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Assessment of ^{90}Sr Concentration in Dental Tissue using Thin-Layer Beta-Particle Detectors and Verification with Numerical Calculations

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Electron paramagnetic resonance (EPR) measurements of tooth enamel can be used as an individual biological dosimeter for external dose assessment. However, the presence of ^{90}Sr in the tooth tissues makes the task of interpreting EPR tooth dosimetry more complicated. The determination of the dose contribution of incorporated ^{90}Sr in calcified tissue to the total dose measured by EPR is one of the main aspects of correct interpretation of EPR tooth dosimetry. In this work, experimental and numerical calculations were performed to convert the measured β-particle dose rate to ^{90}Sr concentration in calcified tissue. The cumulative β-particle dose was measured by exposing artificially contaminated dentin and enamel to thin-layer α-Al_2O_3:C detectors in two different exposure geometries. Numerical calculations were performed for experimental exposure conditions using calculations of electron transport and secondary photons [Monte Carlo n-Particle Transport code version 4C2 (MCNP)™]. Numerical calculations were performed to optimize the sample size and exposure geometry. The applicability of two different exposure conditions to be used in routine analysis was tested. Comparison of the computational and experimental results demonstrated very good agreement.

INTRODUCTION

Electron paramagnetic resonance (EPR) measurement of human teeth is a well-established technique for external γ-ray dose reconstruction for individuals after radiation accidents (*1–3*). However, radionuclides such as ^{90}Sr incorporated in calcified tissue cause additional complications in the interpretation of EPR external dose assessment.

The essence of the EPR technique lies in the assessment of the cumulative dose through the detection of long-lived radiation-induced free radicals in the hydroxyapatite, which is the main component of the calcified tissues (tooth, bone). The free radicals in calcified tissue may be produced from man-made sources such as medical, occupational and accidental exposures as well as from natural radionuclides. When radionuclides from man-made sources are incorporated in the calcified tissue, the internal dose must be subtracted from the cumulative dose assessed using EPR. This problem is of utmost importance for analyzing the effects of the Techa River radiation incident. The Techa River was contaminated from 1949 to 1956 as a result of releases of radioactive materials by the Mayak Production Association (MPA) (*4*). The river was the main source of drinking water for residents of many riverside communities located downstream from the release site. Strontium-90 was the main dose-contributing radionuclide for internal exposure of the Techa River residents; on average, the residents ingested about 1–3 MBq of this nuclide with river water and milk from 1950 to 1952. The biokinetic behavior of strontium and calcium is similar, as are their chemical characteristics. The incorporation of strontium isotopes in bones and teeth has been described in several studies[2] (*5*, *6*). Obviously, for the Urals region, where ^{90}Sr was the main dose-contributing radionuclide, the estimation of human external doses using EPR is impossible without subtraction of the internal dose component (*7*).

The internal dose component contributing to total dose measurements obtained using EPR of enamel can be estimated using current ^{90}Sr concentrations in various parts of dental tissue combined with Monte Carlo modeling of electron transport and biokinetic modeling of ^{90}Sr metabolism in the tooth (*6*, *7*). Therefore, determination of the current ^{90}Sr concentration in the tooth tissues is an important step when assessing of internal dose in tooth enamel.

It has been shown that thin-layer β-particle detectors (α-Al_2O_3:C) can be used to assess the current β-particle dose rate due to the ^{90}Sr concentration present in the tooth tissue (*8*, *9*). However, the conversion of the absorbed dose in the

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[2] I. M. Rasin, *Kinetics of ^{90}Sr retention and forming of tissue doses in growing organisms.* Unpublished Doctoral Thesis, Institute of Biophysics, Moscow University, 1970. [in Russian]

detectors measured for a given exposure geometry and size of sample to the ^{90}Sr concentration in calcified tissue requires the support of numerical calculations and experimental validation. It should also be noted that teeth extracted for medical indications are usually partially destroyed, so the mass of samples to be studied can be significantly smaller than normal. Therefore, there is a need to convert the doses obtained using thin-layer detectors to ^{90}Sr concentration in the tooth tissue such as enamel or dentin for an actual mass of the sample and the exposure geometry of the detectors.

