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Detection limit of electron spin resonance for Japanese deciduous tooth enamel and density separation method for enamel–dentine separation

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ABSTRACT

Electron spin resonance (ESR) dosimetry is one of the most powerful tools for radiation dose reconstruction. The detection limit of this technique using human teeth is reported to be 56 mGy or 67 mGy; however, the absorbed dose of Fukushima residents after the Fukushima Daiichi Nuclear Power Plant (FNPP) accident was estimated to be lower than this detection limit. Our aim is to assess the absorbed radiation dose of children in Fukushima Prefecture after the accident; therefore, it is important to estimate the detection limit for their teeth. The detection limit for enamel of deciduous teeth of Japanese children separated by the mechanical method is estimated to be 115.0 mGy. The density separation method can effectively separate enamel from third molars of Japanese people. As we have collected thousands of teeth from children in Fukushima, the present technique may be useful to examine their external absorbed dose after the FNPP accident.

Keywords: electron spin resonance (ESR); electron paramagnetic resonance (EPR); CO_2^- radical; retrospective dosimetry; Fukushima Daiichi Nuclear Power Plant (FNPP) accident

INTRODUCTION

The Fukushima Daiichi Nuclear Power Plant (FNPP) accident released substantial amounts of radionuclides into the environment. There are serious concerns about the biological effects of radionuclides on humans and animals. From the perspective of the radiation safety, the estimation of the external absorbed dose of Japanese children who lived in Fukushima prefecture during and after the FNPP accident is important. In this work, we utilised electron spin resonance (ESR), also known as electron paramagnetic resonance (EPR) spectroscopy, which is one of the most powerful tools for external absorbed dose estimation.

It is well known that ESR can detect, identify and quantify free radicals. It is commonly used in fields of polymer science [1, 2], dating [3–6], geology [7–9], biology [10–13] and other fields [14–20]. In the field of radiation dosimetry, ESR dosimetry using teeth was applied for the dose reconstructions of the people exposed to the atomic bomb in Hiroshima [21], the nuclear plant accident in Chernobyl [22, 23], nuclear weapons tests, as well as for nuclear industry workers [24–27] and for the FNPP accident in 2011 [28–31], by measuring the long-lived CO_2^- radical in teeth. Thus, ESR dosimetry has been used to investigate the absorbed dose of individuals suffering from severe radiation exposure. However, the detection limit (i.e. the lowest detectable

value) of the ESR technique using human molar teeth is reported to be 67 mGy–561 mGy (measured in three laboratories) [32] or 56 mGy–649 mGy (measured in 14 laboratories, mean value of the detection limit was 205 mGy) [33]. On the other hand, the Fukushima Health Management Survey reported that the maximum individual external does of residents in the Fukushima area after the FNPP accident was approximately 25 mSv [34]. One of our goals is to assess the absorbed radiation dose of children in Fukushima after the FNPP accident via the ESR technique using the deciduous teeth of children living in the ex-evacuation zones of the accident. In addition, the deciduous teeth could be useful for detecting low doses because those should have background doses lower than permanent teeth. Therefore, the estimation of the detection limit of ESR has been a key issue.

Organic materials in dentine can interfere with ESR measurement and bias the ESR-reconstructed dose [26, 35, 36]; thus, it has been necessary to remove dentine by mechanical or chemical means and prepare dentine-free enamel samples. However, from the point of view of the internal absorbed dose, the measurement of the ingested radionuclides (such as ^{137}Cs and ^{90}Sr) is important because it offers the possibility to measure the dose from these radionuclides deposited in the dentine [26, 37, 38]. Thus, it was essential to utilise an enamel–dentine separation technique whereby the dentine could be preserved. Furthermore, as with epidemiological studies that have examined thousands of teeth from children [38] or animals in Fukushima, we needed to prepare and examine a large number of enamel samples. Therefore, it was also essential for our enamel sampling technique to facilitate the efficient separation of enamel from dentine in a short period of time. In the previous work, we applied this density separation method to the teeth of Japanese macaque and successfully prepared dentine-free enamel sample [31].

