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Assessment of the ages of the ancient human and animal bones correlation by specific activity of ^{226}Ra

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ABSTRACT

The paper proposes an age-assessment approach for ancient bones using CR-39 track detectors to measure ^{222}Rn volume activity in equilibrium with ^{226}Ra , then infers ^{226}Ra specific activity in bone and nearby soil. The study applies the method to modern and prehistoric bone specimens and introduces an "age coefficient" based on the bone-to-soil ^{226}Ra ratio. The main claim is that ^{226}Ra accumulation and the derived "age coefficient" correlates with bone age and can be used for practical dating estimates.

Keywords: CR-39 track detector, Volume activity of ^{222}Rn , Specific activity of ^{226}Ra , Ancient bone remains, Radiogenic dating

1. INTRODUCTION

Among the methods of radiogenic dating of archaeological materials, radiometric methods for dating bone finds play an important role. Bone materials have always been of interest to archaeologists, paleontologists and historians, as they contain biological, archaeological and chronological information. Radiogenic dating methods include radiocarbon, potassium-argon, uranium-lead and thorium-lead methods. They are based on the constant rate of radioactive decay of natural radioisotopes in bone materials. The radiocarbon method, using ^{14}C with a half-life of 5570 ± 30 years and maximum beta particle energy of 156.46 keV, has received the most widespread application in archaeology [1].

However, the radiocarbon method allows determining the age not exceeding 50 thousand years [2]. The potassium-argon method is based on the decay of the radioactive isotope ^{40}K with a half-life of 1.45×10^9 years, which decays into ^{40}Ar and accumulates in natural objects. This method is applicable for determining the age of rocks and fossils > 100 thousand years old [3]. There is a work [4], where the authors used potassium-argon dating method and was determined the age of the archanthropus from Selungur cave to be 1.5 million years.

Uranium-lead dating methods are used in geological samples containing uranium isotopes $^{235,238}\text{U}$, $^{206,207}\text{Pb}$ [5]. However, the $^{235,238}\text{U}$, $^{206,207}\text{Pb}$ dating method is not applicable to bone samples. As the results of INAA show, during long-term storage of skeletons in soil layers, migration of some elements from the soil occurs, as a result of which the concentration of uranium in bone remains increases [6]. For example, according to ^{238}U content in bones reached: - in Archanthropus up to 1.5 mg kg^{-1} , in bear up to 24 mg kg^{-1} , mammoths and dinosaurs up to 180 mg kg^{-1} that from 150 to 18000 time more, than that ^{238}U in standard bones (0.01 mg kg^{-1}). This reality is confirmed in [7] that was carry out by using inductively coupled plasma mass spectrometry (ICP-MS) for analysis of the same bones and soils [6]. The concentrations of 64 elements have been detected by ICP-MS.

The ^{238}U are up to two orders of magnitude elevated compared to the soils (e.g. ^{238}U in dinosaur and mammoth bone - 19.6 and 130.1 mg kg^{-1} , respectively and in soils were changes within $1.0\text{-}2.2 \text{ mg kg}^{-1}$). In standard bone made up 0.2 mg kg^{-1} . The authors associated these anomalies with the migration of uranium from the earth into the skeleton as a result of multimillion-year layering. This fact can be seen in [8], where the concentration of ^{238}U in fossil mammoth bones varied from 1 to 1000 ppm while in modern bones the concentration does not exceed 0.1 ppm. The high content of the parent radionuclide ^{238}U in bone remains allows us to measure the specific activities of its daughters ^{222}Rn and ^{226}Ra , that is being radioactive equilibrium in closed of bone-soil system over several million years [3]. Despite its radio-analytical advantages, INAA still lacks the sensitivity required to detect naturally occurring radioisotopes such as ^{222}Rn and ^{226}Ra .

There are several methods for determining ^{226}Ra in the samples under study, one of which is a gamma-spectrometer with HPGe detectors, the energy resolution of which is higher than 1.8 keV at 1332 keV, with the registration of the gamma activity of ^{214}Pb and ^{214}Bi , which are daughters of ^{222}Rn . However, the measurements, it is necessary to have a radioactive equilibrium between the corresponding radionuclides and a system of isolation from the environment [9]. Another method is to record the γ -radiation of Ra-226 with an energy of 186.2 keV. However, the presence of U-235 impurity in the samples can interfere with its γ -peak at 185.7 keV, which complicates the radiochemical extraction Ra-226 [9]. The use of an α -spectrometer to count α -particles of Ra-226 with an energy of 4500 keV is necessary for the radiochemical extraction Ra-226 from bone and soil samples.