The study was aimed primarily at developing a methodology for assessing the ^{90}Sr concentration in actual tooth samples collected from the Techa River population. Numerical calculations were performed to obtain conversion coefficients for various exposure geometry of the detectors and sample size containing various amounts of ^{90}Sr.

A comparison of experimental results and modeling predictions allowed the verification of the experimental results and the assessment of the uncertainty estimate of the ^{90}Sr concentration.

EXPERIMENTAL

Preparation of Artificially Contaminated Dentin and Enamel with a Known ^{90}Sr Concentration and Exposure Geometry of the Detectors

Twenty teeth from an Egyptian dental hospital that had been extracted for medical indications were used as ''zero internal dose'' material. The root dentin and enamel were separated from the tooth body and used as standard material with no ^{90}Sr content. After separation from the roots, the crowns were washed with 0.1 *M* Titriplex III solutions. All the pieces were then treated with 5 *M* NaOH solutions in an ultrasonic bath. The resultant root dentin and enamel grains were crushed. Grains having a size of between 125 and 600 μm were selected and etched with 20% acetic acid. A known quantity of the root dentin grains was immersed in standard ^{90}Sr solutions (10 Bq/ml) to obtain an activity concentration of 5, 12 and 25 Bq/g (for dry samples). Similarly, another known quantity of the enamel grains was immersed in ^{90}Sr solutions (200 Bq/ml) to obtain dry activity concentrations of 5, 10, 20, 40 and 80 Bq/g. The portion of enamel and dentin that was not soaked in standard ^{90}Sr solution was used to monitor the natural environmental background dose rate during the exposure of the samples to thin-layer β-particle TL detectors.

The amount of dental material obtained after chemical and mechanical separation is usually very small, especially from teeth extracted from the Techa River population since they are usually extracted for medical reasons and the donors are relatively old. Therefore, it was essential to optimize the sample size and exposure geometry of the detectors to register the full dose contribution from β-particle-emitting ^{90}Sr (*9, 10*). Two types of sample holder, labeled G1 and G2 in Fig. 1, were used to expose the samples to detectors during storage. The holders are made of tissue-equivalent plastic cylinders with diameters of 10 mm and 6 mm, respectively. A very thin layer of Mylar foil (d = 5.7 μm; surface density: 10 mg/cm^2) was used between the sample and detectors to shield the detectors from α particles emitted from natural radionuclides in dental tissue.

The measurements and numerical calculations were performed for containers filled with various amounts of dentin and enamel for geometries described above for various dental tissues.

Luminescence Measurements of α-Al_2O_3:C Detectors and Calibration

The luminescence properties of the α-Al_2O_3:C detectors developed by the Urals Research Center have been described (*11, 12*), and their applicability to the detection of the β-particle dose rate from natural radionuclides and ^{90}Sr in calcified tissue has been demonstrated by Göksu *et al.* (*8, 9*). Thin-layer α-Al_2O_3:C detectors were purchased from Landauer; their properties have been described in detail by Akselrod *et al.* (*12*). The detectors have a surface density of 2–5 mg/cm^2 that is centered on 0.7-mm-thick aluminum substrate with an inner diameter of 6 mm (*13*). Luminescence emission was recorded with an automatic RISØ-TL-DA-10 reader (without built-in source) using 4-mm-thick blue-transmitting, Corning C 7–59 and Chance-Pilkington heat absorption (HA3) glass optical filters. The individual irradiations of the detectors for sensitivity corrections were performed in a RISØ-TL-DA-12 reader using a built-in ^{90}Sr/^{90}Y β-particle source (555 MBq). Each detector was irradiated automatically under this source (5s) and was removed immediately to the other reader to avoid scattered radiation during measurements (*14*). The measured TL emission intensities were further calibrated after exposure to a ^{90}Sr/^{90}Y source at the Secondary Standard Laboratory facilities