The present study was conducted to: (i) estimate the detection limit of Japanese deciduous teeth, and (ii) evaluate whether the density separation method can be applied for the enamel preparation for human teeth by comparing CO_2^- radical intensities for Japanese molar teeth enamel separated by the density [31] and mechanical separation methods [26, 39].

MATERIALS AND METHODS
Deciduous teeth of Japanese children
This experiment used eight deciduous molar teeth of Japanese children collected before the FNPP accident. Tooth enamel was separated by 'a mechanical separation method' without any chemical treatments. Each tooth was cleaned with distilled water and the dentine was ground away with a dental bur until only the hollow shell of the harder enamel was left [26, 39]. The separated enamel was crushed into small grains with nippers. Enamel grains with a diameter between 0.425 mm and 1.4 mm were obtained by passing them through sieves [40, 41]. The pooled enamel was distributed into seven quartz tubes with an inner diameter and outer diameter of 4 mm and 5 mm, respectively, and a weight of ca. 150 mg.

The seven identical enamel samples were irradiated by ^{60}Co gamma rays up to 1000 mGy (unirradiated, 50 mGy, 100 mGy, 150 mGy, 200 mGy, 500 mGy and 1000 mGy) by cumulative irradiation with a dose rate of ∼130 mGy/h (±1.4%) at the Takasaki Advanced Radiation Research Institute, National Institutes for Quantum and
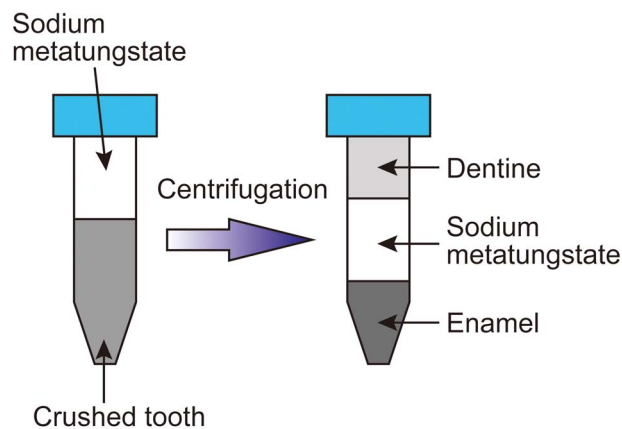

Fig. 1. Enamel was separated from dentine by centrifugation.

Radiological Science and Technology, Takasaki, Japan. A 3 mm-thick polymethylmetacrylate sheet was placed in front of the samples to provide the build-up of secondary electrons.

Before the first irradiation and after each irradiation, ESR measurements for seven samples were performed utilising a model JES-RE2X ESR spectrometer (JEOL, Tokyo, Japan) with a microwave power of 2 mW, a sweep width of ±5 mT, a modulation width of 0.2 mT, a time constant of 0.03 s and a sweep time of 30 s, for 120 scans [26, 27, 42]. To avoid contamination, all the samples were transferred to new quartz tubes before each measurement. The tube was removed from the cavity and shaken, and then placed again into the cavity before every measurement. All the irradiation and measurements were conducted in air at room temperature. Each sample was measured at least four times for each absorbed dose. Observed spectra were deconvoluted by the EPR Dosimetry program [42] to extract the CO_2^- radical intensities.

Third molars of Japanese people
To validate the enamel–dentine separation technique (i.e. density separation method), nine third molars extracted from Japanese people for medical reasons were used. Each tooth was cut into two pieces, and the enamel of one piece of each tooth was separated by the mechanical separation method and crushed into grains as described in the above section for deciduous teeth. The other half of each tooth was first crushed into grains using a cyro-press (Microtex Co. Ltd., Japan). Tooth grains with diameters ranging from 0.425 mm to 1.4 mm were collected by sieving, as described above.

