

Considerations Regarding the Implementation of EPR Dosimetry for the Population in the Vicinity of Semipalatinsk Nuclear Test Site Based on Experience from Other Radiation Accidents

Valeriy SKVORTSOV¹, Alexander IVANNIKOV¹, Dimitri TIKUNOV¹, Valeriy STEPANENKO^{1*}, Natalie BORYSHEVA¹, Sergey ORLENKO¹, Mikhail NALAPKO¹ and Masaharu HOSHI²

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General aspects of applying the method of retrospective dose estimation by electron paramagnetic resonance spectroscopy of human tooth enamel (EPR dosimetry) to the population residing in the vicinity of the Semipalatinsk nuclear test site are analyzed and summarized. The analysis is based on the results obtained during 20 years of investigations conducted in the Medical Radiological Research Center regarding the development and practical application of this method for wide-scale dosimetric investigation of populations exposed to radiation after the Chernobyl accident and other radiation accidents.

INTRODUCTION

Retrospective dose estimation for the population residing in the vicinity of the Semipalatinsk nuclear test site (SNTS) remains an urgent question, because of the need to estimate the risks of radiation effects in the population in order to forecast medical consequences of the radiation exposure. Information about the nuclear tests conducted in SNTS, the radioactive contamination of the adjacent territories, the medical effects, and the results of dose reconstruction have been presented elsewhere.^{1–11)} Dose reconstruction methods based on radioecological modeling provide “geographical averages” or conservative estimates of the doses.¹¹⁾ The retrospective luminescence dosimetry (RLD) method using quartz inclusions in building materials gives estimates of “local doses”, which are related to the locations of sampling of quartz containing materials.^{10,12,13,14)} However in radioepidemiological studies analysis of dose-effect relationships should be based whenever possible on individual dose estimates, i.e., estimates that take into account the actual exposure circumstances of each study subject.¹⁵⁾ There are two

physical methods relevant to retrospective estimation of individual doses. The first method, sometimes called “dose reconstruction”, is based on radioecological modeling with further individualization of dose estimates using data about individual dose-determining factors, such as quantities and sources of food and milk consumed, behavioral factors, conditions of living, relocations, etc.¹⁶⁾ This method has typically been used for dose estimation in radioepidemiological case-control and cohort studies, since such studies require the application of a uniform dosimetry method for all investigated cases and controls. The second method is electron paramagnetic resonance (EPR) spectroscopy of human tooth enamel (EPR dosimetry).¹⁷⁾ Application of EPR tooth enamel dosimetry is limited to subjects with available teeth samples, which can be extracted only for valid medical indications. Nevertheless this instrumental method is very useful for validation of individual doses estimated by dose reconstruction, when such dose estimates can be compared to EPR dose estimates for the same persons.^{18,19)}

To date only a few publications have addressed individual dose estimation by EPR dosimetry for the population of the vicinity of SNTS.^{19,20,21,22)} Further investigation by EPR dosimetry should be continued and expanded in this region. The experience obtained in wide scale application of this method in other regions should be taken into account.^{23,24,25)}

The objective of this paper is to summarize the lessons learned from wide-scale application of EPR dosimetry by the Medical Radiological Research Center (MRRC), Russia, in the Chernobyl-affected area and at other radiation accidents and to identify important aspects of using this method at dose reconstruction in the vicinity of SNTS.

*Corresponding author: Phone: 7(095) 9561440,
Fax: 7(095) 9561440,
E-mail: mrrc@obninsk.ru

¹Medical Radiological Research Center (MRRC), Koroliov str. 4, Obninsk, 249036, Russia; ²RIRBM, Hiroshima University, Kasumi 1-2-3, Minamiku, Hiroshima, 734-8553, Japan.

