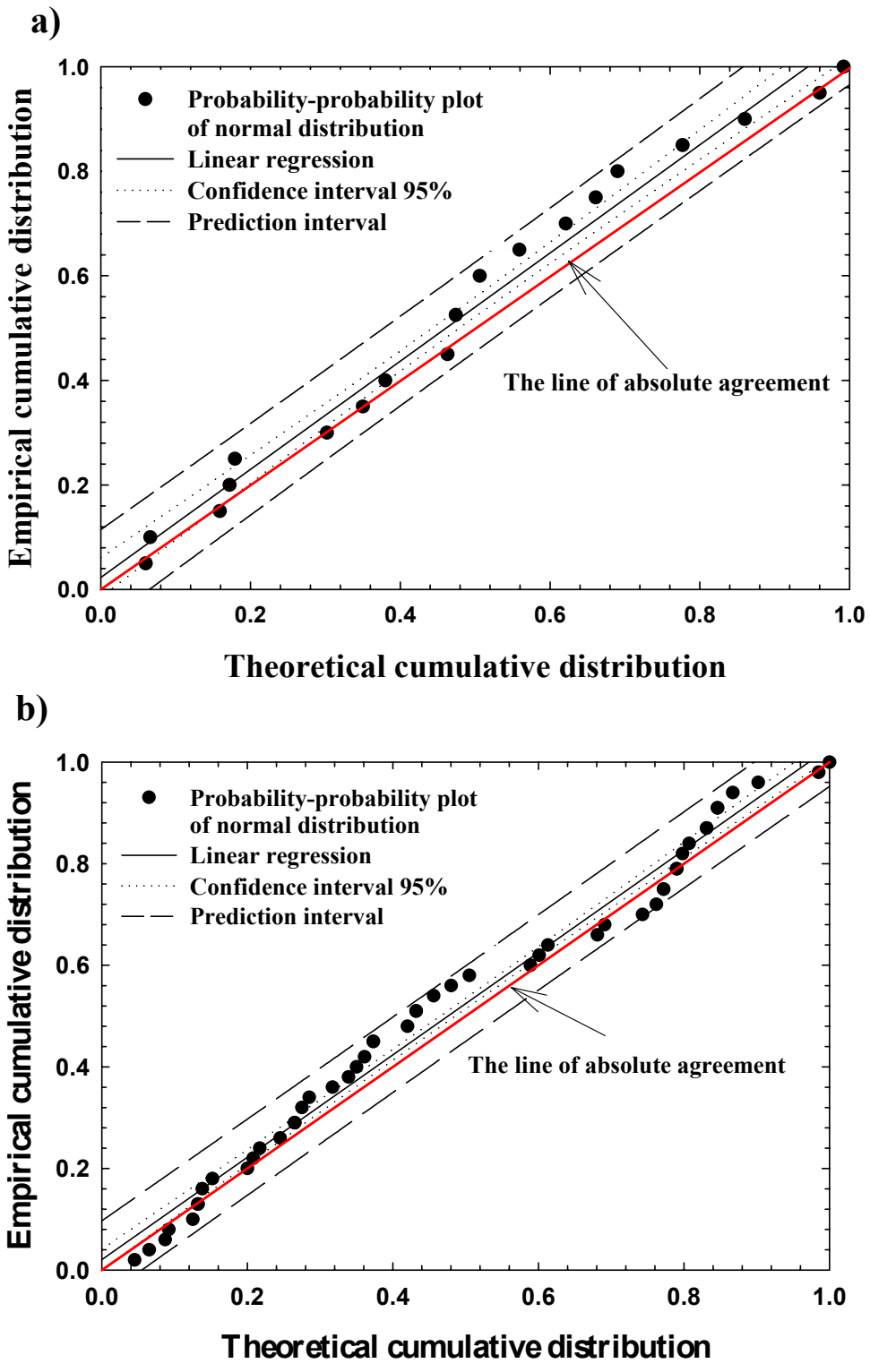

Fig. 10. Comparison of theoretical and empirical cumulative normal distributions for two groups of samples: a) Group 1; b) Group 2.

Comparison of the experimental probability values to a theoretical computation of normal probability (Figs. 10a,b) shows the coincidence of empirical and expected values to be in the range of 11% for the first group and in the range of 8% for the second group. These evaluations are in good agreement with the variations in statistical parameters for the distributions analyzed. So, taking into account standard deviations and range of coincidence with the normal distribution, the uncertainty of reproducibility for each investigated group is 43% (60 mGy for the first and 40 mGy for the second) relative to the average-dose value.

The agreement of this statistical result with standard deviations of individual measurements is demonstrated below (see Section 4.3.1). Each portion of the mixture was measured three times, and has a corresponding average and standard-deviation value. Analysis of dose dependence for these standard deviations has shown no correlation with the background-dose range (Fig. 11).

So, in the range of 70–130 mGy (25^{th}–75^{th} percentiles of estimated dose values in the different portions of the mixture) the reproducibility of measurement results can be assumed to be constant. The average values of standard deviations are (44±22)% for the first mixture and (54±25)% for the second mixture. In both cases it is about 50±25 mGy. All these results are comparable with previous estimation of the reproducibility for the IMP-measurement results. It should be noted that this uncertainty value for reproducibility should be applied only to individual estimates of dose for good quality samples. Additional uncertainty resulting from impurities requires modification of this value. The algorithm for the estimation of such additional uncertainty is currently under investigation.

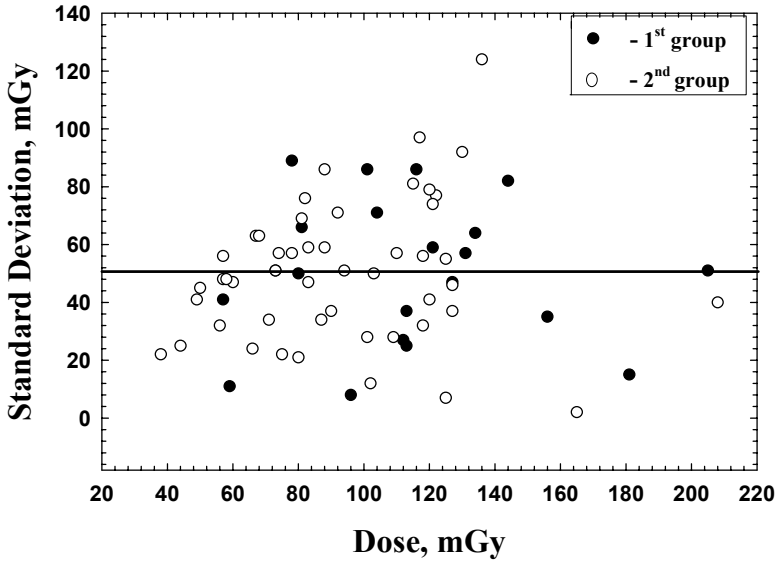

Fig. 11. Dependence of standard deviations of three repeated measurements on estimated average dose. The solid line is the average level of observed standard deviations for doses measured by EPR.

4.2.4. Total uncertainty of EPR measurements for low dose levels

The total uncertainty of individual EPR measurements for low doses has contributions from the following sources:

a) uncertainty of calibration curve;

b) reproducibility error; and

c) uncertainty of spectrum processing.

As shown in Appendix 1, the uncertainty in calibration is not higher than 9%. The reproducibility uncertainty for low dose assessment is about 50 mGy (about 43%). It should be noted that this is an average uncertainty for good-quality samples, but in some cases this value can be higher due to poor spectrum quality. For these cases, this value needs additional individual estimation. Exact estimation of spectrum-processing uncertainty is under investigation now.

However, for current methods based on the results discussed above, it is possible to assume the total uncertainty for analysis of one laboratory data (the same operator) as ±120 mGy and for common analysis of IMP and GSF data as ±140 mGy.

4.3. Improvements in spectrum processing

As shown above, the main factor of uncertainty in the process of spectrum deconvolution is a shift of the g-factor, which is caused by operator arbitrariness. Related to this, two kinds of improvements were made in the method of spectrum processing:

- improvements in spectrum deconvolution\ and
- estimation of deconvolution uncertainty for the new spectrum-processing procedure.

4.3.1. Improvement of the spectrum-deconvolution procedure

Two steps were taken to improve the procedure for deconvolution of EPR spectra. The first step was to change parameters used for the modulation of experimental EPR spectra. Previously, a set of four Gaussian lines was used to deconvolute spectra (Koshta et al. 2000): One line was used for the background signal due to organic radicals, two lines were used for the radiation induced signal (anisotropic and isotropic), and one line was used to modulate signals of unknown origin. The deconvolution procedure was programmed in special EPR-dosimetry software. While working with this software, it was discovered that the background signal was more complicated than expected and that more lines were needed to modulate it. The modulation of the axial anisotropic radiation induced signal also needed some modifications. After improvement, the number of lines used was changed. Also, it became possible to use non-Gaussian lines simulated through the use of external software. The background signals were modulated by three lines: two Gaussian lines and one specific simulated line with g = 2.0046, which appears with a rise in microwave power. The axial radiation-induced signal was simulated with a combination of Gaussian and Lorenzian lines with anisotropic g-factors of 2.0055 and 2.0021 (Callens et al. 1998). The set of lines with new parameters better describes the experimental EPR spectrum than that previously used. An example of a dosimetry-software window with the new deconvolution procedure is shown in Fig. 12.

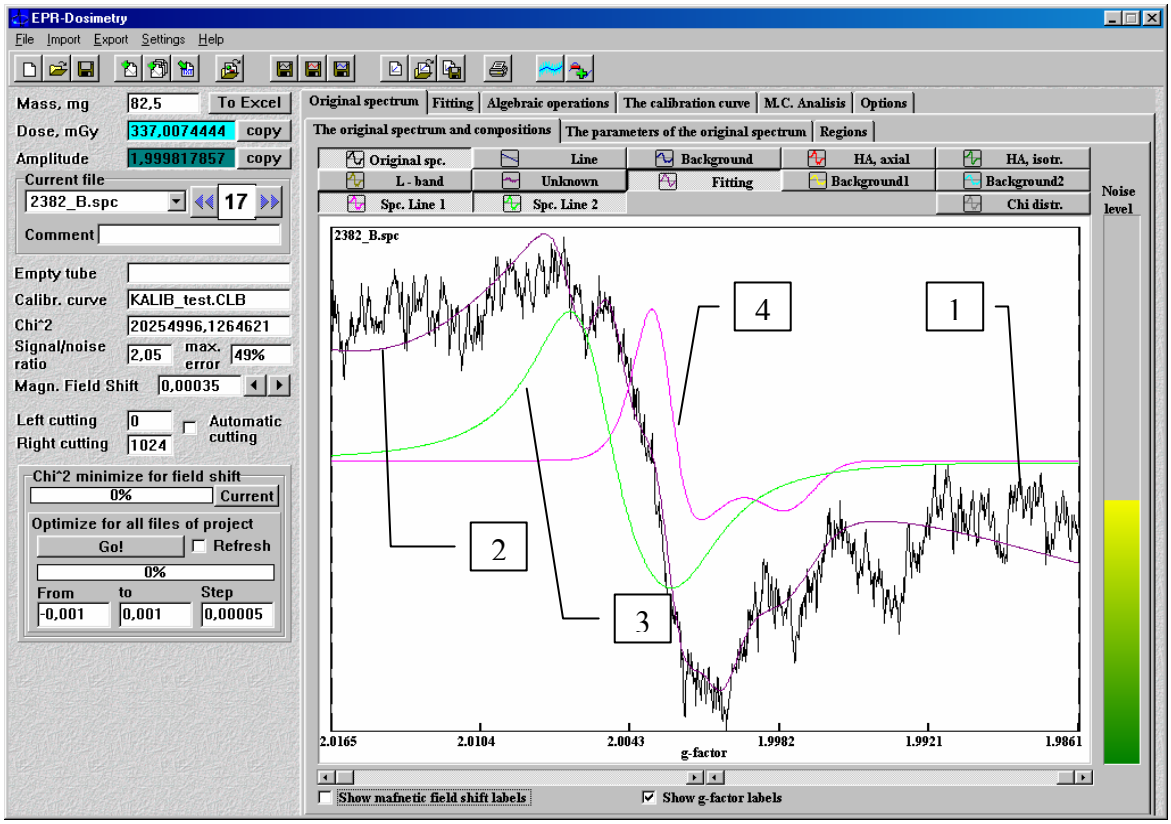

Fig. 12. Example of a custom EPR-dosimetry-software window. Line (1) represents the original EPR spectrum, line (2) the deconvolution result, line (3) the background, and line (4) the radiation-induced axial line. (TC 2382).

The second step was taken to decrease the influence of the operator's choice of deconvolution parameters. The main parameter depending on the operator is the g-factor shift. When using the deconvolution procedure, the operator must select the value of the magnetic shift corresponding to the best fit of a single experimental spectrum. Different operators can select different values according to their experience.

Therefore, automation of that procedure was needed. The automated procedure that was developed uses a chi-square minimization method, i.e., the value of the g-factor shift is selected to minimize the differences between the experimental EPR spectrum and its modulated deconvolution. This automated procedure was tested with spectra from tooth enamel with a range of accumulated doses. The procedure was found to work fine with doses higher than 700 mGy in 100% of cases. For doses lower than 700 mGy the procedure works well in about 70% of cases. The operator's only task is to check that the procedure selected a proper value and to correct it if necessary.

