Data of ESR dosimetry study of population in the vicinity of Semipalatinsk Nuclear Test Site

ZHUMADILOV Kassym[1], IVANNIKOV Alexander[2], STEPANENKO Valeriy[2], ZHARLYGANOVA Dinara[3], ZHUMADILOV Zhaxybay[4], APSALIKOV Kazbek[5], TOYODA Shin[6], ZHUMADILOVA Anara[3], ENDO Satoru[7], TANAKA Kenichi[8], MIYAZAWA Chuzou[9], YAMAMOTO Masayoshi[10], OKAMOTO Tetsuji[11] and HOSHI Masaharu[1]

[1]*Research Institute for Radiation Biology and Medicine, Hiroshima University* kassymzh@yahoo.com

[2]*Medical Radiological Research Center, Obninsk, Russia*

[3]*Astana Medical University, Astana, Kazakhstan*

[4]*Nazarbayev University, Life Sciences Center, Astana, Kazakhstan*

[5]*Scientific-Research Institute for Radiation Medicine and Ecology, Semey, Kazakhstan*

[6] *Okayama University of Science, Okayama, Japan*

[7]*Graduated School of Engineering, Hiroshima University, Japan*

[8]*Center of Medical Education, Sapporo Medical University, Japan*

[9]*School of Dentistry, Ohu University, Koriyama-shi, Japan*

[10] *Low Level Radioactivity Laboratory, K-INET, Kanazawa University, Japan*

[11]*Graduate School of Biomedical Sciences, Hiroshima University, Japan*

Abstract

The method of electron spin resonance (ESR) dosimetry was used to human tooth enamel to obtain individual absorbed doses of population of settlements in the vicinity of the Semipalatinsk Nuclear Test Site (SNTS), Kazakhstan. The distances between investigated settlements and Ground Zero (SNTS) are in the range 70 - 200 km from SNTS. Most of settlements (Dolon, Mostik, Bodene) are located near the central axis of radioactive fallout trace from the most contaminating surface nuclear test, which was conducted in 29, August 1949. The other settlements located close to radioactive fallout trace as a result of surface nuclear tests in 24, August 1956 (Ust-Kamenogorsk, Znamenka, Shemonaikha, Glubokoe, Tavriya, Gagarino), in 7, August 1962 (Kurchatov). Semipalatinsk city was included to investigation as a biggest city which located close to SNTS.

This method was applied to human tooth enamel to obtain individual absorbed doses of residents of Makanchi, Urdzhar and Taskesken settlements located near Kazakhstan-Chinese border (about 400 km to South-East from Semipalatinsk nuclear test site (SNTS) and about 1000 km from The Lop Nor Nuclear Weapons Test Base (China)). Since the ground and atmospheric nuclear tests (1964-1981) at Lop Nor, the people residing in these settlements have believed to be exposed heavily by radioactive fallout. Tooth samples were extracted according to medical reasons in a course of ordinary dental treatment. Kokpekty was chosen as control and was not subjected to any radioactive contamination and located 400 km to the Southeast from SNTS.

1. Introduction

In the period from 1949 to 1962 125 nuclear tests (including 25 near-surface nuclear tests) were conducted at the Ground Zero technical site in the territory of Semipalatinsk Nuclear Test Site (SNTS). The date of 29 August 1949 represents the first nuclear explosion, which contaminated by radioactive fallout a huge territory northeast from the epicenter. The radioactive dust cloud was transferred by the wind and its gradual precipitation formed several radioactive fallout traces [1-3]. There are no data describing the dynamics of the precipitation of the dust from the radioactive cloud on the region adjacent to the SNTS. It is a very complicated procedure to reconstruct the individual and collective radiation doses received by the local population.

On 24 August 1956, the 28th nuclear explosion with 27 kiloton total yield took place, which contaminated with radioactive fallout a huge territory to the east from the hypocenter and near Ust-Kamenogorsk city with radioactive fallout [4, 5]. Reconstruction of individual and collective doses received by the local population is a sophisticated procedure combining data of the radioactive fallout, individual doses measured by ESR, doses reconstructed from retrospective area dosimetry and individual behavior of inhabitants.

The Lop Nor Nuclear Weapons Test Base located in the Malan, Xinjiang Autonomous region of China. China conducted 45 nuclear tests at Lop Nor, including 22 atmospheric and surface tests between 1964 and 1981. According to Gusev et al. [6] about half of the 22 tests were surface tests. Here, three tests in 1964, 1966 and 1971 are explicitly mentioned but only one of them (1966) was expected to affect the population at the Kazakhstan-Chinese border.

