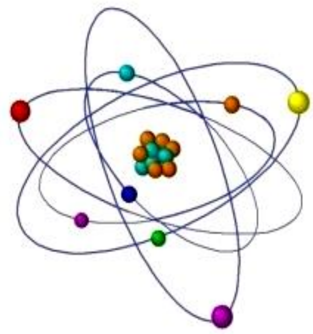
THE SIGNIFICANCE OF RADIOLOGICAL PARAMETERS OF MULTI-LAYERED ARCHAEOLOGICAL SITES IN DATING BIOGENIC ARTIFACTS



[1]Vakhtang Licheli, [2,3]Mikheil Gogebashvili*, [2,3]Nazi Ivanishvili, [4]Eremia Tulashvili, [2]Saba Suladze, [3]Sopho Kalmakhelidze, [2]Grigol Mamniashvili

[1]Iv.Javakhishvili Tbilisi State University, Georgia
[2]Iv.Javakhishvili Tbilisi State University, E. Andronikashvili Institute of Physics, Georgia
[3] Iv.Beritashvili Canter of Experimental Biomedicine, Georgia,
[4] Iv.Javakhishvili Tbilisi State University, Academician N.Kekelidze Materials Research Scientific-Research Institute, Georgia

https://doi.org/10.63465/rrs520258988 *Corresponding author: Email: gogebashvili@gmail.com

ABSTRACT: *The methodological scope of archaeology as a scientific discipline continues to expand in parallel with advancements in modern technology. In recent years, multidisciplinary analytical approaches have gained particular attention, especially in the dating of biogenic artifacts. This study investigates the radioisotopic parameters of a cultural layer dated to the 11th–10th centuries BCE, using the multi-layered archaeological site of Grakliani (Georgia) as a case study. The research highlights the significance of radiological indicators in surrounding soil zones for dating tooth enamel artifacts based on Electron Paramagnetic Resonance (EPR) signals. The analyzed radioisotopic spectrum offers an effective method for assessing layer stability and identifying potential mixing in multi-layered archaeological sites. By examining isotopes from the uranium-thorium series and the specific radioactivity of radioactive potassium, the study proposes an analytical approach that serves two key purposes: first, to determine the integrity or disturbance of cultural layers, and second, to calculate the actual integral dose affecting biogenic artifacts. The latter aspect is particularly crucial for improving the accuracy of dose estimations in EPR signal dating, ultimately leading to more precise chronological assessments of archaeological materials.*

Key words: radiological parameters, multi-layered archaeological sites, ating artifacts

INTRODUCTION

Dating artifacts from archaeological sites remains one of the most significant challenges in the field [1,2]. Various dating approaches exist, relying on physicochemical and biological methodologies [3,4,5]. Among these, the Electron Paramagnetic Resonance (EPR) dosimetry method stands out, as it measures the concentration of radiation-induced radicals in irradiated tooth enamel tissues [6,7,8]. This method offers a relatively low detection threshold for absorbed radiation doses. The intensity of the EPR signal provides crucial information about the cumulative radiation exposure an artifact has experienced over time. Radiation monitoring and dosimetric analysis, including the dating of biogenic artifacts, are based precisely on the examination of signals in the spectra of tooth enamel tissues [9,10]. Notably, the EPR dating method has an extensive overlapping range, spanning several centuries and often exceeding the capabilities of radiocarbon analysis. In EPR dosimetry-based dating, a key issue is the detection of radiation doses accumulated by the studied object from natural radiation sources over centuries. This factor is crucial for determining the artifact's age, as it allows for the experimental assessment of an equivalent radiation exposure level in comparison with radiologically treated materials. Additionally, it helps address significant radiological challenges, such as the continuity of the artifact's location, the uniformity of the soil matrix surrounding the artifact in terms of radiological parameters, and the identification of dose contributions resulting from anthropogenic radionuclide exposure over a specific period.

