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Estimation of radiation doses of population residing nearby area of uranium ore mining and processing in Kazakhstan by tooth enamel EPR spectroscopy

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ABSTRACT

To assess the impact of natural and technogenic radioactivity in uranium mining enterprises on the population, an investigation by the tooth enamel EPR (Electron Paramagnetic Resonance) dosimetry method was conducted for a group of the population living in the Stepnogorsk city (North Kazakhstan) placed near a uranium mining plant. Accumulated radiation EPR doses in enamel were assessed for 96 teeth samples.

From the measured values of EPR doses and the age of tooth enamel since its formation, the individual annual EPR dose rates (DR) were determined with average value of 1.75 ± 0.46 mGy/year corresponding to the hourly DR of 0.20 ± 0.05 μGy/hour (here and below after ± sign the error of the mean is shown). Data on the individual DR were analyzed for samples collected for population of different parts of Stepnogorsk (microdistricts). These data were compared with ambient dose equivalent rate (ADER) of gamma radiation measured by the conventional instrumental dosimetry. No significant differences in DR and ADER between microdistricts were found. The average ADER through the microdistricts of Stepnogorsk was 0.23 ± 0.01 μGy/hour (corresponding to 2.0 ± 0.1 mGy/year), which is consistent with the average DR measured by EPR.

For some tooth donors significantly higher individual EPR doses and DR, exceeding up to several times the bulk level for the investigated group of population were obtained. These high values may be result of local increased radiation level caused by the wastes of uranium mining enterprises.

1. Introduction

The global mining and processing of uranium ore employs a large number of personnel. In addition to personnel exposed to the risk of radiation exposure at work, a large number of people who live in areas near mining and processing sites are exposed to the radioactive effects of uranium mining and processing waste. These are mine and quarry dumps, as well as dumps of processing enterprises.

To assess the level of radiation risks to health, it is of interest to study the radiation impact on the population of areas near uranium mining enterprises. The region of Stepnogorsk city, in Northern Kazakhstan, is of interest as an object of such a survey. The mining and processing of uranium ore is concentrated in this region. Data for this region can be used for a prognosis assessment of the impact of uranium mining enterprises on the population in other uranium mining regions in the world.

The Republic of Kazakhstan ranks one of the first places in the world in terms of uranium reserves and production (15 and 22 %, respectively, in 2022) (Kazatomprom, 2024). About 40 % of the uranium of the former USSR was mined in Kazakhstan (Program, 2001, 2010). Previously, uranium mining in Kazakhstan was carried out mainly by open-pit mining and in mine developments. Currently, mining of uranium ore in Kazakhstan is carried out mainly by in-situ leaching in sandstone-type deposits. Although about 50 % of world production of uranium ore is

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still carried out by quarry and mine methods (Kazatomprom, 2024). After quarrying and mine development, dumps of poor marketable (off-balance sheet) and unprocessed commercial (balance sheet) ore remain, which are sources of radioactivity.

A significant portion of Kazakhstan's uranium production (17 %) takes place in Northern Kazakhstan (Kazatomprom, 2024). The center for processing uranium ore in this region is Stepnogorsk city. The city-forming enterprise is the Stepnogorsk's Hydrometallurgical Plant, where the primary processing of uranium ore is carried out for the purpose of its enrichment.

Waste after enrichment of uranium ore, the so-called tailings dumps are the main radiation-hazardous objects of the uranium mining industry, since during hydrometallurgical processing of ore almost all of the processed mass goes to waste in the form of tailings. With the uranium product, only about 15 % of the total radioactivity of the processed ore is extracted (Beckman, 2010) and a significant part of the original radioactivity remains in the "tails".

At about 20 km to the north of Stepnogorsk there is a large tailings dump of radioactive waste from uranium production containing about 44 million tons of fine radioactive pulp with a total activity of about 5.4 GBq in an area about 8 km^2 (Fig. 1A and B) (Atomic Energy Agency of RK, 1993; Kazymbet et al., 2018).

Potential harm factors include soil contamination with dust containing radionuclides from the surface of tailings dumps with insufficient watering, air pollution with radon and its daughter decay products; pollution of surface and groundwater.

Periodic radiation monitoring is carried out in the vicinity of tailings ponds and dumps and at uranium processing plants in Kazakhstan (Ibrayeva et al., 2020, 2022; Kazymbet et al., 2022). However, for the population such monitoring was carried out only sporadically based on the level of ambient dose equivalent rate (ADER) of gamma radiation measured by conventional dosimeters.

According to published data, the radioactive background in Stepnogorsk is characterized by great spottiness, and ADER ranges from 0.12 to 0.50 μSv/h (Kanaeva, 2010; Dzhamailova, 2011). In this publication it is noted that the increased background was observed only in certain places, namely in the area of the asphalt plant (0.50 μSv/h) and in places where granites come out onto the day surface (0.32–0.50 μSv/h). In the center and western part of the city 0.20 μSv/h were observed, in the southern part 0.125 μSv/h. The average ADER for Stepnogorsk is 0.22 μSv/h. It is noted that the increased radiation background is associated with the use of crushed stone, granite blocks with an increased content of natural radionuclides, ore and ore-bearing dumps as building materials.

In general, a large variation of ADER was observed across the region in vicinity of Stepnogorsk. According to (Ibraeva et al., 2020), in the main part of the residential area of the villages in the northern direction from Stepnogorsk, ADER does not exceed 0.10 μSv/h. However, in the settlement of Aksu 10 km north of Stepnogorsk, spot places with extremely high ADER up to 2.87 μSv/h were found. These places are associated with abandoned mines and dumps.

According to (Kazymbet et al., 2022), in the sanitary zone between the dumps and the residential area, ADER 0.25 - 0.80 μSv/h was observed. On the territory of the Aksu settlement, five local areas with ADER 0.39 - 0.86 μSv/h were found. For the main territory (open area) and the premises of the settlements (Aksu, Kvartsitka, Zavodskoy) located 10 km north from Stepnogorsk, typical ADER 0.06 - 0.16 μSv/h was observed. It is noted that the probable annual effective dose for the population living in local areas of the Aksu settlement can reach up to ~ 6.5 mSv/year, whereas a usual level is 1 mSv/year in radiation free territories.

According to another publication (Brzhanov et al., 2020), within the sanitary zone of the dump area, ADER is found within 0.16 – 0.80 μSv/h and in the dump area up to 17.4 μSv/h.

Thus, in connection with the probable visit to places with elevated radiation levels, significant differences in individual accumulated doses for the population of Stepnogorsk should be expected. Increased doses should be expected especially for the elderly population who lived during the period when intensive open-pit uranium mining was carried out in this region and lower attention was paid for radiation protection.