ESR dosimetry is one of the useful tools for such dose reconstruction [7]. This method can determine the radiation doses retrospectively even more than 40 years after the exposure event. EPR measures the amount of the stable radicals created by radiation exposure in tooth enamel.

2. Materials and methods

From 2000 to March 2005, 97 teeth samples were extracted on the basis of medical indications from adult residents of Dolon, Mostik, Bodene villages, located near the radioactive fallout trace formed as result of the most hazardous nuclear test of 1949 [8, 9], Kurchatov City and Semipalatinsk City, which is located from 70 to 150 km from SNTS [10]. In 2008, 2009, 88 teeth samples have been extracted according to medical reasons from adult residents of Ust-Kamenogorsk city and the Znamenka, Glubokoe, Tavriya, Gagarino and Shemonaikha settlements [11, 12]. For the period from 2008 to 2009, thirty tooth samples were extracted on the basis of medical indications from adult residents of Makanchi, Urdzhar and Taskesken villages, which are located from 100 to 200 km from the Kazakhstan-Chinese border [13, 14]. 8 teeth were collected as controls from the population of Kokpekty village (400 km east of the test site), which was not subjected to any radioactive contamination. A description of the samples is given in the table 1.

Enamel was mechanically separated from dentine using hard alloy dental drills and diamond saws. Dentins were removed carefully with cooling water in order to prevent the sample from heating which can induce an additional ESR signal and significantly change shape of the signal [7]. Tooth enamel was crushed by cutting pliers to chips 0.5–1.5 mm in diameter. Two samples were prepared from buccal and

lingual parts of each tooth.

Table 1. Information about samples from vicinity of the Semipalatinsk Nuclear Test Site

Settlements	Years of analysis	**Measured**	Enamel formed before 1949	Distance from epicenter, km
Dolon	2002	3	3	100
	2004	26	13	
	2005	9	1	
Mostik	2004	10	8	90
	2005	13	4	
Bodene	2004	20	9	90
Semipalatinsk	2002	9	4	150
Kurchatov	2002	7	5	70
Kokpekty	2004	8	-	400
Total		105	47	

The measurements were carried out in the X-band on the ESR spectrometer JEOL JES-FA100 at stabilized room temperature of 21°C. The spectrometer was equipped with a high Q-factor cylindrical TE_{011} cavity model ES-UCX2. The spectrum recording parameters were the same as previously published [15]. Specially designed computer software [16] was used for spectra processing and dose estimation.

3. Results and discussion

The doses for the residents of Semipalatinsk City and Kurchatov City were included in this report from the published data [17]. The experimentally determined dose was considered to consist of two contributions: dose from natural radiation background accumulated during a tooth enamel lifetime and dose received as a result of nuclear tests (excess dose). The last contribution is subject of the interest for present dose reconstruction. At first, the intensity of the RIS was converted into a dose absorbed by enamel D_{en} (expressed in mGy) calibrated using calibration by a ^{60}Co gamma source. Second, excess dose in enamel was determined by subtraction of contribution of the natural background radiation during the enamel existence after its formation from the absorbed dose in enamel.

For the residents of Dolon absorbed doses were found to be in the range from -24 ± 37 to 496 ± 55 mGy. For some doses, negative values were obtained. This is because the measurements were performed near the threshold of sensitivity of the method. It is natural that some of the values become negative according to their statistical distribution determined due to experimental errors. The negative doses probably are the result of the underestimation of uncertainty of the dose assessment.

For all the samples, excess doses were calculated by equation:

$D_{ex} = D_{en} - TA * D_b$,

Where: D_{en} - dose calculated by automatic program, in mGy

TA - teeth enamel age, years

D_b - background dose, 0.8 mGy/year [2, 3]

Uncertainty of dose determination (*Er*) was determined based on semi-empirical formula used in the previous publication [3].

The average excess dose for enamel formed before 1949 for Dolon is 153 ± 54 mGy, for enamel formed after 1949 the average dose is 25 ± 11 mGy. For Mostik excess dose before 1949 is 19 ± 15 and after 1949 is 44 ± 14. For Bodene excess dose before 1949 is 74 ± 40 and after 1949 is 17 ± 10. For Kurchatov average excess dose before 1949 is 11 ± 20 and after 1949 is 9 ± 27 and for Semipalatinsk City average excess dose before 1949 is 145 ± 68 and after 1949 is 74 ± 35. The value of average dose for Semipalatinsk City is close to the average results of Dolon village [10]. One of the explanations is the person worked in place located close to SNTS and another explanation that they were born in village that affected by fallout from Test Site. The bulk of the excess doses are near the sensitivity threshold of the method.