In Electron Paramagnetic Resonance (EPR) dosimetry-based dating, a major challenge is accurately measuring the radiation doses that an artifact has accumulated from natural radiation sources over centuries. This measurement is essential for determining the artifact's age, as it enables researchers to experimentally assess its equivalent radiation exposure in comparison to materials that have undergone radiological treatment. Furthermore, this method tackles significant radiological challenges, such as ensuring the artifact's spatial stability over time, evaluating the uniformity of the surrounding soil matrix in terms of its radiological properties, and identifying contributions from anthropogenic radionuclides over a specific period.

MATERIALS AND METHODS

The research object was the archaeological excavation site of "Grakliani" (Eastern Georgia). The chronological range of the research layers at Grakliani Hill encompasses 1.5–2 million years of human settlement history in this area. In this regard, it is noteworthy that the site comprises 11 cultural layers, which, in turn, form several methodological challenges [11].

Grakliani Hill is a multi-layered archaeological monument that confirms 300,000 years of uninterrupted social development, spanning from the Stone Age to the Roman period. This, in turn, can be used as an adequate model for studying the radiological parameters of various archaeological zones.

Soil samples were taken from functionally different locations of one of the cultural layers. After mechanical processing to achieve powder condition, the samples were dried in a thermostat at a temperature of 105–110°C

The content of radionuclides was determined by gamma-spectrometry (Gamma-Beta Spectrometer, Atomtex-MKC-AT-1315 and Gamma-Spectrometer CANBERRA with liquid nitrogen cooled germanium detector).