Abbreviations: EPR (ESR) – electron paramagnetic resonance (electron spin resonance); SNTS – Semipalatinsk nuclear test site; MRRC - Medical Radiological Research Center; RAMS – Russian Academy of Medical Sciences; RLD – retrospective luminescence dosimetry

MAIN FINDINGS AND DISCUSSION

Principal findings obtained from the application of the EPR dosimetry in exposed populations

Work in the field of EPR dosimetry began about 20 years ago at the MRRC.²³⁾ Since then, large-scale investigations by this method have been performed to support estimation of radiation effects following various radiation accidents. In studies of the consequences of Chernobyl, EPR dosimetry was used for dosimetric evaluation of the population of territories contaminated by radioactive fallout from the Chernobyl accident (about 3000 samples from Bryansk region, Russia) and for populations of uncontaminated (control) territories (about 500 samples, Russia), and for dose estimation among radiation emergency workers in Chernobyl NPP (about 120 samples).^{18,19,24,25)} Examples of distributions of estimated doses caused by radiation from the Chernobyl accident, i.e., additional to background doses, are presented in Fig. 1 for several of the investigated groups. EPR dosimetry has also been applied to personnel of former USSR Navy submarines who were irradiated in nuclear reactor accidents^{25,26)} and to the population of the radioactively contaminated region of the Techa River in South Ural, Russia.²⁷⁾ Some joint investigations of dose estimation using teeth

samples collected in the vicinity of SNTS were performed by MRRC and Hiroshima University.²⁰⁾

Dosimetrical investigation of a population by EPR dosimetry involves the following stages: sample collection, sample preparation, spectra measurement, spectra processing, EPR signal dose response calibration, dose and dose uncertainty estimation, and interpretation of the results. The main findings obtained by MRRC for improving each of these steps are presented below.

Sample collection

About 3500 teeth samples from residents of the Chernobyl-affected area and control (radiation-free) territories were collected and investigated by MRRC. Based on this experience,^{18,19)} we proposed that the following information be collected for every sample. This includes information about the subject; (subject's full name or other unique identifier, address of permanent residence, birth date, gender, residences and relocations during the period since the radioactive contamination of the territory); about the tooth (type of tooth according to its position); about other exposures to ionizing radiation (occupational exposure including time of military service; X-ray medical procedures in the area of teeth, *etc.*, including dates or periods of exposure and doses if possible); and about the collection of the tooth (institution and

