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Results of EPR Dosimetry for Population in the Vicinity of the Most Contaminating Radioactive Fallout Trace After the First Nuclear Test in the Semipalatinsk Test Site

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EPR spectroscopy/Tooth enamel/Dose reconstruction/Semipalatinsk Nuclear Test Site.

The method of electron paramagnetic resonance (EPR) spectroscopy for tooth enamel is applied to individual radiation dose determination to residents of two villages (Dolon and Mostik) in the vicinity of the Semipalatinsk nuclear test site in Kazakhstan. These villages are located near the central axis of the radioactive fallout trace of the most contaminating surface nuclear test conducted in 1949. It is found that excess doses obtained by subtraction of natural background dose from dose absorbed in enamel range up to 440 mGy to residents of Dolon, whose enamel was formed before 1949, and do not exceed 120 mGy to younger residents. To residents of Mostik, excess doses do not exceed 100 mGy regardless of age except for one resident with an extremely high dose of 1.25 Gy. These results are in agreement with the pattern of radioactive contamination of the territory after the nuclear test of 1949 except one case of extremely high dose, which should be additionally investigated.

INTRODUCTION

A long period has passed after the end of aboveground nuclear tests in the Semipalatinsk nuclear test site held by the Soviet Union in 1949–1962. Estimation of health effects of radiation released as result of the tests is still a problem of great concern. One of the regions with the highest risk of exposure to radiation is the territory near the radioactive fallout trace formed as a result of the first test conducted in August 1949. According to data from medical examination, increase in morbidity is observed among the population of that area compared with control territories. The increase is supposed to be a result of exposure to radiation.[1,2)] However, radiation doses to the population in this area are still not well established which makes analysis of the health effects of radiation difficult.

According to archival data on exposure rate measurements in September 1949 after the test[3,4)] and data on isotope contamination of the soil[5)] an axis of the radioactive trace due to the explosion of 1949 is in the immediate vicinity of the villages of Dolon and Mostik. Dolon is located about 100 km away from the site of the explosion and very close (less then 2 km) to the axis of the radioactive trace. Among all settlements in the vicinity of the test site it is the most affected by radiation. Mostik is located near Dolon but about 6 km away from the axis of the radioactive trace[5)]. These villages are of great interest for dosimetric investigation.

There are estimates of average doses of external radiation to these villages based on archival data on exposure rate measurements in September 1949.[1,4)] Local external doses inside some locations in the settlements were estimated by retrospective luminescence dosimetry of quartz-containing building materials.[6)] To make reconstruction of individual doses on the basis of these data information concerning behavior of individuals after the tests is necessary.

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Abbreviations: EPR – electron paramagnetic resonance

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Individual accumulated doses can be tentatively determined long after the exposure to irradiation by the tooth enamel EPR dosimetry method.[7,8] This method has been already used for estimation of individual doses to population in the vicinity of the nuclear test site.[9,10] It has allowed registering increased radiation doses in some settlements. The potentiality of this method makes promising its application for dose reconstruction in the given area. Unfortunately, the tooth enamel EPR dosimetry method has a low sensitivity for application *in vivo*.[8] In the low dose region it can be successfully used only for extracted teeth, and there are problems with their collection. For the population living near the axis of the radioactive trace of 1949, only a few samples were investigated, that is, insufficient for reliable analysis. Among the investigated samples, only three belonged to one resident of Dolon, who lived there at the time of the tests[9].

Due to international collaboration it was possible to collect more samples of tooth enamel of persons lived in Dolon and Mostik at the time of the test of 1949. It enabled carrying-out the present research aimed at estimation of the absorbed doses in tooth enamel in specified groups.

MATERIALS AND METHODS

Sample collection

Teeth of residents of Dolon and Mostik extracted by medical indications were collected during joint Kazakh-Japan medical missions of 2002–2003. In sample tracking forms filled in by dentists, information about teeth donors (name, birth date, address with terms of residence, presence of dental and medical x-ray procedures) and samples (tooth position, date of sampling) was presented. According to information obtained by questioning, no teeth donors were subjected to x-ray procedures of the jaw.

Sample preparation

Enamel samples were prepared by the mechanical method as was described in the previous publication.[9] No additional EPR signals, which can be attributed to mechanically induced strains,[11] were observed.