The new approach was tested using EPR spectra obtained on different teeth from one donor. The donor was a female (Identification Code 46890) born in 1933 who lived in

Gerasimovka (upper Techa) during the years 1951–1957. Enamel samples from seven posterior teeth extracted for medical reasons were prepared and measured at the IMP in 2001–2002 using the old spectrum-processing algorithm; the resulting doses for different teeth were distributed over a very wide range (Table 5). The same spectra (obtained at the IMP) were re-estimated using the new procedure, and, additionally, these samples were measured repeatedly and evaluated at the GSF in 2002.

The results obtained using the old deconvolution procedure demonstrated a large dispersion of doses for the same donor. After the same spectra were re-estimated using the new procedure, the dispersion of doses became significantly smaller (Table 5). This test shows that the new deconvolution procedure can increase the reliability of individual-dose reconstruction on the basis of EPR measurement.

It should be noted that the old and new mean values of the dose estimates are not statistically different. However, the scattering of dose values obtained from the different teeth of the same donor (shown in Fig. 13) demonstrates the potential for large uncertainty in individual-dose assessments obtained on the basis of EPR measurements for the cases if only a single tooth sample from the exposed donor is available. That is why standard deviations of repeated EPR measurements of the same sample (shown in Table 5) can not serve as a "measure of uncertainty" for individual-dose reconstruction.

The new approach results in decreased data scattering such that all estimated doses by the GSF and the IMP are within the statistical error of the mean, assumed above to be ±140 mGy according to the estimated uncertainty for interlaboratory comparison.

The new spectrum-deconvolution procedure works well with spectra from teeth with any accumulated dose, but it is highly appropriate for the low dose range. The role of the operator in the low dose range is very significant, and the automated procedure can significantly reduce the uncertainty. Also, adequate fitting of the background signal is very important in cases when the deconvolution procedure is applied to spectra associated with low doses. Therefore, it is necessary to re-analyze all "old" spectra (available in the

Table 5. Comparison between old and new approaches of spectrum-deconvolution procedure using different tooth measurements for a selected Techa River resident (IC 46890).

Tooth TC	Tooth position	Dose±standard deviation, mGy		
		Old approach	New approach	
		IMP	GSF	IMP
1968	6 upper	197±100	197±11	246±15
1981	4 upper	875±219	240±149	121±22
2382	5 upper	464±116	133±182	460±109
2532	4 lower	49±100	306±49	388±70
2533	5 upper	177±100	169±23	280±109
2534	6 lower	59±100	191±52	261±59
2535	6 lower	197±100	157±23	170±79

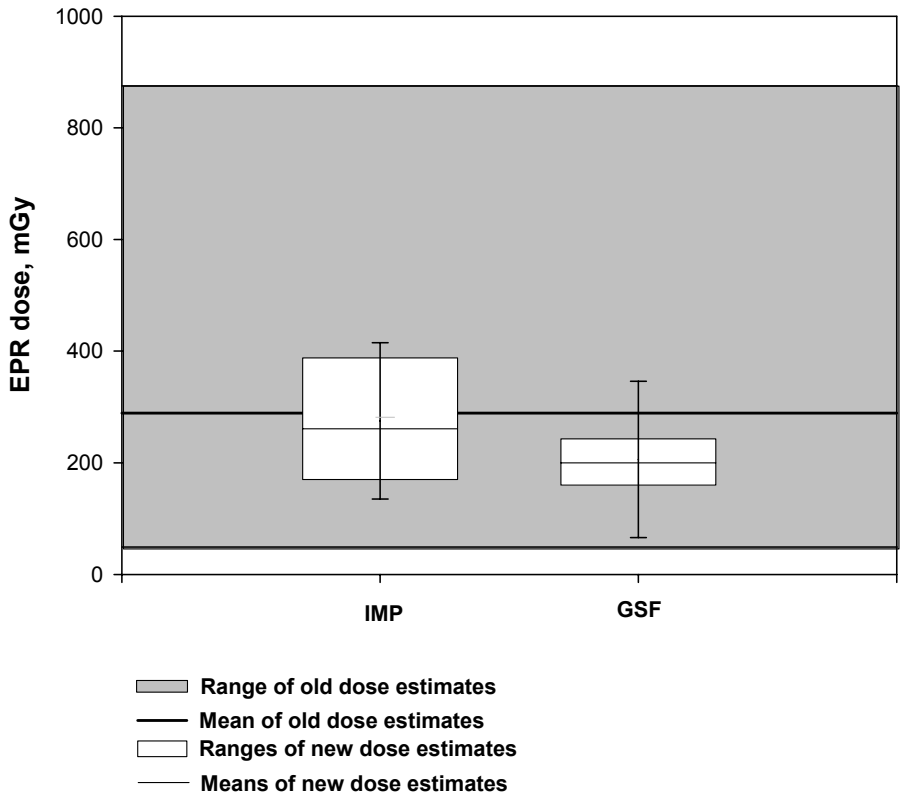

Fig. 13. Results of dose reconstruction using old and new methods of spectra deconvolution. Grey color corresponds to the range of old dose estimations. The bold line is the mean value of old doses. The boundaries of the boxes indicate the range of new dose estimates for the same teeth made at the IMP and the GSF; solid lines within the boxes mark the mean values. Whiskers (error bars) above and below the mean values equal 140 mGy (uncertainty for joint analysis of IMP and GSF data).

IMP and GSF databases) using this new deconvolution procedure. This task has been completed for the "background samples." Samples from exposed donors are currently being re-analyzed.

4.3.2. Estimation of uncertainty for the new spectrum-processing procedures

The shift of the g-factor for each spectrum can differ over a small range, depending on the circumstances under which it was recorded. The distribution of the possible shift was estimated using 28 enamel samples with higher doses and express provision of a radiation-induced peak. For such spectra, the error of estimation of g-factor shift is a negligible quantity. The variations of g-factor shift can be determined for these samples due only to conditions of spectrum recording. The shifts of g-factors were estimated for these spectra. The distribution of shifts was normal (Fig. 14).

The obtained distribution was used to analyze the estimated amplitude stability depending on g-factor variability. This distribution was used to compute the probability of g-factor position. With the use of Monte Carlo methods, the amplitudes were

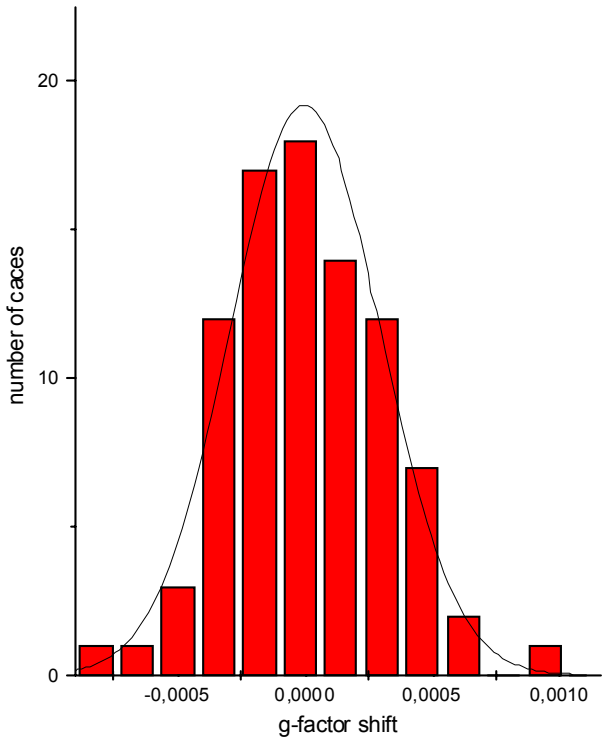

Fig. 14. Distribution of g-factor shifts.

randomly sampled using the g-factor probability distribution to determine the probability distribution of amplitude in the measured spectra.

Five hundred and six samples were tested from the non-exposed and exposed populations in the dose range from 25 mGy to 30 Gy. As a result, it is possible to separate three dose groups based on amplitude-distribution shape:

1. Weak radiation induced signal (Fig. 15a).

The radiation-induced signal is feebly marked. The amplitude distribution is normal (Fig. 15b). In other words, the shift of g-factor in the left direction (smaller value) decreases the amplitude, and g-factor shift in the right direction (higher value) increases the amplitude. Such a distribution is observed for the dose range lower than 450 mGy. Uncertainty of dose estimation is computed based on amplitude-distribution parameters.

For low dose levels, the computed uncertainty varies very significantly depending on spectrum quality and the presence of impurities. The range of uncertainty variations relative to estimated value for individual-spectrum analysis is from 32 % (for doses about 400 mGy) to 1456 % (for doses of tens of mGy).

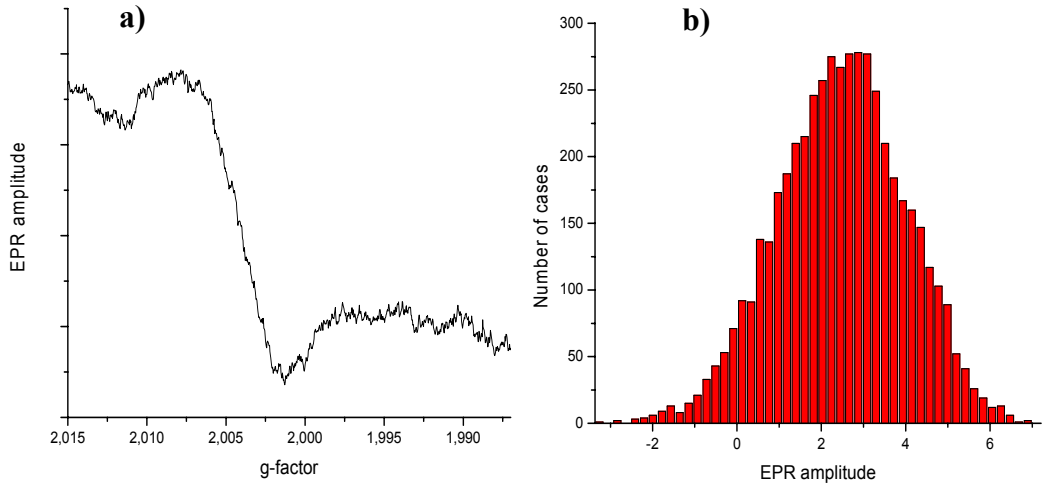

Fig. 15. Weak radiation induced signal; a) example spectrum with weak radiation induced signal; b) amplitude distribution for spectra with weak radiation-induced signals.

2. *Very strong radiation induced signal (Fig. 16a).*

In this case, it is obvious that each shift of g-factor results in a decrease of signal amplitude, because the most likely g-factor position corresponds to the peak position of the spectra. This is typical for the dose range above 700 mGy. The amplitude distribution is shown in Fig. 16b. The uncertainty due to g-factor shift in this case is negligible.

3. *Intermediate reading (Fig. 17a).*

In the dose range from 450 mGy to 700 mGy, where the radiation-induced signal is not very strong but not too weak, the amplitude distribution takes a form intermediate between Cases 1 and 2 (Fig. 17b).

The proposed estimate of the influence of g-factor shift on the value of reconstructed dose is a very important step that is necessary for accurate evaluation of individual-dose uncertainty. In common with the new spectrum-deconvolution process, this Monte Carlo analysis (as an element of uncertainty estimation) allows more reliable evaluation of the results obtained with the EPR method. This uncertainty estimation depends on spectrum quality.

In general, eqn (3) describes the total uncertainty of individual-EPR-dose assessment.

$$\delta = \sqrt{\delta^2_{calibration} + \delta^2_{reproducibility} + \delta^2_{g-factor\ shift}} \qquad (3)$$

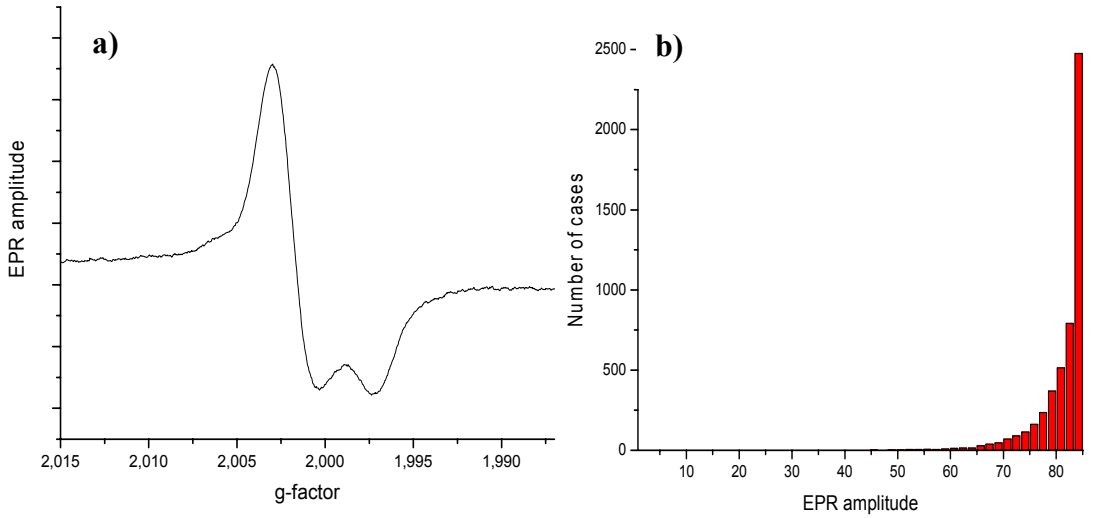

Fig. 16. Very strong radiation induced signal; a) example spectrum with very strong radiation induced signal; b) amplitude distribution for spectra with very strong radiation induced signals.