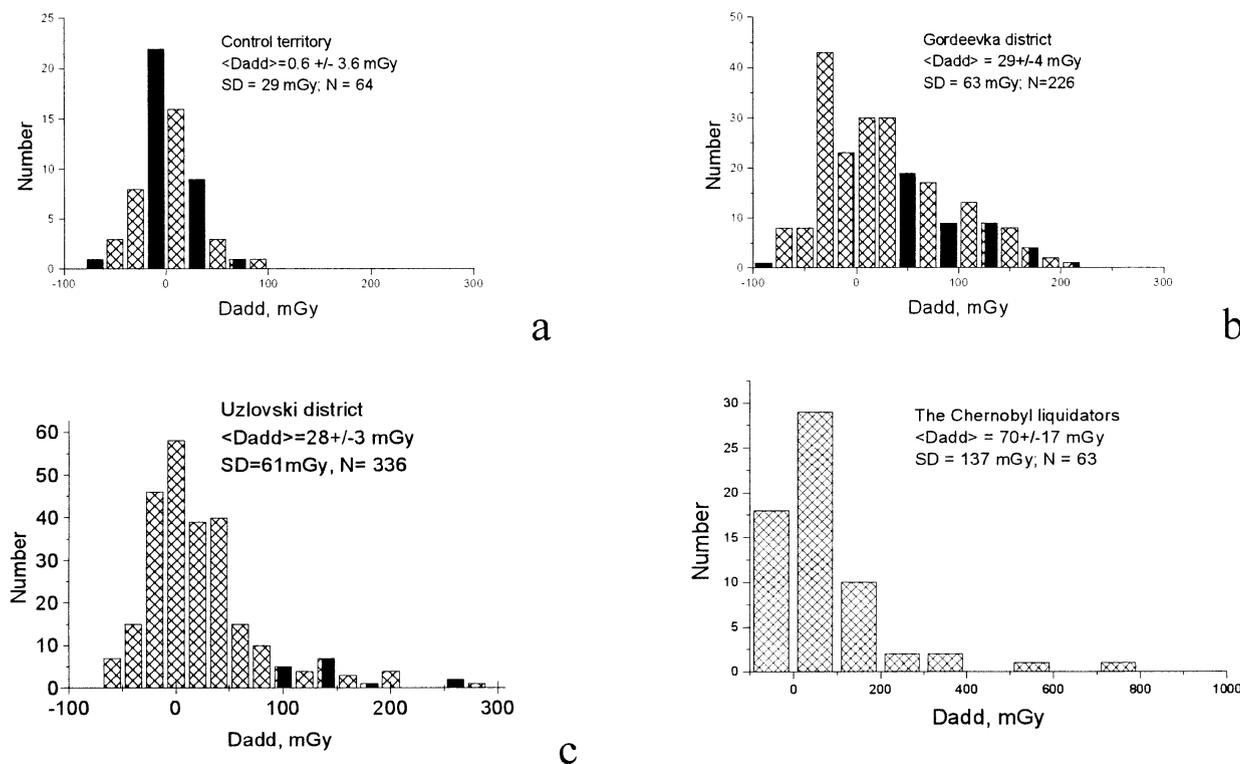


Fig. 1. Examples of statistical distributions of accidental (additional to background) doses (D_{add}): for population of control (not contaminated) territories in Russia (a), for territories of Russia, which were contaminated following the Chernobyl accident - Gordeevsky district of Bryansk region (b), Uzlovski district of Tula region (c), for Chernobyl emergency workers ("liquidators") (d).

person responsible for collection, date of extraction). Also, it is desirable to have additional information about samples, such as description of the extracted tooth (permanent or deciduous, carious cavities, non-carious damage of the tooth enamel, alive or depulped) and information about diseases which might influence the structure of the tooth.

The kinds of information mentioned above can be of great importance in the complicated problem of interpreting measured data. Information about residence during the period of exposure helps in selecting cohorts of interest for dose estimation, and for comparison between results of EPR dosimetry and individual dose reconstruction by radioecological modeling and individual interview.^{16,18,19} Information about tooth position is needed to select teeth that have not been exposed to solar ultraviolet radiation (molar teeth),²⁴ or to ensure that only lingual enamel is used for front teeth. Tooth position must also be known in order to estimate the age of enamel formation, which is necessary to establish the period of dose accumulation. Information about doses from prior occupation or medical exposures can be taken into account in the analysis of the results. The effects on EPR dosimetry of diseases that might influence the structure of the tooth, as well as caries and other tooth diseases, are not well investigated. However, if in the future these effects are found to be important, they can be accounted for in the analysis of the results.

EPR spectra measurement: optimization and standardization of spectra registration parameters

All spectra of the samples collected in the Chernobyl-affected area were recorded at MRRC under standardized