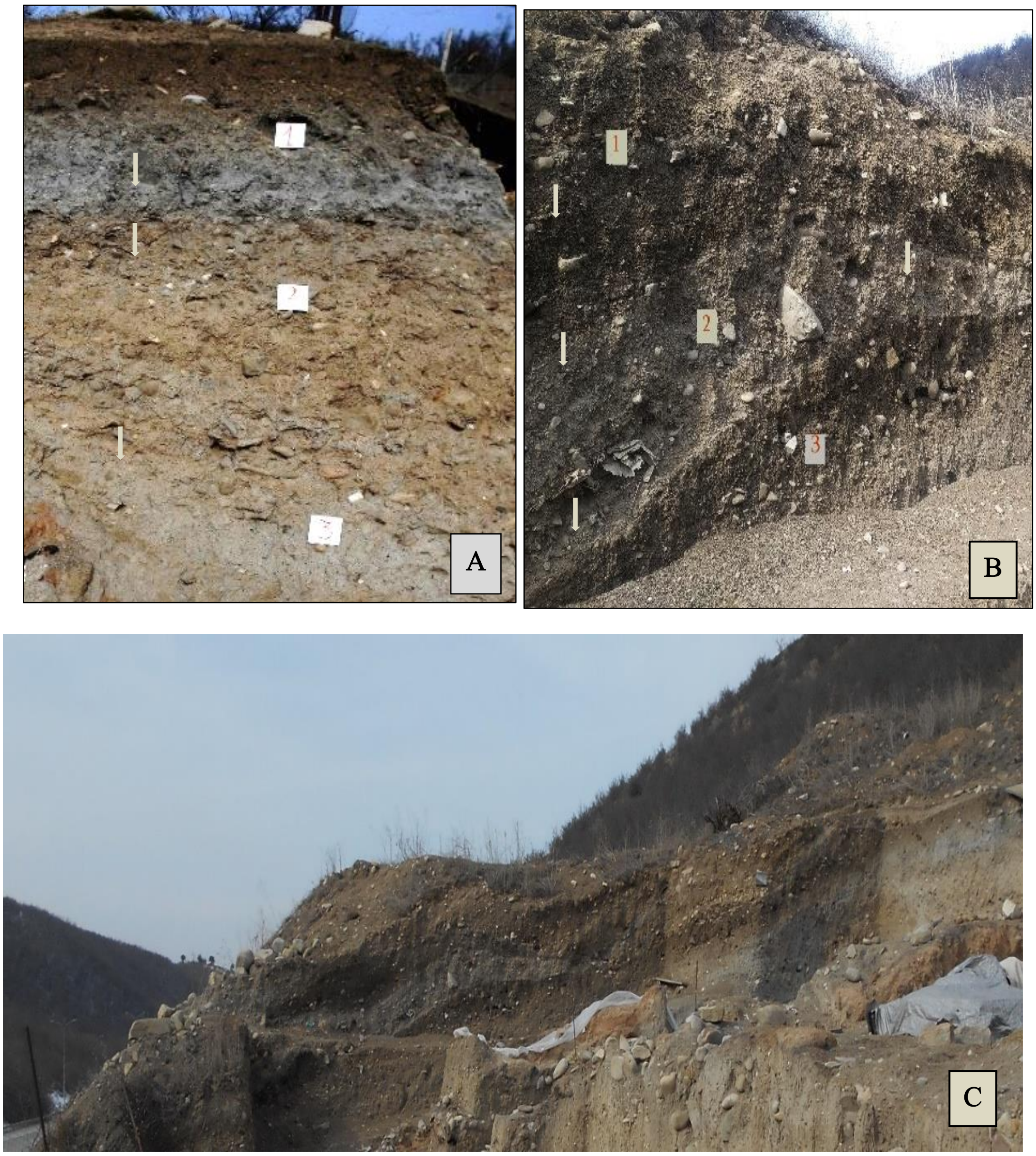
RESULTS AND DISCUSSION

The radiological study of layers in multi-century archaeological sites, particularly those in open areas, has several specific characteristics shaped by the nuances of their formation. A cultural layer refers to the soil deposits at a site of human habitation that preserves material evidence of human activity. The scientific methodology for studying these cultural layers often emphasizes the concept of the "in situ preserved cultural layer." In practical archaeology, the primary goal is to establish a comprehensive list of objective criteria necessary for dating the artifacts discovered. For dating purposes, when tooth enamel is exposed to radiation, it produces a complex, asymmetric Electron Paramagnetic Resonance (EPR) signal. This signal serves as a source of information for determining the structure of the analyzed biogenic artifact and the duration of its presence at a specific location [12]. However, a key issue in this process is assessing the integral dose received by the particular artifact during its prolonged burial and preserved state.

To conduct radiological analysis, we selected a specific layer from the archaeological site of "Grakliani" (Georgia), which is dated to the 11th-10th centuries BCE. Soil samples for radioisotopic analysis were taken from the natural profile of the given area and various locations within the identified cultural layer. Naturally, when using EPR spectrometry for dating, there is a possibility of error due to the influence of anthropogenic radionuclides (technogenic) on the results.

As is well known, since the second half of the 20th century, due to nuclear weapon testing, the presence of technogenic radionuclides has been detected in the atmosphere, the environment, and especially in the soil. Radioactive decay and migration processes have led to the contamination of landscapes with artificial radionuclides. In this regard, the territories of Georgia are no exception [13]. The deposition and distribution of such radionuclides on the Earth's surface are uneven, influenced both by nuclear weapon testing (global dispersal) and during emergencies (such as the Chornobyl and Fukushima nuclear plants). This circumstance, in turn, creates specific methodological challenges in studying the accumulation of artificial radionuclides.

During the radiological study of an archaeological excavation site, it is essential to exclude the factor of technogenic radionuclides. To eliminate the possibility of anthropogenic radioisotope contamination in the excavation areas, surface monitoring of the surrounding soils was conducted. The criterion for anthropogenic radioisotope contamination was ^{137}Cs. The maximum contamination level of this isotope did not exceed 10-15 Bq/kg. Furthermore, the activity of the cesium radioisotope in its localized zone in the soil at depths of 5-10 cm did not exceed these levels, indicating that no technogenic-origin radioisotopes were detected in the studied archaeological zone. This conclusion is supported by the fact that cesium isotopes were not found in any soil samples taken from the vertical profile. The next stage in studying the radiological parameters of soil profiles at various depths involved conducting gamma spectrometric analysis during the normal (horizontal) arrangement of the zones (Fig. 1-A). Additionally, the vertical placement of artifacts is one of the characteristics used for dating specific cultural layers. However, the specificity of the selected object does not exclude the possibility of horizontal shifts in the soil layers. This phenomenon is particularly characteristic of mountainous landscapes. Such horizontal displacement of archaeological zones is also observed in our object (Figure 1-B).