A review of the literature on the measurement of ^{222}Rn and ^{226}Ra in bone samples was showed that radon measurements are mainly carried out using nuclear solid-state track detectors (NSSTDs). The advantages of NSSTD are their simplicity, the ability to over a wide temperature range, under conditions of high gamma and beta radiation fluxes, and the ability to obtain time-integrated information [10]. For measuring the volume activity of ^{222}Rn (VARn) can be use the type of CN-85, LR-115 and CR-39 detectors [10]. Of this group of NSSTD, the CR-39 detector is used very successfully due to detection efficiency of 90%, selectivity and wide energy range of recording α -particles with energies from 0.1 MeV to 20 MeV, as well as the possibility of simultaneously performing mass measurements in biomaterials [11].

There are articles [12-14] in which the α -activity of radionuclides was measured using NSSTD, where the CN-85 track detector was used to irradiate specific α -activity of natural radionuclides ^{210}Pb , ^{210}Po and ^{226}Ra in fresh bones of domestic animals – sheep, cow and chicken [12]. The authors of the articles [13] write that the specific activity of ^{226}Ra (SARa) in bones is $0.26 \div 0.50 \text{ Bq kg}^{-1}$. According to the data [14], SARa for most measured animal bones is $>1 \text{ Bq kg}^{-1}$. ^{226}Ra penetrates to body through the digestive tract with food and water, and accumulates up to 98% (260 mBq kg^{-1}) in bones and up to 2 % (4.1 mBq kg^{-1}) in human teeth [13, 14].

Being a radioactive metal, ^{226}Ra belongs to one of the subgroups of alkaline earth elements and is similar to calcium [13, 14]. The half-life of ^{226}Ra is 1602 year (Y), it emits α - and γ -particles with energies of 4.5 MeV and 186.2 keV, respectively [14]. As a α -decay of the ^{226}Ra is formation to inert radioactive gas radon-222 (^{222}Rn), with a half-life of 3.82 days. The specific α -activity of the one gram ^{226}Ra equal to $3.7 \times 10^{10} \text{ Bq}$ or $\sim 1 \text{ Curi (Cu)}$. Radioactive equilibriums are reached between of ^{222}Rn and ^{226}Ra within 35-40 days at isolated system [13]. However, a literature review reveals at the long-period bedding of skeletons affecting to the specific activity of radium-226 in bone-soil system so far have remained scarce and are poorly understood.

The aim of this work was to study of the variation specific activity of radium-226 in ancient human and animal bones by using of CR-39 track detectors.

2. METHOD

2. 1. Research objects

Prehistoric bones of dinosaur (BD) and bones of southern mammoths of Angren and Kashkadarya (BMA and BMK) [15] were discovered in 1990–2014 on the territory of Uzbekistan. Figure 1 shows tusk and jaw of the southern mammoths that found in Angren and Kashkadarya at the 2013 and 2014 [16].



Figure 1. Tusk and jaw of the southern mammoth.

The samples of bone and its soil were provided by the Geological Museum of the Republic of Uzbekistan. The bone of archanthropus (BA) was presented by the Institute of Archaeology [17]. For comparison and interpretation of the results were taken scalp of the male sheep (BSh) that offering to sacrifice many years ago to Ramadan month (that was buried in 2006 or 2007) and modern bone of cow (BC) from a commercial bazar. As well as, adjacent soils were obtained from the place where the skeletons discovered. The soil that related to the bone was designated by letter *s*, for example BA_s. The bone of archanthropus that found in the Selungur cave of the Fergana region in 1970, belongs to the most ancient objects of the cultural heritage of Uzbekistan [15-18] that shown in Figure 2.



Figure 2. Bone and teeth of an archanthropus were found in the Selungur cave.

The first traces of archanthropus (ancient man) was found in the Selungur Mountains during the so-called Early Paleolithic. Individual stone tools of this period were also found in the Sokh River Valley [18].

The found archaeological bones had different states of preservation in terms of strength and deterioration. Specialists from the Institute of Archaeology of the Russian Academy of Sciences evaluated the natural preservation of osteological materials on a five-point system [19]. Accordingly to [19] the highest score of 5 is a natural state in which the bone tissue retains sufficient strength so that tangible efforts are required to break it, the lowest score of 1 is a state in which the bone tissue crumbles when attempts are made to clean or wash it, a score of 4 denotes bone remains that easily break under directed action but retain their shape if there is no directed impact, especially the surface layer, and scores 3, 2 are for bone remains that, to one degree or another, dry out and crack into smaller fragments after cleaning and washing.

