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EPR tooth dosimetry of SNTS area inhabitants

Sergey Sholom^a, Marc Desrosiers^b, André Bouville^c, Nicholas Luckyanov^c, Vadim Chumak^a, Steven L. Simon^{c,*}

^aScientific Center for Radiation Medicine, Melnikova str., 53, Kiev, Ukraine

^bIonizing Radiation Division, National Institute of Standards and Technology, Gaithersburg, MD, USA ^cDivision of Cancer Epidemiology and Genetics, National Cancer Institute, National Institutes of Health,

6120 Executive Boulevard, Bethesda, MD, USA

Abstract

The determination of external dose to teeth of inhabitants of settlements near the Semipalatinsk Nuclear Test Site (SNTS) was conducted using the EPR dosimetry technique to assess radiation doses associated with exposure to radioactive fallout from the test site. In this study, tooth doses have been reconstructed for 103 persons with all studied teeth having been formed before the first nuclear test in 1949. Doses above those received from natural background radiation, termed "accident doses", were found to lie in the range from zero to approximately 2 Gy, with one exception, a dose for one person from Semipalatinsk city was approximately 9 Gy. The variability of reconstructed doses within each of the settlements demonstrated heterogeneity of the deposited fallout as well as variations in lifestyle. The village mean external gamma doses for residents of nine settlements were in the range from a few tens of mGy to approximately 100 mGy. © 2007 Elsevier Ltd. All rights reserved.

1. Introduction

The accuracy of retrospective dosimetry for inhabitants of the Semipalatinsk area remains a topical problem. Estimates of absorbed dose in the same settlements of this region obtained using various techniques (i.e. EPR dosimetry with teeth, luminescence dosimetry with quartz, and analytical dose reconstruction based on measurements of exposure rate or ¹³⁷Cs ground deposition) may vary several fold (Bailiff et al., 2004; Ivannikov et al., 2006; Stepanenko et al., 2006b). Furthermore, even for the same technique (e.g. model-based dose reconstruction), the estimated doses within a settlement may differ significantly (Stepanenko et al., 2006a). The village of Dolon is a good example, where reported estimates from different techniques cover the range from 0.1 to 2 Gy. This large range highlights the need for additional measurements with high-precision dosimetric techniques and subsequent improvement to the calibration aspects of the calculation techniques.

EPR dosimetry with tooth enamel is known as one of the most accurate and reliable dosimetric techniques (Chumak

et al., 1999; IAEA, 2002). Its distinct advantage is that the accident doses are derived from physical measurements of human tissues. This is especially important for a non-uniformly contaminated area, e.g. the various villages around the SNTS, where fallout clouds from individual nuclear tests varied considerably in their directions of travel and the amount of fallout deposited (Gordeev et al., 2006).

The goal of the present study was the reconstruction of accident doses for SNTS area inhabitants using EPR measurements of teeth. The purpose of the measurements is to support an ongoing epidemiologic study presently being conducted by the US National Cancer Institute (NCI).

2. Materials and methods

2.1. Phases of study

EPR measurements were made in two phases (2002 (see Desrosiers, 2003) and in 2005) at the US National Institute of Standards and Technology (NIST) and the Ukrainian Scientific Center for Radiation Medicine. Doses for 103 persons from nine settlements were reconstructed using EPR measurements.

^{*} Corresponding author. Tel.: +301 594 1390; fax: +301 402 0207. *E-mail address:* ssimon@mail.nih.gov (S.L. Simon).

While similar equipment was used, there were modest differences in techniques and spectra interpretation procedures.

2.2. Teeth

Seventy-two teeth (both molars and front teeth) were measured in 2002 and 32 molars were measured in 2005. Teeth were obtained from inhabitants of eight villages (Dolon, Kainar, Kanonerka, Sarzhal, Novopokrovka, Korosteli, Bolshaya Vladimirovka, Karaul) and one metropolitan city (Semipalatinsk City) following normal extraction for medical purposes. All teeth were formed before 1949, the year of the first nuclear test at the STNS.

2.3. Specifics of measurement techniques

The EPR sample preparation procedures in 2002 included the following steps: separation of the tooth crown; 20 h ultrasonic treatment (Misonix, Inc.) in aqueous 10% KOH at 70 °C; washing, drying, crushing and sieving to a grain size range of 0.1-0.5 mm. Quantification of the radiation-induced signal was determined with a software program for spectral deconvolution of experimental EPR spectra (Koshta et al., 2000). A NIST-calibration curve was used to assess the dose for each tooth enamel sample. A reasonable estimate of the combined uncertainty (one sigma) for the accident doses measured by the 2002 technique is approximately ± 40 mGy for doses lower than 300 mGy.