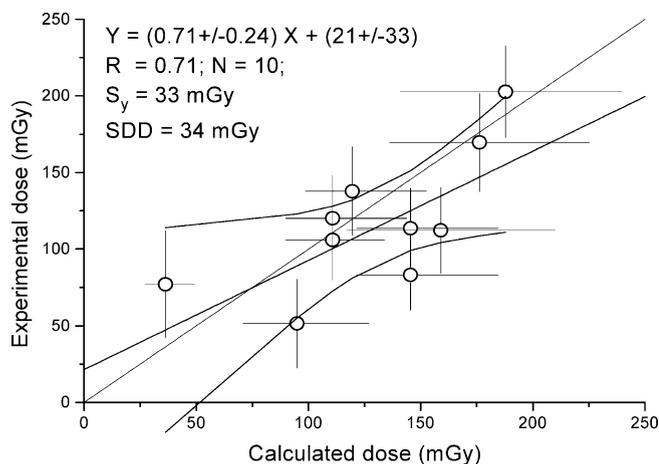


Fig. 2. Accident doses in enamel estimated by instrumental EPR retrospective dosimetry method with tooth enamel (“experimental” dose) versus individual doses estimated by radioecological modeling and individual questioning^{18,19} (“calculated dose”) for inhabitants of the Zaborie village (Bryansk region, Russia), which was highly contaminated following the Chernobyl accident. Dotted lines - linear regression line with 95% confidence interval. Solid line - slope line with zero intercept (added to guide the eye).

conditions of measurements, which were approved in MRRC of RAMS. Optimization of spectra recording parameters (microwave power, modulation amplitude, spectra accumulation time, sample mass) has been described in a recent publication.²⁸ The optimization criterion was minimization of the experimental error of dose estimation, defined as the standard deviation of the arithmetic differences between estimated and nominal doses for a set of samples irradiated with known doses. It should be noted that the optimal recording parameters are not universal and should be found for each specific spectrometer.

As a result of measurements at optimized conditions, the precision of dose determination has reached about 20 mGy for heterogeneous samples (prepared from teeth of different persons) and 15 mGy for homogeneous samples (prepared from pooled enamel). The standard deviation of the differences between EPR doses and results of individual dose reconstruction using radioecological modeling and individual questioning for ten samples collected in the Chernobyl-affected area was 34 mGy^{18,19} (Fig. 2). This result confirms the good agreement between these two methods.

Spectra processing: development of the automatic spectra processing program

A computerized automatic spectra processing procedure was created in order to avoid arbitrariness in spectra deconvolution due to the human element. This program allows isolation of the radiation-induced signal and the native background signal from the whole recorded EPR spectrum using non-linear least squares fitting of the model spectrum. The program automatically calibrates the EPR signal dose response using a preliminary recorded set of spectra from enamel samples irradiated with different doses. Absorbed doses in enamel are estimated automatically using the parameters of the calibration line. Execution of the program creates electronic tables containing the dose estimates along with information about samples and patients, ready for subsequent use in databases and analysis files. The program has been tested using different models of spectra description and modes of operation²⁹ and was found to produce accurate dose estimates.³⁰ However, it remains necessary to identify the mode that minimizes the influence of impurities of unknown origin contributing to the native background signal.

Application of non-destructive methods of dose estimation

It is desirable to keep all tooth samples after dose estimation because it may be necessary to repeat sample measurements in order to check previously obtained results, or to re-assay samples after further development of the EPR dosimetry method. Therefore a non-destructive method of sample investigation using the universal calibration coefficient is applied at MRRC instead of the dose-addition method.¹⁷

The mean square variation of individual radiation sensitivity measured for 50 samples from adult donors is about 10%,^{31,32} which agrees with that from other publications.¹⁷ This variation of sensitivity can be incorporated into the uncertainty of dose estimation. In the low dose range (up to 0.5 Gy) this contribution does not markedly increase the uncertainty of estimated doses because it is small in comparison with other sources of error.

Estimation of radiation dose absorbed in enamel

In the non-destructive method of sample investigation using the universal calibration coefficient, dose absorbed in enamel is estimated from the equation:

$$D_{en} = (1/Slope) (RIS/m) K_{en} - D_{int}, \quad (1)$$

Where:

RIS/m – intensity of radiation-induced signal normalized by sample mass;

$Slope$ – slope of a calibration line obtained by linear regression of the radiation-induced signal intensity normalized by the sample mass versus doses in calibration samples;

D_{int} – intercept value of the calibration line.

K_{en} – correction coefficient accounting for the difference in energy dependence of enamel and the dosimeter using measured doses on irradiated calibration samples.