The enamel was then separated from dentine by the density separation method [31, 43–45] using sodium metatungstate (Wako Pure Chemical Ind., Tokyo, Japan) [46, 47]. The principle behind this floating method is that when tooth grains including both enamel and dentine particles are put into sodium metatungstate solution with a density of 2.8 g/cm³, the enamel (i.e. with a density of 2.8 g/cm³–3.0 g/cm³) sinks, while the dentine (i.e. with a lower density of 2.0 g/cm³–2.1 g/cm³) floats in the solution (Fig. 1). Thus, the enamel and dentine form distinct layers after centrifugation of the solution.

Enamel could be separated by removing the dentine layer with a pipette, after which the sodium metatungstate solution was discarded. If the tooth grain is from the enamel–dentine interface, some dentine

may remain in the enamel grain. To make sure to prepare the dentine-free enamel sample or if we observe an irregular ESR signal with the remained dentine [31], treatment of the enamel grains with NaOH solution [31, 32, 48] is effective to remove the remaining small amount of dentine in the tooth grain and to observe an appropriate ESR signal. No irregular signals were observed in this work, we did not treat the samples chemically.

The collected enamel was washed several times with distilled water and dried at 40°C overnight, then they were used for ESR measurements. Each enamel sample was placed in a quartz tube with an inner and outer diameter of 4 mm and 5 mm, respectively, and a weight of ca. 100 mg, then measured at least five times to estimate the CO_2^- radical intensities.

RESULTS AND DISCUSSION
Typical tooth ESR spectrum and spectrum deconvolution

Figure 2(a) shows the typical ESR spectrum of a 500 mGy-irradiated deciduous tooth from a Japanese child (green), fitted spectrum (black) and deconvoluted spectra of radiation induced radicals which are CO_2^- radical (red) and native radical (blue). The ESR spectrum consists of at least five components, including background components, radiation induced components and some unknown component [31, 42]. The major components are the radiation induced components shown as red and blue signals in the figure. One can see that the summation of several functions (fitted spectrum) is similar to the original spectrum. Figure 2(b) shows the ESR spectra for the same deciduous tooth with different absorbed dose. With increasing the absorbed dose, the sharp peak due to CO_2^- radical appears on the shoulder of the ESR spectrum, therefore, it is important to reduce the native radical component due to dentine to extract the CO_2^- radical intensity.

The relationship between the CO_2^- radical intensity and the absorbed dose for deciduous tooth enamel of Japanese children separated by the mechanical separation method

The dose response curve of the CO_2^- radical intensity is shown in Fig. 3. Plots represent the mean value of the CO_2^- radical intensity for each sample, and the error bars represent the standard deviation for each sample. The CO_2^- radical intensity increases with increasing dose. The coefficient of determination, R^2, for CO_2^- radical vs dose represented by a solid line is estimated to be 0.989, indicative of the clear linear relationship between the CO_2^- radical intensity and the absorbed dose. According to the definitions [32, 33, 49–51], the detection limit of this work (represented by dashed lines in the figure) is estimated to be 115.0 mGy.

The detection limit of 115.0 mGy is higher than the lowest reported values of 67 mGy [32] and 56 mGy [33] for human molar teeth. This is maybe due to the larger sample-to-sample variation in deciduous teeth [52]. We estimated the absorbed dose for about 10 deciduous teeth, however, we could not find teeth which show the absorbed dose obviously higher than the detection limit. Further study is required to improve the detection limit for deciduous teeth enamel separated by the density separation method with appropriate chemical treatments
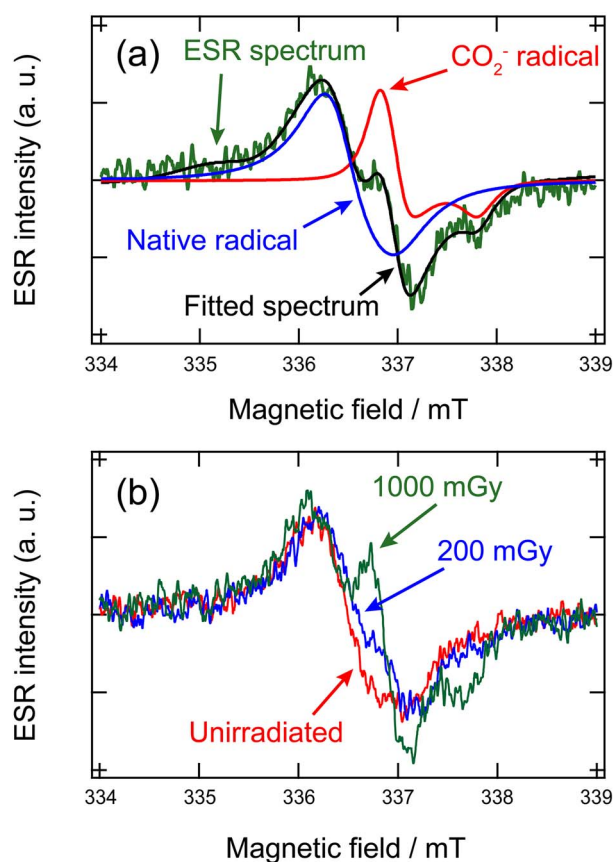