Samples were prepared from lingual and buccal sides of teeth. The sides of teeth were not marked on the samples immediately in the course of teeth extraction. Therefore, qualified dentists identified the sides of teeth before sample preparation according to tooth appearance. It should be noted that for some of teeth such identification should be accepted as not reliable because of high destruction by caries.

Samples for calibration were collected in the Semipalatinsk region in territories not subjected to radioactive contamination in the period of the nuclear tests at persons of known age whose teeth enamel were formed after the end of the surface tests (after 1962). According to questioning, these teeth were not subjected to medical x-ray procedures. Enamel from several teeth was mixed together to obtain pooled samples, from which seven aliquots of 100 mg were prepared. Spectra of all samples used for preparation of pooled enamel were recorded before irradiation to ensure that there were no increased radiation-induced signal and impurity signals.

Irradiation of calibration samples

Irradiation of the calibration samples was performed using gamma rays from a ^{60}Co source at RIRBM, Hiroshima University. Dose control was performed with the use of the tissue equivalent dosimeter. Enamel was placed in thin paper packs between two 4-mm plastic plates as buildup material. Under such conditions of irradiation, dose absorbed in enamel is close to dose measured by a tissue equivalent dosimeter.[12] The standard uncertainty of the radiation dose is estimated as 3%. Three samples were not irradiated; the other four samples were irradiated with nominal doses of 100, 200, 300 and 500 mGy. Before measurement, samples were stored over one week in order to eliminate transient signals.[13]

Spectrum measurement

EPR spectra were recorded at room temperature in the X-band using a spectrometer (JES-FA100, JEOL Ltd., Japan), supplied with a resonator cavity ES-UCX2 characterized by an unloaded quality factor value of 7440. Microwave power was 2 mW, sweep width 10 mT, sweep time 30 sec, receiver time constant 30 ms, number of spectra accumulation, 40 times, modulation amplitude 0.3 mT. These recording conditions were selected as result of optimization giving the best accuracy of dose determination for the type of spectrometer used.[14]

The third and fourth lines of the EPR signal of a marker sample containing Mn^{2+} permanently mounted in the cavity were recorded together with enamel spectra. These marker signals were used for correction of the spectra magnetic field position at spectra processing and as amplitude reference during correction of sensitivity due to sample mass effects.

The same quartz sample tube of 3-mm inner diameter (JEOL) was used for all samples. The tube was positioned in the cavity so that the center of the sample coincided with a center of the cavity. In a spectrum of the empty tube no significant spurious signals were observed, and so no special efforts were undertaken for correction on the empty tube signal.

Spectra were recorded four times for each sample. After recording of each spectrum, the tube with sample was removed from the cavity and shaken, and then it was placed back with the tube rotated to an arbitrary angle.

Spectra processing

Spectra were processed with the use of a computer program based on the previously described one[15,16] by the least squares fit of a model spectrum to the experimental spec-

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trum. The program was used in the mode in which the native signal in enamel is composed of wide and narrow components consisting of linear combinations of the derivative Gaussian functions. At fitting, vertical position and field shift of the model spectrum, amplitudes of the radiation-induced signal and the native signal were varied. The width of the narrow component and amplitude of the wide component of the native signal were also varied to fit individual shapes of the native signal. Repeatedly recorded spectra of each sample were added together after correction of the field shift of each spectrum using the field position of the marker signals.

Calibration

Examples of spectra recorded for the calibration samples with results of spectra processing are presented in Fig. 1. If these spectra and results of fitting with those presented in the previous publication[9] are compared, it is seen that in the present work the noise level in spectra is less and the quality of spectra fitting is higher. That is achieved because of the averaging of repeatedly recorded spectra.

Parameters of the calibration line were determined as a slope and an intercept of the regression line of the plot of RIS amplitude normalized by the sample mass and by the amplitude of the marker signal versus the nominal doses. It was performed in the automatic mode of the processing program for the set of spectra recorded for the calibration samples. Precision of calibration was characterized by a value of 15 mGy estimated by root mean square of the radiation-induced signal amplitudes deviation from the regression line converted to dose units by normalizing by its slope. Standard uncertainty of the intercept was 10 mGy; relative standard uncertainty of the slope was 2%.