Surface contamination of the test bones was cleaned by swab treatment with ethanol. The test samples were then stored in a separate room for 40–50 days to remove moisture and residual

^{222}Rn activity, and to achieve radioactive equilibrium between ^{226}Ra and ^{222}Rn . The bone and soil fragments were crushed, mixed and sifted through a sieve with diameter of 2 mm to obtain a homogeneous powder sample.

2. 2. The measuring method of the VARn and the SARa

The samples of bone and soil a mass of 10 g was placed in a plastic cup. The CR-39 detector of $2 \times 2 \text{ cm}^2$ is hanged at 13 cm from sample to registration α -particles of ^{222}Rn only, shown in Figure 3 [20].

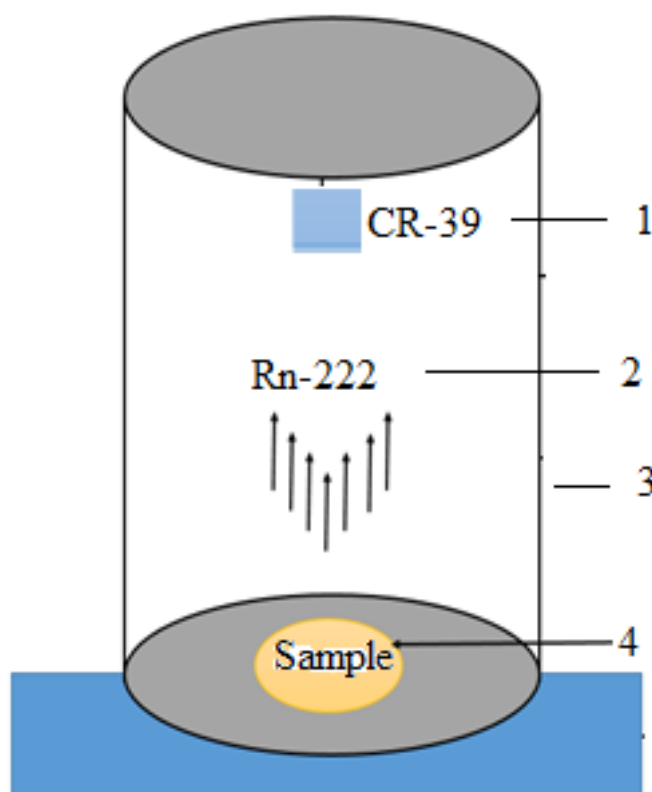


Figure 3. Schematic view of the plastic chamber for registration α -particles of the ^{222}Rn and its daughter products: 1–CR-39 track detector; 2–atoms of ^{222}Rn ; 3–plastic chamber; 4–sample of the bone or soil.

To protect against external atmospheric influences, the outer circle of the chamber was isolated with a thin layer of sealant. Fifteen chambers were exposed simultaneously for 35 days. To account for the contribution of the background to measurements were taken three chambers without samples, simultaneously. After exposure, the CR-39 detectors were removed for etching in a 6M NaOH solution at a temperature of 70°C for 7 hours. The number of α -particle tracks per $1 \times 1 \text{ cm}^2$ of the CR-39 area was counted from the monitor screen of the optical setup [20]. The view of α - tracks from the mammoth (1) and sheep (2) bones a shown in Figure 4.