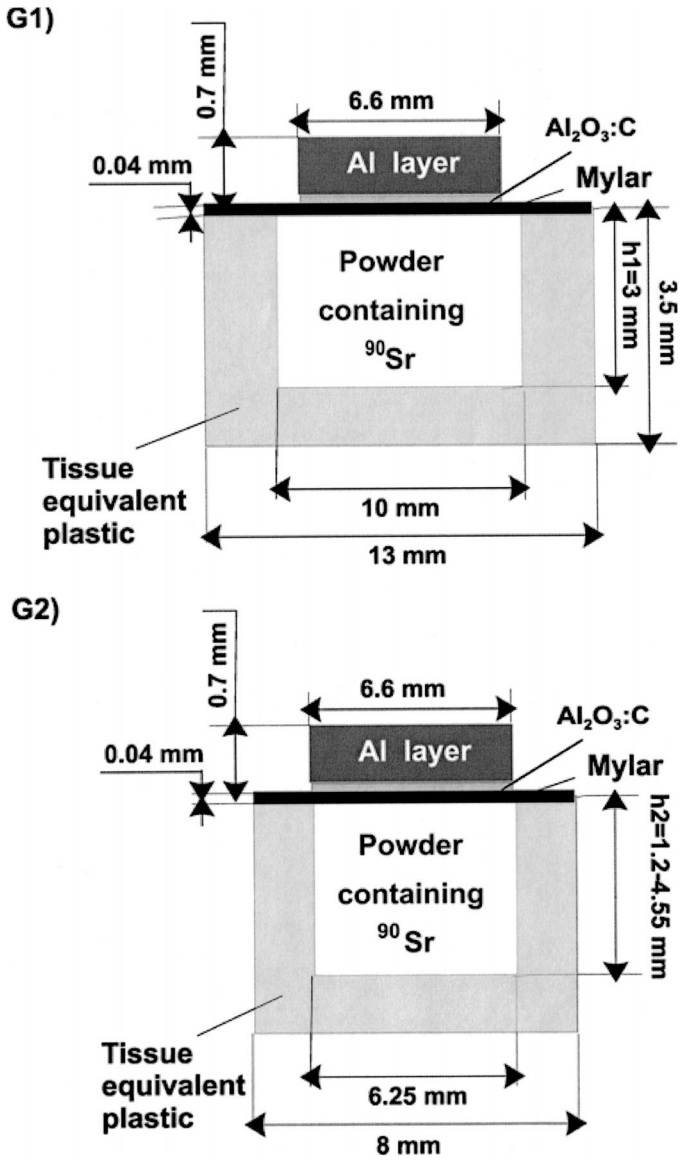

FIG. 1. The two exposure geometries (G1 and G2) of the detectors used for experiments and numerical calculations. The detectors are exposed to 200 mg of dental tissue placed in a tissue-equivalent cylinder-shaped container in G1, and the detectors are exposed to varying amounts of sample placed in a tissue-equivalent container by changing the height of the cylinder in G2.

TABLE 1
Chemical Composition and Density of Simulated Media (*6, 7*)

Element	Atomic number	Fraction by weight			
		Dentin	Enamel	α-Al_2O_3:C	Mylar and tissue-equivalent plastic
Hydrogen	1	0.0124	0.00195	0	0.101
Carbon	6	0.0278	0.0168	0.0424	0.111
Neon	7	0	0	0	0.026
Oxygen	8	0.437	0.109	0.448	0.762
Fluorine	9	0.00957	0.0002	0	0
Sodium	11	0.00738	0.0104	0	0
Magnesium	12	0.0105	0.00540	0.000034	0
Aluminum	13	0	0	0.50231	0
Silicon	14	0.00000670	0.0000462	0.000368	0
Phosphorus	15	0.162	0.276	0	0
Chlorine	17	0.000738	0.00545	0	0
Potassium	19	0.000670	0.00462	0	0
Calcium	20	0.331	0.570	0.00137	0
Titanium	22	0	0	0.000566	0
Chromium	24	0	0	0.000613	0
Iron	26	0.00000670	0.0000385	0.0032	0
Nickel	28	0	0	0.000348	0
Copper	29	0	0.000154	0.000151	0
Zinc	30	0.000172	0.000247	0	0

(SSDL) at the GSF using a 74 MBq ($^{90}Sr/^{90}Y$) plaque source, where in the dose in air was corrected for the ambient temperature and air pressure at the time of exposure. Prior to the calibration measurements, α-Al_2O_3:C (β-TLD) detectors were annealed at 400°C for 15 min to remove all the residual charges in deep traps, and the signal intensity was further stabilized by successive irradiation and heating to 400°C at a rate of 2°C/s five times (*8*).