The intercept value was corrected by the dose absorbed in

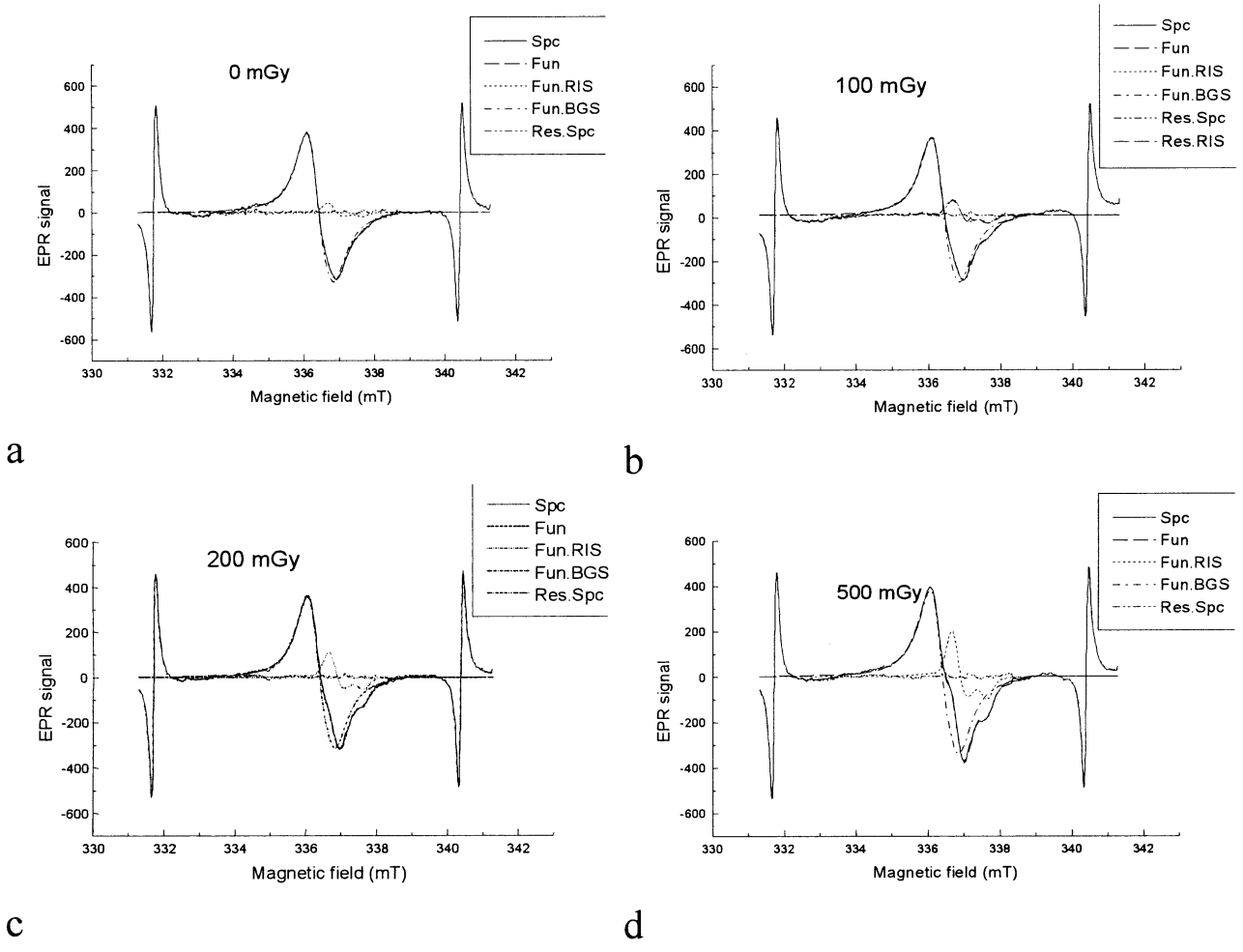

Fig. 1. Examples of EPR spectra of the calibration samples and results of spectra processing. The spectra presented on the plots are obtained after averaging of four repeatedly recorded spectra. Dose values are given in the plot panels. Curve notations: *Spc* experimental spectrum; Fun modeled spectrum; *Fun.RIS* modeled radiation-induced signal; *Fun.BGS* modeled native background signal; *Res.Spc* residual spectrum obtained after subtraction of the modeled spectrum from the experimental spectrum.