Parameters $Slope$ and D_{int} should be determined for each specific set of conditions for spectra measurement and spectra processing. The absorbed doses in the calibration samples should be defined in units of dose absorbed in enamel. In this case K_{en} equals unity. Dose due to the natural background radiation accumulated in the calibration samples should be estimated from the age of enamel used for preparation of the samples, and subtracted from the total irradiation dose. For example, if the average age of the enamel is 30 years, then background accumulation should be 24 mGy if it is assumed that the annual contribution from natural background dose is 0.8 mGy per year

The excess (or additional) dose caused by the radiation exposure of interest (D_{ex}) is then determined from D_{en} by subtraction of the accumulated dose due to the natural background radiation:

$$D_{ex} = D_{en} - Enamel\ Age \times dD_{nat} / dt \quad (2)$$

Where:

$dD_{nat} / dt = 0.8\text{--}1.0$ mGy per year – dose rate of the natural background radiation;

$Enamel\ Age$ – age of enamel since its formation up to the time of measurements. This is determined according to the subject's age and the age of enamel formation, which differs for various types of teeth.¹⁷

Uncertainty of dose determination

The uncertainty of dose determination (Er) is estimated according to the following equation with empirical param-

eters:³¹

$$Er^2 = Er_1^2 + (Er_2 / (m/100))^2 + (Er_3 D_{en})^2 \quad (3)$$

Where:

$Er_1 = 20\text{--}30$ mGy – constant contribution to the uncertainty due to variation of the individual EPR signal line shape and the uncertainty of the intercept value of the calibration line;

$Er_2 = 20\text{--}30$ mGy – parameter characterizing the sample mass dependent contribution to the uncertainty caused by spectrometer noise;

$m/100$ – sample mass, normalized to a standard value (100 mg);

$Er_3 = 0.12$ – parameter characterizing the dose dependent contribution to the uncertainty due to variation of individual radiation sensitivity (relative variation about 10%), uncertainty of dose assessment in the calibration procedure (3%) and uncertainty of the estimated slope of the calibration curve (3%).

In practice, the total error of dose estimation in the dose range of 0 – 500 mGy, depending on the sample mass, is in the range of 30–80 mGy, which has been confirmed by the results of the international intercomparison.³⁰

EPR signal dose response calibration

The process of EPR signal formation in enamel samples irradiated for calibration was investigated experimentally and theoretically using Monte Carlo simulation of photon and electron transport.³³ The EPR signal response of tooth enamel to dose in air was measured experimentally in the

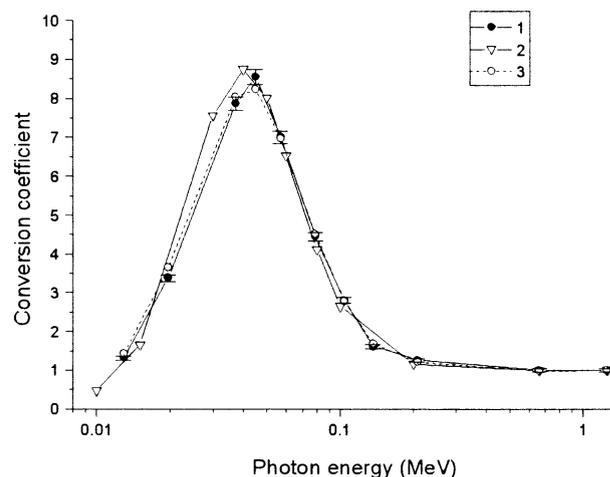


Fig. 3. Comparison of experimental EPR signal dose response of enamel with the results of estimation by the Monte Carlo method: Experimental energy dependence of the radiation-induced EPR signal in enamel per unit dose absorbed in air normalized to the value at 1.25 MeV. (1) Calculated energy dependencies of the conversion coefficient from the dose in air to the dose absorbed in enamel calculated for mono-energetic photons (2) and for real photon spectra used at irradiation (3) normalized to their values at 1.25 MeV.