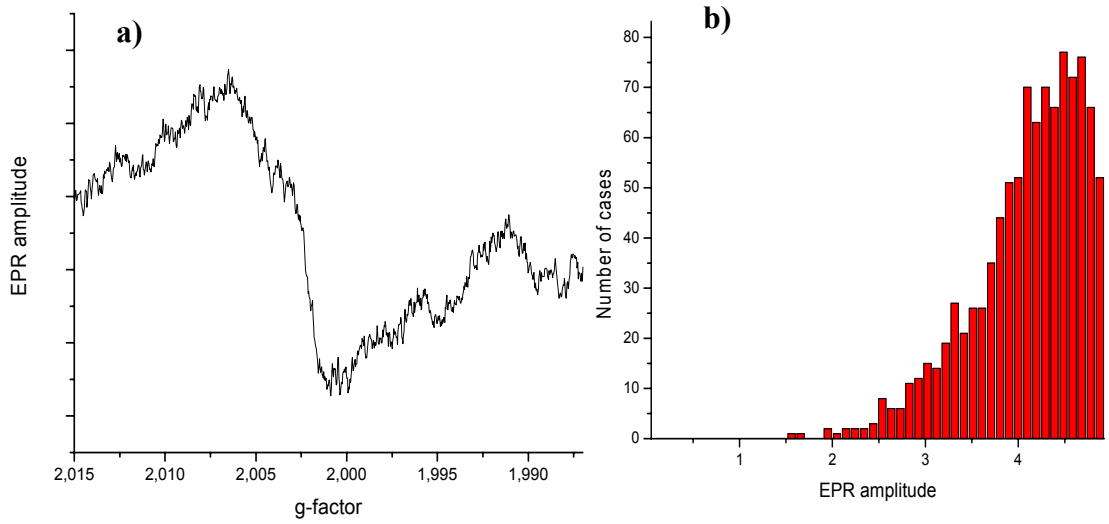

Fig. 17. Intermediate reading of radiation induced signal; a) example spectrum with intermediate strength of radiation induced signal; b) amplitude distribution for spectra with intermediate reading of radiation induced signals.

An example of the application of eqn (3) for the assessment of total uncertainty for individual measurements is shown in Table 6 for a selected Techa River resident (the same as described above in Table 5). As can be seen, the total uncertainty of individual

Table 6. Application of new approaches of estimating uncertainty for individual measurements of tooth enamel for a Techa River resident (IC 46890). The results shown were obtained at the IMP.

Tooth TC	Tooth position	Dose, mGy	Total uncertainty, mGy
1968	6 upper	246	113
1981	4 upper	121	343
2382	5 upper	460	369
2532	4 lower	388	282
2533	5 upper	280	322
2534	6 lower	261	197
2535	6 lower	170	142

dose evaluated on the basis of a single EPR measurement within the range 120–460 mGy varies 46–280%. These values are significantly larger than the standard deviations shown in Table 5. The value of uncertainty depends on the quality of the spectrum and on the mass of sample.

The new methodological approach described in this section was applied to the results of EPR measurements of teeth obtained from donors who lived in the non-contaminated areas of the Urals region. These data were used for the estimation of background-EPR-dose levels described in the next section.

5. ESTIMATION OF BACKGROUND-EPR DOSES

Accurate knowledge of the background-radiation dose is a critical element of EPR studies of exposed populations. The radiation-background dose for an individual consists of several components, including external contributions from natural radioactive background (terrestrial and cosmic), medical diagnostics, and an internal contribution from natural radionuclides accumulated in calcified tissues. Moreover, some portion of "background dose" measured by the EPR method is attributable to an intrinsic signal (which can be a reflection of systematic error of EPR-spectrum analysis or, probably, addition of a non-radiation signal). The amplitude of the intrinsic signal for the GSF EPR-tooth-dosimetry system was evaluated on the basis of investigation of deciduous teeth from one donor as equivalent to 60 mGy (Appendix 1). It has been assumed that this bias is inherent to the GSF EPR-measurement system, and this value should be subtracted from all GSF EPR measurements in order to extract the radiation-induced component of the EPR signal. Investigation of an intrinsic signal was not performed at the IMP laboratory in which permanent teeth (having low levels of background-radiation dose) were used to receive a universal calibration curve (Appendix 1). Nevertheless, taking into account the similarity of EPR techniques used in both laboratories, we can assume that the same (or very close) bias is inherent to the IMP EPR-measurement system. However, the nature of the intrinsic dose component is not yet clear.

There have been a number of studies of EPR-background dose in different geographical regions (Romanyukha et al. 1999; Ivannikov et al. 2000). The studies clearly demonstrate that the dose rate from natural background radiation varies widely among different geographic regions. For example, the estimate for the annual (external) dose rate of Kaluga Oblast in Russia is about 1 mGy per year (Ivannikov et al. 2000). This is reasonable, because geographic factors can influence background-dose levels due to natural radioactivity, radon, and global radioactive fallout. Within a population, individual-dose variation takes place due to biological variety. Moreover, differences in background doses measured on anterior and posterior teeth were detected by Ivannikov et al. (1997; 2000).

Significant dependence of the level of EPR signal on tooth position has been found as a result of the induction of paramagnetic centers in front teeth by the UV component of solar light. Solar light induces in the buccal enamel of front teeth an EPR signal with the same properties as the radiation sensitive signal; on average the doses absorbed in front teeth (positions 1–3) are found to be about 200 mGy higher than for inner teeth (positions 1–8). The depth of the exponential distribution of the solar induced paramagnetic centers was determined to be about 300 μm. Such a thick enamel layer cannot be removed by surface etching. Therefore, it was recommended at the end of the 1990s to use the enamel of back teeth or the enamel from the inner side of the front teeth for the purposes of dose reconstruction.

However, before this recommendation was made, EPR measurements for the Techa River residents had been provided using total enamel from incisors. Also, in some cases the mass of the inner side of the front teeth is too small for evaluation of the EPR signal. It should be noted here that teeth from donors who lived in the upper reaches of the Techa River represent the best opportunity for validation of external dose, as such teeth would have a higher proportional signal from extra external exposure. As discussed in Degteva et al. (1997), it is very difficult to collect samples from this most important group of persons, because they were evacuated from their initial places of residence. Therefore, all teeth available from this important group of persons should be measured, and appropriate background levels (including the enamel from inner and outer sides as well as samples of total enamel of incisors) must be evaluated.

This section is devoted to the results of investigation of teeth that were received from non-exposed donors from the Urals rural population (background teeth). The main purpose is evaluation of average background-EPR dose for anterior and posterior teeth of persons within an age range similar to the range in age for members of the ETRC. In this report we apply the term “EPR dose” to indicate EPR signal in total (without the separation of its components related and not related to ionizing irradiation). This is reasonable, because the radiation dose absorbed in tooth enamel of the persons exposed due to their residence on the Techa River is evaluated as the difference between their EPR dose and the average background-EPR dose for respective types of tooth samples. Because the background EPR dose contains the same contribution of non-radiation sources, such subtraction allows us to avoid the problem of separate evaluation of the intrinsic signal and the contribution from ultraviolet light.

For the estimation of background-EPR dose, two approaches have been used. The first is mathematical averaging of individual measurements. This approach was applied

to first molars. This kind of tooth (position 6 in the denture) has maximal mass of enamel in comparison with teeth from other positions. So, the majority of first molar samples have enough mass of enamel to perform EPR measurements on an individual basis. It was decided to try to investigate individual variability in background levels using this group of teeth.

The second approach is physical averaging (using mixtures of enamel of teeth from several donors). Such an approach does not allow investigation of the variability of individual levels, but avoids limitations in mass for individual teeth. Such physical average measurements have been done for second premolars and different fractions of incisor enamel (lingual, buccal and whole enamel).

In order to increase the reliability of results, samples measured in two laboratories (GSF and IMP) were used predominantly. The absence of a systematic shift between the measurements from these two laboratories (demonstrated in our Milestone 2 Report [Shishkina et al., 2001]) allows consideration of the results obtained in these two laboratories as equivalent measurements.

5.1. Evaluation of background dose for posterior teeth

Two kinds of posterior teeth (1st molars and 2nd premolars) have been used in the current investigation. Mathematical averaging of individual measurements was used for the 1st molars, and physical averaging (mixes of enamel) was used for the 2nd premolars.

5.1.1. Estimation of background dose for 1st molars

Estimates of background doses for first molars (position 6 in the denture) have been provided based on analyses of measurements made using visual criteria of sample selection. Fig. 18 demonstrates the cumulative probabilities of experimental results of background-dose distributions obtained at the IMP and the GSF. Statistical parameters of these distributions are shown in Table 7.

Table 7. Statistical parameters of background-dose distribution for 1st molars obtained in two laboratories.

Parameter	IMP value, mGy	GSF value, mGy
Mean	191	143
Standard deviation	81	114
Median	171	121
Minimum	68	9
Maximum	470	542
5th percentile	83.5	19
95th percentile	340	333

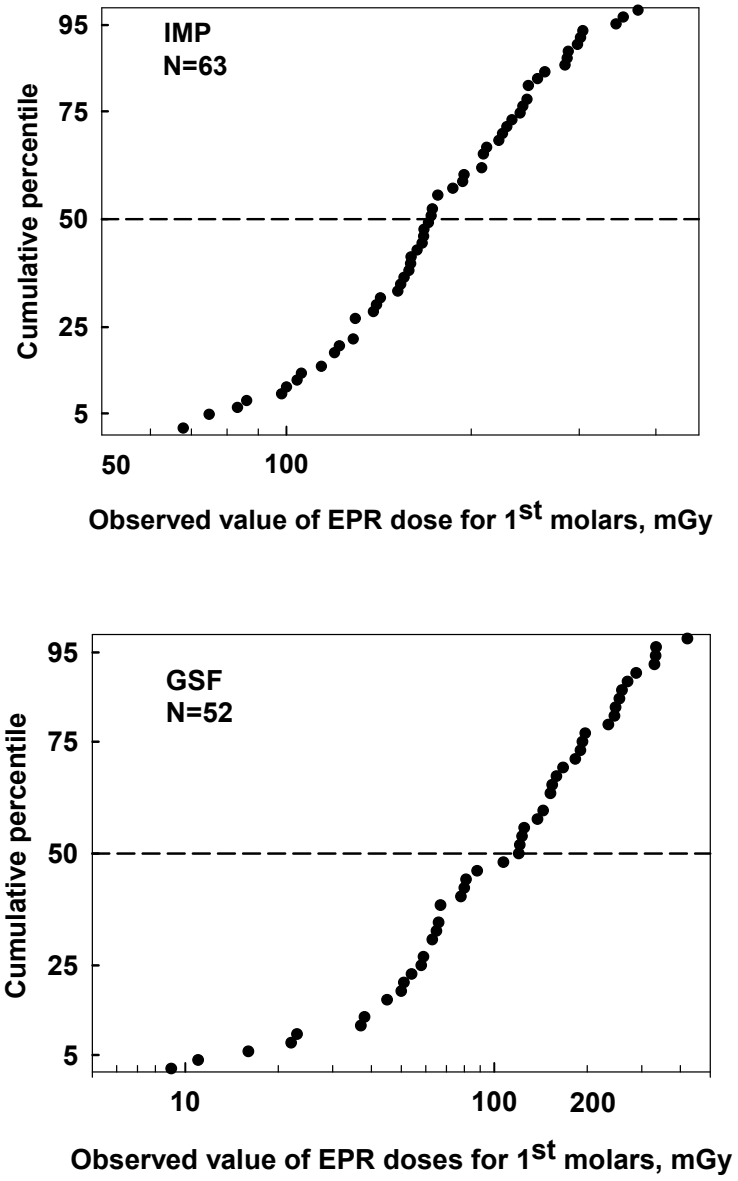

Fig. 18. The distributions of background doses measured in 1st molars. Upper panel: IMP results; Lower panel: GSF results (for both laboratories the results shown include the intrinsic signal).

The average value obtained from the IMP results is slightly higher than the average for the GSF results. The difference can be explained by the significant uncertainty of the EPR method in the low dose range. An attempt to estimate age dependence for background doses is shown in Fig. 19. Because all measured samples were of the 6th position, the tooth age was approximately equal to the age of donor.

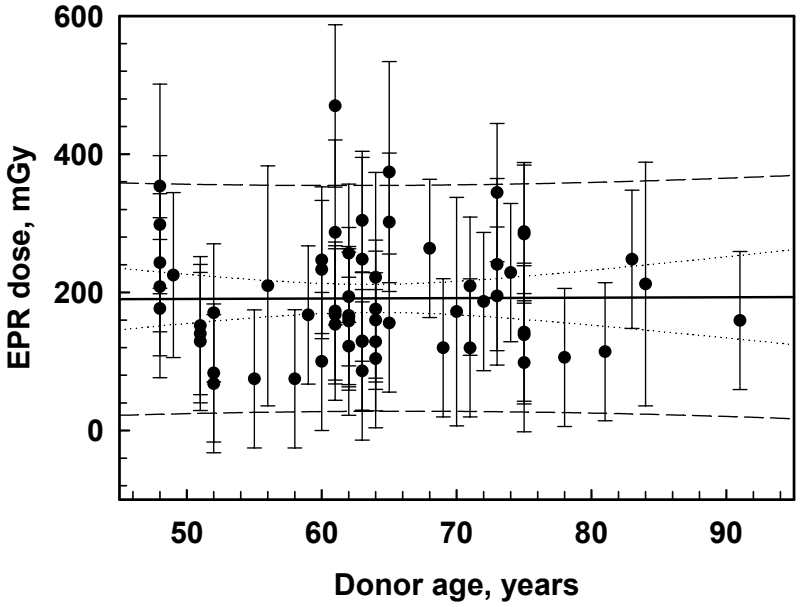

Fig. 19. Age dependence of background doses measured in 1st molars. The solid line is the line of linear regression. The dotted lines are 95% confidence intervals of the regression. The dashed lines are 95% prediction intervals of the regression corresponding to the 95% confidence intervals of the deviation among individual EPR results.