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Table 1. Enamel formation age for different types of teeth taken as average of the published age ranges.[7)]

Tooth position	1	2	3	4	5	6	7	8
Tooth type	1st incisor	2nd incisor	Canine	1st premolar	2nd premolar	1st molar	2nd molar	3rd molar
Enamel completed (years)	4	4	6	5	6	3	7	12

the calibration samples due to natural background radiation during the age of enamel existence since its formation, to obtain the undisturbed bias dose level in the following way. Based on known ages of teeth donors, mass of enamel taken from each donor and enamel formation age according to teeth position (Table 1), the average mass weighted age of enamel was estimated as 30 years. Annual background dose was taken as 0.8 mGy yr^{-1}, which is typical for the Semipalatinsk region rural areas[17)] Thus, for correction on the background dose accumulated in calibration samples, the intercept value obtained at calibration was decreased by 24 mGy to obtain the unbiased dose level.

Dose determination

The absorbed dose in enamel was experimentally determined from the amplitude of the radiation-induced signal normalized by the sample mass, by the marker signal amplitude, by the slope of the calibration line and after subtraction of the undisturbed bias dose level. No correction on the energy dependence of enamel sensitivity to photons or calculation of the effective dose was performed because of unknown photon energy distribution in the contaminated territory. Excess dose in enamel caused by technogenic radiation was determined by subtraction of the contribution caused by the natural background radiation during the age of enamel existence after its formation from the experimental dose absorbed in enamel. The age of the enamel was determined by subtraction of the age of tooth enamel crown formation (according to Table 1) from the age of the person at the time of measurement. The annual natural background dose was taken as 0.8 mGy yr^{-1}.

Evaluation of uncertainty

The standard uncertainty of the absorbed dose determination (U) was evaluated with the use of a semi-empirical formula analogous to that used in the previous publications:[9, 16)]

$$U^2 = U_1^2 + (U_2 D_{en})^2 + U_3^2 \quad (1)$$

Where: U_1 = 25 mGy – contribution caused by signals of impurities in enamel, variation of the BGS line shape and uncertainty of the bias dose level;

U_2 = 0.12 – dose dependent contribution caused by variation of enamel sensitivity and uncertainty of the calibration line slope;

D_{en} – absorbed dose in enamel (in mGy) determined by EPR method according to calibration;

U_3 – contribution caused mainly by random spectral noise and instability of the spectrometer. It is determined at the spectra processing stage from the uncertainty of the radiation-induced signal amplitude by converting to dose units according to the slope of the calibration line.

RESULTS

Elevated doses for buccal enamel for some teeth with positions from 1 to 3 were obtained. These results were attributed to the effect of solar ultraviolet on front teeth;[18)] they are rejected from the analysis. Only results of dose determination for premolar and molar teeth were used, and they are presented in Table 2. Uncertainties of the absorbed doses in lingual (inner) and buccal (outer) enamel are given by parameter U_3 in Equation 1 obtained as result of spectral processing. For one sample obtained from a Mostik resident, relatively high doses of about 1.3 Gy were obtained for both halves of the tooth (Table 1). This is a molar tooth, and doses are high in the enamel of both sides of the tooth. Therefore, this high dose may be due to radiation effects, rather than effects of solar ultraviolet.

To control precision of dose estimation, correlation between the absorbed doses obtained for lingual and buccal enamel was analyzed (Fig. 2). The result with extremely high dose is excluded from this analysis. Mean square deviation between doses in lingual and buccal enamel is 65 mGy. To increase the accuracy, averaging of the results obtained for outer and inner enamel weighted by sample mass was performed. A plot of doses absorbed in enamel obtained as result of averaging versus the year of enamel formation is presented in Fig. 3. The uncertainties of absorbed doses are evaluated by Equation 1 taking sample mass weighted mean square average of parameters U_3 for lingual and buccal enamel.

If it is assumed that contribution to the mean square deviation between results obtained for outer and inner enamel is given by random uncertainty of both sets of measurements and taking into account the rules of statistical analysis,[19)] the average random uncertainty of both sets of results for outer and inner enamel may be estimated by the value of $65/\sqrt{2} = 44$ mGy. For averaged results obtained for outer and inner enamel the average random uncertainty of individual doses is $44/\sqrt{2} = 33$ mGy. This value is close to the

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Table 2. Results of dose determination for population of Dolon and Mostik.