However, for the population of these territories the real doses accumulated during their whole life are unknown. The method of dose determination using EPR spectroscopy of tooth enamel samples (EPR dosimetry) allows determining individual doses accumulated over a long period since the formation of tooth enamel. Principles and application of this method are described in a number of publications,. They are summarized in the reviews (Fattibene and Callens, 2010; Bailiff, Sholom, McKeever, 2016) and in official publications (IAEA Report - 2002, 2002; ISO 13304–2, 2020).

We used this method for wide scaled EPR dose measurements for population of other territories radiation-contaminated after the Chernobyl accident and the Semipalatinsk nuclear tests (Ivannikov et al., 2002, 2004; Zhumadilov et al., 2007, 2009, 2013). The results of a pilot survey using the EPR dosimetry method are for a relatively small group

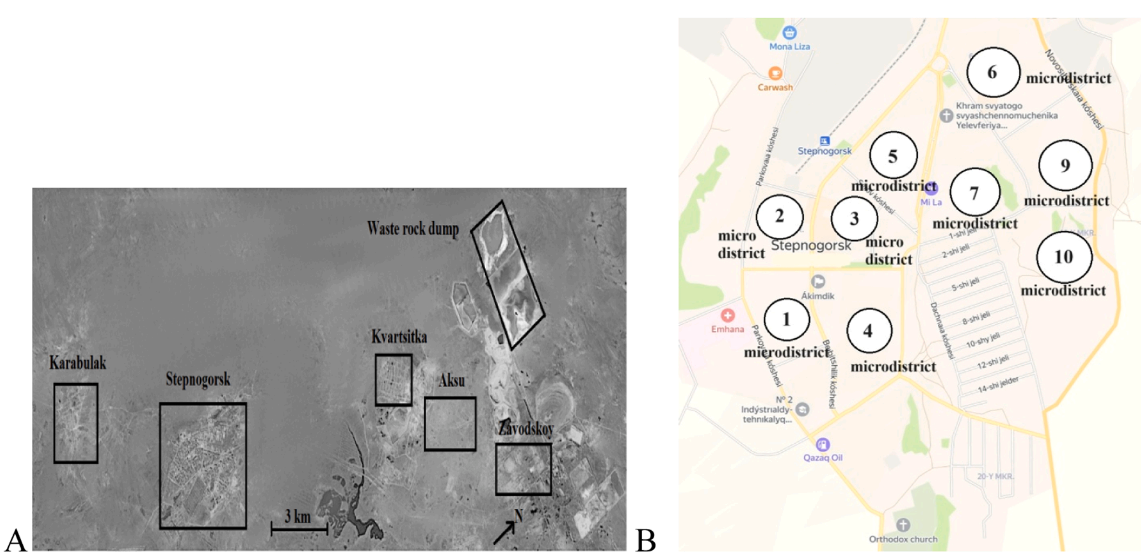

Fig. 1. A - plan of the surrounding area of Stepnogorsk and location of the waste rock dumps; B - plan of Stepnogorsk with the numbers designating microdistricts (Adapted from -Google, 2024).

of the Stepnogorsk population presented in a recent publication (Zhumadilov et al., 2015). No excess EPR doses were found for the group of the Stepnogorsk hydrometallurgical plant personnel compared to the rest of the population. The results of the EPR dose assessment for the uranium miners in this region of Northern Kazakhstan are presented in another publication (Zhumadilov, 2017). Significantly higher EPR doses compared to the population of Stepnogorsk were observed for them.

The aim of this work is to study the contribution of natural and man-made radioactivity to radiation doses for an extended group of the population of Stepnogorsk living near uranium mining enterprises.

Currently, the bulk of uranium mining is carried out by open-pit mining and in mines in different countries (Beckman, 2005). There are huge areas occupied by rock dumps after uranium mining and ore processing. Therefore, the results of a study of the influence of uranium mining on radiation doses to the population of Stepnogorsk are relevant for assessing dose loads on the population living in the vicinity of other developed uranium deposits.

2. Materials and methods

2.1. Sample collection and preparation

Collection of samples of teeth removed for medical reasons was carried out from the population of Stepnogorsk in local dental clinics. For each sample, the information has been provided on the type of the teeth (the position of the teeth in jaw) and individual information about the donors of the teeth (name, date of birth, address, profession, X-ray diagnostic procedures). Of the total number of samples collected, only 96 teeth samples had sufficient enamel content and were used for dose measurement. Typical sample mass of about 100–120 mg was used.

From the information obtained as a result of the survey and questionnaire, the collected samples belonged to the population that was not directly involved in the mining and processing of uranium ore.

According to the administrative division, Stepnogorsk is divided into microdistricts, which are used as the address of residents. Therefore, it is convenient to analyze data depending on the place of residence in microdistricts. The plan of Stepnogorsk with the designations of microdistricts is presented in Fig. 1C.

Sample preparation, EPR spectra measurement and dose determination were carried out in accordance with the recommendations of the IAEA-2002 and ISO-2020 (IAEA Report - 2002, 2002; ISO 13304–2, 2020) (Ivannikov et al., 2002, 2004;. Zhumadilov et al., 2009, 2013).

The enamel samples were prepared mechanically by cleaning the enamel from dentin using dental burs. The samples were prepared separately for enamel obtained from the inner (lingual) and outer (buccal) sides of the teeth. The samples were crushed to granules 0.5–1.0 mm in size using hard alloy nippers.

2.2. EPR spectra measurements, calibration and dose determination

The EPR spectra measurements were performed using the EPR spectrometer ESP 300E (Bruker) operating in the X-band at microwave power of 10 mW and at recording conditions that exclude distortion of the spectra (modulation amplitude 3 mT, receiver time constant 162 ms, scan time 2 min) with scan width of 80 mT and 10 accumulations. Together with the spectrum of enamel sample, the signal of the manganese oxide marker built into the resonator was measured.

The radiation-induced signal (RIS) amplitude in enamel was obtained by subtracting a simulated model native signal (measured for an unirradiated enamel sample) after fitting its amplitude and width to the measured native signal. This fitting was carried out using the spectrometer software under operator control. The RIS amplitude was converted into experimental EPR dose, *Depr*, using the following formula:

$$Depr = K \times Aris/(M \times Am) - Do \quad (1)$$

where: K – dose response coefficient obtained at calibration; *Aris* - RIS amplitude; M – sample mass; *Am* – marker signal amplitude; *Do* – reference correction value obtained at calibration.