Monte Carlo Modeling

The Monte Carlo calculations for the transport of electrons and secondary photons were performed with the MCNP™ code, version 4C2 (*15*). The transport calculations took into account in a fairly accurate way the diffusion and slowing down of all types of radiation in the electron-photon cascade established in the media.

The calculations were performed for two modeled geometries (G1 and G2) as shown in Fig. 1 to assess the energy deposition in the detectors. The chemical compositions of the simulated media are listed in Table 1. It was assumed that radionuclides inside the volume of the simulated medium (tooth powder) were distributed uniformly and that the electron emission from ^{90}Sr and its daughter product ^{90}Y was isotropic within the source cavity. The fractional weight of metal impurities in the simulated medium was so small that variation in its chemical composition of the simulated media did not play an important role in the computed results. The uncertainty of the computational results was estimated based on the variation of model parameters such as the density of the dental tissue powder and the density of the detector layer.

The dependence of energy deposition in α-Al_2O_3:C on the mass of the sample is calculated using powder samples filling 12 different containers (G2) having a cylindrical shape with various heights ranging from 0.2 to 4 mm (Fig. 1). All the results were based on a sample of 40×10^7 histories of emitted β particles.

EXPERIMENTAL RESULTS

Determination of Dose Conversion Coefficients (DRcc) for Enamel and Dentin using Thin-Layer β-Particle Detectors (β-TLDs)

The dose conversion coefficient defined as the ratio between the current absorbed dose rate measured using β-TLDs and ^{90}Sr concentration (DRcc = mGy year^{-1}/Bq g^{-1}) was assessed using dentin and enamel prepared in the laboratory with a known amount of ^{90}Sr. The dose rate was obtained by exposing a thin-layer α-Al_2O_3:C detector to enamel or dentin placed in the two different sample holders (G1 and G2) shown in Fig 1. The radiation dose response of the detectors exposed to the standard calibration source (2.8 MBq ^{90}Sr β-particle source at the Secondary Standard Dosimetry Laboratory) was shown to be linear in the relevant dose range from a few micrograys to 5 Gy (*8*). The radiation dose response and TL glow of thin-layer β-particle (α-Al_2O_3:C) detectors exposed for 1 week to enamel powder placed in a sample holder containing various amounts of ^{90}Sr are shown in Fig. 2. The standard deviation of the slope was found to be 6% from the fitted curve that is expressed as $Y = (5761 \pm 358)X + (3210 \pm 257)$.

The sample holder in the G1 exposure geometry contained about 200 mg of sample when fully filled with dentin or enamel. The G2 geometry was designed in such a way that it was possible to change the amount of material (dentin or enamel) contained in the sample holder by varying the height of the container. Experiments were performed using two geometries and placing various amounts of material in the containers. The variation of measured absorbed dose with the mass of the sample placed in two different types of container and exposure geometry (G1 and G2) is plotted in Fig. 3. The saturation dose value on the *y* axis was used to assess the approximate sample size to be placed in sample holders to detect the full β-particle dose. The natural photon background inside the lead shield where detectors were exposed to dental tissue was monitored by placing the detectors on pure quartz.

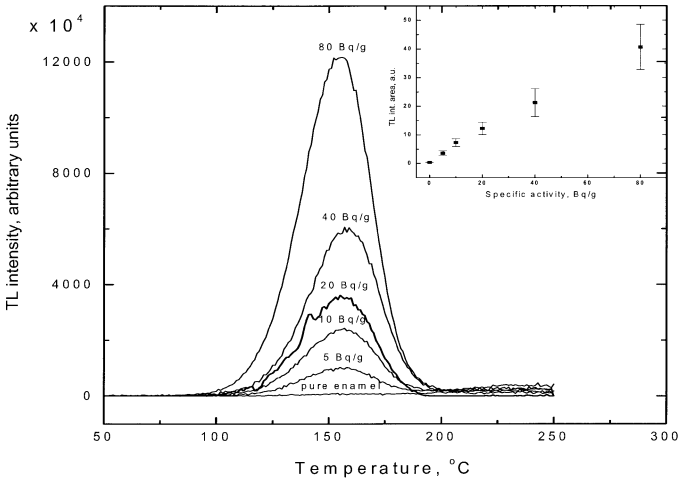

FIG. 2. TL glow curves obtained by exposing the detectors to enamel powder containing a known amount of ^{90}Sr. Inset: radiation dose response of detectors.