For control samples, excess doses are from –66 ± 39 up to 24 ± 39 mGy, for Dolon from -74 ± 38 up to 440 ± 106 mGy, for Mostik from –64±32 up to 119± 51, for Bodene from –50 ± 38 up to 356 ± 58 mGy, for Kurchatov from -47±85 up to 56±42 and for Semipalatinsk City from 0±46 up to 268±79. Low doses were found for the group with enamel formed after 1962, the end of atmospheric nuclear tests. The dose values for the group having enamel formed before 1962 are consistent with estimations based on the official registered data indicating high levels of the fallout in the period 1949-1962. The experimentally measured individual doses can be compared with data of dose reconstruction, which were shown in previous publication [8, 9] and which amount was about 0.5 Gy for Dolon.

For Tavriya and Gagarino villages all studied samples have been formed before the date of the nuclear test. For other settlements teeth samples were divided into two parts: before and after nuclear explosion. Low mean excess doses have been found for Gagarino residents, while the highest mean excess dose has been determined for Shemonaikha and Ust-Kamenogorsk residents (Table 2).

Table 2. Information about samples from the settlements located close to radioactive fallout trace as a result of surface nuclear tests in 24, August 1956 in the Semipalatinsk Nuclear Test Site.

Settlements	Population	Archival dose (mSv) [18]	ESR average excess dose (mGy)	ESR maximal excess dose (mGy)
Znamenka	-	25	41±46	268
Glubokoe	11,192	10-15	36±31	83
Tavriya	4,280	10-15	17±36	54
Ust-Kamenogorsk	298,700	80	20±25	120
Gagarino	1,038	10-15	-24±33	47
Shemonaikha	17,000	0.1	17±37	110
Kokpekty	5,301	<0.1	0.01±33	24

Given the small number of tooth samples investigated, the deduced doses should not be seen as representative for the whole population in the selected villages. The maximum dose obtained for samples from Shemonaikha was not expected, due to its large distance from the radioactive trace, but may be due to radioactivity released by some of the uranium enterprises located there. The maximum dose obtained for Znamenka village [17] can confirm that this village locating close to radioactive trace. The other

settlements included in the study also have uranium enterprises, except for Tavriya and Gagarino, where agriculture is prevalent. Average excess doses in the latter two settlements are consistent with estimations based on the official registered data indicating high levels of fallout in the period 1949-1962 [18].

The individual excess dose determinations for different years of enamel formation are shown in table 3. The average excess dose for Makanchi is 62 ± 28 mGy. For Urdzhar, the average excess dose is 64 ± 30 mGy. For Taskesken, the average excess dose is 49 ± 27 mGy and for Kokpekty village, the average excess dose is -19 ± 36 mGy. The average excess doses for the investigated settlement are higher than for the control village.

Table 3. Archival data of external dose estimation [6] and results of the study of the Lop Nor nuclear test site influence to the Makanchi, Urdzhar, Taskesken settlements [14].

#	Date of explosions	External dose (mGy)		
		Makanchi	Urdzhar	Taskesken
1	28.12.1966	6.2	5.33	4.61
2	17.6.1967	220.0	196.0	166.0
3	27.6.1973	341.0	308.0	262.0
4	ESR max excess dose	123.0	118.0	107.0
5	ESR average excess dose	62.0	64.0	43.0

The dose values are consistent with estimations based on the official registered data indicating high levels of the fallout in the period 1966-1981 [19]. This dose estimation is an estimation of external dose only. Some difference between ESR dose estimation and data from Table 1 can be explained by a shielding factor (staying inside house) and a behavior factor (resident's location during the tests and migration).

According to a previous study [6] the population of Makanchi, Urdzhar and Taskesken were not heavily exposed by the Chinese test site or the Semipalatinsk test site. Some results of this study are consistent with archival data [19]. Retrospective analyses were made of the formation of the radiation situation in the population points of the South-east district of the Semipalatinsk region as a result of nuclear weapons testing. In table 3, data of dose estimations from 1967 to 1981 are shown. The territories of the Makanchi, Urdzhar and Taskesken districts were contaminated 11 times by local radioactive fallout from atmospheric, surface and underground explosions conducted at the Lop Nor test site. This was confirmed by the appearance of freshly produced fission products, particularly iodine radioisotopes, ^{90}Sr and others in the environment.