Fig. 2. (a) Typical ESR spectrum, deconvoluted spectra and a fitted spectrum for a 500 mGy-irradiated deciduous tooth from a Japanese child. (b) ESR spectra for the same deciduous tooth irradiated with different dose.

to accurately quantify small amounts of radicals generated by doses less than 100 mGy.

Comparison of the CO_2^- radical intensities for Japanese molar teeth enamel separated by the density and mechanical separation methods

Tooth enamel consists of ∼96% inorganic matter (mostly hydroxyapatite), 3% water and less than 1% organic matter (protein matrix). In contrast, dentine contains 70% inorganic matter, and 30% organic matter and water [26]. As mentioned previously, it is essential to collect the enamel free from dentine, because the organic materials in dentine interfere with the ESR measurement and biases the ESR-reconstructed dose. Although a chemical method using NaOH/KOH can easily separate enamel [26, 32, 33, 48], the dentine is lost in this method. We are also interested in the internal absorbed dose due to ^{137}Cs [53] and ^{90}Sr [54–56], so it is necessary to keep the dentine. Therefore, neither the mechanical separation method nor the chemical method is suitable for our purposes. Furthermore, these methods, especially the mechanical separation method, could not be applied for such epidemiological studies whereby a large number of enamel samples are examined, because it
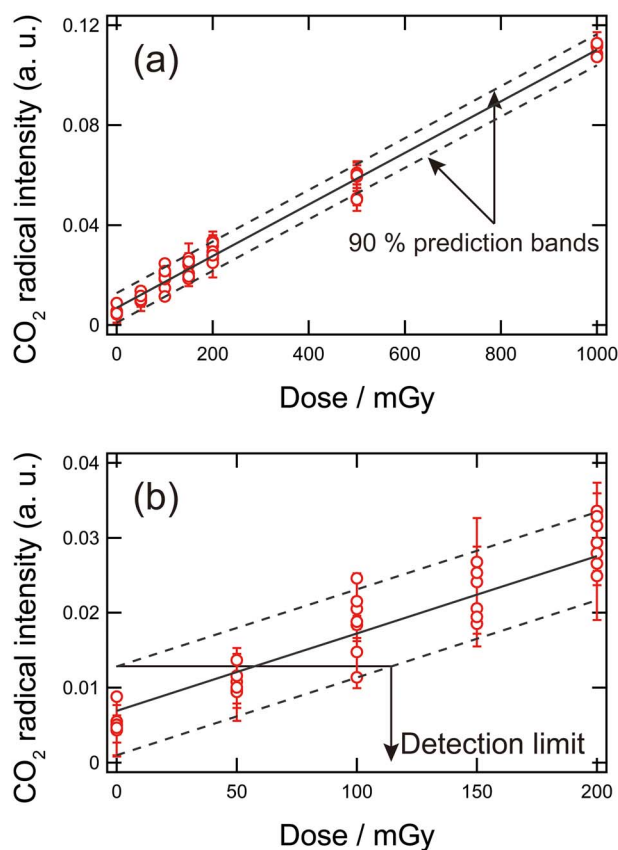
Fig. 3. Dose response curves of enamel sample; CO_2^- radical intensities vs absorbed dose (a) from 0 mGy to 1000 mGy and (b) from 0 mGy to 200 mGy. Solid lines represent the results of the linear least-squares fit. Dashed lines represent the 90% upper and lower prediction bands.