A - Soil profile of zone A in normal horizontal arrangement
B, C - Soil profile of zone B in case of horizontal arrangement disturbance
(The top three levels are marked with numbers; the boundaries of the soil horizons are indicated with arrows)

Based on the above, it is clear that the precise radiological characterization of the soils surrounding the discovered artifact can provide additional information regarding the duration of the artifact's presence at the site. For a model study of such situations, we selected specific archaeological locations based on the discovered objects and structures with various functional characteristics. "The following locations were selected for the study: 1- Vertical distribution of the natural soil zones (Fig. 1); 2- Agricultural zone (presumably a dairy production site) (Fig. 2-4); 3- Location of the residential building (Fig. 2.5); 4- Area designated for religious purposes (Fig. 2-6); and 5- Location adjacent to the clay furnace (Fig. 2-7).

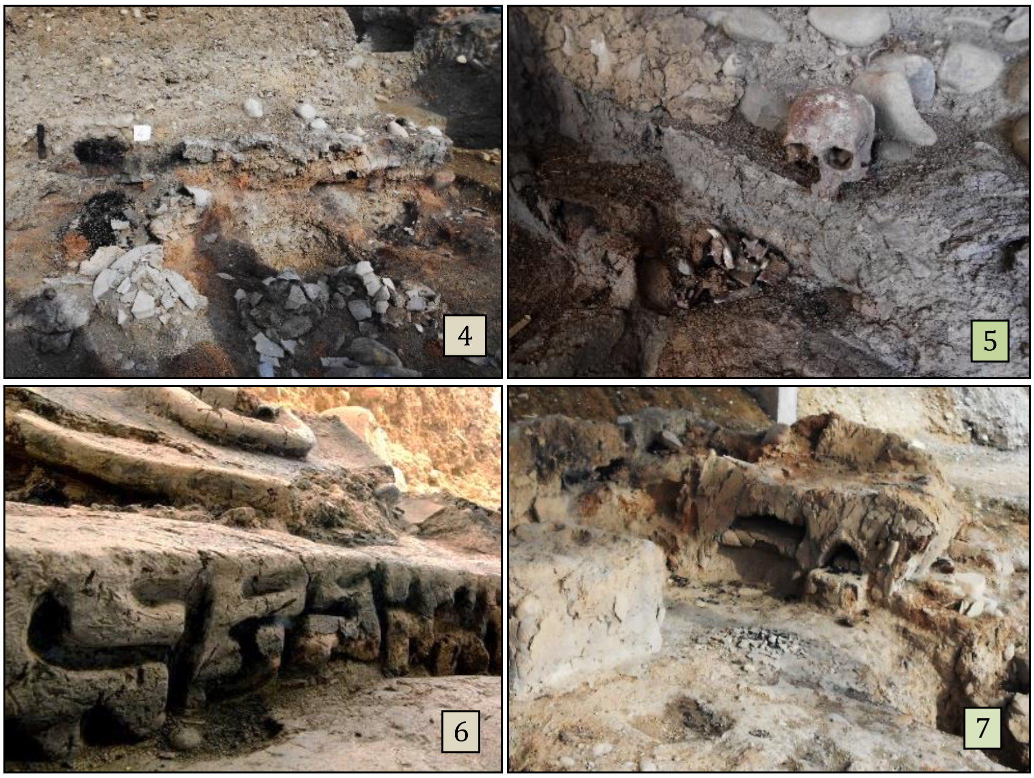


Fig. 2. 4 - Agricultural zone (presumably a dairy production site); 5 - Location of the residential building; 6 - Area designated for religious purposes); and 7 - Location adjacent to the clay furnace

The results of the gamma spectrometric analysis of these locations are shown in Figure 3. Each sample has a distinct isotopic composition, represented by groups of isotopic families in the diagram. In the ^{238}U family, the isotope ^{226}Ra was detected only in the upper horizon of the soil vertical profile. At the same time, relatively high activity of this isotope was observed in the 4th**,** 5th**,** and 7th samples from the study area. This phenomenon can be attributed to specific agricultural practices in the areas studied. Additionally, the absence of the radium isotope in the sample from the sixth location, which is believed to have religious significance, may provide further evidence (Fig. 3-A).