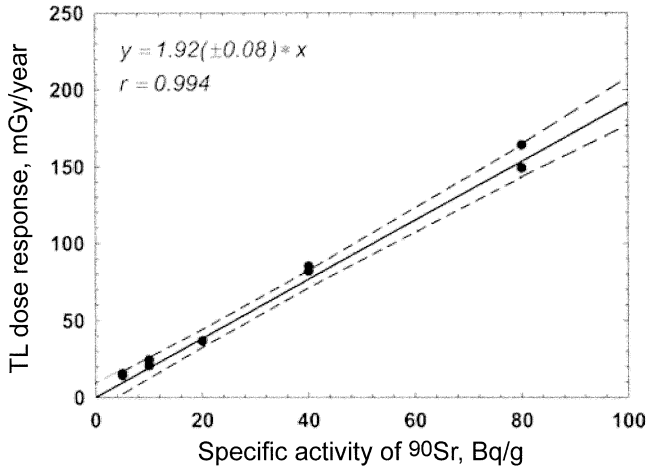

FIG. 4. Variation of measured absorbed dose with ^{90}Sr concentrations in enamel.

The measured absorbed dose in the detectors exposed to samples in the G2 geometry as a function of the specific activity is shown in Fig. 4. From the slope of the curve the average conversion coefficient was found to be equal to 1.92 ± 0.06 mGy/year for enamel. Similar experiments were performed for dentin in the G1 and G2 geometries. The uncertainty in the conversion coefficient obtained for dentin was larger than that for enamel since the total amount of sample that could be used in this experiment was significantly less than that used for the enamel. The conversion factors for dentin and enamel measured in the two geometries (G1 and G2) are listed in Table 2.

RESULTS OF MONTE CARLO CALCULATIONS

Simulation of Experimental Conditions

The numerical calculations were performed using various chemical compositions and density variations that were normally inherent in each modeled medium (*13*, *16*, *17*) (Table 1). It was found that for a constant density, the variation of chemical composition did not exert any significant influence on the calculated dose rate since the fractional weight contribution of metal impurities was insignificant. The uncertainty of the computational results was estimated based on the variation of model parameters such as density of the tooth tissue powder and density of the detector layer. It must be noted that in this type of detector, the β-particle sensitive layer is so thin that a very large (up to 40%) variation in the density of the detectors can be observed within a very short range (*11*). The calculations are performed to take into account such a density variation in the detector surface. It was found that such a variation in density caused a 3.5% variation in the calculated dose rate. The computational relative error was found to be no higher than 2.5%. The uncertainty in the tooth powder density variation was estimated to be 2% for enamel, 4% for crown dentin, and 6% for root dentin. Figure 5 shows the calculated dose conversion coefficients (mGy/year per Bq/g of ^{90}Sr/^{90}Y) as a function of the powder density for the two different geometries. Overall the relative model uncertainty

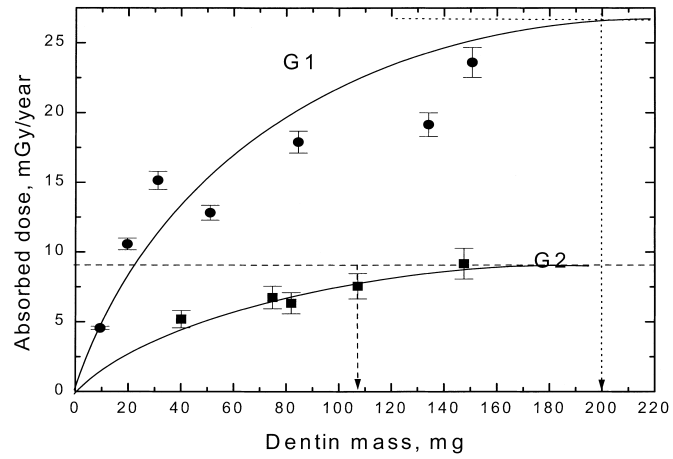

FIG. 3. Variation of measured absorbed dose with the mass of the dentin containing a known amount of ^{90}Sr for two exposure geometries (G1 and G2).

