Tooth enamel ESR dosimetry for Hiroshima ‘black rain’ zone residents
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ABSTRACT

Electron spin resonance (ESR) dosimetry was applied to human tooth enamel in order to obtain individual absorbed doses for victims of the Hiroshima bomb who lived in the ‘black rain’ area. The so-called ‘black rain’ fell in the form of precipitation on the western part of Hiroshima city and the northwestern suburbs within a few hours after the explosion of the atomic bomb on 6 August 1945, and exposed the population in this area. Only three tooth samples were collected from this area. Since the teeth were located at positions 1, 2 and 4, only the lingual portion was used for the analysis. The results showed that the excess dose after subtracting natural radiation for one (position 4; hh1) was background, for the second (position 2; hh2) it was 133 mGy, and for the other (position 1; hh3) it was 243 mGy. Based on these results, we further investigated the radiation dose attributed to dental X-rays and head CT scan. Such dose of the hh3 radiographic examination was estimated to be 57–160 mGy, which implies an additional exposure around 135 mGy after subtraction. On the other hand, the dose data of hh1 after subtracting dental X-rays was negative. This may mean that such additional doses are an overestimation. In addition, the effect of sunlight should be considered, which is the same direction of overestimation. As a result, the residual dose of 140 mGy suggests the inclusion of radiation from the ‘black rain.’

Keywords: electron spin resonance (ESR); tooth enamel; Hiroshima; retrospective dosimetry; absorbed dose

INTRODUCTION

The USA, with the consent of the UK as laid down in the Quebec Agreement, dropped nuclear weapons upon the Japanese cities of Hiroshima and Nagasaki in August 1945, during the final stage of World War II. The two bombings, that killed at least 129 000 people, remain the only use of nuclear weapons for warfare in history.

On 6 August the USA dropped an uranium gun-type atomic bomb (Little Boy) on Hiroshima. Three days later, on 9 August, the USA dropped a plutonium implosion-type bomb (Fat Man) on the city of Nagasaki. Within the first two to four months of the bombings, the acute effects of the atomic bombings killed 90 000–146 000 people in Hiroshima and 39 000–80 000 in Nagasaki; roughly half of the deaths in each city occurred on the first day. During the following months, large numbers died from the effect of burns, radiation sickness, and other injuries, compounded by illness and malnutrition. As it was known that the so-called ‘black rain,’ which may not have been dark in color, but simply contained radioactivity, fell on the western part of Hiroshima city, as well as on its northwestern suburbs, within a few hours after the explosion of the atomic bomb over Hiroshima at 8:15 on 6 August 1945. Many people who were exposed to radiation due
to ‘black rain’ later suffered from various diseases [1]. Purpose of this study is to estimate doses of collected tooth samples and try to explain origin of estimated doses.

Electron spin resonance (ESR) spectroscopy or electron paramagnetic resonance (EPR) spectroscopy is a widely used way of measuring and characterizing electronic materials [2, 3]. It is also used in biomedical analysis and imaging [4] because radicals can be identified and quantitatively determined through this method. The ESR phenomenon occurs based on the Zeeman effects of the unpaired electron in radicals; it can be observed by the resonance absorption of the magnetic wave at the specified frequency. The ESR frequency of the magnetic wave is proportional to the applied direct current (DC) magnetic field.

The ESR spectrum is provided by a frequency sweep of the magnetic wave or a sweep of the DC magnetic field strength. However, sweeping the DC magnetic field is widely used in most cases because the resonator that is designed to make resonance at the specific frequency is used to detect the small absorption of the electromagnetic power in the resonance [5]. In ESR spectroscopy on the frequency axis, the control system of the DC magnetic field can be eliminated, and this enables development of the radical sensors which integrate the ESR measurement circuit.

ESR dosimetry is one of the tools for retrospective individual dose reconstruction [5–10]. This method can help to estimate the individual absorbed doses more than 50 years after the exposure event. ESR can measure the number of radicals created by ionizing radiation exposure in tooth enamel. The detection threshold of this method is relatively low, i.e. approximately 30 mGy in terms of absorbed dose.

### MATERIAL AND METHODS

Japanese volunteers and scientists from Japan, Russia and Kazakhstan made an additional effort to determine individual absorbed doses of those exposed to nuclear atomic bomb victims, by means of the ESR method. A total of three tooth samples of two atomic bomb survivors living in Hiroshima were included in the study (Table 1). The subjects were residents of the ‘black rain’ area who lived in the Yasu-Minami district near the border between the heavy rain area and the light rain area of ‘black rain’ area [1]. The place is located about 9.8 km north/northwest of the hypocenter in Hiroshima, so, they have no direct exposure to the Hiroshima atomic bomb.