It is obvious from Fig. 19 that there is no apparent age dependence for the donors' age range from 45 to 95 years (1910–1953 birth years). This is perhaps due to the scatter in the data, which reflects uncertainties contributed by individual variability in dose and also by the measurement process itself.

5.1.2. Estimation of average-background dose for 2nd premolars

In as much as an age dependence of background doses based on individual measurements was not detected, we have tried to use another approach that minimizes the uncertainty contributed by the variability of individual-background doses. For this purpose, EPR measurements were performed using mixtures of enamel of 2nd premolars (position 5 in the denture) obtained from different donors. The mixtures were prepared for teeth of donors subdivided into five age groups. Each mixture was subdivided into a number of portions having masses optimal for EPR measurements. A description of the groups and the measurement results are shown in Table 8.

As can be seen from Table 8, we have used mixtures of enamel according to five age groups. Within each age group the mixed enamel was divided to a number of portions, which were measured, and the results were averaged (the number of portions was determined by total mass of mixed enamel). Distributions of dose in each age group were normal with standard deviations approximately half of those for "individual data" (Table 7).

Table 8. EPR measurement results for 2nd premolars (position 5) teeth using mixtures of enamel separated according to age groups.

Group number	Birth-date range	Number of mixed teeth in the group	Number of portions	Average birth year for group	Weighted average dose and std. dev, mGy
1	1918–1925	17	20	1922±3	182±42
2	1926–1935	38	50	1930±3	115±42
3	1936–1940	19	16	1938±1	142±48
4	1941–1949	32	26	1945±3	110±26
5	1950–1953	22	25	1952±1	117±31

Fig. 20 is a demonstration of the age dependence of the weighted average doses. As can be seen, there is a tendency for the background dose to increase with the age of the donors (correlation coefficient is equal to 0.72 with the probability $p = 0.1$). The average dose estimated for all groups is 133±30 mGy.

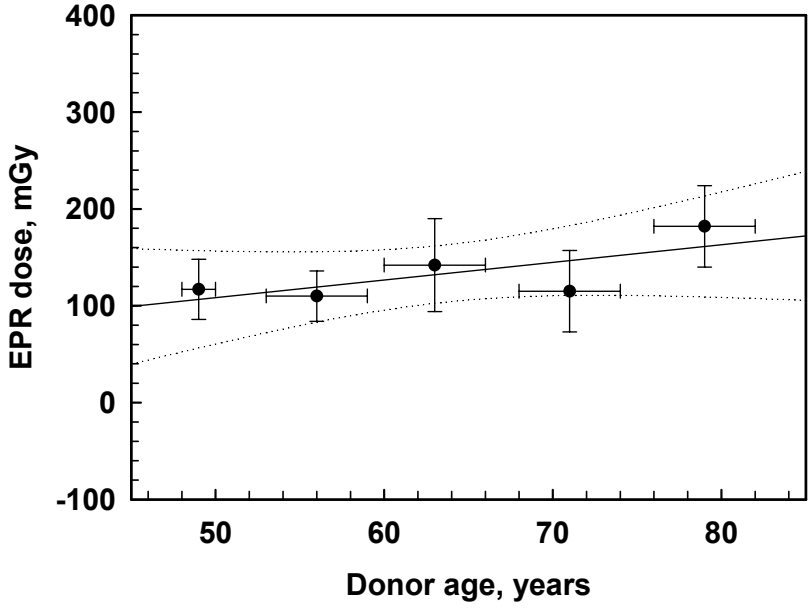

Fig. 20. Age dependence of background EPR dose for 2nd premolars (5th tooth position). Horizontal bars are standard deviations of the average age in each group. Vertical bars represent standard deviations of the average dose in each group. The regression is shown as solid line and 95% confidence intervals are indicated by dashed lines.

5.2. Evaluation of background dose for anterior teeth

The following factors complicate individual-dose reconstruction using anterior (front) teeth:

– ultraviolet radiation and

– small mass of enamel.

Ultraviolet irradiation (UV) can make a significant contribution to the radiation-induced EPR signal. This fact was discovered when significant deviations in doses reconstructed using incisors of exposed people were found (Ivannikov et al. 1997). Experiments with tooth enamel exposed to UV confirmed the hypothesis of creation of free radicals by solar light (Ivannikov et al. 1997). To take into account the contribution of UV radiation, the enamel of incisors was separated into two parts: front (lingual) part and back (inner or buccal) part. "Dose" absorbed in the front part of the enamel includes a contribution by UV radiation. Dose in the back part does not include this contribution, but, unfortunately, the mass of the inner part of incisor enamel is relatively small, which makes dose reconstruction on the basis of individual-tooth samples difficult and sometimes impossible. Therefore, mixtures of enamel obtained for lingual and buccal parts of incisors were used for the evaluation of background dose.

5.2.1. Lingual fraction of incisor enamel

Mixtures of enamel from 20 teeth divided into four age groups (five teeth in each group), and enamel of seven teeth mixed without age separation, were used. A description of the groups and measurement results are given in Table 9.

In contrast to the similar test for 2nd premolars, the number of portions for each group was very limited due to the small mass of enamel of each incisor. The uncertainties of weighted average dose in the groups are higher in comparison with the results obtained for posterior teeth (Table 8). The results obtained at the IMP and the GSF are comparable to each other. An age dependence of background doses for lingual enamel of incisors is not found (Fig. 21). The average value of dose estimated for all groups of data is equal to 182±48 mGy.

Table 9. EPR measurement results for incisor teeth using lingual mixes of enamel.

Group number	Age range	Number of mixed teeth in the group	Number of portions	Average age in group	Weighted average dose in group, mGy	
					GSF	IMP
1	1926–1929	5	1	1928±1	179±100	156±127
2	1932–1939	5	1	1937±3	117±100	194±100
3	1943–1947	5	1	1945±2	115±100	215±100
4	1951–1955	5	1	1952±2	115±180	174±100
5	1928–1952	7	4	1942±11	-	228±100

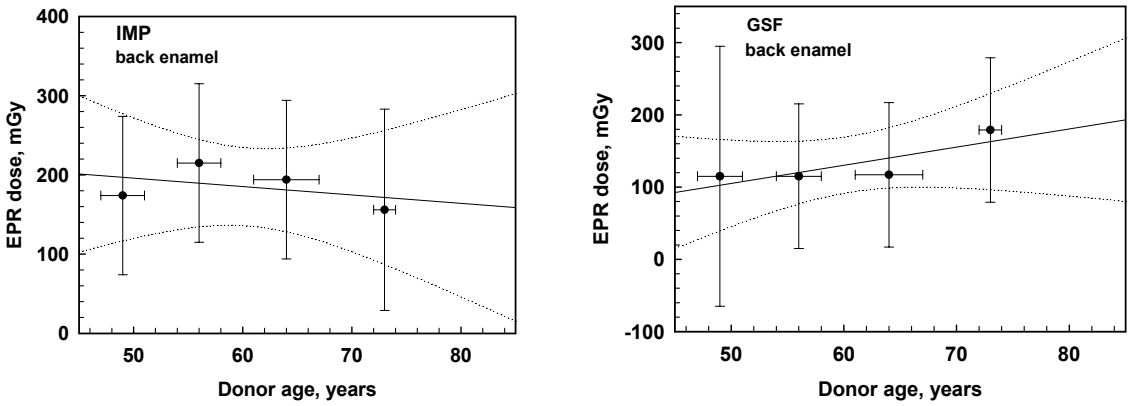

Fig. 21. Age dependence of background EPR dose for lingual enamel of incisors. Horizontal bars are standard deviation of average age in group. Vertical bars represent standard deviations of average dose in group. Regressions are shown as solid lines and 95% confidence intervals are indicated by dashed lines.

5.2.2. Buccal fraction of incisor enamel

The same 27 incisors as above divided according to the same age groups were used for the investigation of background dose for the buccal fraction of enamel. A description of the groups and measurement results are demonstrated in Table 10. Age dependence of background doses for buccal enamel of incisors is also not found (Fig. 22).

As can be seen, the situation for the measurement of the buccal fraction of tooth enamel is similar to that with the lingual fraction, but the levels of dose are relatively higher. The average dose value estimated for all groups of data is equal to 294±79 mGy.

Table 10. EPR measurement results for mixtures of buccal enamel from incisors.

Group number	Birth year range	Number of mixed teeth in the group	Number of portions	Average birth year in group	Weighted average dose in groups, mGy	
					GSF	IMP
1	1926–1929	5	2	1928±1	164±112	181±100
2	1932–1939	5	2	1937±3	361±111	452±124
3	1943–1947	5	1	1945±2	162±100	187±100
4	1951–1955	5	2	1952±2	245±136	243±114
5	1928–1952	7	5	1942±11	-	383±100

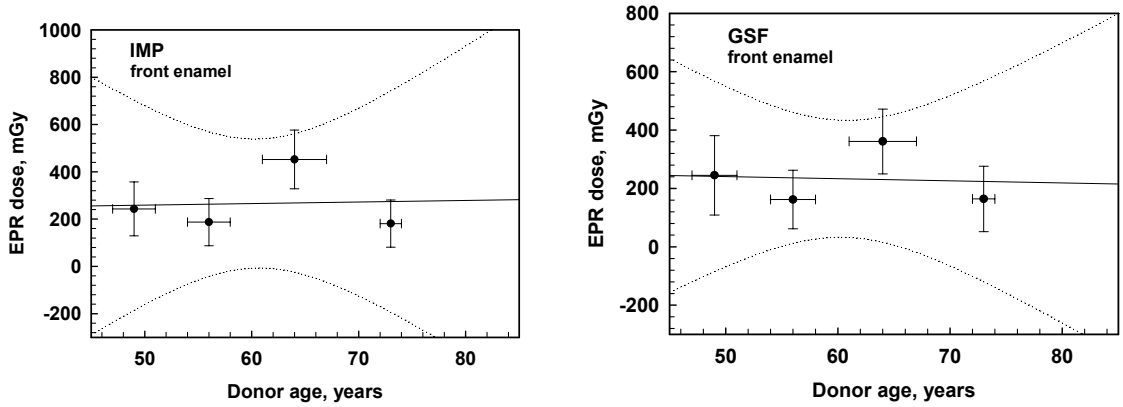

Fig. 22. Age dependence of background-EPR dose for the buccal enamel layer from incisors. Horizontal bars are standard deviations of average age in group. Vertical bars represent standard deviations of average dose in group. Regressions are shown as solid lines and 95% confidence intervals are indicated by dashed lines.

5.2.3. Combined enamel of incisors

After performing EPR measurements, the lingual and buccal fractions of enamel for the same 27 incisors (divided to the same age groups) were mixed together. A description of the groups and measurement results are demonstrated in Table 11. The age dependence of background doses for total enamel of incisors is demonstrated in Fig. 23.

Table 11. EPR measurement results for incisors using combined mixes from buccal and lingual samples of enamel.

Group number	Birth year range	Number of mixed teeth in the group	Number of portions	Average birth date in group	Weighted average dose in groups, mGy	
					GSF	IMP
1	1926–1929	5	2	1928±1	217±100	221±100
2	1932–1939	5	2	1937±3	179±100	198±100
3	1943–1947	5	1	1945±2	154±100	168±100
4	1951–1955	5	2	1952±2	164±125	152±100
5	1928–1952	7	9	1942±11	-	277±100

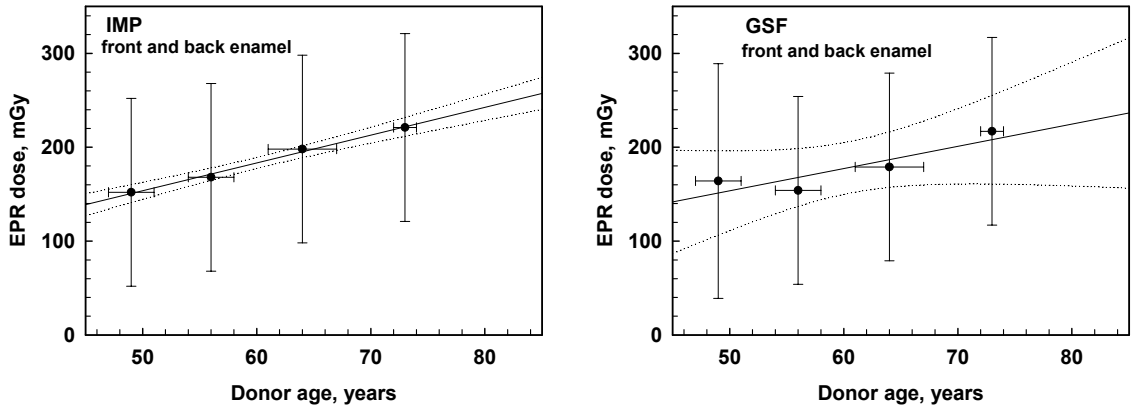

Fig. 23. Age dependence of background-EPR dose for combined enamel of incisors. Horizontal bars are standard deviations of average age in group. Vertical bars represent standard deviations of average dose in group. Regressions are shown as solid lines and 95% confidence intervals are indicated by dashed lines.

As expected, the levels of dose for combined enamel are intermediate between the results obtained for the lingual and buccal portions. An interesting fact is that the data for the combined incisor-enamel samples show a dependence of background dose with age of donors (Fig. 23). Nevertheless, because of the absence of age-dependences for the other samples, this age-dependence is not taken into account in the current investigation. The average dose value estimated for all groups of data is equal to 214±55 mGy.