photon energy range 0.015–1.25 MeV using X-ray irradiation, and ^{137}Cs and ^{60}Co sources. Doses absorbed in enamel were calculated by simulation of photon and electron transport under the same conditions used in the real experiment. The experimental EPR signal response of the enamel normalized to dose in air was compared with the calculated dose in enamel (Fig. 3). The ratio of the calculated absorbed doses in enamel to the experimental EPR dosimetric response was found to be nearly uniform for all photon energies investigated (Fig. 4). Therefore absorbed doses in enamel calculated by Monte Carlo simulation under other

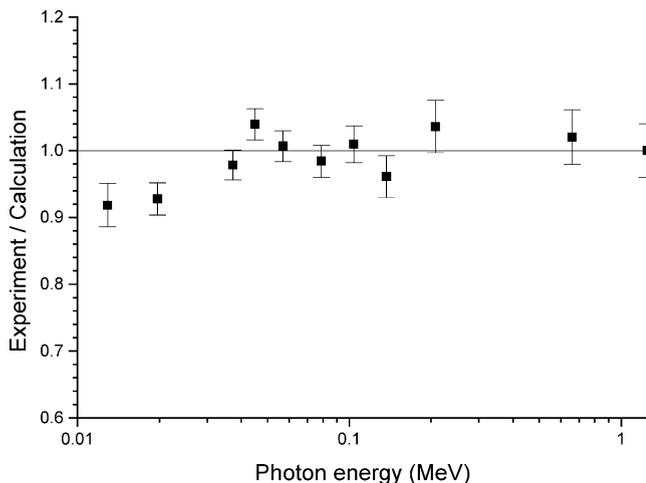


Fig. 4. Energy dependence of the ratio of experimental EPR signal intensity response to the calculated absorbed dose in enamel (corrected on the real photon spectra and normalized to 1.25 MeV).

conditions of irradiation (for example, in the human phantom) can be used for assessment of enamel EPR response. Calculations of conversion coefficients from absorbed dose in enamel to doses in whole body and different organs described in the following section are based on this assumption.

Relationship between dose absorbed in enamel and doses in whole body and organs

Measurement of the EPR signal provides the possibility of estimating absorbed dose in enamel. However, for estimation of health consequences of ionizing irradiation of population residing in the contaminated territories, it is necessary to estimate whole body doses and doses in organs caused both by external and internal radiation.

It is known that irradiation of the population in the Chernobyl-affected area was caused mainly by the isotopes ^{134}Cs and ^{137}Cs , giving comparable (but not equal) contributions of external and internal radiation to the whole body dose and doses to organs, and that dose distributions among different organs are relatively homogenous.¹⁸⁾ The relationship between dose in enamel and external and internal whole body doses due to these isotopes was estimated by the Monte-Carlo method for the mathematical phantom of an adult human³⁴⁾ with an added dental component consisting of enamel and dentine. For the external irradiation, the isotropic gamma-component was considered, while for the internal irradiation both the gamma- and beta-components were taken into account. As result of these calculations, the following equation for the incremental dose in enamel (D_{ex})

Table 1. Conversion coefficients, K, from the external dose in whole body and the external doses in some organs to the external dose in enamel calculated for gamma spectra estimated in different periods in the territories contaminated after the Chernobyl accident.

