Retrospective Dose Assessment of Inhabitants in the Contaminated Areas of Russia by EPR Measurement of Tooth Enamel

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Abstract

Results of wide-scale dose reconstruction with the use of EPR spectroscopy of tooth enamel are presented for the population living in the radioactive contaminated territories of Bryansk region of Russia. The population of radiation-free territories of neighboring Kaluga region was examined as the control group. The excess doses caused by radiation contamination were determined after subtraction of the contribution into EPR signal in tooth enamel due to the action of the natural background radiation during the lifetime of teeth. The average excess doses were determined for the groups of population formed according to the place of residence. The average values were determined with accuracy ranging from 4 to 25 mGy depending on the number of individuals included into the groups and on the scattering of individual results. The average excess doses highly varied for different places of residence and ranged up to 70 mGy. In general, the average doses of external exposure of the population obtained with EPR spectroscopy of teeth enamel were found to be consistent with the results based on other methods of direct dosimetry and retrospective dose reconstruction methods. Essential exceeding of the individual doses above the average level within the population groups was observed for some persons. That gave a possibility to detect the individuals with overexposure and to include them into groups of increased radiation risk for medical monitoring.

Introduction

It is known that tooth enamel exhibits the features of individual dosimeter and it may be used for assessment of individual accumulated doses in accidental and uncontrolled situations. The stabile paramagnetic centers are formed in calcified matrix of tooth enamel after its exposing to ionizing radiation. These centers can be detected using the spectroscopy of electron paramagnetic resonance (EPR), and the individual accumulated doses can be estimated.

The method of EPR dosimetry was successfully applied for dose reconstruction at uncontrolled irradiation [1-5].

This method of EPR dosimetry has advantages that tooth enamel accumulates the results of radiation influence during a whole period of existence of tooth enamel after its formation. Hence, EPR dosimetry with tooth enamel is suitable for dose reconstruction many years after the exposure. This technique is of particular interest in case when the results of conventional dosimetry are not available (e.g. in accidental circumstances), especially many years after the event, and when doses were accumulated during long period of irradiation. Just the case is dose reconstruction for inhabitants in the contaminated areas after the Chernobyl accident. A disadvantage of the method is that there are problems at sample collection, because it is usually possible to obtain only teeth extracted for health reasons. So, it is often difficult to collect enough samples from the population group of concern.

In this work, the results of EPR dosimetry for the population living in the south-west territories of Bryansk region subjected to radioactive contamination after the Chernobyl accident are summarized and analyzed basing on our previous publications [4-7]. As a control groups, the population of uncontaminated territories of the north part of Kaluga region was investigated.

All measurements were carried out with the same methodical approach adopted as standardized in routine measurements, rigorously the same for all enamel samples. That permitted to make a combined comparative statistical analysis at the examination of all obtained results which were considered to have the same systematic uncertainties.

Materials and methods

Tooth samples were collected in local dental clinics as extracted during an ordinary dental practice. Information about patients and teeth were provided by dentists in special questionnaires together with the samples.

The optimized methodical approach developed in MRRC and adopted as standard for all routine measurements was used for investigation of all enamel samples. Detailed description and basing of the methodical approach are given in the previous publications [4,8], which meets the requirements of State Standard of Russian Federation (1995) [9]. The essential features of the methodical approach used for dose reconstruction are the following.

Enamel samples were prepared by removing dentine from a crown of tooth using hard alloy dental drills at low rotation speed and were cut to pieces of about 1.0 mm. Samples with mass in the range 60 - 120 mg were assumed to be suitable for measurements. The EPR spectra were recorded at room temperature with ESP-300-E spectrometer (Bruker, Germany) using a standard rectangular probe cavity in X-band at microwave power of 10 mW. Modulation amplitude is 0.3 mT, sweep width 8 mT. The time of recording of each spectrum with 16-time accumulation was 45 min. The amplitude of the 4-d line of marker sample containing MnO permanently mounted near the bottom of the cavity was used as a signal intensity reference.