Sample code	*Pers. code*	*Res. code*	*Birth year*	*Tooth pos.*	*En. form.*	m_{buc}	D_{buc}	U_3	m_{lin}	D_{lin}	U_3	D_{exc}	U
313	3	1	1957	7	1964	93	90	17	77	110	16	68	28
314	4	1	1933	7	1940	49	139	30	27	98	63	–38	66
315	7	1	1946	4	1951	94	70	22	86	45	20	16	29
316	16	1	1948	6	1951	111	–7	23	120	46	28	–6	35
317	18	1	1950	5	1956	91	81	29	58	–11	42	–3	47
318	24	1	1938	7	1947	91	314	26	64	338	27	283	52
339	1	1	1947	8	1959	127	48	15	117	17	16	–3	26
340	2	1	1947	7	1954	105	114	13	98	105	13	70	27
341	5	1	1947	5	1953	62	84	24	44	12	31	8	37
342	6	1	1943	7	1950	122	41	22	108	86	23	21	32
344	9	1	1942	7	1949	95	55	18	96	90	19	29	29
346	7	1	1946	5	1952	69	66	19	73	32	19	8	28
350	14	1	1941	7	1948	107	43	12	94	144	39	50	45
351	17	1	1942	5	1948	51	124	37	29	92	31	64	39
352	19	1	1930	6	1933	109	378	24	67	613	86	440	106
353	20	1	1935	8	1947	66	121	22	112	112	14	72	28
354	21	1	1957	6	1960	101	109	14	82	151	18	96	31
355	27	1	1939	7	1946	150	81	13	143	92	10	41	25
356	28	1	1930	7	1937	63	98	52	53	115	32	54	40
358	3	1	1957	8	1969	71	75	27	85	75	19	48	29
359	23	1	1924	4	1929	59	404	25	51	416	22	351	57
360	26	1	1928	4	1934	14	496	84	11	487	70	430	93
312	37	2	1931	5	1937	65	1317	7	20	1289	23	1250	159
321	30	2	1938	8	1950	77	85	29	77	75	21	37	30
322	32	2	1936	7	1943	130	114	14	93	187	28	102	39
323	33	2	1939	6	1942	72	78	21	52	–31	42	–26	47
324	34	2	1932	6	1935	107	54	20	125	58	12	1	24
326	36	2	1952	7	1959	151	43	11	126	77	16	25	27
327	39	2	1958	8	1970	82	113	28	95	63	25	62	34
329	41	2	1962	6	1965	39	118	29	16	153	48	105	54
331	43	2	1939	8	1953	78	142	32	54	52	37	57	44
332	44	2	1962	4	1967	49	191	21	24	105	43	119	51
333	45	2	1929	4	1934	79	105	28	53	102	24	48	33
334	46	2	1959	8	1971	56	49	18	39	–20	43	–11	47
335	47	2	1946	8	1958	139	52	12	26	52	37	16	43
337	49	2	1934	7	1941	138	60	29	91	221	62	91	68
338	50	2	1940	7	1947	69	54	32	73	73	20	19	29
357	38	2	1931	4	1936	131	49	15	92	62	20	2	29
361	30	2	1938	7	1945	69	76	34	54	126	28	54	37

Notations and abbreviations: *Sample code* – sample code according to the sample collection of RIRBM; *Pers.code* – personal code according to the questionnaire form; *Res.code* – residence code (1 – Dolon; 2 – Mostik); *En.form.* – year of enamel formation ; m_{buc} and m_{lin} – mass of samples (in mg), D_{buc} and D_{lin} – experimental absorbed doses in buccal and lingual enamel respectively; U_3 – contribution to the uncertainty of buccal and lingual doses according to Equation 1; D_{exc} – excess dose; U – its uncertainty. Doses and uncertainties are expressed in mGy.