Organs	Place	K values for different year after the Chernobyl accident			
		1987 year	1988 year	1989 year	1990 year
Whole body	Meadow	1.23	1.24	1.21	1.20
	Tillage	1.41	1.36	1.27	1.27
	Forest	1.24	1.25	–	1.18
	Indoor	1.53	1.55	1.42	1.38
Thyroid	Meadow	1.18	1.20	1.17	1.16
	Tillage	1.34	1.30	1.23	1.22
	Forest	1.19	1.20	–	1.14
	Indoor	1.44	1.45	1.35	1.31
Red bone marrow	Meadow	1.10	1.11	1.08	1.08
	Tillage	1.24	1.20	1.13	1.12
	Forest	1.18	1.106	–	1.06
	Indoor	1.33	1.35	1.20	1.21

caused by radioactive contamination following the Chernobyl accident was obtained:¹⁸⁾

$$D_{ex} = 0.95 D_{ext134} + 0.97 D_{ext137} + 0.46 D_{int134} + 0.35 D_{int137} \quad (4)$$

Where:

D_{ext134} and D_{ext137} – external, D_{int134} and D_{int137} – internal contributions to the whole body dose caused by the isotopes ^{134}Cs and ^{137}Cs respectively.

Conversion coefficients relating external whole body and external organ doses to the external dose in enamel were calculated for the same phantom under conditions of external isotropic irradiation by photons having real spectra estimated for different conditions (contamination of meadow, tillage, forest, indoor) in Chernobyl-contaminated territories. These coefficients are presented in Table 1. To obtain the external dose in enamel, the whole body and organ external doses should be multiplied by these coefficients. The conversion coefficients range from 1.1 to 1.5 for different organs under various conditions of irradiation. As is evident from Eq. (4) the conversion coefficients for external irradiation by the cesium radionuclides are smaller than in Table 1. This difference is explained by the fact that in real conditions the photon spectrum is modified by scattering and absorption in the ground and in air. As a result, the low energy component of the photon spectrum became more intense. This leads to increased external doses in enamel relative to external dose in soft tissue because enamel has a relatively higher absorption coefficient in the low energy range.³³⁾

Effect of solar ultraviolet radiation on tooth enamel

Analysis of EPR dosimetry for the population of the Chernobyl-affected territories and radiation-free (control) territories have revealed significant variation of doses, with average doses several times higher for the front teeth compared to the teeth of the back part of the jaw.^{17,24,32)} This effect was attributed to induction of paramagnetic centers in tooth enamel by the ultraviolet radiation component of sunlight. These solar light-induced centers are distributed in the sur-

face layer of about 0.3 mm in the outer (buccal) enamel of front teeth (positions 1-3 in the jaw), which is too thick to be removed. Therefore, only enamel of molar teeth or of the inner (lingual) side of front teeth is recommended for dose measurements.

Age-dependencies of absorbed doses in enamel for populations of control and contaminated territories

In regression analysis of EPR dose versus age of enamel for samples from uncontaminated (control) territories, it has been found that the slope of the linear regression line is in good agreement with the natural dose rate.^{25,31)} This suggests that biological elimination of radiation induced paramagnetic centers in enamel of teeth *in vivo* is not detectable at least for periods up to 50–60 years after irradiation and can be neglected in dose assessment. Negligible biological elimination over 50 years was also observed in an analysis of correlations between EPR doses and doses estimated from measured activity of ^{90}Sr in enamel for population of the Tcha river region South Ural, Russia, which was contaminated by radioactivity after the accident of 1958.²⁷⁾

Considerations regarding application of EPR dosimetry to the population in vicinity of SNTS

Considerations regarding the application of EPR dosimetry to the population in the vicinity of SNTS are summarized in Table 2. One group of these considerations concerns the collection and documentation of samples. Careful documentation is very important in order to ensure correct analysis of doses estimated by EPR dosimetry, and in order to provide comparison of EPR dosimetry estimates with the results obtained by individual dose reconstruction using radioecological modeling and individual questioning and the RLD method,^{10,18,35)} taking into account high heterogeneity of radioactive fallout around SNTS.^{14,36)} The long interval that has passed since the period of atmospheric tests creates problems in identifying and locating teeth donors for sample collection. Assuming the age of enamel formation is 3–12 yrs, those with permanent teeth at the time of the first nucle-

Table 2. Main considerations regarding the implementation of EPR dosimetry method for population in vicinity of SNTS.