It is known that an EPR spectrum of irradiated dental enamel is composed of two overlapped principal signals: the radiation induced signal (RIS) and the radiation-independent native background signal (BGS) described elsewhere [1,10]. In order to separate the RIS and determine its intensity, the basic software of the build-in computer was used for spectra processing. The spectrum of children's unirradiated enamel was used as the reference spectrum to simulate BGS. The amplitude, field position and width of the reference BGS were manually adjusted under operator control to fit the low-field part of the measured spectra, where RIS was not revealed. The pure RIS was obtained after subtraction of the simulated BGS. The intensity of RIS was measured as the amplitude of its low-field maximum.

The intensity of RIS was converted into units equivalent to exposure dose at calibration according to the following formula:

 $R = A K_{cal}, \qquad (1)$

where:

 $A(g^{-1})$ - intensity of the RIS normalized by sample mass and by intensity of the marker signal;

 K_{cal} (*mGy g*) - calibration coefficient corresponding to average slope of enamel EPR response to radiation versus exposure dose. This coefficient was taken as reciprocal of an average slope of calibration dependencies measured for several enamel samples exposed to a direct collimated beam of ⁶⁰Co gamma-radiation source in dose range 0 - 500 mGy. The exposure dose was measured as air kerma by an air equivalent dosimeter. Calibration with ¹³⁷Cs source gives approximately the same estimation for radiation sensitivity of enamel [11].

The intensity of RIS expressed in units equivalent to exposure dose at calibration was converted to dose absorbed in enamel according to the formulae:

$$D_{en} = (R - D_i) K_{air-en} , \qquad (2)$$
 where:

 $K_{air-en} = 0.99$, conversion coefficient of air equivalent exposure dose at calibration to units of dose absorbed in enamel. In the region of relatively high photon's energy, which were used at calibration, it is determined by the ratio of mass-energy absorption factors of the enamel to that of the air, and its value is close to unity [12].

 D_i - measurement bias arising from intercept value of the calibration dependence of the RIS intensity versus irradiation dose. It is sometimes called initial (or intrinsic) signal [13,14]. This value is specific for measurement conditions and spectra processing procedure. The value of this parameter is mainly determined by difference of the BGS lineshape between the spectra of samples under investigation and the reference spectra used for simulation of the BGS. Its average value can be determined from calibration dependencies after correction on the contribution of natural background radiation. In the case of methodical approach used for spectra processing in this study, its average value was adopted as 65 mGy with a standard deviation of individual values estimated as about 20 - 30 mGy [4].

The additional dose (or excess dose) values caused by additional irradiation due to radiation contamination were determined by subtracting the contribution from natural background radiation accumulated during the lifetime of the tooth (age of the tooth from its formation up to the measurement for dose reconstruction):

 $D_{add} = D_{en} / K_{en} - TA \cdot D_n , \qquad (3)$

where:

 $K_{en} = 1.2$ - correction factor on the energy dependence of enamel sensitivity taken for radiation of ¹³⁷Cs contaminated soil [13].

 $D_n = 0.8 \ mGy \ y^{-1}$ - the annual contribution to dose in enamel due to accumulation of the natural background radiation corresponding to typical value in radiation free territories of central Russia;

TA - age (lifetime) of the tooth (in years).

The lifetime of the tooth was obtained by subtraction of the average age of tooth formation for a given tooth position from the age of a person at the moment of measurement. The ages of tooth formation were determined according to published data [13,15].

The values of the calculated excess doses in some cases may be negative following subtraction of the measurement bias and the accumulated background radiation. This is a result of uncertainty in the RIS amplitude determination near the threshold of sensitivity. These negative values should not be rejected as having no physical meaning, but should be used for subsequent statistical treatment to get true average values and distribution parameters.