Fig. 4. Comparison of the CO_2^- radical intensities between the density separation method (red) and the traditional mechanical separation method (blue) by using same tooth. Nine teeth were subjected to the comparison and both methods show the same intensities within a standard deviation.

would have been extremely time-consuming to mechanically separate enamel from dentine.

In the present work, we applied the density separation method to separate enamel that preserves dentine and requires little time for sample preparation. Figure 4 shows the results of the comparison test; the vertical axis represents the CO_2^- radical intensities of enamel separated by the floating method (red) along with those separated by the traditional mechanical separation method (blue) for nine teeth. The observed CO_2^- radical intensities for enamel of Japanese molar teeth separated by the density and the traditional mechanical separation methods were consistent within the standard deviation of the remeasurements (error bars) as shown in Fig. 4, suggesting that we can obtain the enamel sample by the density separation method equivalent to the mechanical separation method without losing dentine. Although we were concerned about possible chemical contamination of the teeth due to sodium metatungstate, the results indicate that no contamination was observed.

The advantages of the density separation method are the following: (i) no special skill is required, (ii) it is operator independent, (iii) it can separate enamel in parallel with dentine, high quality enamel samples with less organic materials can be obtained by simply increasing the
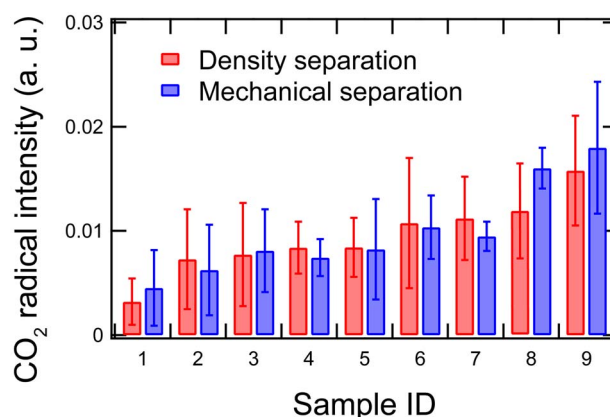
sodium metatungstate density accordingly, (v) it takes little time for sample preparation (working hours reduces to 1/10 compare to the mechanical separation method), and (vi) moreover, this method can be applied to teeth of animals such as macaques [31], racoons [57] and mice, which are difficult to process with the mechanical separation method because of the small size and structural complexity of their teeth.

In subsequent research, we will attempt to estimate the absorbed dose of wild raccoons in Fukushima prefecture and discuss the relationship between the biological effects and the absorbed doses. In addition, preparing more precise dose response curves for Japanese deciduous teeth and estimating the absorbed dose less than 100 mGy of the children in Fukushima prefecture is our next important work.

SUMMARY

We have estimated the detection limit of Japanese deciduous teeth is 115.0 mGy, which is higher than the lowest reported values of 67 mGy [31] and 56 mGy [33] for human molar teeth, maybe due to the larger sample-to-sample variation in deciduous teeth. We successfully demonstrated that the separation technique using sodium metatungstate is moderately useful for enamel–dentine separation for Japanese molar teeth, and has advantages such as ease of separation, operator independence, and the ability to separate large amounts of enamel in parallel and obtain enamel. We are going to apply the density separation method for Japanese deciduous teeth to prepare more precise dose response curve and estimate the absorbed dose of children in Fukushima in an epidemiological context.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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REFERENCES