5.3. Comparison among background doses for different tooth positions

The average-background dose estimated for 1st molars on the basis of combined GSF and IMP data is equal to 167±34 mGy. This is not significantly different from the average dose for 2nd premolars evaluated as 133±30 mGy. Because a statistically confirmed age dependence was not found for both kinds of teeth, we can use in our further investigations the average value for 1st molars and 2nd premolars as a nominal level of background dose for posterior teeth. This level is equal to 150±24 mGy. It should be noted that this estimate is close to a value of 140±50 mGy, which is the average EPR-background dose for posterior teeth of persons aged 56–70 years residing in the Kaluga Oblast in western Russia (Skvortsov et al. 2000).

For anterior teeth, nominal levels of background dose can be estimated on the basis of average values obtained for incisors as 182±48 mGy for the lingual fraction of enamel; 294±79 mGy for the buccal fraction of enamel; and 214±55 mGy for total-incisor enamel. It should be noted that the value for the lingual (inner) part of incisor enamel is slightly higher than the value for posterior teeth. This could be explained by the difference in the crystalline size and orientation within enamel of molar and incisor teeth (such properties of enamel can influence sensitivity to ionizing radiation). Also, it should be noted that our estimate for total-incisor enamel is slightly lower than the analogous estimate by Skvortsov et al. (2000) who received 298±50 mGy for the Kaluga residents of ages 61–70 years. Nevertheless, these estimates are comparable to each other.

6. ^{90}SR CONCENTRATION IN TOOTH TISSUES BY TL-CONTACT BETA DETECTION

As discussed above, the task of validation of external dose for the Techa River residents consists of several subtasks. These subtasks are EPR measurements of cumulative absorbed dose in the tooth enamel of donors from the Techa River region, EPR measurements of tooth enamel of unexposed donors (background-level determination), and evaluation of the effect of incorporated radionuclides (internal component of the dose to enamel) (Shishkina et al. 2002). Obviously, in the Urals region where ^{90}Sr was the main dose-producing radionuclide, the validation of external dose is impossible without subtraction of the internal dose component.

There are several existing methods that can be used for the investigation of ^{90}Sr content and distribution in tooth tissues. These include radiochemistry; thermoluminescent (TL)-contact detection; autoradiography; and photostimulated luminescence (PSL). The possibilities of these methods and the advantages/drawbacks of their use for reconstructing the internal component of dose in tooth tissues were discussed in our Unscheduled Report (Shishkina et al. June 2002).

The most widely known methods of measuring the concentration of radionuclides, such as radiochemistry or beta counting, have disadvantages for our current investigation. Radiochemical analysis is destructive of the investigated samples. The method of beta counting is not destructive; however, the masses of our samples are typically too small for reliable estimates of concentration. In contrast, TL-contact-beta detection is nondestructive and reliable (Göksu et al. 2002). Moreover, this method is relatively simple and can be applied for routine measurements.

Because measuring ^{90}Sr concentrations in individual-tooth tissues is a significant problem, an important element of our current investigations of tooth internal dose has been the determination of the concentration of ^{90}Sr in tooth tissues using this new method of contact-TL-beta counting. TL-beta detection of ^{90}Sr concentration was proposed at GSF (Göksu et al. 2002).

The TL-contact-detector method was considered as very promising, but this method was not calibrated at the time of our Unscheduled Report. In this section we describe this method of calibration. An approach for calibration of another promising method, the PSL method (also called the Fuji-plate method), is described in Appendix 2.

In order to use the TL-contact method, it was necessary to provide a calibration and a mass correction (taking into account the mass of the sample). Monte Carlo simulations supported the experiment. The following tasks were performed:

1. Verification of model computations using a calibration source;
2. Model estimation of TL response as a function of mass of contaminated sample;
3. Improvement of experimental conditions; and
4. Estimation of current ^{90}Sr concentration in tooth tissues for Techa River residents.

In order to verify the comparison of the calibration source and the model, the physical system and the model are first described.

6.1. Materials and methods used in thermoluminescence measurements and calibration of α-Al_2O_3:C detectors

Thermoluminescence measurements and individual calibrations of the detectors were carried out using two automated TL readers. Thermoluminescent emission was measured using 4 mm thick, blue transmitting, Corning C 7-59 and Chance-Pilkington heat absorption (HA3) glass filters with a Risø-TL-DA-10 reader, without the built-in calibration source (to avoid bremsstrahlung). The individual irradiations of the detectors for sensitivity corrections were performed in a Risø-TL-DA-12 with a built-in beta source of 555 MBq ^{90}Sr/ ^{90}Y. Each detector was irradiated automatically under this source (5 s) and removed immediately to the other equipment for detection of luminescence emission. The measured TL-emission intensities were further converted to absorbed radiation dose using the Secondary Standard Development Laboratory (SSDL) facilities at the GSF with a 74 MBq (^{90}Sr/ ^{90}Y) plaque source, where the absorbed dose rate in α-Al_2O_3:C at the irradiation position was found to be 455 μGy s^{-1}.

Luminescence intensities were measured by heating the detectors to 400°C at a rate of 2°C s^{-1}; the TL-peak maximum obtained was around 200°C. The fresh α-Al_2O_3:C dosimeters were annealed at 900°C for 15 min to remove all residual charges in deep traps, and signal intensity was further stabilized by successive irradiation and heating to 400°C at a rate of 2°C s^{-1} five times before they were used for routine measurements.

6.2. Monte Carlo simulation of experimental conditions

The experimental conditions were reconstructed in the description of model geometry with extreme accuracy. Fig. 24 (a,b) demonstrates the sizes and shapes of models, where a) is an initial geometry corresponding to the experiment described in Göksu et al. (2002), and b) is a new geometry corresponding to more useful experimental conditions. In both cases, the tooth-tissue powder had a cylindrical shape inside the tissue-equivalent cavity. The composition assumed for the simulated media used in the calculations is given in Table 12.

The average values and range of mass densities for powder of enamel and dentin and for the detector layer that were taken into account are shown in the last row of Table 12.

A uniform, isotropic source of electron emission from ^{90}Sr/^{90}Y decay was assumed in the source-cavity cylinder. Thus, it was assumed that the radionuclide uniformly occupies the tooth-powder volume so that the emitted radiation must cross into the thin layer of α-Al_2O_3:C detector to deposit energy there. No other sources of radiation were taken into account.

The Monte Carlo calculations of transport of electrons and secondary photons were done with the MCNP code, version 4C2 (Briesmeister 2000). The transport physics takes into account in a rather accurate way the diffusion and slowing down of all radiations in the electron-photon cascade established in the media.

The correspondence of experimental and computational results was tested on the basis of the initial geometry (Fig. 24 a). For interpretation of the experiment described in Göksu et al. (2002), computations were performed for enamel, root and crown dentin using the initial geometry.

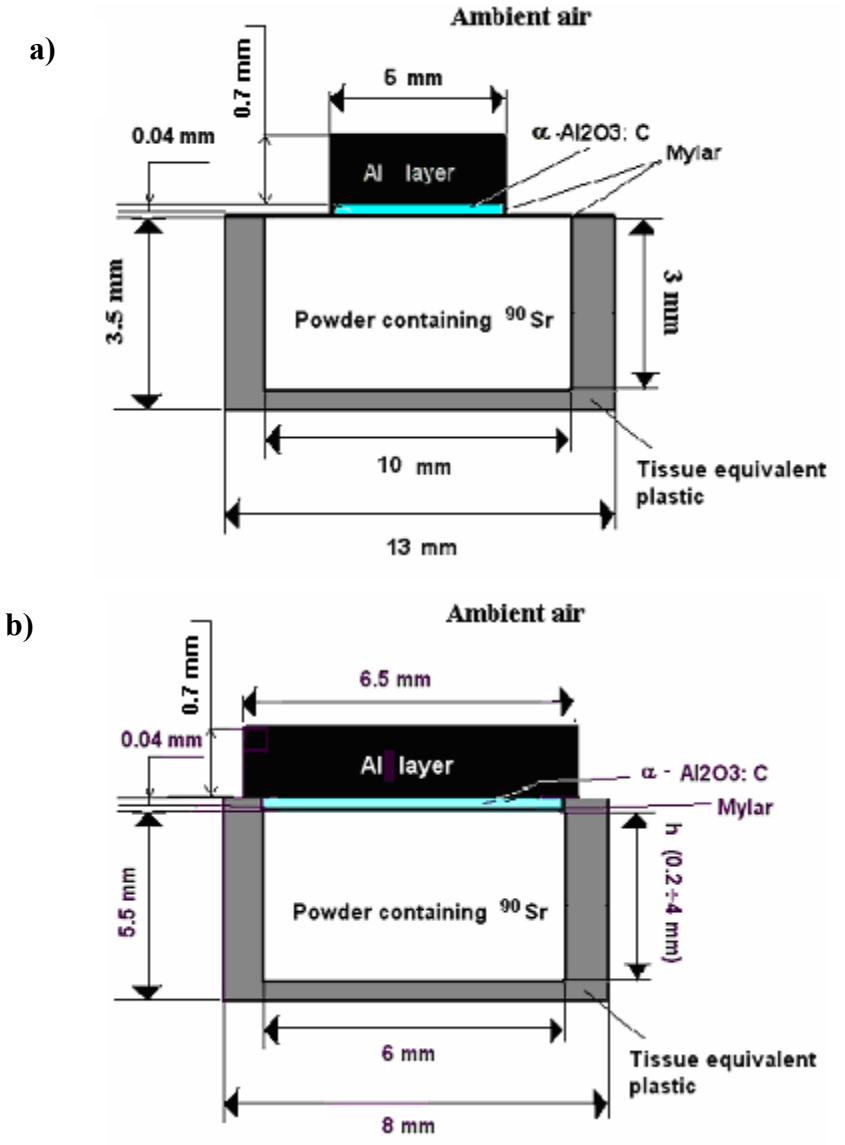

Fig. 24. Geometric description of simulation for detector calibration: a) initial geometry; b) new geometry.

Table 12. Chemical composition and density of simulated media.

Element	Atomic number	Fraction by weight			
		Dentin	Enamel	α-Al_2O_3:C	Mylar and tissue-equivalent plastic
H	1	0.0124	0.00195	0	0.101
C	6	0.0278	0.0168	0.042422	0.111
N	7	0	0	0	0.026
O	8	0.437	0.109	0.44799	0.762
F	9	0.00957	0.0002	0	0
Na	11	0.00738	0.0104	0	0
Mg	12	0.0105	0.00540	0.000034	0
Al	13	0	0	0.502941	0
Si	14	0.00000670	0.0000462	0.000368	0
P	15	0.162	0.276	0	0
Cl	17	0.000738	0.00545	0	0
K	19	0.000670	0.00462	0	0
Ca	20	0.331	0.570	0.001367	0
Ti	22	0	0	0.000566	0
Cr	24	0	0	0.000613	0
Fe	26	0.00000670	0.0000385	0.0032	0
Ni	28	0	0	0.000348	0
Cu	29	0	0.000154	0.000151	0
Zn	30	0.000172	0.000247	0	0
Density, ρ, g cm^{-3}		Crown dentin 1.1±0.07; Root dentin 1.01±0.07	1.41±0.07	0.875±0.375	Mylar 1.75 Tissue-equivalent plastic 0.975

Because the metal impurities of the simulated media amount to an insignificant weight fraction of the media (Table 12), the variation of chemical composition does not have any significant effect on the computational results. Therefore, the uncertainty of the computational calibration was estimated based on variation of model parameters, such as density of tooth-tissue powder and density of the detector layer.

For estimation of the powders' mass dependence of energy deposition in Al_2O_3, calculations were performed for 12 different powder-cylinder heights in the range from 0.2 to 4 mm (Fig. 24 b). All results are based on a sample of 40,000,000 histories of emitted beta particles.

6.3. Verification of model computations using a calibration source

The experimental situation and the modeled system are described in this section.

6.3.1. Experimental calibration results

A two-stage calibration is performed to convert dose in the detector to the concentration of ^{90}Sr in dentin. For the first stage, a ^{90}Sr-beta source from the Secondary Standard Laboratory of GSF (nominal activity of 84 mCi) is used to convert the TL readings to absorbed dose in the detector. Then dentin obtained from a tooth of an unexposed donor is used to prepare standard samples with a known concentration of added ^{90}Sr. These standard samples are measured with beta detectors in a geometry described in Fig. 24a to obtain the conversion factors from ^{90}Sr concentration to absorbed dose as measured in thin beta detectors.

The TL-detector reading is dose rate in the detector due to current radionuclide concentration. Comparison of source activity and TL reading for this experimental condition allows estimation of the conversion factor from dose rate in the detector to units of ^{90}Sr concentration. The experimentally estimated conversion factor is 2 (mGy y^{-1}) per (Bq g^{-1}) where the concentration (Bq g^{-1}) is in the units of ^{90}Sr activity.

6.3.2. Monte Carlo computation results

Beta emission was simulated for powders of enamel, crown dentin, and root dentin distributed uniformly in a cylinder of 10-mm diameter (Fig. 24a). The mass densities of tissue powder were assumed to correspond to average values of density. The results of the computation are shown in Table 13. All results are given in terms of absorbed dose rate per unit specific activity.

The uncertainty of the modeling result was estimated by variation of model parameters. The height of the source was fixed at 3 mm according to the conditions of the calibration experiment.

The variation of chemical composition was not found to have significant influence on the results. For example, for the same density, the enamel and dentin compositions (Table 12) resulted in a 0.7% difference in the calculated dose rate in the detector layer.