The activity of isotopes in the ^{232}Th family is characterized by defined information content. This radioisotope was detected only in the upper horizon of the soil profile and was virtually absent in other locations of the studied zone. In this case, this fact can be interpreted as an indicator of the archaeological zone's stability, which may have been caused by the absence of mixing between the upper soil layer and the deeper layers (Fig. 3-B). Among the samples analyzed, the isotopes of the ^{235}U family exhibited the lowest activity (Fig. 3-C).

In terms of activity, ^{40}K occupies a particularly significant position. As shown in the diagram of Figure 3-D, in some samples, the activity of this isotope reached 450 Bq/kg, which is significantly higher than the activity of the other presented radioisotopes. Considering that the total activities of isotopes from the ^{238}U (^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi) and ^{232}Th (^{228}Ac, ^{210}Pb, ^{212}Bi) families do not exceed 60 and 38 Bq/kg, respectively, the radioactivity of ^{40}K emerges as the major component of the total radiation dose.

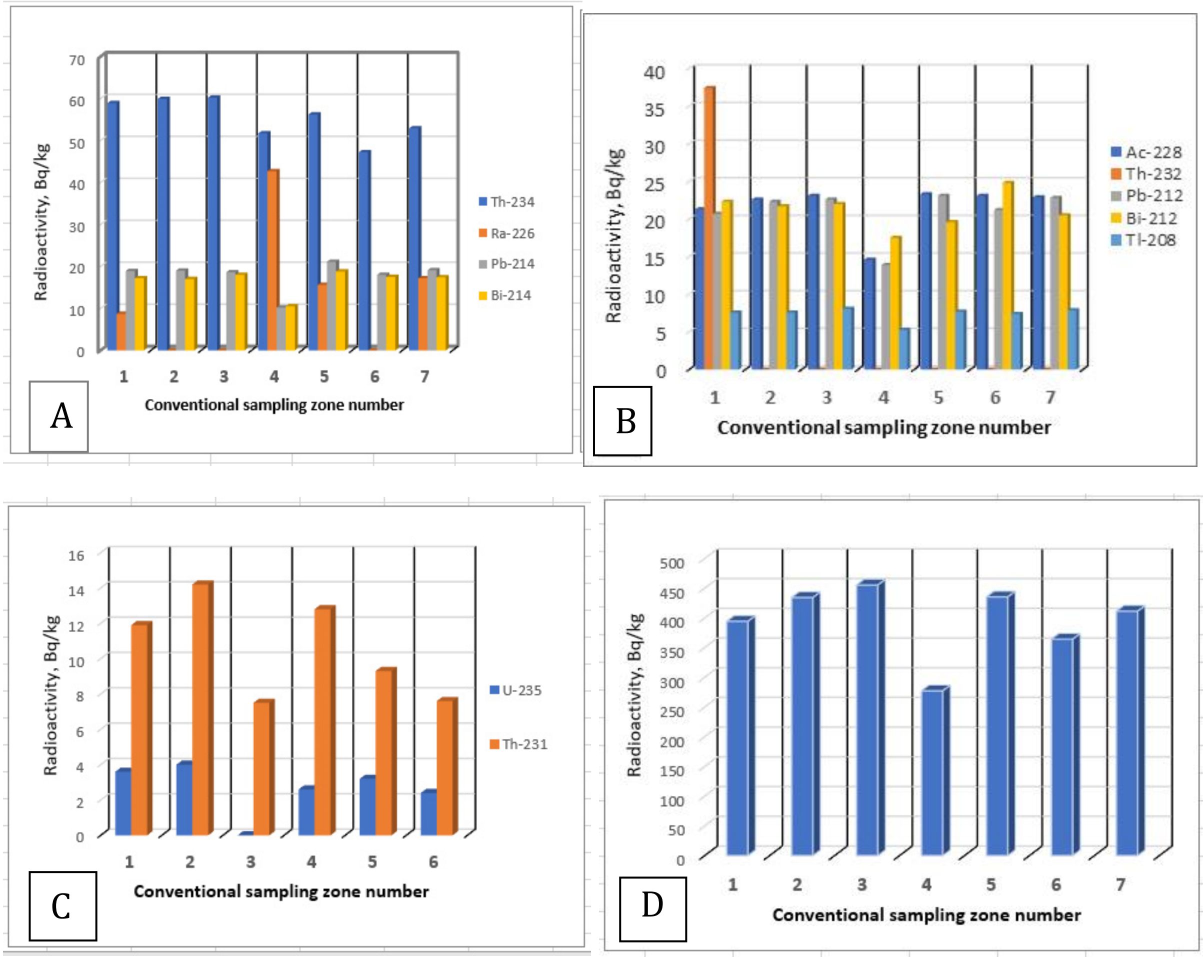


Fig. 3 Distribution of different family isotopes in the soil samples of the studied archaeological site