1. Oshima A, Ikeda S, Katoh E et al. Chemical structure and physical properties of radiation-induced crosslinking of polytetrafluoroethylene. *Radiat Phys Chem* 2001;62:39–45.
2. Hill DJT, Thurecht KJ, Whittaker AK. A study of the radiation chemistry of poly(chlorotrifluoroethylene) by ESR spectroscopy. *Radiat Phys Chem* 2003;68:857–64.
3. Ikeya M. Dating a stalactite by electron paramagnetic resonance. *Nature* 1975;255:48–50.
4. Ikeya M, Miki T. Electron-spin resonance dating of animal and human bones. *Science* 1980;207:977–9.
5. Toyoda S, Ikeya M. Thermal stabilities of paramagnetic defect and impurity centers in quartz: basis for ESR dating of thermal history. *Geochem J* 1991;25:437–45.
6. Brooks AS, Helgren DM, Cramer JS et al. Dating and context of 3 middle stone-age sites with bone points in the upper Semliki valley, Zaire. *Science* 1995;268:548–53.
7. Weiss BP, Kim SS, Kirschvink JL et al. Ferromagnetic resonance and low-temperature magnetic tests for biogenic magnetite. *Earth Planet Sci Lett* 2004;224:73–89.
8. Kumagai Y, Nakamura N, Sato T et al. Ferromagnetic resonance spectroscopy and rock magnetic characterization of fossil coral skeletons in Ishigaki Islands, Japan. *Geosciences* 2018;8:1–11.
9. Tanaka K, Muto J, Yabe Y et al. Effect of fracture on ESR intensity using a low-velocity rotary shear apparatus. *Geochronometria* 2021;48:205–14.
10. Anzai K, Saito K, Takeshita K et al. Assessment of ESR-CT imaging by comparison with autoradiography for the distribution of a blood-brain-barrier permeable spin probe, MC-PROXYL, to rodent brain. *Magn Reson Imaging* 2003;21:765–72.
11. Adhikary A, Kumar A, Khanduri D et al. Effect of base stacking on the acid-base properties of the adenine cation radical [A•⁺] in solution: ESR and DFT studies. *J Am Chem Soc* 2008; 130:10282–92.
12. Oka T, Yamashita S, Midorikawa M et al. Spin-trapping reactions of a novel gauchetype radical trapper G-CYPMPO. *Anal Chem* 2011;83:9600–4.