TABLE 2
Comparison of Experimental Conversion Factors with Numerical Calculations for Various Dental Tissues for Exposure Geometries of the Two Detectors

		Conversion factor (DRcc = mGy year^{-1}/Bq g^{-1})	
Geometry	Tissue	Experimental	Numerical calculations
G1	Enamel		2.28 ± 0.16
	Root dentin	2.03 ± 0.06	2.04 ± 0.15
	Crown dentin		2.06 ± 0.14
G2	Enamel	1.92 ± 0.06	1.91 ± 0.14
	Root dentin	1.70 ± 0.3	1.70 ± 0.13
	Crown dentin		1.76 ± 0.13

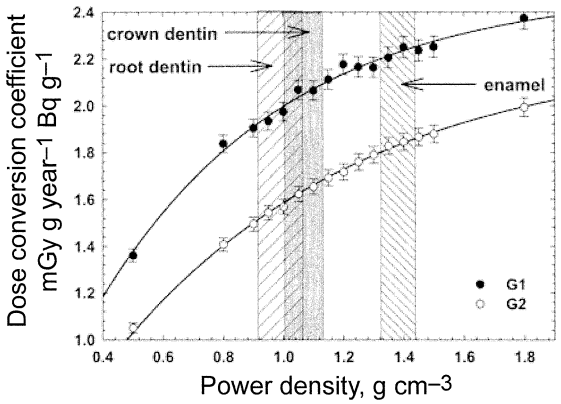

FIG. 5. Variation of dose conversion coefficients (DRcc) with the density of dental tissue obtained using Monte Carlo simulation. Solid lines represent the regression of the exponential rise approximation for computational results. The ranges of the density variation in root, crown and enamel are confined between the dotted, long dashed, and short dashed lines respectively.

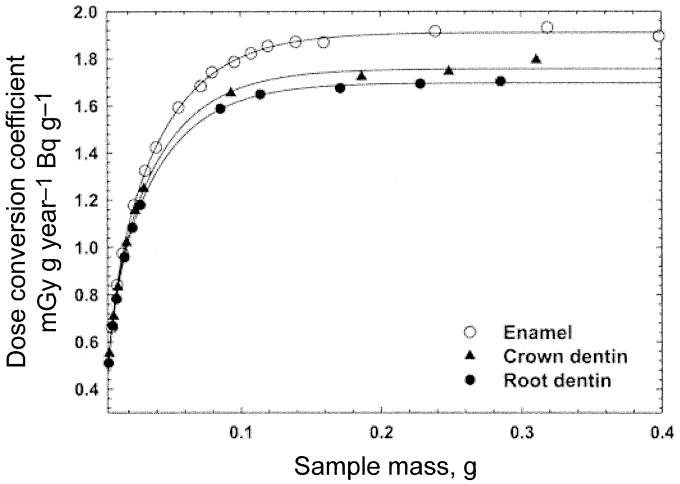

FIG. 6. Variation of the conversion coefficients with the mass of enamel (○), crown dentin (▲), and root dentin (●) calculated using Monte Carlo simulations for measurement geometry (G2).

was assessed to be no higher than 7.5%. The numerically calculated conversion factors for different tooth tissues (in amounts equal to 200 mg or higher) are summarized in Table 2, and the details are discussed below.

Numerical calculations were further performed using Monte Carlo calculation for the correction of variations in the mass in the cavity having G2 geometry. Figure 6 includes the results of Monte Carlo calculations for geometry G2 with enamel and crown and root dentin. The fitted curves have a shape described by an equation having two exponential build-up terms. The calibration coefficients for each tissue type (k) were found to be described by the following expressions:

Enamel:

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

Crown dentin:

$$k_{cd} = 0.59(1 - e^{-160m}) + 1.17(1 - e^{-26.8m}), \quad (2)$$

Root dentin:

$$k_{rd} = 0.59(1 - e^{-160m}) + 1.11(1 - e^{-26.9m}), \quad (3)$$

where m is the mass (in grams) of sample exposed to the detectors.