Calibration of enamel radiation sensitivity was performed using a Co-60 gamma source and a tissue equivalent dosimeter. Therefore, the dose measured by the EPR can be considered as a measure of the dose absorbed in the tissue. We did not make a correction for energy dependence since the energy spectra in the areas where the samples were collected are unknown.

For calibration, pooled enamel samples were used obtained from teeth (molars) mixed in equal proportions from several teeth donors of known age living in areas free from radioactive contamination.

The *Do* is resulted from the difference between the shape of the model signal used for simulation and real native signal shape. This value was determined as the difference between the cutoff value of the calibration curve (RIS amplitude expressed in dose units extrapolated to zero applied dose) for the calibration samples and the contribution of natural background radiation to these samples. This contribution was estimated as accumulated over the period after the formation of the enamel used for the calibration samples preparation. For that, average age of the teeth donors' tooth enamel and the annual dose of external radiation 1.0 mGy/year corresponding to the typical natural gamma dose rate were taken into account.

Enamel age was determined as difference between age of teeth's donors and age of enamel formation, defined according to the recommendations of the IAEA report (IAEA Report - 2002, 2002).

For the posterior teeth (molars and premolars, teeth positions 4–8), the RIS amplitudes obtained for the enamel of both sides of the teeth were used to determine doses. For the anterior teeth (incisors and canines, positions 1–3), only the results of measurement for the enamel obtained from the inner side of the teeth were used to avoid the effects of solar ultraviolet illumination, which occurs on the outer side of anterior teeth (Ivannikov et al., 1997; IAEA Report 2002).

2.3. Error estimation

In accordance with the previously published methodology (IAEA Report -2002, 2002; Ivannikov et al. 2000, 2004), one of the main contributions to the error of EPR dose determination is caused by signals of impurities in enamel of undefined origin, which vary for different samples. This contribution is difficult to determine directly; it can be estimated indirectly as about 30–40 mGy (not depending on the sample mass), as the minimum error of dose estimation achieved on the basis of calibrations and interlaboratory comparisons for the heterogeneous samples prepared from different teeth (Ivannikov et al., 2000, 2004; Fattibene et al., 2011).

Another important contribution to the dose determination error is uncertainty in determining the amplitude of the RIS because of the low-frequency noise in the spectrum. Such noise is caused by the unstable parasite signals and the time-depending distortion of the spectrometer baseline shape. It depends on the type and the technical state of the EPR spectrometer – on its sensitivity and the base line stability.

The contribution of the low-frequency noise may be estimated by its average amplitude in the spectrum region near the sample signal. For the spectrometer used and for the conditions of measurement used this contribution was about 30–40 mGy in terms of dose in enamel. However such determination of this contribution is not reliable since these noises may overlap with the signal of enamel.

In this work, the contribution of low-frequency noise, *Enoise,* was estimated basing on the difference in the determined EPR doses for enamel samples obtained from the outer and inner sides of the same tooth, *Dout* and *Dinn*, respectively.

Basing on the assumption that the enamel composition on both sides of teeth is the same, impurities in the enamel give the same contribution to the dose determination error, which is neutralized when *Dout* and *Dinn* are subtracted from each other. Therefore, it was believed that the difference between *Dout* and *Dinn* is mainly due to the low-frequency noise, and the contribution of the low-frequency noise, *Enoise*, to the error of each individual measurement can be determined from this difference. It is known, that when determining the error of the difference between two values, the squared error of the result equals to the sum of the squared of the values. Therefore, *Enoise* was determined using the ratio:

$$Enoise = \ (Dinn - Dout) \ / \ \sqrt{2} \tag{2}$$

The contribution of the uncertainty of individual radiation sensitivity of enamel to dose determination is about 10 % of the determined dose (IAEA report - 2002, 2002; Ivannikov et al. 2000). In the range of low doses it can be neglected.

The error of individual dose determination, *Eepr,* was estimated by the ratio:

$$Eepr = \left[Enoise^2 + Eimp^2 + (Esens * Depr)^2\right]^{1/2} \tag{3}$$

where: *Eimp*=40 mGy - contribution to the error caused by impurities in enamel, taken as indirectly estimated value as described above; *Enoise* – contribution caused by noises of the spectrometers as it is determined by Eq. (2) for each pair of individual measurements; *Esens* = 0.1 – relative contribution to the error caused by variation of the radiation sensitivity of individual samples; *Depr* – measured dose.

The EPR dose accumulation rate (DR) in enamel was determined by dividing EPR dose by the age of enamel. Accordingly, the error of DR was estimated from the dose determination error dividing it by the age of enamel.

2.4. Measurement of the ambient dose equivalent rate

Measurement of the ambient dose equivalent rate (ADER) of gamma radiation was carried out using a dosimeter- radiometer MKS-SRP-08A with gamma unit BDPS-25–01A (Amplituda, Russia). This dosimeter is certified with a relative error ±15 % and supplied with a control source of gamma radiation cesium-137 for verification. The gamma unit consists of a scintillation detector NaI (Tl). The energy dependence of the sensitivity in the range from 50 to 3000 keV relative to the energy of 662 keV gamma radiation of cesium-137 is not more than ±25 %. It is calibrated to measure the ADER of photon radiation (MKS dosimeter instruction, 2021)

The ADER measurements were carried out in the immediate vicinity of the entrances of residential buildings in different microdistricts of Stepnogorsk according to the administrative division shown in Fig. 1B (by the three measurements were performed in each microdistrict). These buildings were not obligatory the same as the houses in which the tooth donors reside.

Data processing was performed with the use Origin6.0 (Microcal) and BioStat5.0 (AnalistSoft) programs. Linear regression parameters were calculated taking into account the error-weighted data. Mean values and their uncertainty were also calculated for error-weighted data. The calculation of probability of coincidence of the two mean values was carried out by the Student's *t*-test using an online calculator (Ref. Student's *t*-test online calculator 2024). Throughout the paper after the ± sign, the error of the mean or the error of the individual value is indicated.

3. Results and discussion

3.1. Determination of EPR dose values and analysis of their reliability

The RIS values were measured in the enamel samples obtained from the outer and inner sides of teeth, and the EPR dose values *Dout* and *Dinn*, respectively, were estimated using Eq. (1) in accordance with the calibration relationship.