13. Kumagai J, Masui K, Itagaki Y et al. Long-lived mutagenic radicals induced in mammalian cells by ionizing radiation are mainly localized to proteins. *Radiat Res* 2003;160:95–102.
14. Bernhard WA, Close DM. DNA Damge dictates the biological consequences of ionizing irradiation: the chemical pathways. In: Mozumder A, Hatano Y (eds). *Charged Particle and Photon Interactions with Matter: Chemical, Physicochemical, and Biological Consequences with Applications*. New York: Marcel Dekker, Inc., 2004, 431–70.
15. Yokoya A, Akamatsu K. EPR spectrometer installed in a soft X-ray beamline at SPring-8 for biophysical studies. *Nucl Instrum Methods Phys Res Sect A-Accel Spectrom Dect Assoc Equip* 2001;467: 1333–7.
16. Nakagawa S. ESR spectral change of radicals produced in L-alanine-3,3,3-d3 and L-alanine-d4. A new pathway to produce the de-hydrogen radical and the hydrogen exchange reactions of the de-amino radical. *Radiat Phys Chem* 2019;165:108406–1–7.
17. Oka T, Yokoya A, Fujii K. Electron paramagnetic resonance study of unpaired electron species in thin films of pyrimidine bases induced by nitrogen and oxygen K-shell photoabsorption. *Appl Phys Lett* 2011;98:103701–1–3.
18. Oka T, Yokoya A, Fujii K. Lifetime of the unpaired electron species in calf thymus DNA thin films induced by nitrogen and oxygen K-shell photoabsorption. *Int J Radiat Biol* 2012;88:884–7.
19. Oka T, Yokoya A, Fujii K et al. Unpaired electron species in thin films of calf-thymus DNA molecules induced by nitrogen and oxygen K-Shell Photoabsorption. *Phys Rev Lett* 2012;109: 213001–1–5.
20. Oka T, Yokoya A, Fujii K et al. Substituent effect on the yield of unpaired electrons in DNA bases studied by electron paramagnetic resonance. *Appl Phys Lett* 2018;113:243701–1–4.
21. Ikeya M, Miyajima J, Okajima S. Electron-spin-resonance dosimetry for atomic-bomb survivors using shell buttons and tooth enamel. *Jpn J Appl Phys Part 2 - Lett* 1984;23:L697–L9.
22. Skvortsov VG, Ivannikov AI, Stepanenko VF et al. Application of EPR retrospective dosimetry for large-scale accidental situation. *Appl Radiat Isot* 2000;52:1275–82.
23. Gualtieri G, Colacicchi S, Sgattoni R et al. The Chernobyl accident: EPR dosimetry on dental enamel of children. *Appl Radiat Isot* 2001;55:71–9.
24. Romanyukha AA, Ignatiev EA, Ivanov DV et al. The distance effect on the individual exposures evaluated from the soviet nuclear bomb test at Totskoye test site in 1954. *Radiat Prot Dosim* 1999;86:53–8.
25. Iwasaki M, Miyazawa C, Chida T et al. Dental ESR dosimetry of a medical physicist who received occupational radiation exposure for almost 40 y. *Health Phys* 2002;83:534–8.
26. IAEA. *Use of Electron Paramagnetic Resonance Dosimetry with Tooth Enamel for Retrospective Dose Assessment*. Vienna, Austria: IAEA, 2002.
27. Zhumadilov K, Ivannikov A, Psalikov KN et al. Radiation dose estimation by tooth enamel EPR dosimetry for residents of Dolon and Bodene. *J Radiat Res* 2006;47:A47–53.
28. Harshman A, Toyoda S, Johnson T. Suitability of Japanese wild boar tooth enamel for use as an electron spin resonance dosimeter. *Radiat Meas* 2018;116:46–50.

29. Toyoda S, Murahashi M, Natsuhori M et al. Retrospective ESR reconstruction of cattle tooth enamel doses from the radioactive nuclei released by the accident of Fukushima Dai-ichi atomic power plants. *Radiat Prot Dosim* 2019;186:48–53.

30. Todaka A, Toyoda S, Natsuhori M et al. ESR assessment of tooth enamel dose from cattle bred in areas contaminated due to the Fukushima Daiichi Nuclear Power Plant accident. *Radiat Meas* 2020;136:106357–1–6.

31. Oka T, Takahashi A, Koarai K et al. External exposure dose estimation by electron spin resonance technique for wild Japanese macaque captured in Fukushima prefecture. *Radiat Meas* 2020;134:106315–1–4.

32. Wieser A, Fattibene P, Shishkina EA et al. Assessment of performance parameters for EPR dosimetry with tooth enamel. *Radiat Meas* 2008;43:731–6.

33. Fattibene P, Wieser A, Adolfsson E et al. The 4th international comparison on EPR dosimetry with tooth enamel part 1: report on the results. *Radiat Meas* 2011;46:765–71.

34. Ishikawa T, Yasumura S, Ozasa K et al. The Fukushima health management survey: estimation of external doses to residents in Fukushima prefecture. *Sci Rep* 2015;5:12712–1–11.

35. Wieser A, El-Faramawy N, Meckbach R. Dependencies of the radiation sensitivity of human tooth enamel in EPR dosimetry. *Appl Radiat Isot* 2001;54:793–9.

36. Fattibene P, Callens F. EPR dosimetry with tooth enamel: a review. *Appl Radiat Isot* 2010;68:2033–116.

37. Wieser A, Romanyukha AA, Degteva MO et al. Tooth enamel as a natural beta dosemeter for bone seeking radionuclides. *Radiat Prot Dosim* 1996;65:413–6.

38. Takahashi A, Chiba M, Tanahara A et al. Radioactivity and radionuclides in deciduous teeth formed before the Fukushima Daiichi Nuclear Power Plant accident. *Sci Rep* 2021;11:10335–1–11.