Due to the very thin detector, variation in the density of the detector in the range of 0.875±0.375 g cm^{-3} (according to Akselrod et al. 1990) reflected a variation in the dose rate of only about 3.5%. The computational relative error was not higher than 2.5%. Uncertainty in the density of tooth-powder density was estimated as 2% for enamel; 4% for crown dentin; and 6% for root dentin (Fig. 25). In total, uncertainty in the model results was estimated to be not greater than 7.5%.

Table 13. Computed results of dose rates in the α-Al_2O_3:C detector for initial geometry.

Source powder	Average source density, g cm^{-3}	Dose rate in the α-Al_2O_3 detector, mGy y^{-1} per Bq g^{-1}
Enamel	1.41	2.28±0.16
Crown dentin	1.10	2.06±0.14
Root dentin	1.01	2.04±0.14

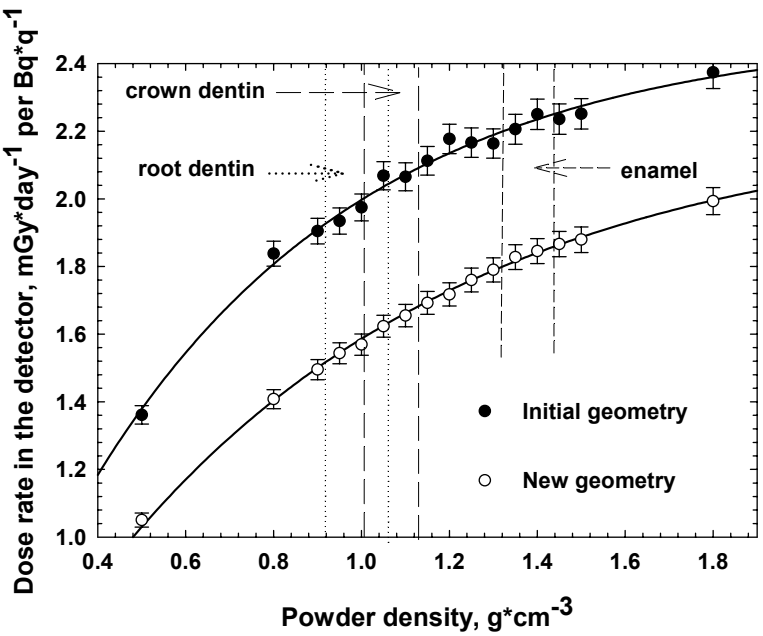

Fig. 25. Dose rate in the detector depending on variations in powder density (results of Monte Carlo simulation). The initial geometry corresponds to the experimental calibration situation. The new geometry corresponds to an actual experimental condition. The solid lines are regression lines of exponential approximation. The dotted lines indicate the range of variation in the density of root dentin. The lines with long dashes indicate the range of variation in the density of crown dentin. The lines with short dashes indicate the range of variation in the density of enamel.

Computational results are in complete agreement with experimental results. The difference among computed dose rates for different tissues is not significant and can be ignored. Detection of beta radiation is very sensitive to any variations in experimental geometry. A verified computational technique now allows the use of calculated conversion factors for future work with ^{90}Sr detection under other experimental conditions without the need for additional calibration experiments.

6.4. Improvement of experimental conditions and model estimation of the dependence of TL response on the mass of the contaminated sample

In practice, the limitation of actual masses of samples of tooth tissues made it impossible to fix the geometry of the exposure of the sample to the detector. Therefore, new smaller cavities were used for routine measurements (Fig. 24b). The advantage of the new cavities is in the lower likelihood of losing source-powder material and in the simple regulation of source-cavity height (which is critical, taking into account the amount of investigated material).

For the new geometry, the Monte Carlo-based calculational estimates of the dependence of TL response due to height of enamel, crown dentin, and root dentin were made. Fig. 26 illustrates the computational results. On the abscissa, the sample mass

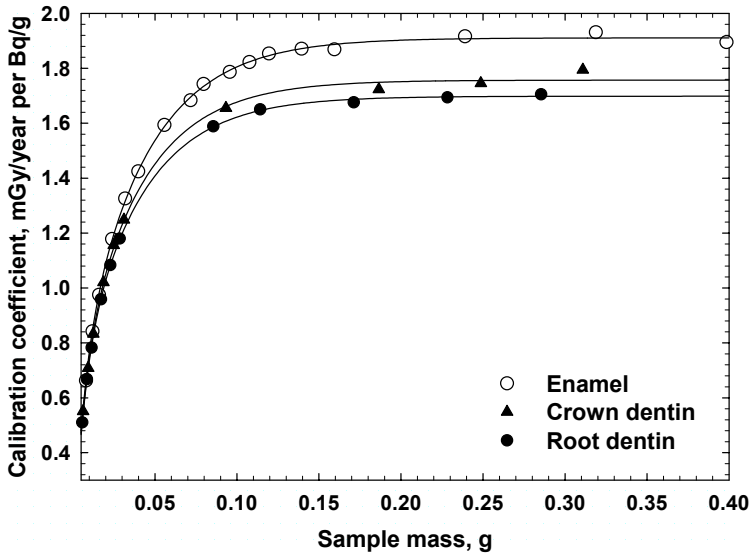

Fig. 26. The dependence of the calibration coefficient on sample mass in the new geometry. The symbols represent results of Monte Carlo simulations. The solid lines are fits to the data.

(which is proportional to sample height) is distinguished because the application of the computational results is more useful in terms of mass. On the ordinate axis, the dose rate in the TL detector is shown for 1 Bq g^{-1} of ^{90}Sr concentration; that is, the calibration coefficient depends on sample mass.

The fitted curves may be described by equations of exponentials with two terms. The calibration coefficients (k) can be formulated as following:

1. Enamel:

$$k_e = 0.59(1 - e^{-160m}) + 1.32(1 - e^{-25.3m}) \quad (5)$$

2. Crown dentin:

$$k_{cd} = 0.59(1 - e^{-160m}) + 1.17(1 - e^{-26.8m}) \quad (6)$$

3. Root dentin:

$$k_{rd} = 0.59(1 - e^{-160m}) + 1.11(1 - e^{-26.9m}) \quad (7)$$

where m is mass (g) of the sample that contains the ^{90}Sr being measured.

The first term is the same in all equations, because the source mass (~40 mg) is practically constant. For smaller masses (a very thin layer of radiation source <0.4 mm), the first term dominates. The value 0.4 mm corresponds to three average track lengths (mean free paths) for electron activity in the source media.

7. DISCUSSION

Concern about the reliability of the EPR method arose when a large scattering was found of doses measured on different teeth obtained from the same donor. Such scattering might be explained by different reasons, such as uncertainty in the EPR method, difference in background dose for different tooth positions, different ^{90}Sr concentrations in different teeth, or possible x-ray exposure of some teeth that was not recorded in dental records. The first three possibilities were investigated for the current report.

7.1. EPR methodology

Detailed investigation of uncertainties of the EPR method currently used at the GSF and the IMP for dose reconstruction for Urals residents was presented in Section 4 of this report. For the first time, the contributions to the total uncertainty due to operator arbitrariness, difference in equipment used in these two laboratories, and reproducibility errors were evaluated. Improvements in the spectrum-deconvolution procedure were suggested that permitted a significant reduction in the first component of total uncertainty.

Investigations have shown that conventional EPR dosimetry with old equipment does not allow for the accurate assessment of individual doses at the levels needed for members of the ETRC. Teeth from these persons are typically not in good condition from the standpoint of EPR spectroscopy, as the teeth have been extracted for reasons of dental health and are frequently carious and limited in mass; some teeth also contain metal impurities that may affect the "radiation-induced" signal (Shishkina et al. 2001; Shishkina et al. 2002).

As shown above, the uncertainties in the EPR method of measuring dose can be reduced further. In spite of spectra-deconvolution improvements that allow increased accuracy in dose estimation, current uncertainties of EPR dosimetry are too large for low dose levels. It is possible to obtain more precise dose estimates by increasing the ratio of signal to noise, which can increase the reproducibility of measurement results and reduce the detection limit. Fortunately, the sensitivity of EPR instruments has increased by one order of magnitude recently, due to new designs of the critical components [Bruker ELEXSYS EPR spectrometer (Maier 1997)]. Improvements in the methods of analysis can now also be used to describe uncertainty in the measurement results, and this correspondingly increases the reliability of EPR measurements. Thus, for future measurements, it is very important that laboratories be considered that have new equipment and that have access to improvements in all steps of the overall technology, including methods of signal extraction.

Scientists in charge of Project 1.1 have determined that the most qualified, available laboratory to undertake the required EPR-validation measurements is the EPR laboratory of the Instituto Superiore di Sanità (ISS) in Rome, Italy. Background information regarding the selection of the ISS in contained in Shishkina et al. (2002). The ISS has achieved excellent results in international intercomparison exercises, the ISS has the most modern spectrometer available, and the methods of EPR-spectrum deconvolution used at

the ISS have been recommended highly by specialists at the US National Institute of Standards and Technology (NIST).

A proposal entitled "Pilot study of the application of the electron paramagnetic method of reconstruction of low doses from the population living on the Techa River" has been submitted to the U.S. Department of Energy. This proposal would allow the involvement of ISS colleagues in a joint study aimed at the investigation of the feasibility and preparation of a detailed protocol for the EPR reconstruction of individual doses at low levels (<400–500 mGy).

7.2. Background-EPR dose

Evaluation of background-dose levels for the Urals population and of low level external doses for Techa River residents is a very important problem in the framework of validation of external dose for members of the ETRC. Background levels of EPR signals in enamel were investigated using 218 teeth (63 molars, 128 premolars and 27 incisors) obtained from donors lived in non-contaminated rural areas of the Urals region. Thirty-three percent of background samples were repeatedly measured both at the IMP and at the GSF. An age dependence was not found for background doses in the investigated age range of donors (birth years 1910–1953 corresponding to ages 45–95 years). Average-background doses were estimated for posterior teeth as 150±24 mGy. For anterior teeth, average values obtained for incisors were 182±48 mGy for the lingual fraction of enamel, 294±79 mGy for the buccal fraction of enamel, and 214±55 mGy for total (lingual and buccal portions combined) enamel. It should be noted that the contributions by UV radiation and the 'intrinsic signal' were not subtracted, and therefore these values should not be interpreted as 'a dose received due to exposure from background sources of ionizing radiation.' It is supposed that during the estimation of doses for Techa River residents the influence of non-radiation factors will be simply eliminated by subtraction of the total-background-EPR signal (without the separation of its components related and not related to ionizing irradiation).

The most detailed investigation of background doses was done by specialists of the Obninsk Medical Radiobiological Research Center (MRRC) for non-exposed donors of the Kaluga Oblast of Russia (Skvortsov et al. 2000). According to estimates by the MMRC workers, the average-background doses (evaluated without subtraction of 'intrinsic signal') for posterior teeth in the age range of 56–70 years is 140±50 mGy and for anterior teeth in the age range of 61–70 years is 298±50 mGy (measurements of whole enamel). Our results (obtained predominantly for mixes of enamel powder of similar teeth from different donors) described here are in good agreement with data from the workers at the MMRC. The "physical averaging approach" used here allowed reduction of the uncertainty contributed by the variability of individual-background levels. Nevertheless, future investigations using the most modern spectrometer and modern methods of EPR-spectrum deconvolution are strongly desirable to receive more precise evaluations of background-EPR levels and their individual variability for the Urals population.

7.3. ^{90}Sr concentration in tooth tissues

Evaluation of the enamel-dose component contributed by ^{90}Sr distributed in tooth tissues is also a very important task for the validation of external doses calculated with the TRDS-2000. The two luminescence methods (thermoluminescent-contact detection and photostimulated luminescence) were considered as very promising, but these methods had not been calibrated previously. In the current report, we have described a calibration for the thermoluminescent (TL)-contact-detection method. The experiment with the calibration source demonstrates good agreement with computational estimates of dose rate in the TL α-Al_2O_3:C detector. However, it should be noted that, taking into account the high importance of this task, this experiment is planned to be repeated with more accurate source preparations. Also, we have described an approach for calibration of the photostimulated luminescence (PSL) method (Appendix 2), which can give the volume distribution of ^{90}Sr concentration through dental tissues (Romanyukha et al 2002).

Based on estimates of ^{90}Sr specific activity in the tooth tissues of Techa River residents, it is possible to estimate the component due to internal dose in enamel. An example of such an evaluation can be done on the basis of preliminary investigations of the teeth from Techa River donors using the TL method.

Twenty-two samples of tooth enamel were measured using the improved experimental condition (Section 6.4). It was found that enamel of 21 teeth, which had been mature in the period of ^{90}Sr intake (1950–1956), gave TL response close to the background level, but one tooth with a high level of ^{90}Sr (in which the enamel was calcified in 1950), can be used for preliminary evaluation of ^{90}Sr content. The inverse value of the calibration coefficient evaluated on the basis of eqn (5) was used for the evaluation of ^{90}Sr+^{90}Y concentration. The absorbed dose due to ^{90}Sr was evaluated by assuming a single intake in 1950 and subsequent decrease of ^{90}Sr concentration in enamel due to radioactive decay. Table 15 demonstrates the results of evaluation of ^{90}Sr+^{90}Y concentration and integral dose calculated on the basis of TL measurements in comparison with the value of dose measured by the EPR method. As can be seen, the high value of EPR dose in this sample can be explained by high beta-exposure from ^{90}Sr incorporated in enamel. This result confirms the findings described by Tolstykh et al. (2003) that high levels of both ^{90}Sr concentration and absorbed dose are observed in tooth tissues, if the intake of ^{90}Sr occurs during the period of tooth calcification.