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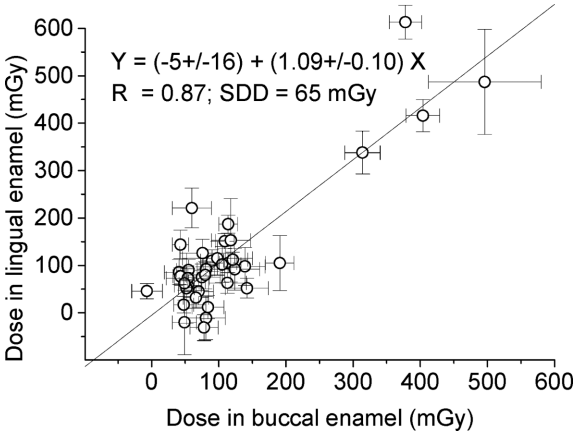

Fig. 2. Correlation between the experimental absorbed doses in lingual and buccal enamel for different samples. Parameters of the regression line with standard uncertainties are presented on the plot panel. Notations: *R* correlation coefficient; *SDD* mean square deviation between doses in lingual and buccal enamel.

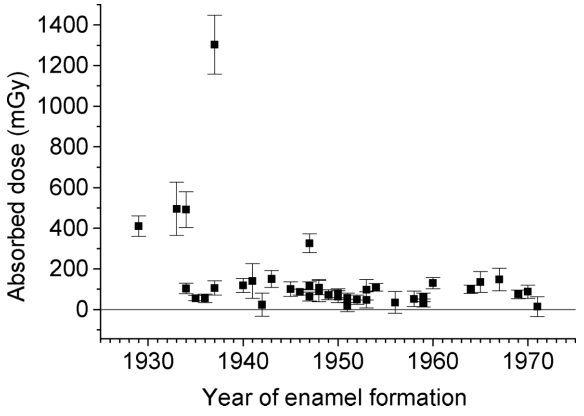

Fig. 3. Dependencies of the absorbed dose in enamel (averaged for both sides of teeth) on the year of enamel formation for residents of Dolon and Mostik.

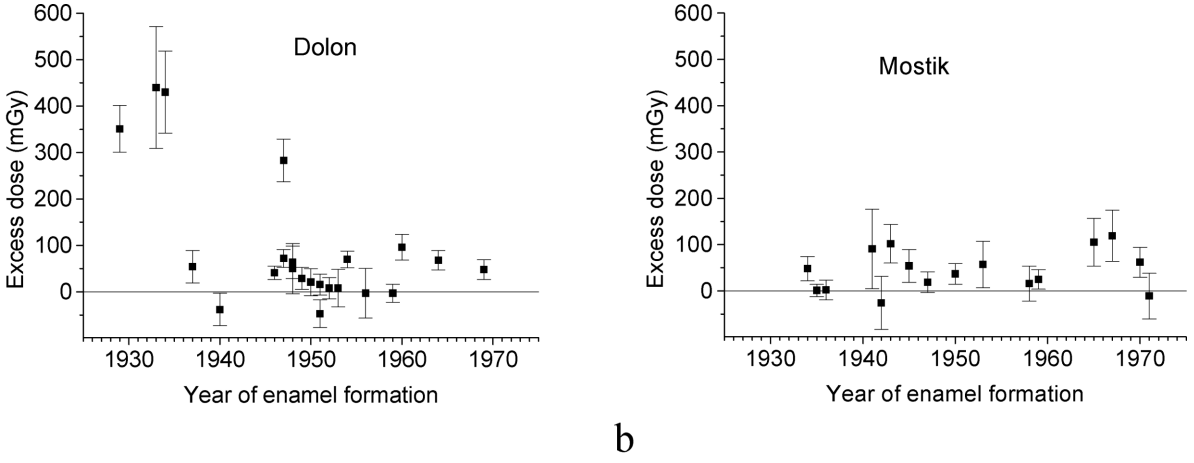

Fig. 4. Dependencies of excess dose in enamel on the year of enamel formation for Dolon (a) and Mostik (b). The high dose of about 1.25 Gy for the person from Mostik is not included in the plot.

average of total uncertainties calculated according to Eq.1 for absorbed dose to enamel averaged for lingual and buccal enamel (38 mGy), which confirms these calculations

Excess doses caused by accidental radioactivity are determined by subtraction of contribution of natural background radiation from dose absorbed in enamel (Table 2). Uncertainties of the excess doses are taken as being the same as absorbed doses averaged for lingual and buccal enamel. Dependencies of the excess doses to residents of Dolon and Mostik on the year of enamel formation are presented in Fig. 4. Negative dose values appeared in the course of mathematical processing. These values should be considered as doses below the uncertainty of dose determination. As it is seen from the figures, all of them are within 2σ uncertainty corresponding to the 95% confidence interval.