4. Conclusions

Higher average excess doses were determined in Dolon and Semipalatinsk city for residents whose tooth enamel was formed before 1949. Results of dose estimation from Dolon samples are in agreement with the fact that this village is located closer to the axis of the radioactive trace. A result from

Semipalatinsk needs special investigation.

Compared to samples from the distant Kokpekty village which chosen as controls, higher average excess doses have been obtained for Znamenka, Ust-Kamenogorsk city and Shemonaikha. This is in agreement with the fact that Znamenka village and Ust-Kamenogorsk city is locating close to the axis of the radioactive trace, but Shemonaikha is locating on the distance about 70 km from it. It is necessary to note, that the investigated area is well known for its active uranium processing plant. This may explain that higher values have also been found for samples from Shemonaikha, which is located about 70 km from the center line of the radioactive trace. At Tavriya and Gagarino no uranium enterprises exist and, accordingly, the measured doses are consistent with independent estimates of external doses from the fallout that can be found in the literature.

Calculated external doses to the population of Makanchi, Urdzhar and Taskesken from the three main dose-forming explosions are about three times larger than maximal excess doses estimated by the ESR method, but it should be noted that the number of investigated tooth samples is insufficient to make a final conclusion about the influence of the Lop Nor Test site explosions on the population of the Semipalatinsk region near the Chinese border, and required additional investigation. One of the main problems with this kind of study is following the migration of the population that moves from the investigated region into some other part of the Semipalatinsk area.

References

1. Hoshi, M., Toyoda, S., Ivannikov, A., Zhumadilov, K., Fukumura, A., Apsalikov, K., Zhumadilov, Z.S., Bayankin, S., Chumak, V., Ciesielski, B., De Coste, V., Endo, S., Fattibene, P., Ivanov, D., Mitchell, C.A., Onori, S., Penkowski, M., Pivovarov, S.P., Romanyukha, A., Rukhin, A.B., Schultka, K., Seredavina, T.A., Sholom, S., Skvortsov, V., Stepanenko, V., Tanaka, K., Trompier, F., Wieser, A., Wolakiewicz, G., 2007. Interlaboratory comparison of tooth enamel dosimetry on Semipalatinsk region: Part 1, general view. Radiat. Meas. 42, 1005-1014.
2. Ivannikov, A., Zhumadilov, K., Tieliewuhan, E., Jiao, l., Zharlyganova, D., Apsalikov, K.N., Berekenova, G., Zhumadilov, Zh., Toyoda, Sh., Miyazawa, C., Skvortsov, V., Stepanenko, V., Endo, S., Tanaka, K. and Hoshi, M., 2006. Results of EPR Dosimetry for Population in the Vicinity of the Most Contaminating Radioactive Fallout Trace After the First Nuclear Test in tne Semipalatinsk Test site. J. Radiat.Res. 47, A39-A46.
3. Zhumadilov, K., Ivannikov, A., Apsalikov, K.N., Zhumadilov, Zh., Toyoda, Sh., Zharlyganova, D., Tieliewuhan, E., Endo, S., Tanaka, K., Miyazawa, C., Okamoto, T. and Hoshi, M., 2006. Radiation Dose Estimation by Tooth Enamel EPR Dosimetry for Residents of Dolon and Bodene. J. Radiat.Res. 47, A47-A53.
4. Gordeev, K., Vasilenko, I., Lebedev, A., Bouville, A., Luckyanov, N., Simon, S.L., Stepanov, Y., Shinkarev, S., Anspaugh, L., 2002. Fallout from nuclear tests: dosimetry in Kazakhstan. Radiat. Environ. Biophys. 41, 61-67.
5. Gordeev, K., Shinkarev, S., Ilyin, L., Bouville, A., Hoshi, M., Luckyanov, N., Simon, S.L., 2006. Retrospective dose assessment for the population living in areas of local fallout from the Semipalatinsk nuclear test site Part I: External exposure. J. Radiat. Res. 47, A129-136.
6. Gusev, B.I., Kurakina, N.N., Sekerbaev, A.Kh., 2008. Cancer mortality in populations in Kazakhstan subjected to irradiation from Nuclear Weapons Testing in China. Technical report, DTRA 01-03-D-0022. ITT Corporation Advanced Engineering & Science 2560 Huntington Avenue Alexandria, VA 22303-1410.
7. IAEA Report, 2002. Use of electron paramagnetic resonance dosimetry with tooth enamel for

retrospective dose assessment. Report of a coordinated research project. IAEA-TECDOC-1331. Vienna.