A semi-empirical formula for the standard error of estimation dose absorbed in enamel was used:

$$Er^{2} = Er_{I}^{2} + (Er_{2} / (m/100))^{2} + (Er_{3} D_{en})^{2}, \qquad (4)$$

where:

Er - overall uncertainty of dose determination;

 $Er_1 = 30 \ mGy$ – constant contribution to the error due to individual variation of the enamel properties (BGS lineshape variation and impurity signals);

 $Er_2 = 30 mGy$ – parameter specifying the sample mass dependent contribution to the error due to instability and low frequency component of electronic noises of the spectrometer;

m/100 = sample mass (in mg) normalized by the standard value of 100 mg;

 $Er_3 = 0.15$ – coefficient specifying the contribution due to individual variation of the enamel EPR dose response and to uncertainties in exposure dose determination at the calibration.

The formula was derived under the following considerations. There should be a constant contribution to the error of RIS amplitude determination due to variation of the individual sample BGS line shape. In addition, there should be a contribution due to uncertainty of the RIS amplitude determination, which is caused by a low frequency noise component of EPR spectra. This contribution is

specific for the sensitivity of the spectrometer used and for the precision of the spectra processing procedure. Since the RIS value is normalized by a sample mass to obtain a dose value, this contribution should be inversely proportional to sample mass. There also should be a dose dependent contribution due to variation of enamel sensitivity and uncertainties of dose response calibration experiments [13,16]. It should be emphasized that the values of the parameters in this formula are specific for experimental conditions. These parameters were also determined in calibration experiments for methodical approach similar to that used in this study [17,18].

The methodical approach of MRRC used in this study was tested at international intercomparison [12], and it was proved to reconstruct doses with uncertainty within 32 mGy in the dose region 0 - 800 mGy. A modification of this methodical approach with the use of specially designed automatic computer software was also tested [17,18], and the uncertainty of 25 mGy was achieved in the dose region 0 - 500 mGy.

So, the uncertainty of individual dose measurement in the range 200 - 300 mGy will be about 30 - 50 mGy for the sample mass range used. The close level of uncertainty is observed in the deviation of results around the regression line for dose-age dependence obtained for the control population in uncontaminated territories [4].

The specifici fearture of this method appears when it is applied to for groups of population ; the averaging of results reduces random errors. Accuracy of averaged value of the determined dose for the group of population (at N ~ 50 - 100 persons) may be estimated as $Er_N = Er / N^{1/2} \sim 5 mGy$.

Results and discussion

The collection of tooth samples for EPR measurements was started in 1993. The structure of population groups and the number of analyzed samples are presented in Table 1. All measured EPR spectra in digital form, the result of measurement of RIS values together with questionnaire information on patients were entered in a computer database, and used for subsequent interpretation and statistical treatment. The tooth enamel samples are also preserved and are available for repeated duplicate measurements.

Population groups	Number of measurements for each group		
Radiation contaminated territories of Bryansk region	2518		
Among them:			
Gordeevka district	625		
Zlynka district	384		
Klimovo district	423		
Klyntsy district	382		
Klyntsy city	280		
Krasnaya Gora district	163		
Novozybkov district	81		
Starodub district	180		
Control territories (uncontaminated territories of Kaluga region)	420		
The Chernobyl liquidators	122		
Total number of measurements	3060		

 Table 1. Structure of population groups and numbers of measurements by tooth enamel EPR spectroscopy for different population groups.

In the course of investigations, the essential contribution into RIS due to the action of ultraviolet component of solar light was revealed in tooth enamel from front part of jaw (incisors and canines, teeth with positions 1 - 3) [4,19,20]. Therefore, it was concluded that only results obtained for back teeth (premolars and molars, positions 4 - 8) are suitable for interpretation in terms of exposure dose. Part of measurements (about one third) was performed for front teeth or for teeth with unknown position in the former period, when the effect of solar light on front teeth has not been discovered yet. Wherefore, the results for front teeth and for teeth with unknown positions were rejected and are not considered here.