DISCUSSION

Relatively high doses are detected in some residents of Dolon whose enamel was formed before the period of the most contaminating nuclear test of 1949. Excess doses of this group ranged up to 440 mGy, and average dose for 11 samples, whose enamel was formed before 1949, is 161 ± 53 mGy. For the other of residents of Dolon, whose enamel was formed after 1949, doses are found to be essentially lower. For this group (11 samples), individual excess doses are in the range up to 96 mGy and average dose is 25 ± 12 mGy. High doses to the elder group may be due to the result of exposure to the radioactive fallout following the nuclear test. Lower doses observed for the younger group of population are explained by the radioactivity from the fallout

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after the nuclear tests decaying relatively rapidly, and the main contribution to the dose to population being formed within one year[4].

For residents of Mostik, excess doses varied from zero to 119 mGy (with the exception of one person, for whom an extremely high dose of about 1.25 Gy was obtained). If exclude this dose from the analysis, an average excess dose for 16 samples from Mostik appeared to be 44 ± 11 mGy. Lower doses for residents of Mostik compared with Dolon may be due to the fact that this settlement is located at a higher distance from the radioactive trace axis.

Estimated individual doses may be compared with doses reconstructed by other authors. The dose of external radiation calculated basing on archived data on exposure rate measurements[4] is equal to 2.2 Gy.[1] Local external doses in the air obtained by the retrospective luminescence dosimetry method[6,20] for several locations within Dolon village[21] are in the range from 420 mGy to 504 mGy[6,20,22,23] (average value is equal to 484 ± 75 mGy[24]). Recent calculations (based on archival exposure rate measurements) on the local external dose in the air for Dolon taking into account the narrow width of the radioactive trace gives a dose value for the same locations equal to 645 ± 70 mGy, which is consistent with dose estimations obtained by other methods of dose calculation about 500–600 mGy.[25,26]. These values correspond with maximal doses in the range of 0.30–0.44 Gy obtained by EPR dosimetry for residents of Dolon with enamel formed before 1949. For the population of Mostik, dose reconstruction on the basis of archived data, gives an average dose of about 150 mGy,[2] which is also consistent with the results of EPR dosimetry.

For dose reconstruction on the basis of archived data it was supposed that stay at the contaminated area during the period of radioactivity decay was permanent and residents received the maximal external dose. The specified values give an estimation of maximal average doses received by the population proceeding from the condition that they were exposed outdoors during the entire period of action of radiation, taking into account a certain shielding factor caused by the part of the time that the people stayed inside buildings.

EPR dosimetry allows for estimating individual doses, which are formed in dependence on individual behavior and possible individual displacements. The individual doses can differ from the average dose because of peculiarities of individual behavior. Some of the people in the period of the tests and after the passage of the radioactive trace and the radioactive fallout could stay in places with lower or higher level of radiation or to spend part of the time indoors. Therefore, individual doses for high-contaminated area such as Dolon, can be lower in comparison with doses reconstructed based on archived data on radioactive contamination, because part of the time a person could spend in a shelter indoor or in less contaminated territories. It is to be noted that comparison dose estimates by the ESR tooth enamel dosimetry method compared with computed mean doses in the settlement provides the possibility to estimate the value of the shielding and behavior factor of dose reduction for inhabitants of Dolon village, which was found to be equal 0.28+/–0.068.[24]

Otherwise, for residents from the less contaminated area (Mostik) located in the vicinity of the more-contaminated area (Dolon), doses can be higher, just as was observed for one person with high dose permanently residing in Mostik. To explain this anomalous high dose, additional individual information on places of location of that person in the period of the tests (perhaps, in more-contaminated territories of the nuclear test site) and probable additional irradiation (professional, medical) is required.

CONCLUSION

Results of the present investigation allow one to estimate individual doses to residents of two villages located near the most contaminated radioactive fallout trace after the nuclear test. Higher doses were detected in Dolon; they are in agreement with the fact that this settlement is located closer to the axis of radioactive trace. It should be noted that the number of samples investigated is insufficient to draw final conclusions about the level of exposure to population on the basis of the results of EPR dosimetry. It is necessary to investigate more samples of tooth enamel formed before 1949 for residents permanently staying in affected areas in the period of the tests.

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