Medical histories and records of X-ray procedure of the jaw and CT examinations were found in the Dental Hospital. Tooth samples were cut to their buccal and lingual parts. For these teeth, the buccal part was not included to the dose estimation procedure because their position within the oral cavity did not exclude the possibility of sunlight exposure, as the latter could affect the ESR signal [6, 11]. Enamel was mechanically separated from dentine using hard alloy dental drills and diamond saws. Dentin was removed carefully with a low-speed rotation drill machine, in order to prevent the sample from heating as the latter can induce a mechanically-induced signal and significantly change its shape [11]. The tooth enamel was crushed by cutting pliers to chips of 0.5–1.5 mm grid. Enamel samples for calibration curve were prepared from molars collected as control from the population of Hiroshima city (Table 1). Tooth enamel powder prepared from different teeth was pooled together and split into aliquots of 100 mg. The resulting aliquots were irradiated to doses of 0, 100, 200, 300, 500 mGy, respectively, from a collimated 60Co source at Hiroshima University [12, 13]. The standard uncertainty of the radiation dose is estimated as 3%.

The ESR measurements were carried out at least 10 days after irradiation and sample preparation, so that all transient radiation- and mechanically-induced signals have faded out or come to an equilibrium state [6]. All measurements were performed at stabilized room temperature of 21°C using an X-band ESR spectrometer JEOL JESFPA100 equipped with a high Q-factor cylindrical TE011 cavity model ES-UCX2, and keeping the spectrum recording parameters similar to those previously reported [14]. A specially designed computer software [10, 15] was used for extraction of the radiation-induced signal (RIS) from the total ESR spectrum and determination of its intensity (or peak-to-peak amplitude). The ESR spectrum of irradiated enamel was divided into the RIS and the background signal (BGS) by applying a non-linear least square fitting of the model spectrum describing RIS and BGS in an analytical form. The spectrum processing procedure has been applied with description of the BGS by an asymmetric narrow component and a wide component composed of superposition of Gaussian derivative functions. A fitting window of 3.0 mT width was used for all the enamel spectra (the left border of the fitting window was −1.0 mT and the right border +2.0 mT relative to the maximum of the BGS) [14, 16]. The parameters of the calibration signal were determined as the slope and intercept of the regression line of the RIS amplitude, normalized by the samples mass and by the amplitude of the marker signal, plotted versus the nominal doses. The fitting procedure was performed in the automatic mode of the processing program. All intercept values were corrected by the dose absorbed in the calibration samples from natural background. Based on the age of the tooth samples’ donors, the mass of enamel and the enamel formation age that depends on tooth position [6], the intercept value obtained at calibration was estimated at 24 mGy [12], and was used to obtain the unbiased dose level.

On Figs 1 and 2 provide examples of calibration curves for four times repeated spectra measurements (Fig. 1a), and after averaging over the results obtained from four repeated spectra measurements (Fig. 1b), and example of calibration spectrum with a dose of 500 mGy (Fig. 2), respectively [12, 17].

In principle, the experimentally-determined dose is considered to consist of several contributions: (i) the dose from natural radiation background accumulated during the lifetime of the tooth enamel; (ii) the dose from any dental X-ray examination; and (iii) the dose from solar UV light as well as the dose received as a result of nuclear tests (excess dose). The latter contribution is the subject of interest for

### Table 1. Tooth enamel samples and donors’ descriptions

| No | Sample code | Birth year | Age of Enamel formation | Year of Enamel formation | Tooth position |
|----|-------------|------------|-------------------------|--------------------------|----------------|
| 1  | hh1         | 1937       | 02.06.1937              | 1942                     | 4              |
| 2  | hh2         | 1940       | 02.07.1940              | 1944                     | 2              |
| 3  | hh3         | 1940       | 02.07.1940              | 1944                     | 1              |
the present dose reconstruction effort. The intensity of the RIS was converted to a dose absorbed by the enamel, $D_{en}$ (expressed in mGy) calibrated using a $^{60}$Co gamma source. The excess dose in the enamel was determined by subtraction of the natural background radiation accumulated after enamel formation from the absorbed dose in the enamel [18].

As shown in Table 1 sample number hh1 has been collected from one person, while samples hh2 and hh3 from another person. For this analysis we used only the lingual side of teeth, because of the effect of UV upon buccal side. The lifetime of the tooth enamel was obtained by subtraction of the average age of the tooth formation for a given tooth position from the age of a person at the moment of measurement.