The steps for EPR measurements used in 2005 technique including sample preparation, spectral deconvolution, and uncertainty propagation are described elsewhere (Sholom et al., 2006; Sholom and Chumak, 2003). The additive-dose method was applied to all samples using additional irradiation with a calibrated ¹³⁷Cs gamma source. The uncertainties in the estimates of the accident doses were derived from an analysis of the individual uncertainties for the buccal and lingual components, the background dose and the calibration coefficient; the uncertainties were estimated to be approximately ±30 mGy for doses lower than 300 mGy and approximately ±10% of the dose estimate for doses higher than 300 mGy.

The EPR spectra were obtained using a Bruker ESP 300E spectrometer (2002 technique) and a Bruker ECS 106 spectrometer (2005 technique) with a cylindrical 4108 TMH microwave cavity. The spectra were recorded with the following experimental parameters for the 2002 and the 2005 technique, respectively: magnetic field sweep, 5.0 and 10 mT; microwave power, 16 and 10 mW; modulation frequency, 100 kHz (for both techniques); modulation amplitude, 0.32 and 0.4 mT; number of scans, 81 and 120. Three to five EPR spectra were taken for each sample in order to estimate the stochastic component of uncertainty. The sample was shaken before recording each spectrum to randomize the enamel grains. A typical spectrum is shown in Fig. 1 for a sample that had received a dose of about 300 mGy. In this figure, the dots are the experimental measurements while the solid line is the best fit to the data.



Fig. 1. Example of spectrum for a tooth sample that received approximately 300 mGy. Dots represent the experimental measurements while the solid line is a best fit to the data.

Since the quantity of interest is absorbed dose, the equivalency of the 2002 and 2005 measurement phases is derived from the traceability of the dosimetry for each phase to the NIST-calibrated gamma source that serves as the US national reference. Differences that may arise from the measurement technology are captured in the uncertainty assigned to each measurement phase. The 2002 and 2005 measurement phases similarly accounted for the accumulation of background radiation dose by assuming 1 mGy per year of external gamma dose.

3. Results and discussion

Seventy-six lateral teeth and 27 front teeth were measured from 103 inhabitants of eight villages and one city near to the SNTS. Summary statistics of the accident doses reconstructed by EPR methods, as described earlier, are presented in Table 1.

A comparison of dose estimates to lateral teeth, stratified by village of residence at the time of tooth extraction, distinguished two groups based on similar median doses. One group was defined by village-median doses in the range of 30–60 mGy (Kanonerka, Korosteli, Sarzhal, Semipalatinsk, Kainar) and the other with village-median doses of 90–100 mGy (Novopokrovka, Dolon, Karaul, and Bolshaya Vladimirovka) (see Fig. 2). EPR measured doses, however, were quite variable within each village, likely reflecting several different phenomena including spatial heterogeneity of the deposited radioactive fallout as well as individual behavioral and lifestyle differences, e.g. the amount of time spent in and outdoors, and differences of construction materials of residences. Coefficients of variation (CVs, in %) within villages ranged from 50% to 200%.

Consistent with the high CVs, the distributions of estimated doses at each village (lateral teeth) were highly positively skewed. Several villages had a single estimated dose much higher than the other doses received in that village. This was true, for example, for Kanonerka (median = 58, max =1273 mGy), Dolon (median = 89, max =1788 mGy), Sarzhal (median = 44, max =1054 mGy) and Semipalatinsk

Table 1											
Summary	statistics of	accident	doses (mGy)	estimated by	EPR by	village a	nd by	position i	n mouth	(lateral	or front)

	Bolshaya Vladimirovka	Dolon	Kainar	Kanonerka	Karaul	Korosteli	Novopokrovka	Sarzhal	Semipalatinsk City
Lateral teeth									
Number of samples	5	13	8	7	12	11	5	12	2^{a}
Minimum	30	20	0	12	0	0	5	0	5
Maximum	146	1790	345	1270	306	171	92	1050	44 ^a
Mean	98.4	222.0	68.9	230.0	129.0	70.5	63.4	159.0	24.5
Median	105.0	89.0	31.5	58.0	88.5	58.0	83.0	44.0	24.5
Std. error	21.8	131.0	40.6	174.0	28.0	17.5	16.5	85.9	19.5
CV (%)	49.4	213.0	167.0	200.0	75.0	82.4	58.1	187.0	112.0
Front teeth									
Number of samples	5	2	2	5	2	1	7	3	_
Minimum	156	66	25	17	119	156	49	0	_
Maximum	354	308	326	138	179	156	226	65	_
Mean	249.0	187.0	176.0	90.8	149.0	156.0	149.0	21.7	_
Median	250.0	187.0	176.0	92.0	149.0	156.0	152.0	0.0	_
Std. error	34.8	121.0	151.0	20.9	30.0	_	22.9	21.7	_
CV (%)	31.3	91.5	121.0	51.5	28.5	-	40.6	173.0	-