Table 15. Comparison of computed dose due to ^{90}Y/^{90}Sr in enamel with EPR dose for a tooth of a Techa River donor (IC 11317).

Donor IC	Year of birth	Tooth position	Concentration of ^{90}Sr+^{90}Y, Bq g^{-1}	Computed internal dose in enamel, Gy	EPR dose measured at GSF, Gy
11317	1947	5	96±18	13.5±2.5	13.96±0.11

It must be noted that the TL method is currently under further development. Moreover, additional work is now underway for more precise measurements of radionuclide concentration and estimation of TL background. Additional work is also being performed in order to calibrate and improve measurements of ^{90}Sr in tooth tissues by the method of photostimulated luminescence (PSL).

7. CONCLUSIONS

Currently, estimates of individual dose based on available methodology of EPR measurements is impossible in the dose range lower than 200 mGy. Only statistical analyses, that is, averaged measurements of many teeth, can be applied for this dose range. For the current method, it is possible to estimate that the total uncertainty for statistical analysis of low dose data from one laboratory (the same operator) as ±120 mGy and for the analysis of combined data from the IMP and the GSF as ±140 mGy.

New improvements in the analysis of uncertainty related to the deconvolution of the EPR signal related to estimates of individual dose allow for the consideration of measured doses in three groups:

<450 mGy with high uncertainties depending on spectrum quality. A normal distribution of EPR-amplitude probability is a typical result for this dose range. For each spectrum, the width of the distribution characterizing the uncertainty in the deconvolution must be estimated individually. In this dose range, the uncertainty in the deconvolution process is the main factor in total uncertainty.

450–700 mGy. A truncated normal distribution of EPR-amplitude probability is typical for this dose range. For each spectrum, the width of the distribution characterizing the uncertainty in the deconvolution must be estimated individually. In this dose range, the deconvolution uncertainty is a significant fraction of the total uncertainty.

>700 mGy. The uncertainty in the deconvolution process is insignificant. In this dose range, the uncertainty in reproducibility (due to signal anisotropy) is the main fraction of total uncertainty.

Average-background doses were estimated for posterior teeth as 150±24 mGy. For anterior teeth, average values were obtained for incisors of 182±48 mGy for the lingual fraction of enamel, 294±79 mGy for the buccal fraction of enamel, and 214±55 mGy for total enamel. An age dependence was not found for background doses for persons in the rural population of the Urals region in the investigated age range of donors (birth years 1910–1953 corresponding to ages 45–95 years).

Evaluation of the dose component in enamel contributed by ^{90}Sr distributed in tooth tissues is also a very important task for the validation of external doses calculated by the TRDS-2000. The thermoluminescent (TL)-contact detection and photostimulated luminescence (PSL) methods were considered as very promising, but these methods had not been calibrated previously. A new method of calibration for TL-contact-beta counting is presented for the evaluation of the concentration of ^{90}Sr in tooth tissues.

Further improvement of this method is currently underway to determine ^{90}Sr-activity levels corresponding to radionuclide retention in the teeth of Techa River residents for whom exposure was completed in 1950. An approach for the calibration of the PSL method (that allows mapping the distribution of ^{90}Sr in teeth) is described in an appendix.

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APPENDIX 1

DESCRIPTION OF EPR-DOSIMETRY METHODS USED AT THE GSF AND THE IMP

Sample preparation at the IMP

A chemical method is used at the IMP for the preparation of tooth-enamel samples for EPR measurements. This method is based on the treatment of teeth in a concentrated solution of alkali with use of an ultrasound bath. During this procedure, the organic base of dentin is removed, and the dentin is separated from the enamel and dissolves. Raising the temperature to 60^0 C speeds up the process.

A 5*M* solution of KOH was used. This alkali is more intensive than the traditionally used NaOH and results in better removal of dentin.

The procedure for preparation of enamel from molars includes the following steps:

1. Remove caries and fillings from the tooth with stomatologic equipment.
2. Separate the crown from the root with a diamond saw.
3. Wash in distilled water in an ultrasound bath for 5 min.
4. Treat in a 5*M* KOH solution in ultrasound bath at 60^oC for 15 hours.
5. Wash in distilled water in an ultrasound bath for 15 minutes three times.
6. Dry at 35^oC at room pressure.
7. Remove remaining dentin with a dissecting needle and/or low speed, hard alloy, rotary grinder.
8. Crush in a mortar, and select grains with size less than 0.25 mm.
9. Etch in 20% acetic acid for 5 min.
10. Wash three times in distilled water for 5 min.
11. Dry at 35^oC at room pressure.

Front teeth were prepared using the same method, but after Step 2 the crown was separated into front and back parts, which were treated separately.

Sample preparation at the GSF

Investigators at the GSF use a method similar to that used at the IMP, but they use NaOH instead of KOH. However, because a more powerful ultrasound bath is used at the GSF for sample preparation, use of NaOH gives good results.

The following steps are used:

1. Remove tooth fillings and indications of diseases (black spots) on the crown with a diamond drill. Cut off and halve the crown with a circular saw blade. For incisor and canine teeth only the lingual side is used for further treatment.

2. Wash the crown pieces in a test tube for 15 min with 0.1*M* Titriplex III solution (ca. 10 ml) in the ultrasound bath. Wash the crown pieces in a test tube for 5 min with water (ca. 10 ml) in the ultrasound bath.

3. Treat the crown pieces in a test tube with 5*M* NaOH solution (ca. 10 ml) at 40°C for 15 hours in an ultrasound bath.

   3.1 Wash the crown pieces three times for 15 min with fresh water (ca. 10 ml) in the ultrasound bath.

   3.2 If the water appears milky at the last washing, repeat step 3.1.

   3.3 Rinse the crown pieces in a test tube with ethanol (ca. 10 ml).

   3.4 Dry the crown pieces under vacuum for 30 min at 40°C.

   3.5 Remove any remaining dentin with a dissecting needle and/or low speed, hard alloy, rotary grinder.

4. Grind the crown pieces and select by sieving grains of size 125–600 μm.

5. Etch the enamel powder in a test tube for 5 min with 20% acetic acid (ca. 10 ml).

   5.1 Shake the test tube for five seconds in a mixer at the beginning and after three minutes of etching.

   5.2 Rinse the powder sample immediately in a test tube with water and shake the tube in a mixer for 5 s. Pour off the water 10 s after shaking. Repeat the rinsing three times.

6. Wash the powder sample in a test tube with ethanol (ca. 10 ml) for five minutes. Shake the test tube in a mixer for five s every minute. Before pouring off the ethanol, wait at least 10 s after the last shaking.

6. Dry the powder in the test tube under vacuum for 30 min at 40°C.

EPR measurements at the GSF and the IMP

A standard homodyne X-band electron spin resonance spectrometer ERS-231 produced in the former German Democratic Republic was used for measurements at the IMP. The spectrometer was connected to a PC/AT-486 computer to automate the spectrum-recording procedure and for computerized spectra processing.

A Bruker cavity with working mode TE_{102} was used. The sensitivity of the spectrometer was calibrated with a standard probe consisting of Mn^{2+} with ZnS. EPR measurements were conducted at room temperature. The following parameters were used at the IMP for spectra recording:

Magnetic field sweep:	10 mT
Magnetic field-modulation frequency:	100 kHz

Magnetic field-modulation amplitude:	0.45 mT
Gain:	100 000
Microwave power:	13 mW
Number of scans:	30

A standard Bruker ECS 106 spectrometer operating in the X-band was used for measurements at the GSF. EPR measurements were conducted at room temperature. The following parameters were used at the GSF for spectra recording:

Magnetic field sweep:	5 mT
Magnetic field-modulation frequency:	50 kHz
Magnetic field-modulation amplitude:	0.145 mT
Gain:	125 000
Microwave power:	25.3 mW
Number of scans:	40

According to both the IMP and the GSF protocols, the spectrum of each sample is measured three times. On the basis of these repeated records the average value and standard deviation of reconstructed dose are evaluated.

Spectrum analysis at the IMP and the GSF

Special software that utilizes a deconvolution procedure was developed for spectrum analysis at the IMP and the GSF (Fig. A1). The deconvolution method is based on computer-spectrum modeling using a set of Gaussian lines with fixed line widths and g-factors. Line amplitudes in this procedure are variables. The resulting line amplitude corresponding to the radiation-induced EPR signal and is used as the dose response.

Parameters of Gaussian lines were selected *a priori*. The one parameter dependent on the operator is the so-called g-factor (or magnetic field) shift, caused by fluctuations of circumstances (such as room temperature, humidity etc.) or instability of the spectrometer. The objective of the operator is to minimize differences between base and result spectra by selecting an appropriate g-factor shift value. For sufficiently large doses (0.7 Gy or higher), the selection is not difficult, but for the low dose range it is not easy to select this parameter. Different selected values result in differences in the deconvolution results, and therefore in values of reconstructed doses.

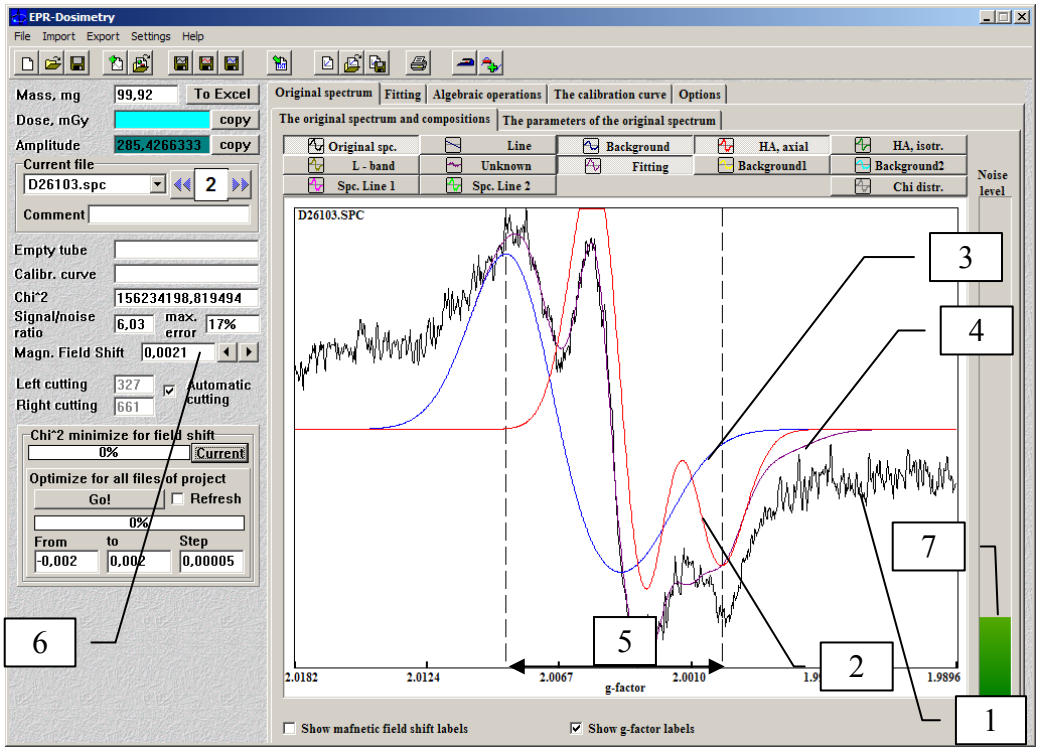

Fig. A1. Use of custom spectrum-deconvolution software for EPR measurements on tooth enamel. Numbers in boxes illustrate: (1) base EPR spectrum; (2) deconvolution result; (3) and (4) components of the deconvoluted spectrum (only some components are shown); (5) the spectrum region used for deconvolution; (6) g-factor shift value – the basic operator-dependent parameter; and (7) spectrum-quality scale (shows magnitude of noise at the wings of the spectrum relative to the amplitude of the radiation-induced signal).

Additional lines appearing in the spectrum due to sample impurities make the g-factor shift-selection procedure much more difficult for the operator. They are also significant in the low dose range, where the amplitude of the radiation-induced signal and the amplitudes from the impurity signals are comparable. It is difficult to perform spectrum deconvolution for such samples, and sometimes it is not possible.

The spectrum-deconvolution program is constantly being improved. So, a criterion of signal-to-noise ratio was suggested for estimation of spectrum quality. Noise amplitude is estimated at the wings of the spectrum and compared with the amplitude of the radiation-induced-signal. If this ratio is lower than unity, then the quality of the spectrum may be low.

Sample calibration at the GSF and the IMP

The primary method for sample calibration used at the GSF and the IMP is the universal calibration-curve method. The calibration curve is obtained by averaging the irradiation curves of several samples with low values of base absorbed dose. The main advantage of this method is speed compared with other methods. Moreover, in using this method, samples are not destroyed during the measurement procedure. The calibration procedure is included in the custom EPR spectrum-deconvolution software.

To receive a calibration curve it is necessary to irradiate enamel samples (having zero or at least a very low level of initial radiation dose) several times with known doses (using standard a source of ^{60}Co), and to measure the amplitudes of EPR signals before irradiation and after each cycle of irradiation. The dependence of EPR-signal amplitude on ^{60}Co-equivalent dose is named 'calibration curve.' The calibration curves obtained at the IMP and the GSF are shown in Fig. A2.