The results of measuring *Dout* and *Dinn* depending on the age of the enamel for all samples are presented in Fig. 2A. For clearance, some points outside the main data set are connected by a dotted line and some sample numbers are indicated. For comparison, a line with a slope of 1.0 mGy/year is presented, corresponding to the typical dose rate of natural background radiation for radioactively clean territories and residential places of Kazakhstan (0.11 μGy/hour) (Kazymbet et al., 2018; Ibrayeva et al., 2020, 2022). For clearance, a dotted line connects some points outside the main data group and some sample numbers are indicated. The results for the samples #105 and #162 are noteworthy; the EPR doses for these samples are significantly higher than the doses for the other samples. The sample numbers marks the data points for these samples on the plots.

In Fig. 2B the results of dose determination are presented for the samples in which doses were measured only for the enamel of one side of the tooth. For this group of samples a linear regression is calculated, parameters or which are shown on plot panel.

Negative EPR dose values were obtained for some samples; these are the resulted values caused by large absolute errors for small doses. These negative values cannot be excluded at the statistical analysis, as they must be taken into account to obtain unbiased estimates.

To control the correctness of dose estimation and to assess the measurement error, a comparison was made of the EPR dose values measured for the samples prepared from the inner and outer sides of teeth, *Dout* and *Dinn* respectively (Fig. 3A). Differences between doses, ΔD=*Dout* – *Dinn,* were analyzed depending on the average dose for the both sides (Fig. 3B), in accordance with the recommendations of the publication (Altman et al., 1983). For ΔD, the standard deviation, SD was estimated at 67 mGy. The mean of ΔD is 7 ± 8 mGy, which is close to zero within the error of its determination.

As follows from Fig. 3B where there are only a few data points little outside the ±2SD level, corresponding to the 95 % confidence interval. Therefore, it can be concluded that there are no outliers and the observed data scatter for ΔD is due to random variation of Dinn and Dout because of uncertainties of RIS measurements.

The histogram of the distribution of ΔD is presented in Fig. 3C. The distribution of ΔD is fitted by a normal distribution with parameters corresponding to the mean value and standard deviation. According to the normality test of Shapiro-Wilka (Glantz, 2017), the normality is accepted. This indicates that the observed scatter of ΔD is due to the random measurement error and there are no outliers in the determination of dose due to any unaccounted phenomena.

For ΔD, the root mean square deviation of 67 mGy was estimated. Basing on the estimated SD value for ΔD, the average for all pairs of samples (outer and inner enamel) contribution to the error caused by the spectrometer's noises, according to Eq. (2) is estimated as $67/\sqrt{2} = 48$ mGy. When the average error of individual dose measurement according to Eq. (3), was estimated at about 70 mGy.

For the samples for which EPR doses only for one side of teeth (outer or inner enamel) were measured (Fig. 2B), the standard deviation from the mean was estimated as 65 mGy. This value is close to the error of single measurement determined for the pairs of samples. Therefore, it was accepted that there are no outliers among these dose measurements and all these results were used at subsequent data analyzes. These data were assigned an error equal to the average error for individual measurements, that is 70 mGy.

Of particular interest is the question of the reliability of the measurements with increased doses for samples #105 and #162. For these samples, ΔD are 50 and 140 mGy respectively, which lay within the double standard deviation, 2SD = 140 mGy, which corresponds to a confidence level of 95 %. Therefore, these data points are not outliers, and the corresponding doses can be considered reliable with error obeyed to normal statistical distribution.

To estimate variation of EPR doses for different types of teeth, dependence of doses on tooth positions was analyzed (Fig. 4). It is clearly seen that for samples #105 and #162 significantly higher doses are observed. The contribution to increased doses may be due to some

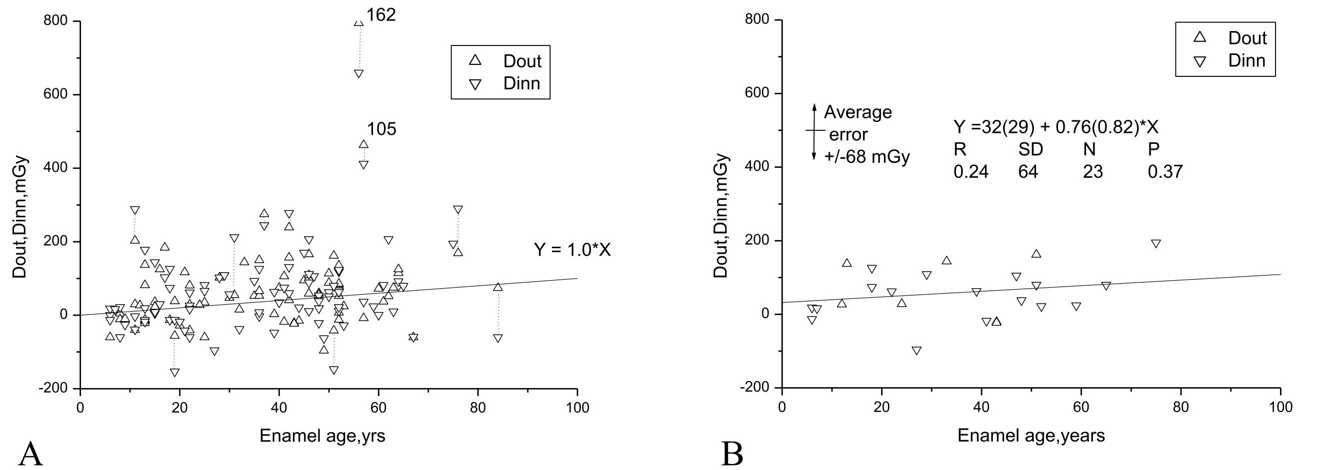

Fig. 2. EPR dose values measured for enamel from outer and inner sides of teeth (*Dout* and *Dinn*, respectively) plotted versus the enamel age: A – for all samples; B – for the samples in which doses were measured only for the enamel from one side of the tooth.