^aA third tooth from Semipalatinsk was measured with a value of 8890 mGy, however, the dose was not considered to be a SNTS-related accident dose (see text for discussion) and, for that reason, is not included in these summary statistics.



Fig. 2. Box plot of accident dose (mGy) in lateral teeth from persons living in nine villages near to the Semipalatinsk nuclear test site. Note that five outlier data points are not shown, four that are considered as plausible accident doses (Dolon: 1800 mGy, Kainar: 350 mGy, Kanonerka: 1275 mGy, Sarzhal: 1050 mGy) and one value (Semipalatinsk City: 8890 mGy) that was likely a result of medical radiotherapy. Each gray bar includes 50% of data, median is shown as a horizontal line within the bar, end caps on vertical lines from each gray box show minimum and maximum values except for outliers.

City (median = 44, max = 8894 mGy). Grouping of the villages by village-mean dose rather than by median dose would give different groups because the mean values are greatly affected by the few very high values (greater than 1 Gy), while the median dose values for each village presented (Table 1) are more stable estimates of central tendency. The four very high values (one each in the villages of Dolon, Kanonerka, Sarzhal, and Semipalatinsk City) are clear outliers. These values are not considered to be measurement outliers (because the precision of the measurements at those dose levels is $\pm 10\%$ or less), but rather to be non-typical doses that should be verified by other types of information. At the present time, two possible reasons for these outliers have been considered. The first is the likelihood of having received radiotherapy to the head and neck; the person from Semipalatinsk whose dose is close to 9 Gy is probably an example of such a case. A second explanation is that the heterogeneity of radioactive fallout after nuclear tests, resulted in "hot spots" at which the individual spent significant amounts of time. This is likely to be the case for the three persons from Kanonerka, Dolon and Sarzhal.

About one third as many front teeth as lateral teeth were measured (using only corresponding lingual parts). CVs for front teeth were nearly equivalent for equal sample sizes. Comparing median front teeth doses to median lateral teeth doses, stratified by village, indicated that lingual parts of front teeth generally received higher exposures by a factor of 2 to 3. Because the front and lateral teeth measured were not from the same individuals, however, it is not possible to deduce a precise difference in exposure of front teeth compared to lateral teeth. Front teeth are generally considered as less reliable than lateral teeth for EPR dosimetry since the EPR signal from front teeth can be significantly influenced by exposure to UV radiation (even for lingual parts, Sholom et al., 2001).

There still remains some discrepancy between EPR reconstructed doses and doses reconstructed either from historical exposure rate measurements or from contemporary soil ¹³⁷Cs measurements, though it is difficult to generalize the level of disagreement since there are also differences in reconstructed estimates from different investigators. The village of Dolon has received the most attention and comparisons can be best made for that location. Based on environmental measurements, reconstructed doses in air have been reported in the range of 466–780 mGy (Imanaka et al., 2006; Stepanenko et al., 2006a) and whole-body doses have been estimated to be about 500 mGy (Simon et al., 2006). In comparison, the tooth doses derived from our EPR measurements for lateral teeth from Dolon (median = 89 mGy, mean = 221 mGy, max = 1.8 Gy) are substantially smaller, and whole-body doses estimates as low as 180 mGy have been obtained for Dolon using measurements of stable chromosome aberrations, i.e. by FISH (Chaizhunusova et al., 2006). Further study may explain the reasons for these differences.

4. Conclusions

External doses to teeth from exposure to residual fallout radioactivity from the SNTS were determined by the EPR technique for inhabitants of nine settlements located near to the Semipalatinsk nuclear test site. The nuclear tests-related doses ranged from zero up to approximately 2 Gy, excluding one case of about 9 Gy that in all likelihood was the result of radiotherapy as a medical treatment.

A high variability among teeth from within each village was found while differences between village average doses were no more than a factor of two. Estimates of external dose for Dolon made by model-based dose reconstruction techniques are several times greater than the village median or village average EPR dose estimates; these differences are yet to be explained.

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