**RESULTS AND DISCUSSION**

The absorbed doses with their uncertainties were obtained as result of the spectra processing as described above. In order to minimize the effects of any unknown X-ray diagnostic contribution to the dose, and to reduce effect of solar light, only the results obtained for the lingual part have been used for further analysis [18]. In Table 2, individual excess dose determinations for different years of enamel formation are shown. The age of tooth enamel formation was determined according to data from previous publications [5, 6]. For one dose, negative value was obtained. This is not unexpected since the measurements were taken near the method’s sensitivity threshold. Naturally, some values become negative as a result of their statistical distribution, determined due to experimental errors. Negative doses are possibly the result of underestimating the uncertainty in the dose estimate. A unique source may be the individual variation in the background dose component, which is estimated from measurements of reference samples collected in different places [18].

The excess dose caused by ionizing radiation—probably due to the atomic bomb test—has been determined by subtracting the contribution from natural background radiation (equation 1):

$$D_{ex} = D_{en} - TA \times D_{b}$$

where $D_{en}$ is the absorbed dose was calculated by the evaluation software (mGy), $TA$ is the tooth enamel age (y), and $D_{b}$ is the background dose of 0.8 mGy/y [19].

The corresponding uncertainty of dose determination was determined based on a semi-empirical formula used in [14, 16].

The standard uncertainty in dose determination ($E_r$) has been determined using a semi-empirical formula similar to that used in the previous edition (equation 2):

$$E_r \approx E_{r1}^2 + E_{r2}^2 + (E_{r3}D_{en})^2$$

where: $E_{r1} = 25$ mGy is the contribution caused by extraneous
signals in the enamel spectrum, variations in the FS line shape and the uncertainty of the offset level during calibration. In this equation, $E_{r3} = 0.12 \text{ mGy}$ and it represents the dose-dependent contribution caused by variation in enamel sensitivity and uncertainty in determining the calibration of the calibration line. Moreover, $D_{en}$ is the ESR-determined experimental dose absorbed by the enamel. $E_{r1}$ is a contribution caused mainly by noise in the spectra and by the instability of the spectrometer; this parameter varies for different samples and increases with decreasing sample mass. In this work, $E_{r1}$ is determined on the basis of the error in determining the RIS amplitude during spectral processing by converting it to dose units using the calibration of the calibration straight line. The dependence of this parameter on the sample mass is characterized by the following equation (equation 3):

$$E_{r1} = E_{r1,m} / (m/100)$$  \hspace{1cm} (3)

where: $E_{r1,m} = 20 \text{ mGy}$; $m/100$ is the mass of the sample (in mg) reduced to 100 mg \[12,17\].

For the dental X-rays and other computed tomography (CT) examinations, the examination records of the Dental Hospital were examined. The results found in the records showed that the survivor hh1 had undergone a panoramic six times and a dental X-ray seven times, while the survivor hh2/hh3 had received a head CT 13 times and a dental X-ray nine times.

Assuming: (i) a dental X-ray dose of 1 mGy per one exposure \[20\]; (ii) a panoramic dose of 2.5 mGy; (iii) an X-ray enamel sensitivity of six to eight times \[21,22\]; and (iv) a head CT dose of 0.3–6 mGy per dose \[23\], hh1 had received a dose range of 142–189 mGy. For the hh2/hh3 survivor, the corresponding dose range was calculated at 58–150 mGy.

In the case of hh1, the total dose after subtraction was found to be negative. One reason is that the dental X-rays may not have been in the exposure area. In such cases, the doses to the teeth will be much smaller. In addition, the effect of sunlight should be considered, which is the same direction of overestimation. As the other reason, there may have been some other problems with the ESR dose estimation; however, the causes were not known.

On the other hand, in the other samples of hh2 and hh3, hh2 was almost background and hh3 had a remainder of around 140 mGy, therefore, this could suggest an exposure dose by the ‘black rain.’

**CONCLUSION**

Individually-absorbed doses were measured for residents of the Hiroshima City who were affected by radiation from the ‘black rain’ soon after the explosion of an atomic bomb. They are not directly exposed to the Hiroshima atomic bomb. In one sample, hh1, the residue minus the dental X-ray exposures was negative. In hh2, the dose was close to the background radiation. On the other hand, in hh3, the residue was found to be around 140 mGy, which may imply the exposure to Hiroshima ‘black rain.’ Further studies are necessary, since the sample size is not yet sufficient. This study showed importance in continuation of it in further collections of biomaterial including bone samples.

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**CONFLICT OF INTEREST**

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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