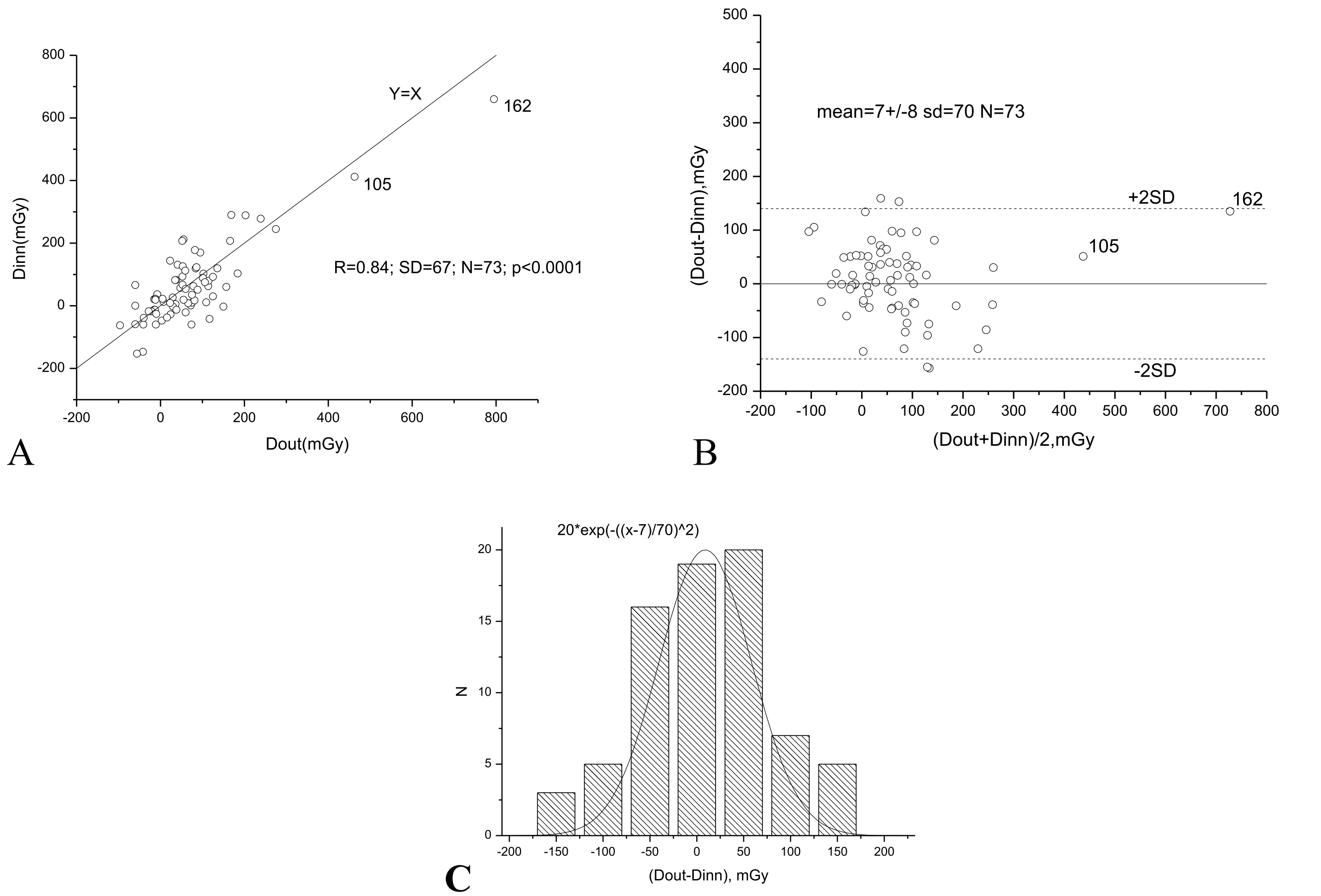

Fig. 3. Comparison between dose values measured for outer and inner enamel: A - the relationship between doses measured for outer and inner enamel. For clarity, a line with a unit slope is added; B - the difference between the doses measured for outer and inner enamel versus the average dose for both sides (the dotted lines show levels of mean ±2 SD); C - histogram of the distribution of difference in doses measured for the outer and inner enamel. A plot of normal distribution with the estimated parameters for these data is added.

technogenic irradiation at medical exposure, or it may be the result of a significant excess of the local dose rate at the place of residence or work compared to the typical average level of background radiation.

For the three samples from inner side enamel with tooth position 2 somewhat higher doses are observed**.** This values and their data scattering are not significantly different in comparison with dose values for other teeth positions. Therefore, it was accepted that EPR doses for these samples are not outliers and they were used the further analysis.

3.2. Analysis of the dose dependence on the enamel age

For further analysis, the results of measurement for all samples were used. For samples for which the doses in enamel for both sides of the tooth were measured, the EPR dose, *Depr*, was determined as the average

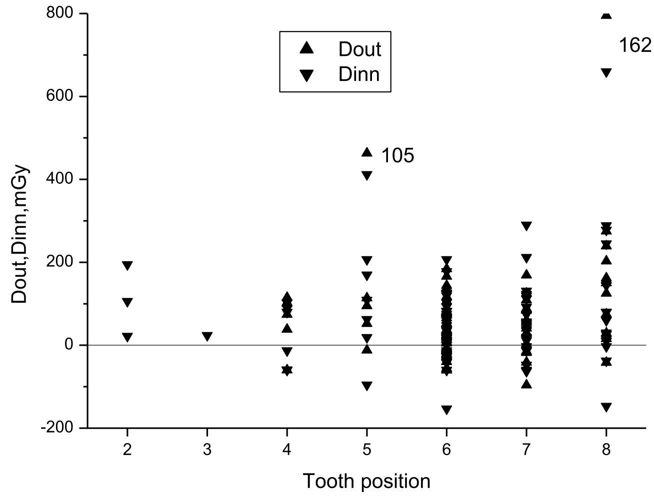

Fig. 4. Dependence of the EPR dose values for outer and inner enamel on the tooth position. Sample numbers are shown for the pairs of points with excess values.

for both sides. The error was estimated using Eq. (3), dividing it by $\sqrt{2}$. Since for the single sample average error od dose was estimated at 70 mGy, when the average error for these doses is 50 mGy. For samples, for which the doses for enamel on only one side of the tooth were measured, *Depr* was taken equal to this dose, and the error was determined equal to the average error value for the enamel of the remaining samples, which was estimated at 70 mGy.

The dependence of *Depr* on the age of enamel is shown in Fig. 5. There are two outlier points that stand out significantly beyond the main bulk of data (samples #105 and #162). The results of calculating the parameters of the linear regression weighted by errors, estimated for all samples, and also without taking these outlier points into account, are shown. The regression line itself and the boundaries of the confidence intervals are shown in this figure for all samples.

The reliability of the linear regression is characterized by the estimated uncertainty of its parameters (slope and intercept), as well as the standard deviation, SD from the regression line, which are indicated on the graph panel. The values of the correlation coefficient, *Rcor*, and the significance coefficient of its assessment, *P*, are also indicated (the smaller *P*, the higher the reliability of the *Rcor* assessment).