8. Imanaka, T., Fukutani, S., Yamamoto, M., Sakaguchi, A., Hoshi, M., 2005. Width and Center-axis Location of the Radioactive Plume That Passed over Dolon and Nearby Villages on the Occasion of the First USSR A-bomb Test in 1949. J. Radiat. Res. 46 (4), 395-399
9. Stepanenko, V.F., Hoshi, M., Dubasov,Yu.V., Sakaguchi, A., Yamamoto, M., Orlov, M., Bailiff, I.K., Ivannikov, A.I., Skvortsov, V.G., Kryukova, I.G., Zhumadilov, K.S., Apsalikov, K.N., Gusev, B.I., 2006.A gradient of radioactive contamination in Dolon village near SNTS and comparison of computed dose values with instrumental estimates for the 29 August, 1949 nuclear test. J. Radiat.Res. 47, A149-A158.
10. Zhumadilov, K., Ivannikov, A., Apsalikov, K., Zhumadilov, Zh., Zharlyganova, D., Stepanenko, V., Skvortsov, V., Berekenova, G., Toyoda, S., Endo, S., Tanaka, K., Miyazawa, C., Hoshi M., 2007. Results of tooth enamel EPR dosimetry for population living in the vicinity of the Semipalatinsk nuclear test site. Radiat. Meas. 42, 1049-1052.
11. Zhumadilov, K., Ivannikov, A., Zharlyganova, D., Zhumadilov, Z., Stepanenko, V., Apsalikov, K., Ali M.R., Zhumadilova, A., Toyoda, S., Endo, S., Tanaka, K., Okamoto, T., Hoshi M., 2009. ESR dosimetry study on population of settlements nearby Ust-Kamenogorsk city, Kazakhstan. Radiat. Environ. Biophys. 48, 419-425.
12. K. Zhumadilov, A. Ivannikov, D. Zharlyganova, Z. Zhumadilov, V. Stepanenko, S. Abralina, L. Sadvokasova, A. Zhumadilova, S. Toyoda, S. Endo, T. Okamoto, M. Hoshi, 2011. ESR dosimetry study for the residents of Kazakhstan exposed to radioactive fallout on 24, August 1956. Radiat. Meas. 46, 793-796.
13. Vanmarcke, H., 2000. Annex C: Exposures to the public from man-made sources of radiation. UNSCEAR 2000: Sources of Ionizing Radiation, pp. 158-291.
14. K. Zhumadilov, A. Ivannikov, D. Zharlyganova, V. Stepanenko, Z. Zhumadilov, K. Apsalikov, S. Toyoda, S. Endo, K. Tanaka, C. Miyazawa, T. Okamoto and M. Hoshi, 2011. The Influence of the Lop Nor Nuclear Weapons Test Base to the population of the Republic of Kazakhstan. Radiat. Meas. 46, 425-429.
15. Zhumadilov, K.S., Ivannikov, A.I., Skvortsov, V.G., Zhumadilov, Zh.S., Endo, S., Tanaka, K., and Hoshi, M., 2005. Tooth enamel EPR dosimetry: selecting optimal spectra registration parameters and effects of sample mass on sensitivity. J. Radiat.Res. 46 (4), 435-442.
16. Ivannikov, A.I., Trompier, F., Gaillard-Lecanu, E., Skvortsov, V.G. and Stepanenko, V.F., 2002. Optimization of recording conditions for the electron paramagnetic resonance signal used in dental enamel dosimetry. Radiat. Prot. Dosim. 100, 531-538.
17. Ivannikov, A.I., Zhumadilov, Zh., Gusev, B.I., Miyazawa, Ch., Jiao, L., Skvortsov, V.G., Stepanenko, V.F., Takada, J. and Hoshi, M., 2002. Individual dose reconstruction among residents living in the vicinity of the Semipalatinsk Nuclear Test Site using EPR spectroscopy of tooth enamel. Health Phys. 83 (2), 183-196.
18. Logachev, V.A., et al., 2008. Nuclear tests in the USSR and their influence on the health of the population of the Russian Federation. M., IzdAT, 470 pp.
19. Gusev, B.I., 1990. Estimation of external exposure doses in the population on the territories of Makanchi and Urdzhar districts of the Semipalatinsk region formed due to Chinese nuclear tests, A Transitional Report (Archives of Kazakh Scientific Research Institute for Radiation Medicine and Ecology). Semipalatinsk.