A-Family ^{238}U (^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi); **B**-Family ^{232}Th (^{228}Ac, ^{210}Pb, ^{212}Bi, ^{208}Tl); **C**-Family ^{235}U (^{231}Th, ^{235}U); **D** – ^{40}K

(The numbering of the study zones in Figures 3 and 4 corresponds to the location in Figures 1 and 2

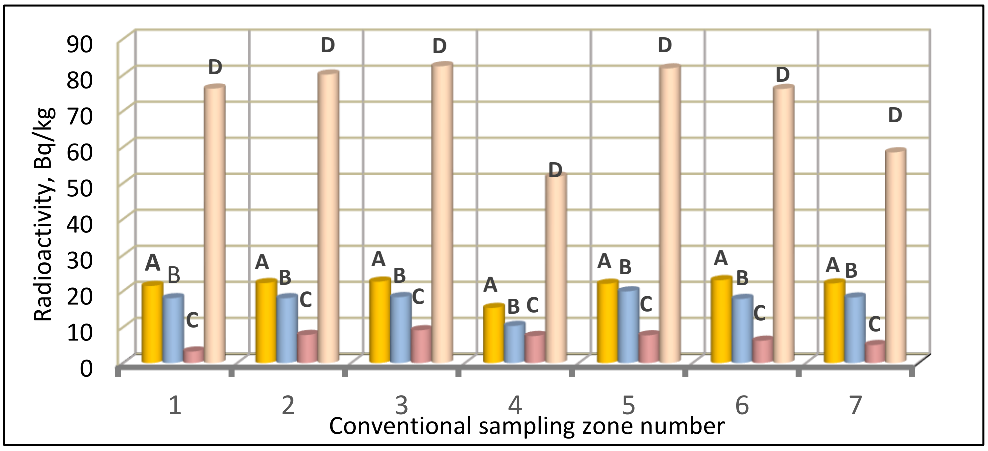


Fig.4. Radioactivity of uranium-thorium family radioisotopes in soil samples from different locations of the archaeological site

A-Family ^{232}Th (^{228}Ac, ^{210}Pb, ^{212}Bi, ^{208}Tl); **B**-Family ^{238}U (^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi); **C**-Family ^{235}U (^{231}Th, ^{235}U); **D** - Radium equivalent activity

Thus, by analyzing the spectra of different isotopic families, it is possible to obtain additional characteristics of a multi-layered archaeological site. However, the key factor for dating through EPR analysis is determining the total radiation dose in the artifact's in situ fixed state. As shown in the data presented in Figure 4, the total radioactivity of the uranium-thorium family isotopes in each sample does not exceed 20 Bq/kg. To obtain the total values resulting from various types of radioisotopic irradiation, the equivalent activity of the radium isotope was used [14]. This criterion significantly improves the accuracy of the EPR signal analysis methodology for artifact dating.