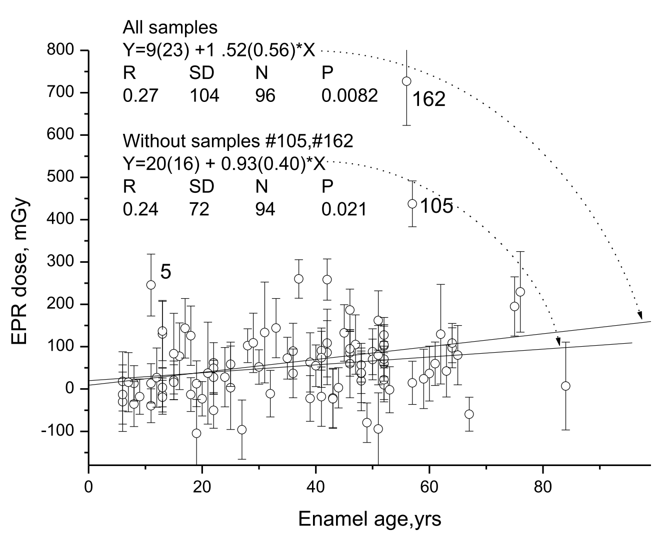

Fig. 5. Dependence of the EPR doses on the enamel age. Corresponding regression line is shown on the plot. The parameters of regression line are shown as calculated for all samples and without outliers samples #105 and #162.

The relatively low values of *Rcor* are due to the relatively small number of data points and the high data scattering of the EPR doses in relation to the annual rate of dose accumulation. However, the estimated low value of $P < 0.05$ indicates the reliable estimation of *Rcor* and a trend towards increasing dose with age.

More informative is the value of the slope of regression line (1.52 ± 0.56 mGy/year for all samples and 0.93 ± 0.40 mGy/year without samples #105 and #162). This value gives an idea of the average annual dose accumulation rate for a given group of population.

The slope value obtained without outliers is close to the typical annual dose for the radiation uncontaminated territories, corresponding to dose rate 0.11 mGy/h. The intercept value 20 ± 16 mGy is not differed significantly from zero within its error. This indicates that the baseline used in estimating the dose from the RIS was determined correctly at calibration. The somewhat higher than zero value of this parameter is partly due to the systematic error of the bias parameter *Do* in Eq. (1).

Individual EPR doses vary greatly. These variations are partly due to random error, and partly due to differences in the individual irradiation level. However, it is impossible to isolate this contribution to the measured values masked by the large random error of individual measurements. Only the values for samples #105 and #162 can be recognized as reliable increased doses, for which the estimated dose values (437 mGy and 727 mGy, respectively) significantly exceed the error of their estimation (about 50 mGy and 140 mGy, respectively) and significantly differed from the mean dose for other samples. According to the Student criterion, the parameter of probability of coincidence of these doses with the mean dose, $P < 0.01$.

According to the questionnaires, for the samples of donors with high doses (#105 and #162) it is not indicated, that they worked in industries related to the mining and processing of uranium ore. If to exclude the error factor, then, probably, the high doses for them can be due to their temporary stay in places with an increased radiation level. Given their age (over 60 years), they lived during the active functioning of the uranium mining industry in this region. At that time, little attention was paid to radiation contamination by waste, and the radiation load on the population and personnel was higher.

3.3. Assessment annual and hourly EPR dose rate

It is impossible to judge about the intensity of radiation exposure over a long period only by the value of accumulated EPR dose, since the ages of tooth donors are different. Therefore, it is reasonable to estimate the annual dose rates (DR), based on which it is possible to judge the intensity of radiation exposure. To obtain the individual annual DR for each sample and their errors, the individual EPR doses and their uncertainties were divided by the enamel age. The dependence of the individual DR on the age is presented in Fig. 6.

The following feature should be noted for the obtained DR values and their errors. Since the doses and their errors are divided by the age of the enamel, at small ages the error increases greatly, inversely proportional to the age. The age dependences of the doubled average error in the positive and negative directions, superimposed on the average DR are shown in Fig. 6.

The average error of DR was determined based on the average error of dose estimation, about 50 mGy, by dividing by age. These dependencies define the limits (95 % confidence interval) within which DR variations can be considered random, caused by their error. For samples #5, #105 and #162, DR significantly deviate from these limits. Therefore, these values are not caused by the random deviations due to the errors, and their increased values, in contrast to the average DR, can be considered statistically significant (for the sample #5, the probability of lack of difference, $P = 0.07$, for #105 and #162, $P < 0.01$).

The average DR, obtained as the arithmetic mean weighted by the error values for all samples, is 1.75 ± 0.45 mGy/year, which

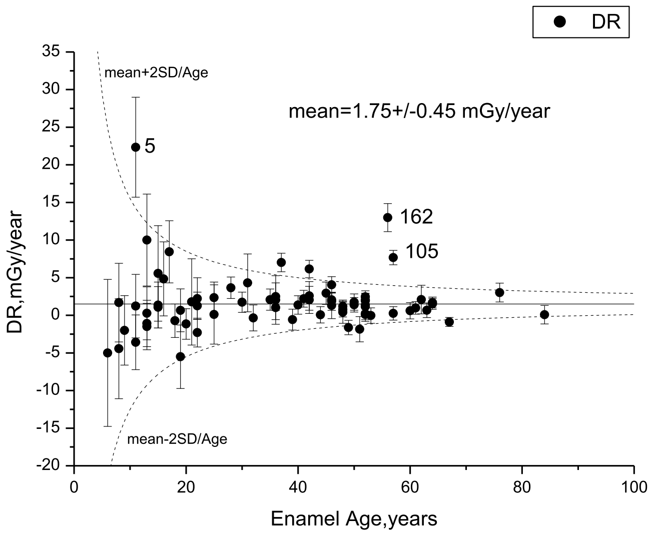

Fig. 6. Dependence of the individual annual EPR dose rate on the enamel age. The dotted lines represent the age dependences of the double estimated average error in the positive and negative directions, superimposed on the mean DR. The mean value is presented as determined by averaging weighted by errors for all samples.

corresponds to 0.20 ± 0.05 μGy/h. If to exclude from the calculation the increased DR values for the three samples, which go beyond the double error, then the average DR is reduced to 1.50 ± 0.20 mGy/year (0.17 ± 0.02 μGy/h).

The observed elevated values of DR can be considered as outliers. However, these data can probably be explained as being caused by increased radiation exposure of the individuals from whom these samples were obtained.

For the sample #5, the obtained DR is suspiciously high (almost 10 times higher than the average level and it was obtained with the large relative error. For samples #105 and #162, the excess of DR over the average level is from 3 to 5 times. Such differences can be expected for the territory of Stepnogorsk. As noted in the publication (Kanaeva 2010; Dzhumailova 2011), in the territory of Stepnogorsk, according to conventional dosimetry data, ADER varied up to 4 times in different places of this city. In the surrounding area, in the residential area, in some locations ADER exceeded the normal level up to 20 times. Therefore, it is possible that the observed elevated DR for donors of these samples may be caused by some accidental irradiation or by long staing in places with elevated radiation levels.