Circumstances	Problems to be solved
A long time has passed since the period of atmospheric tests. Heterogeneity of contamination and relatively short duration of irradiation are typical in the case of nuclear tests.	Problems with sample collection and documentation are possible. It's necessary to take into account relocations of investigated subjects. Tooth age and position of the tooth have to be defined precisely.
Enamel radiation sensitivity isn't known for local conditions. Line shapes for native background and radiation-induced EPR signals aren't known for local conditions.	These factors should be investigated in order to avoid systematic errors at spectra processing and dose evaluation procedures.
Non-homogeneous external and internal irradiation of the body are typical following nuclear tests. The radionuclide inventory differs from the Chernobyl situation	Conversion coefficients from enamel doses to whole body and organ doses have to be assessed accounting for specific conditions of irradiation.

ar test are now over 70 years old. Relatively few persons of such age are alive now, and special efforts should be contact them, obtain their informed consent, and collect samples, as part of their routine medical or dental care. Dose accumulation occurred in conditions of variable radioactive contamination during relatively short periods (about one year) after each nuclear test.¹⁻⁸⁾ Therefore, it is important to collect information about personal relocations in the periods of highest exposure after tests. Also, it is important to define precisely the age of enamel according to tooth position. The age of enamel formation varies from 3 years for the first molar (position 6) to 12 years and over for the wisdom teeth (position 8).¹⁷⁾ The effect of solar ultraviolet radiation on the front teeth has been observed in the temperate climate in the areas close to Chernobyl. In the vicinity of SNTS, which is characterized by higher solar radiation (due to lower latitude and higher number of daylight hours), this effect is expected to be more significant, and precautions should be taken to avoid possible artifacts connected with the effect of solar ultraviolet light on teeth.

A second group of considerations concerns problems connected with the analysis of EPR spectra to estimate dose absorbed in enamel. Radiation sensitivity of tooth enamel and the parameters of EPR spectra of enamel may vary among geographically or ethnically distinct populations because of differences in nutrition. This question should be analyzed in comparison with population of other regions.

The third group of considerations concerns interpretation of the absorbed doses in enamel to obtain estimates of whole body doses and doses in organs taking into account external and internal irradiation. The radionuclide composition of contamination and geometry of irradiation, which leads to specific radiation spectra, vary between different sites and differ from the Chernobyl situation. Irradiation from the radioactive cloud and from surface contamination is believed to predominate in the vicinity of SNTS, whereas in the Chernobyl-affected area the main contribution to external irradiation was caused by relatively long-lived isotopes of cesium in the ground. Also, in the conditions of SNTS, other radionuclides contributed to irradiation,⁷⁾ and heterogeneity of irradiation is possible. The calculations of conversion factors from doses in enamel into doses in whole body and in organs must account for all these aspects.

Finally it should be noted that existing EPR dosimetry method has a natural limitation: the teeth samples are been sampled only in a case of medical indications. Future developing of EPR dosimetry (like "in vivo" EPR dosimetry³⁷⁾) is very promising, but in the case of the "in vivo" method the problem of achieving the high sensitivity still remains unsolved.

CONCLUSIONS

Application of the EPR tooth enamel individual dosime-

try method in the vicinity of SNTS is important for comparison with individual retrospective dose estimation based on radioecological modeling and individual interviews, which are used in radioepidemiological studies.

Further developing of the EPR dosimetry method should be performed in order to adopt this method for territories around SNTS:

-To investigate the "solar effect" and sensitivity of tooth enamel samples which were collected from locations near SNTS;

-To estimate conversion factors from doses in enamel to doses in whole body and in different organs accounting for the radionuclide inventory following nuclear tests, different geometry of irradiation (from the ground surface, from the radioactive cloud), energy of irradiation, and nonhomogeneous irradiation of the human body;

-To investigate line shapes of background and radiation induced EPR signals from local teeth enamel samples.

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