Overall, the conducted research demonstrated that determining radiological parameters provides a relatively complete picture for characterizing multi-layered archaeological sites. Specifically, the radioisotopic spectrum from different locations during excavations justifiably reflects the potential for horizontal movement of soil masses. This is particularly relevant in cases where archaeological excavations are carried out in seismic and unstable landscape zones [15]. Furthermore, the increase in dosimetric parameters has the potential to significantly improve the accuracy of the EPR method in dating biogenic artifacts.

REFERENCES

[1]. Thilo Rehren, Efthymia Nikita. Archaeological Science. Encyclopedia of Archaeology (Second Edition). Volume 1, 2024, Pages 123-132.

[2]. Elisa Crabu, Caterina Fenu, Giuseppe Rodriguez, Giuseppa Tanda. Dating archaeological sites by seriation. Journal of Cultural Heritage. Volume 72, March–April 2025, Pages 42-47.

[3]. Grzegorz Piotr Guzik, Wacław Stachowicz, Jacek Michalik. Chapter 6 - Aspects of the EPR study on mineralized tissues as related to medicine, dosimetry, and dating. Experimental Methods in the Physical Sciences. Volume 50, 2019, P.115-127.

[4]. Öykü Ataytür, Cécile Gautheron, Adriana Horbe, Thierry Allard. Thermal stability of artificial radiation-induced defects in kaolinite: Enhancing EPR dating protocol. Applied Clay Science. Volume 252, 2024, 107349.

[5]. Shigueo Watanabe, Nilo F. Cano, Alvaro B. Carvalho-Júnior, Jorge S. Ayala-Arenas, Carlos D. Gonzales-Lorenzo, T.K. Gundu Rao. Dating of carbonate covering cave paintings at Peruaçu, Brazil by TL and EPR methods. Applied Radiation and Isotopes. Volume 153, 2019, 108847.

[6]. H.S. Sullasi, M.B. Andrade, W.E.F. Ayta, M. Frade, M.D. Sastry, S. Watanabe. Irradiation for dating Brazilian fish fossils by thermoluminescence and EPR technique. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms. Volume 213, 2004, p.756-760.

[7]. Shuchen Wang, Zhenlin Jia, Hongshan Gao, Desheng Xue, Baotian Pan. Impact of microwave power on equivalent dose (De) evaluation in ESR dating. Radiation Measurements. Volume 177, 2024, 107231.

[8]. Anne R. Skinner. Electron Spin Resonance Dating. Encyclopedia of Geology. 2021, P.153-163.

[9]. Paola Fattibene, Freddy Callens. EPR dosimetry with tooth enamel: A review. Applied Radiation and Isotopes. Volume 68, Issue 11, 2010, Pages 2033-2116.

[10]. Alfredo Coppa, Rainer Grün, Chris Stringer, Stephen Eggins, Rita Vargiu. Newly recognized Pleistocene human teeth from Tabun Cave, Israel. Journal of Human Evolution. Volume 49, Issue 3, 2005, Pages 301-315

[11]. Vakhtang Licheli. The second stage of the Grakliani Culture. Wonders Lost and Found. A Celebration of the Archaeological Work of Professor Michael Vickers. Oxford: Archaeopress. P. 48-58.

[12]. Licheli V., Chikvaidze E.N., Gavasheli TA., Mamniashvili G.I. Gegechkori T.O., Gogebashvili M.E., Ivanishvili N.I. Electron paramagnetic resonance method for dating of archeological sites in Georgia. Journal of Radiobiology and Radiation Safety, Vol.1, No2, 2021, p.65-70.

[13]. Kiazo Nadareishvili at all. Effect of Chernobyl disaster on the radioecological situation in Transcaucasia. Radiation studies, Georgia, Tbilisi, 1991, Vol.VI, p.132-151.

[14]. Beretka, J., Mathew P.J. Natural Radioactivity of Australian Building Materials, Industrial Wastes and By-products. Health Physics 48(1), 1985, p. 87-95.

[15]. Iain Stewart. Archaeological and Cultural Records of Active Tectonics. Treatise on Geomorphology (Second Edition). Volume 2, 2022, Pages 13-33