These equations allowed calculation of the calibration factors taking into account the sample mass. The first term is the same in all the equations, because for a source mass smaller than 40 mg (a very thin layer of radiation source <0.4 mm), the mass dependence of three calibration curves practically coincided, as shown in Fig. 6. In other words, for such small source thickness the differences in density and chemical composition between the calcified tissues investigated had a very negligible effect on the cumulative dose obtained using thin-layer TL detectors. The value 0.4 mm corresponded on average to three average track lengths (mean free paths) for β-particle spectra ($E_{av} = 0.56$ MeV), corresponding to an equilibrium state of $^{90}Sr/^{90}Y$ in the source medium. Theoretically the source whose thickness corresponded approximately to 3 mean free paths can be described as a plate source. In practice it is impossible to obtain a uniform plate source from a biological tissue. Moreover, the value of 0.4 mm is equal to the average grain size of enamel powder.

The computed dose rates obtained using crown and root dentin were not found to be significantly different, so that in practical routine measurements the difference may be ignored. The second terms of Eqs. (1–3) define the tissue difference for calibration coefficients.

DISCUSSION

The assessment of internal dose in enamel due to the presence of ^{90}Sr in various parts of the tooth tissue is a very important task supporting the EPR dosimetry for the Techa River radiation incident. The two luminescence methods suggested by Göksu *et al.* (*9*) and Romanyukha *et al.* (*18*) were considered to be very promising but required further efforts to convert measured absorbed dose or photostimulated images in detectors to ^{90}Sr concentration in calcified tissue. In this work, it is shown that by combining the numerical calculations with experimental results obtained using thin-layer β-particle detectors (β-TLDs), it was possible to convert the measured absorbed dose to ^{90}Sr concentration in particular tissue.

It should be noted that the detection of low-level ^{90}Sr concentration using β-TLDs is not a simple task, due to the strong dependence of the absorbed dose in dosimeters on detector grain size and thickness, exposure geometry and sample size. In this study, the dentin and enamel extracted from fresh teeth were prepared with a known level of ^{90}Sr contamination by soaking the samples in ^{90}Sr solutions. Careful preparation of the samples, even if carried out un-

der highly controlled conditions by a scientist with considerable experience in the field of radiation chemistry, does not guarantee the uniform distribution and penetration of the ^{90}Sr in the dental tissue. Therefore, it was considered essential to include Monte Carlo calculations of electron transport to simulate the exact exposure and detection conditions of the thin-layer β-particle detectors and thus to validate the conversion coefficients obtained experimentally.

It can be seen from Table 2 that the computational results were in good agreement with the experimental results. A verified computational technique now allows the use of calculated conversion factors for the detection of ^{90}Sr concentration as a routine experimental procedure. It was shown experimentally and numerically that the β-particle detection efficiency was very sensitive to the exposure geometry of the detectors and the sample mass (Figs. 3, 5 and 6). In routine work, after separation of the organic fraction, real human teeth may yield only 10–300 mg of calcified tissue, depending on the extent of tooth damage. In such cases the smaller cavities (such as G2 geometry) were found to be more suitable for routine measurements of calcified tissue.

CONCLUSIONS

In this work it has been shown that by combining the absorbed dose measurements obtained using β-TLDs with the numerical calculations, the ^{90}Sr concentration in dental tissue can be assessed for the experimental conditions used, such as the exposure geometry of the detectors and the sample size. It was further demonstrated that by using the results of numerical calculation, the minimum required sample mass can be reduced to as little as 40 mg, for samples containing more than 0.3 Bq/g of ^{90}Sr in the calcified tissue.

It must be noted that, when the β-TLD measurements are required for the detection of low-level ^{90}Sr concentration ($<$1 Bq/g) in the dental tissue, knowledge of the β-particle-emitting natural radionuclide in the calcified tissue is a prerequisite for this kind of study. The natural β-particle dose rate from the teeth that are collected from the population of uncontaminated settlements along the Techa River is currently under investigation.

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