The dynamics of RIS formation in tooth enamel was analyzed to examine its dependencies on the



Fig. 1 e

Fig. 1 f

Fig. 1. Dependencies of the RIS in enamel on tooth age for different groups of population. (a) control territory; (b - f) radioactive contaminated territories: (b) Gordeevka district (137 Cs soil contamination density ranges from 1 to 41 Ci km⁻²); (c) settl. Gordeevka, (5 to 40 Ci km⁻²); (d) settl. Myrny (Gordeevka district), (9 to 85 Ci km⁻²); (e) settl. Zlynka (10 to 70 Ci km⁻²); (f) settl. Vyshkov (Zlynka district) (9 to 45 Ci km⁻²). Parameters of the appropriate linear regressions are given on the figures (SD - standard deviation from regression line, N - number of measurements). The RIS values are expressed in relative units corresponded to exposure dose units at calibration with ⁶⁰Co gamma-radiation source. Note that the measurement bias is not subtracted from the RIS.

age of teeth. Such dependencies on the examples of groups of population living in different territories are presented in Fig. 1.

It should be emphasized that the RIS values in these plots should not be interpreted as absolute values of doses, because the measurement bias was not subtracted at conversion of RIS to dose units according to the formula (1). Only results for adults are considered in order to avoid complication. The results for the control territory (Fig. 1 (a)) have clear tendency to grow with the age. This is well accounted for by accumulation of the effect of natural background radiation with annual accumulated dose about 1 mGy. Such clear tendency is not observed and the data scattering is much higher in Fig. 1 (b)





(a) control territory; (b - f) radioactive contaminated territories: (b) Gordeevka district; (c) settl. Gordeevka; (d) settl. Myrny; (e) settl. Zlynka; (f) settl. Vyshkov. Parameters of distributions are given on the pictures ($<D_{add}>$ - average value; SD - standard deviation of the measurement; N - number of measurements).

for the population of Gordeevka distrct as a whole. For the population of individual settlements in Gordeevka district (Fig. 1 (c)-(f)), however, some tendencies in the course of the RIS with age can be found. Data scatterings in individual settlements are smaller in comparison with that for the whole Gordeevka district but higher in comparison with the control territory.

The observed slopes of appropriate linear regression can not be explained by accumulation of the effect of natural background radiation. In some cases it is too high and in some cases, on the contrary, it becomes negative. Such high data scattering and complicated behavior of age dependencies is obviously caused by additional contribution into exposure due to radioactive contamination of territory, which was different for individuals of different ages.

The interpretation of the obtained results is rather complicated and ambiguous. High data scattering may be caused by differences of professional occupation and high heterogeneity of radiation contamination within the bounds of settlements. It was found that results of dose rate measurements after the accident varied by factors up to 20 in different points of the same settlement (IAEA Technical Report, 1992, p.186) [21]. The differences in the slope of age dependencies may be caused by peculiarities of professional occupation and migration behaviour of population of different age groups. According to the analysis of migration of the population in radiation contaminated territories of Bryansk region [22], over 60% of population of some districts have been changed because of migration during the time period after the Chernobyl accident. It is known that in Myrny and Zlynka settlements especially high migration of the population had taken place and effective countermeasures against radioactive overexposure of population have been undertaken.

Results of RIS measurements were interpreted in terms of additional expose doses D_{add} due to

	Ν	$<\!\!D_{add}\!\!>$,	SE,	SD,	$Q^{137}Cs$,	$Q^{137}Cs$,
		mGy	mGy	mGy	$kBq m^{-2}$	Ci km ⁻²
Bryansk region:						
Gordeevsky district	226	29	4	63		
Myrny	73	8	5	43	1120	30.2
Gordeevka	34	51	9	60	760	20.7
Tvorishino	15	51	14	54	380	10.3
Kozhany	13	4	10	36	1400	37,7
Strugova Buda	10	23	25	56	296	8.0
Klintsy city	150	28	12	48	130	3.6
Klintzovsky district	114	30	8	48		
Smotrova Buda	16	40	16	62	200	5.4
Smolevichi	16	40	16	62	100	2.7
Gulevka	11	19	16	48	260	7.0
Zlynkovsky district	104	50	8			
Zlynka	28	28	7	38	1000	26.8
Vyshkov	25	71	13	66	1000	27.1
Krasnogorsky district	26	6	8	40		
Klimovsky district	34	12	7	32		
Klimovo	11	26	15	40	270	7.4
Kaluga region (control territories):						
Borovsky district	64	1	4	29	<4	<0.1

Table 2. Average additional doses determined using tooth enamel EPR spectroscopy.