39. Skvortzov VG, Ivannikov AI, Eichhoff U. Assessment of individual accumulated irradiation doses using epr spectroscopy of tooth enamel. *J Mol Struct* 1995;347:321–9.

40. Iwasaki M, Miyazawa C, Uesawa T et al. Effect of sample grain size on the CO_3^{3-} signal intensity in ESR dosimetry of human tooth enamel. *Radioisotopes* 1993;42:470–3.

41. Ivannikov AI, Trompier F, Gaillard-Lecanu E et al. Optimisation of recording conditions for the electron paramagnetic resonance signal used in dental enamel dosimetry. *Radiat Prot Dosim* 2002;101:531–8.

42. Koshta AA, Wieser A, Ignatiev EA et al. New computer procedure for routine EPR-dosimetry on tooth enamel: description and verification. *Appl Radiat Isot* 2000;52:1287–90.

43. Brik A, Radchuk V, Scherbina O et al. Metamorphic modifications and EPR dosimetry in tooth enamel. *Appl Radiat Isot* 1996;47:1317–9.

44. Sholom SV, Haskell EH, Hayes RB et al. Influence of crushing and additive irradiation procedures on EPR dosimetry of tooth enamel. *Radiat Meas* 1998;29:105–11.

45. Bailiff IK, Stepanenko VF, Goksu HY et al. Comparison of retrospective luminescence dosimetry with computational modeling in two highly contaminated settlements downwind of the Chernobyl NPP. *Health Phys* 2004;86:25–41.

46. Gregory MR, Johnston KA. A nontoxic substitute for hazardous heavy liquids - aqueous sodium polytungstate (3Na2WO4.9WO3.H2O) solution. *N Z J Geol Geophys* 1987; 30:317–20.

47. Krukowski ST. Sodium metatungstate: a new heavy-mineral separation medium for the extraction of conodonts from insoluble residues. *J Paleontol* 1988;62:314–6.

48. Güttler A, Wieser A. EPR-dosimetry with tooth enamel for low doses. *Radiat Meas* 2008;43:819–22.

49. Currie LA. Nomenclature in evaluation of analytical methods including detection and quantification capabilities (IUPAC recommendations 1995). *Pure Appl Chem* 1995;67:1699–723.

50. ISO/TC 69/SC 6. 11843-1:1997 Capability of Detection - Part 1: *Terms and Definitions.* 1997;1–10.

51. ISO/TC 69/SC 6. ISO 11842-2:2000. Capability of Detection - Part 2: Methodology in the Linear Calibration Case. 2000;1–20.

52. Murahashi M, Toyoda S, Hoshi M et al. The sensitivity variation of the radiation induced signal in deciduous teeth to be used in ESR tooth enamel dosimetry. *Radiat Meas* 2017;106:450–4.

53. Fukuda T, Kino Y, Abe Y et al. Distribution of artificial radionuclides in abandoned cattle in the evacuation zone of the Fukushima Daiichi Nuclear Power Plant. *PLoS One* 2013;8: 1–7.

54. Koarai K, Kino Y, Takahashi A et al. Sr-90 specific activity of teeth of abandoned cattle after the Fukushima accident - teeth as an indicator of environmental pollution. *J Environ Radioact* 2018;183:1–6.

55. Koarai K, Kino Y, Takahashi A et al. Sr-90 in teeth of cattle abandoned in evacuation zone: record of pollution from the Fukushima Daiichi Nuclear Power Plant accident. *Sci Rep* 2016;6:24077–1–9.

56. Koarai K, Matsueda M, Aoki J et al. Rapid analysis of Sr-90 in cattle bone and tooth samples by inductively coupled plasma mass spectrometry. *J Anal At Spectrom* 2021;36:1678–82.

57. Mitsuyasu Y, Oka T, Takahashi A et al. Examination of application of external exposure dose assessment method for individual raccoons living in difficult-to-return areas. *KEK Proc* 2020;144–9.