3.4. Analysis of the distribution of EPR doses and of the average annual EPR dose by different locations of the city (microdistricts)

It is of interest to study the radiation background in different parts of the city. As noted above, Stepnogorsk is divided into microdistricts, the numbers of which are indicated in the addresses. Moreover, the numbers of the microdistricts increase in the north-eastern direction, namely in the direction of the location of the uranium mining dumps.

It is impossible to judge the level of radiation exposure by the value of the accumulated EPR dose, since the ages of tooth donors are different. Therefore, we analyzed differences between microdistricts in terms of the annual dose rate.

The annual individual EPR dose rate for different microdistricts is presented in Fig. 7A. The number 0 denotes the group of measurements for which the microdistrict numbers are unknown. According to the multi-factor analysis of variance (ANOVA and the Kruskal-Wallis nonparametric multi-factors test), no significant difference was observed between the mean and the median DR for microdistricts. Such conclusion was obtained for both the full data and the outlier-excluded data.

The Fig. 7B shows the average weighted by the error values of DR for each microdistrict. For clarity, in order to highlight data with increased accuracy, the size of the points is shown proportional to the reciprocal value of the error. That is, contribution of the data points (weight factors) when assessing the weighted average corresponds to their area. Visually, it can be seen that the average DR with a large statistical weight tend to increase with an increase in the microdistrict number, that is, in the direction to the north-east.

3.5. Comparison of EPR dose rates with ADER obtained by conventional instrumental dosimetry

The ambient dose equivalent rate levels, ADER, were measured in the residential area of Stepnogorsk in various microdistricts using a conventional dosimeter. In each microdistrict three measurements were performed. The average value for all measurements is 0.23 ± 0.01 μSv/h, corresponding to the annual dose rate 2.0 ± 0.1 mSv/year. Results of all ADER measurements and the values averaged for each microdistrict are presented in Fig. 8. As it can be seen visually, there is a tendency of ADER to increase for microdistricts with numbers 6 to 10. These microdistricts are placed in the north-east part of the city, as it is seen in Fig. 1C. It is just in this direction where the waste dumps of uranium production are located and the increased radiation level is observed. For this dependence, very small correlation coefficient R, is obtained. The observed weak correlation is probably due to the small sample number

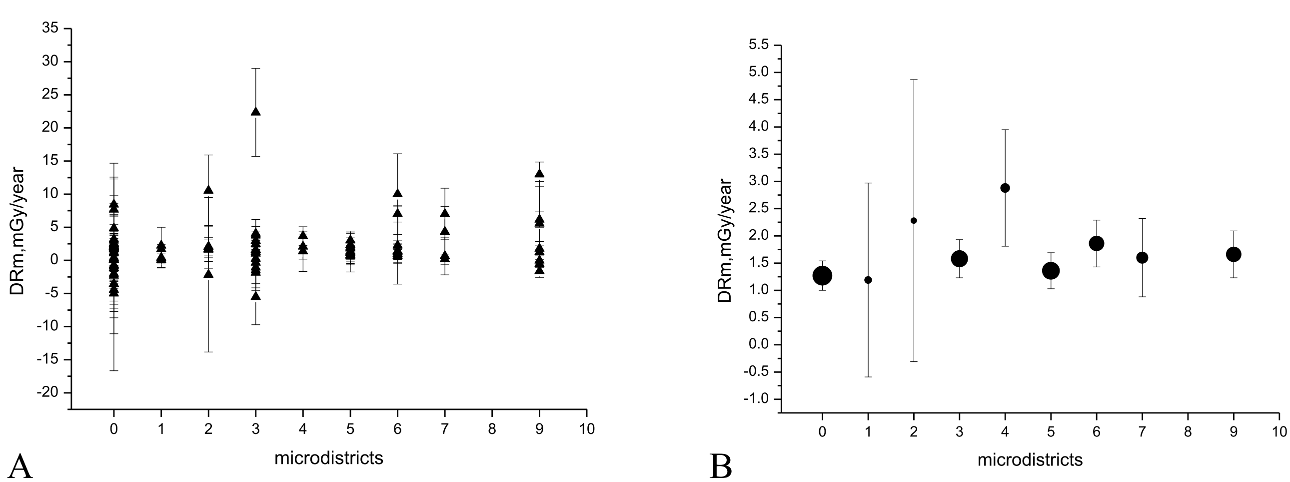

Fig. 7. The annual EPR dose rates, estimated for various microdistricts: A - individual dose rates for all samples, B - Mean dose rates for different microdistricts with indication of the errors of the mean in the form of bars, as well as with the size of the points proportional to the reciprocal error.

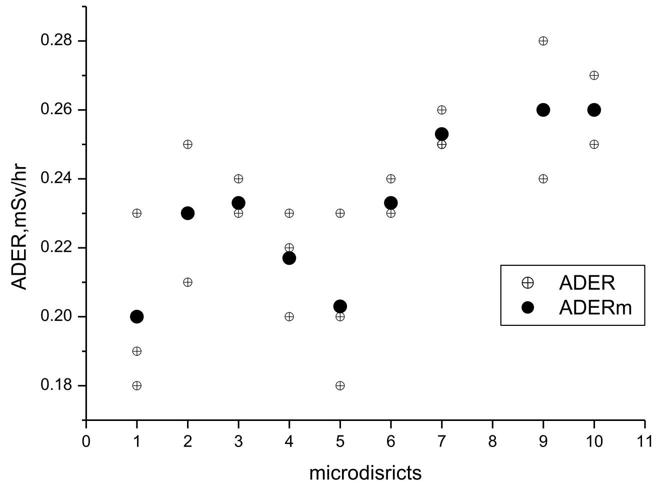

Fig. 8. The results of measuring of the ambient dose equivalent rate (ADER) near residential premises in different microdistricts versus the microdistrict number (data notations: blank points - separate measurements, dark points - mean values for microdistricts).

for each microdistrict and the high scattering of the EPR doses in relation to the small observed differences in ADER between microdistricts.

The EPR dose rates grouped by microdistrict were compared with the average ADER obtained for each microdistrict Fig. 9. For the averaged DR by microdistricts, an unclear dependence on the values of ADER averaged by microdistricts is observed. It became clearer if to show the data by points with a size connected with their accuracy, inversely proportional to the error. Their area corresponds to the statistical weight used in calculating the linear regression weighted by data errors. For these data, the weighted linear regression with a slope close to unity and $R = 0.32$ was obtained, at $P = 0.41$. This indicates the presence of a connection in the form of a trend between the averages of DR and ADER for the microdistricts.