Only posterior teeth with positions 4 - 8 of adults are used. Results are corrected on initial intrinsic signal 65 mGy, energy dependence correction factor 1.2 and on the accumulated natural background radiation 0.8 mGy per year during time of tooth existence.

N - number of measurements; $\langle D_{add} \rangle$ average additional dose, *SE* - standard error of the average; *SD* - standard deviation of the measurement; *Q* - density of contamination of soil by ¹³⁷Cs according to (Reference book, 1992)

radioactive contamination according to the formulae (3). Examples of histograms of additional dose distribution for population of different territories are presented in Fig. 2. The parameters of distributions are given on figures. The standard width of distribution of the control territory (29 mGy) gives the approach to estimate the constant contribution of natural background to the overall uncertainty of dose reconstruction in low dose range, if we can assume that all persons were subjected to natural background radiation in the same conditions. It is clearly seen in Fig. 2 that the width of distributions and average values are higher for the population of contaminated territories in comparison with ones for the control territory. The contribution into these parameters is given partially by random error of individual measurements and the rest - by differences between individual excess doses. The average values show the possibility to estimate the level of radiation effect upon the population of given territory with relatively high accuracy.

The averaged values of additional doses determined from the intensities of RIS with taking into account correction parameters for different groups of population are presented in Table 2. Only those settlements where more than 10 measurements have been made are included into the table. The values for average level of radioactive contamination of settlements by ¹³⁷Cs were taken from Reference book (1992) [23]. It should be mentioned that average values of radioactive contamination are determined with high uncertainties because of small number of measurements in every settlement and large variation of the local contamination.

The dependence of the average dose versus the average level of contamination is presented in Fig. 3. The initial part of this dependence is well described by the weighted linear regression with the slope 0.068 + 0.010 mGy per kBq m⁻² (2.4 + 0.4 mGy per Ci km⁻²). This value is well consistent with one obtained by analytical method for accumulated external dose during 8 years in the radioactive contaminated territory. In Technical Report IAEA (1992, p246) [21] the value of the appropriate coefficient was estimated for Bryansk region as 1.6 mGy per Ci km⁻² for the first 4 years after the accident.

As can be seen in Fig. 3, the results for Zlynka, Myrny and Kozhany settlements are not included within the calculation of regression line. Average doses in these three settlements have anomalous low values. Relatively low doses may be explained by a result of countermeasures which were undertaken in these settlements having extremely high level of contamination. It should be mentioned that the analogous effect was observed for the results of direct exposure doses measurements by individual dosimeters (ECP 10 Report 1996, pp104-110) [24], and it was explained by high value of shielding factor due to countermeasures in high contaminated territories.





Parameters of the weighted linear regression are presented on the figure.

Conclusions

Regularities in formation of individual doses for the population residing radiation free and radiation contaminated territories as a function of teeth age and ¹³⁷Cs contamination density have been traced. Statistical analysis of results allowed to detect the contribution into accumulated exposure dose due to the action of the natural background radiation and due to radioactive contamination of territory.

In general, the average doses of external exposure of the population obtained with EPR spectroscopy of teeth enamel are consistent with the results based on other methods of direct and retrospective dosimetry. Relatively low average dose values obtained for some settlements may be explained by peculiarities of behaviour and migration of population. It makes possible to appreciate the efficiency of countermeasures undertaken for prevention of overexposure of population.

The significant exceeding of the individual doses above the average level within the population groups is observed for some persons. This permits to choose the individuals with high radiation risk among the population of radioactive contaminated territories and also to detect the overexposure among the Chernobyl liquidators. Received results of measurement of individual doses are used for formation of groups of increased radiation risk for subsequent medical and dosimetric control.

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