The calibration curve at the IMP was obtained using five enamel samples prepared from permanent teeth (first lower molars) of donors who lived in non-contaminated areas of the Urals region. The slopes of these lines correspond to the radiation sensitivity of enamel. The intercept of the line with the ordinate axis is determined by the sum of the 'intrinsic signal' and the dose of background radiation absorbed in permanent tooth. Both components can vary from sample to sample; therefore it is impossible to evaluate the 'intrinsic signal' from calibration curves obtained with permanent teeth. With consideration of this circumstance the resulting IMP calibration curve (violet line in

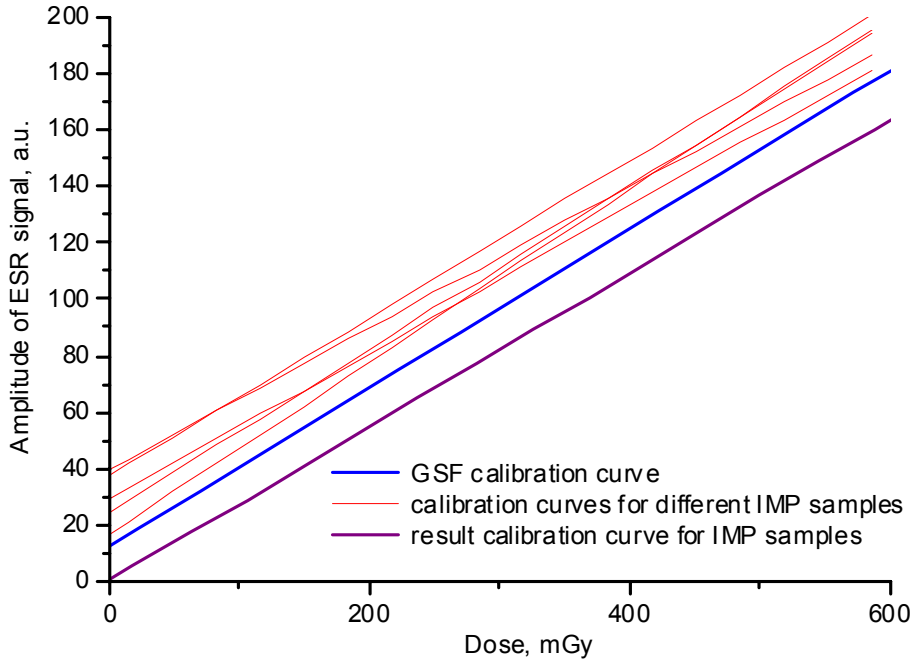

Fig. A2. Comparison of the GSF- and the IMP-calibration curves.

Fig. A2) was obtained by averaging the slopes of individual lines and assuming zero amplitude of the EPR signal at zero added irradiation dose.

The GSF-calibration curve (Fig. A2, blue line) was obtained by estimation of dose response for a deciduous tooth from a German donor. It has been assumed that the background-radiation dose in such sample is negligible, and the intercept of calibration curve can be associated with the 'intrinsic signal'. The intrinsic signal of the GSF tooth-dosimetry system was evaluated as equivalent to 60 mGy. Therefore, the GSF calibration curve allows the exclusion of the intrinsic signal from all EPR measurements, but, strictly speaking, this estimate reflects the bias inherent in the GSF-EPR-measurement system.

As can be seen from Fig. A2, the slopes of the IMP and GSF calibration curves are practically the same. For a more in-depth analysis of radiation sensitivity of the samples used in the IMP- and the GSF-calibration procedures, five additional background samples of first lower molar teeth prepared at the IMP were irradiated and measured at the GSF (red lines in Fig. A2). It easy to see (Fig. A2) that the lines have a similar slope, which indicates similar radiation sensitivities of the samples used for calibration.

Nevertheless, a distinction between the calibration procedures at the IMP and the GSF exists. It consists in subtraction of an intrinsic signal equal to 60 mGy for the GSF-calibration method (in the IMP-calibration procedure, the intrinsic signal is not subtracted). This difference can influence the estimation of background-radiation doses. However, it does not influence estimation of external doses from residents on the Techa River, because in the subtraction of the background dose from the total signal the difference due to value of the intrinsic signal is eliminated. However, in our investigation of the comparison of results for background dose, all measurements carried out at the GSF were taken without adjustment for the intrinsic signal.

APPENDIX 2

APPROACH FOR CALIBRATION OF THE PSL METHOD

Prepared jointly with Dr. S.M. Prigarin

**Institute of Computational Mathematics and Mathematical Geophysics,
Siberian Branch of Russian Academy of Sciences, Pr. Acad. Lavrentieva, 6,
Novosibirsk, 630090, Russia**

1. INTRODUCTION

Most physical experiments are performed to get a quantitative evaluation of some characteristics of a studied object. However, usually the experimental results, *w,* do not directly give the required quantity, *f*, but they present only some reflection of *f* :

$$Af = w \ . \tag{A1}$$

The operator *A* depends on the nature of the investigated process and the experimental conditions. With use of available measurement results it is necessary to make a conclusion about the value of *f*.

The quantitative evaluation of ^{90}Sr distribution in the tooth body using Fuji image plates is a pertinent example of the problem mentioned. Fuji plate mapping for ^{90}Sr detection was proposed by Romanyukha et al. (2002). The Fuji plate uses the method of Photo Stimulated Luminescence (PSL) to indicate the detection of beta radiation. Scanning is used to create an image that simulates a "photograph."

The imaging plate is a flexible image sensor in which bunches of very small crystals (grain size about 5 μm) of photostimulable phosphor of barium fluorobromide containing a trace amount of bivalent europium as a luminescence center, formulated as $BaFBr:Eu^{2+}$, are uniformly coated on a polyester support film.

The ions of Eu^{2+} lose electrons and transform to Eu^{3+} under the influence of ionizing radiation. Under the influence of light, the inverse process of Eu^{2+} recombination results in the emission of photons. The exposed imaging plate is scanned with a laser beam of red light by the plate being moved with high accuracy in a phosphor reader. The image resolution is 0.01 mm^2.

The scanned image is digitized and presented as a table of numbers. The unit of detected values is PSL (photo stimulated luminescence).

An analysis of preliminary investigations was made in Shishkina et al. (2002). It was shown that ^{90}Sr mapping in teeth with an imaging plate gives one the unique opportunity to create a fast, relatively inexpensive and very sensitive method for screening a population for possible ^{90}Sr intake. However, this method reveals a number of problems related to calibration and the interpretation of results. To solve the problems it is necessary to improve the technique and to refine the existing calibration.

The main conclusions made of the preliminary study and the analysis of the experience of investigators (Romanyukha et al. 2992) at the US National Institute of Standards and Technology (NIST) can be formulated as follows:

1. For such a sensitive method, which is able to detect a systematic shift of about 9%, it is important to use a much more accurate calibration than that proposed in Romanyukha et al. (2002). Moreover, it is necessary to take into account the shadow effect for thick samples.
2. In the halves of teeth that have been studied, ^{90}Sr is distributed non-uniformly both in the horizontal (plane of contact with the detector) and in the vertical (perpendicular to the tooth cross-section plane) directions. And the sample height itself in the vertical direction was not the same. This makes interpretation of the

results obtained with tooth halves impossible. It is preferable to use thin slices of tooth (~0.4–0.5 mm) for minimization of shadow effects and to achieve approximate homogeneity of the distribution of ^{90}Sr in the vertical direction.

3. The non-uniform distribution of ^{90}Sr in the tooth body and the high variability of geometric parameters influence the complicated superposition of shadows that is presented in the PSL image. It is not possible to determine a linear relationship of the PSL response in terms of the concentration of ^{90}Sr in the tooth. To solve this problem the use of electron-transport modeling is essential.
4. One more problem, also related to the high sensitivity of the film, is a widespread shadow reproducing the contours of strontium-containing fragments and hampering superposition of the real tooth contour onto the image. While the crown is close to symmetrical in the radial direction, this is not the case in the axial direction. Thus, a displacement of the contour with respect to the image can take place, and this can dramatically affect the description of the radionuclide distribution in the tooth.

Therefore, the problem of using the PSL plates for quantitative evaluation of ^{90}Sr distribution in the tooth body requires additional accurate experimental and theoretical investigations to test and calibrate the method. The aim of this document is to describe a joint working plan of research on calibration of the PSL method.

2. GENERAL TASK FORMULATION

Henceforward, we assume that the detector is a segment of photoluminescent plate with area equal to the image resolution (0.01 mm^2), and the source is a fragment of a thin slice of similar area.

The directional comparison of PSL response in the detector and ^{90}Sr concentration in the adjacent source (under experimental conditions) is not reasonable, because electron scattering results in significant influence of each source to a set of detectors. In other words, a detected image is a superposition of responses due to each of many sources.

Under the assumption that the detector response is proportional to cumulative absorbed energy, Monte Carlo methods can be used to calculate the energy absorption in the detector due to ^{90}Sr+^{90}Y disintegration spectra for different sources (by taking into account the distance from the source to the detector, density of emissive media, and density of the masking circle). The value of the absorbed energy from source i emission (a_i), estimated by Monte Carlo simulation, is proportional to operator A in eqn (A1), and it can be normalized with a conversion factor, η, from MeV to PSL units.

The conversion factor, η, is independent of type or character of exposure and depends only on the properties of the photostimulated plates. This results in the necessity of a fading correction $\eta(t)$.

Hence, the system of equations in eqn (A1) can be rewritten in the following way:

$$w = \eta(t) \sum_{i=1}^{n} a_i f_i \quad , \tag{A2}$$

where n is the number of sources.

The number of equations in system (A2) is redundant, because the number of detectors is less than the number of sources due to electron scattering causing an extensive shadow area around the sample outline.

According to the above, the solution of Fuji image-plate calibration must include the following steps:

1. evaluation of optimal experimental conditions;
2. accurate estimation of physical characteristics for the investigated detector (such as result reproducibility and fading range);
3. evaluation of uncertainty for detection of the PSL response;
4. estimation of energy absorption in the detector due to ^{90}Sr+^{90}Y disintegration spectra (a_i) for different sources and estimation of uncertainty;
5. estimation of the conversion factor from MeV to PSL units [$\eta(t)$] and its uncertainty; and
6. implementation of the inverse problem solution for eqn (A2) that includes verification of the filtering algorithm and estimation of uncertainty.

In other words, the quantitative assessment of radionuclide concentration using the PSL (Fuji plate) method is a nontrivial problem that requires multidisciplinary research including different experimental and theoretical methods of physics and mathematics.

3. EXPERIMENTAL WORK

3.1. Sampling

We propose to use molars from Techa River donors of different ages (different radionuclide localization) and some teeth from non-exposed donors (as a test of natural radionuclide detection and background-level estimation).

3.2. Sample preparation

Preparation of tooth samples includes two steps: mechanical cutting of the tooth using a diamond saw and hand polishing of the slices to reduce the thickness to 0.4–0.5 mm. It should be noted that the thin calcified material is very fragile and that the preparation of a single sample can require about two hours.

A standard sample (with uniform and well-known ^{90}Sr concentration) with regular shape will be prepared to estimate the conversion factor [$\eta(t)$] and to make a preliminary verification of the solution of eqn (A2).

3.3. Evaluation of optimal experimental conditions

The careful development of coordinate representation and merging of data for sources and detectors is very important. For this step we propose to use optical densitometry to describe the geometry of the tooth tissue. It is necessary to develop the

procedure for fixation of sample slices in the same position for densitometry and PSL measurements in order to minimize uncertainty in the merging of coordinate data.

3.4. Estimation of optimal time for exposure and estimation of physical characteristics for the investigated detector

We propose to use a prepared standard sample with uniform and well-known concentration of ^{90}Sr and of regular shape. For this case, the PSL response due to this sample exposure will have a radial distribution that decreases from the center to the boundary. Estimation of the variation in results for equal distances from the center will demonstrate the dependence of PSL response on measurement reproducibility. On the basis of this result, it will be possible to determine the detection limit for PSL response and the uncertainty of the method due to reproducibility.

3.5. Estimation of time dependence of PSL response for fading correction and measurements of tooth samples

Measurements of the background levels of natural radionuclides can be done using samples from non-exposed donors of the Urals. Then, the final step can be measurements of tooth samples from Techa River residents.

4. MONTE CARLO COMPUTATION

4.1. Modeling of experimental conditions for standard source exposure

Computation of the radial distribution of energy deposition in the detectors enables one to compare computational and experimental results and evaluate the conversion factor.

4.2. Modeling of experimental conditions for exposure of tooth slices

This task involves significant simplification and evaluation of the main factors influencing detector response from different sources of irradiation. As a result, the value of the transition operator (a_i) will be estimated. Variation of parameters related to geometry and density in the model will make it possible to evaluate the uncertainty of model computations.

5. MATHEMATICAL INTERPRETATION OF RESULTS

The inversion of eqn (A2) is a problem of the ill-posed type, as the experimental results and the computed transition operator (a_i) can be specified only approximately (with some uncertainty), and the errors of measurements are stochastic. Therefore, it is possible to evaluate only some pseudo-solution that can significantly differ from the true radionuclide concentration (i.e., there is an unstable solution of the factor of measurement reproducibility). This pseudo-solution can lose the physical sense that is intrinsic for the ideal case with no uncertainties.

Many methods are available to solve ill-posed problems of this kind (e.g., Tikhonov and Arsenin 1977). However, there is no universal or standard way and for each case the approach must be found individually. Additionally, a priori information

about solution properties, observed experimental data, and the transition operator must be taken into account to find a regular solution of the problem.

6. ANTICIPATED PECULIARITIES OF THE PROBLEM

Algorithms of image processing are very labor intensive and time consuming, because of the large dimensions of the data arrays. Thus, significant computational resources will be needed. Also, the observed images are very noisy. Therefore, it is vital to use nontrivial stochastic numerical methods for image processing.

Special methods developed for image processing and analysis can be applied (Pratt 1978; Winkler 1995) in this case, if more simple and traditional methods fail.

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