Thus, there is very weak correlation between dose rates obtained by the conventional dosimetry and by EPR. The reason of that is very high data scattering for DR and the small number of data for each microdistrict measured by EPR. In addition, DR and ADER were measured for different objects and are difficult to compare directly. DR were measured for peoples, which do not stay in same place, and ADER were measured in fixed locations.

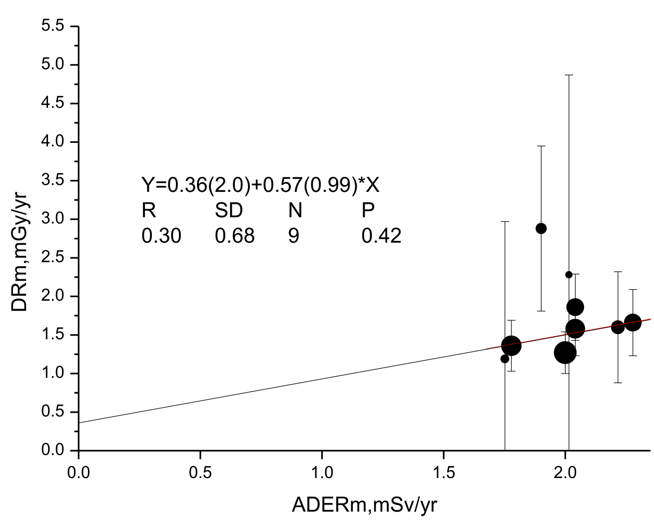

Fig. 9. The relationships between the mean annual EPR dose rates and the average ADER for the microdistricts, and the results of calculation of the linear regression weighted by errors. The sizes of data point are shown proportional to the errors of mean, which are shown by bars.

However, mean dose rates determined by EPR for all samples (1.75 ± 0.45 mGy/yr or 0.20 ± 0.05 μGy/hr) and measured by the conventional dosimeter (2.0 ± 0.01 mGy/yr or 0.23 ± 0.01 μSv/hr) are very close with each other (the two means are not different with probability of disagreement $P < 0.05$). After excluding the outlier results for the three samples, the mean DR is appeared to be 1.50 ± 0.30 mGy/yr (or 0.17 ± 0.03 μGy/hr). It is also consistent with average ADER ($P < 0.14$). The observed difference is within the error limits of both methods, given that the systematic error of the data of each method is at least 15 %.

The average ADER for Stepnogorsk obtained from our data according to the instrumental dosimetry is about 23 μSv/h, which is consistent with the average value of 22 μSv/h for Stepnogorsk in accordance with the data from the publications (Kanaeva 2010; Dzhumailova 2011). As it is noted in these publications, for the individual locations of Stepnogorsk, a strong spread of ADER ranging from 0.12 to 0.50 μSv/h was observed. Moreover, differences in the average ADER for different parts of the city were observed. In the center and western part of the city the average ADER 0.20 μSv/h was recorded, in the southern part - 0.12 μSv/h.

For the actual values of the individual dose rate to which residents of Stepnogorsk are exposed, there may be significant differences due to differences in individual exposure conditions (influence of different shielding and behavior factors). However, for the measured DR, the actual dose rate values are masked by variations caused by random errors of their estimation, inherent in the EPR method. Although for the majority of measurements, with the exception of outliers, an average value of DR is observed which corresponds to the normal background level of 0.11 μSvh.

Only for some measurements, the significantly high DR values are observed. Namely, for samples #105 and #162, the DR values were about 0.7 μGy/h and 1.5 μGy/h respectively, which significantly exceed the bulk normal level. These high DR may be caused by some accidental exposure or by prolonged staying in locations with high radiation level.

The EPR method of dose determination with tooth enamel provides a unique opportunity to determine retrospectively accumulated doses over a long period for a person directly, rather than through dose reconstruction based on the indirect measurements by conventional dosimetry. This is its value and justification for use, despite the existing shortcomings. Potential limitations and disadvantages of the EPR method of dosimetry on tooth enamel are the large error in estimating the individual dose and, accordingly, the large error in estimating the dose accumulation rate.

Therefore, to obtain reliable data in the range of low doses close to background values, measurements for one patient are not enough, but it is necessary to obtain data for a sufficiently large group of population and analyze by averaging in order to obtain an acceptable error of the result. Individual doses and annual dose rates were obtained with a large error. However, when averaging across population groups, the accuracy increases significantly, and the average values make possible to judge with confidence about the level of radiation exposure.

As a result of the conducted studies, the average dose accumulation rate for the population group of Stepnogorsk was estimated by the EPR method. Despite significant individual error, the average value of DR for the all population group was obtained with an accuracy of about 20 % for the average DR close to the normal background level. This error value is comparable with the uncertainty of conventional dosimeters (about 15 %), which can only estimate local dose rate for a limited period of time.

4. Conclusions

The use of the EPR dosimetry method for tooth enamel made it possible to estimate individual accumulated doses, as well as the average annual dose rate over a period of almost the entire life of the patient. This is unique data, which are impossible to obtain by other methods.

The annual dose rate obtained by EPR averaged for all investigated group of population of Stepnogorsk or averaged for the limited territories of this city are consistent with the data on ambient dose equivalent rate obtained by the conventional dosimetry in these territories.

For the individual EPR doses and dose rates, there is a significant scatter, much of which is due to measurement error. Only for the two persons elevated values were obtained, which can be confidently interpreted as being due to increased radiation exposure.

For the majority of the surveyed population of Stepnogorsk, based on the estimated doses, in general there is no significant impact of uranium mining operations and their waste on the accumulated doses of radiation and annual dose rates. However, for several people, significantly increased accumulated doses, as well as average annual dose rates of radiation were obtained. It is possible that these increased values are due to local technogenic radiation exposure in certain places that they visited or where these people were for a long time.

CRediT authorship contribution statement

Kassym Zhumadilov: Writing – review & editing, Writing – original draft, Supervision, Investigation, Funding acquisition, Conceptualization. **Alexander Ivannikov:** Writing – original draft, Investigation. **Murat Kassymzhanov:** Writing – original draft, Investigation. **Assel Bagramova:** Investigation. **Issa Zhantore:** Investigation. **Sarsenova Samal:** Investigation. **Valeriy Stepanenko:** Writing – review & editing. **Artem Khailov:** Investigation. **Masaharu Hoshi:** Writing – review & editing, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envc.2025.101098.

Data availability

Data are presented in the Supplement and will be made available on request.

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