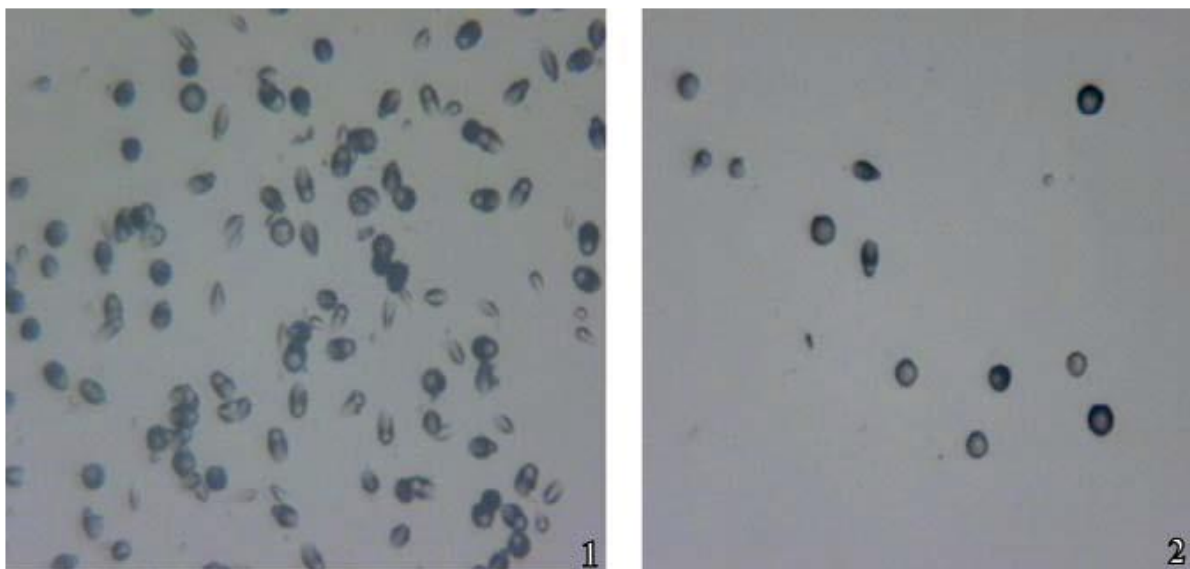


Figure 4. View of α -particles of ^{222}Rn from bones of mammoth–1 and sheep–2. It is clearly seen the number of α -tracks in the mammoth bone is many times more than number of α -tracks in the sheep bone.

3. RESULTS OF MEASUREMENTS

3. 1. The volume activity of ^{222}Rn

The α -track density of the samples is a main indicator that determines the specific activity of ^{226}Ra by measuring of the volume activity of ^{222}Rn . The VARn is the amount of spontaneous decay of ^{222}Rn in 1 m^3 of air and the A_{Rn} (Bq m^{-3}) values are determined by the formula:

$$A_{Rn} = \rho/K \quad (1)$$

where ρ is the α -particle track density in CR-39, track $(\text{cm}^2 \cdot \text{day})^{-1}$; K is the calibration coefficient, track $(\text{cm}^2 \cdot \text{day})^{-1}(\text{Bq m}^{-3})^{-1}$. The measuring chamber was calibrated and the K values were equal to 0.17 ± 0.03 track $(\text{cm}^2 \cdot \text{day})^{-1}(\text{Bq m}^{-3})^{-1}$ [20]. Evidently, the VARn in bones varies in range $13.0 - 5313 \text{ Bq m}^{-3}$ at the track density in a $2.2 - 2766 \text{ tr}(\text{cm}^2 \cdot \text{d})^{-1}$ and the VARn in soils varies in range $226 \pm 52.9 - 269 \pm 63 \text{ Bq m}^{-3}$ at the track density in a $39 - 46 \text{ tr}(\text{cm}^2 \cdot \text{d})^{-1}$ (№2–6, Table 1), with exception of the Angren mammoth.

3. 2. The specific activity of radium-226

The S_{Ra} is the specific activity of radium, Bq kg^{-1} is determined by the formula (2) [21]:

$$S_{Ra} = A_{Rn} \cdot V/m \quad (2)$$

where A_{Rn} is the volume activity of radon-222, Bq m^{-3} ; V is the effective volume of the exposure chamber, m^3 ($h = 15 \text{ cm}$, $d = 9.0 \text{ cm}$; $V = 9.54 \cdot 10^{-4} \text{ m}^3$); m is the mass of the samples under study ($m = 10 \text{ gr}$). The error in determining of the specific activity of ^{226}Ra in the samples under

study does not exceed 25% and consists of errors in determining the calibration coefficient $\leq 15\%$, counting α -particle tracks $\leq 10\%$, and taking into account the radon back diffusion coefficient $\leq 15\%$ [21-23]. The measured values of α -tracks, A_{Rn} and S_{Ra} for bones and its soils are listed in Table 1 by Roman numerals.

Table 1. The kind of bones with soils, α -track density, VARn and SARa.

Nº	Bone and soil	ρ , tr $(\text{cm}^2 \cdot \text{d})^{-1}$	A_{Rn} , Bq m^{-3}	S_{Ra} , Bq kg^{-1}
I	BC	2.2	13.0 ± 3.1	6.45 ± 2.1
	BC _s	40	235.3 ± 55.1	22.4 ± 6.0
II	BSh	30	182.4 ± 43.0	17.4 ± 3.5
	BSh _s	40	235.3 ± 55.1	22.4 ± 6.0
III	BA	140	812 ± 190.4	77.5 ± 15.3
	BA _s	39	226 ± 52.9	21.6 ± 4.7
IV	BMK	508	2970 ± 696.4	283.3 ± 70.8
	BMK _s	46	269 ± 63	25.7 ± 5.9
V	BMA	2766	16180 ± 3794.2	1543.6 ± 257.2
	BMA _s	239	1398 ± 327.8	133.4 ± 27.1
VI	BD	916	5313 ± 1245.8	506.9 ± 106.2
	BD _s	41	239.8 ± 56.2	22.8 ± 4.7

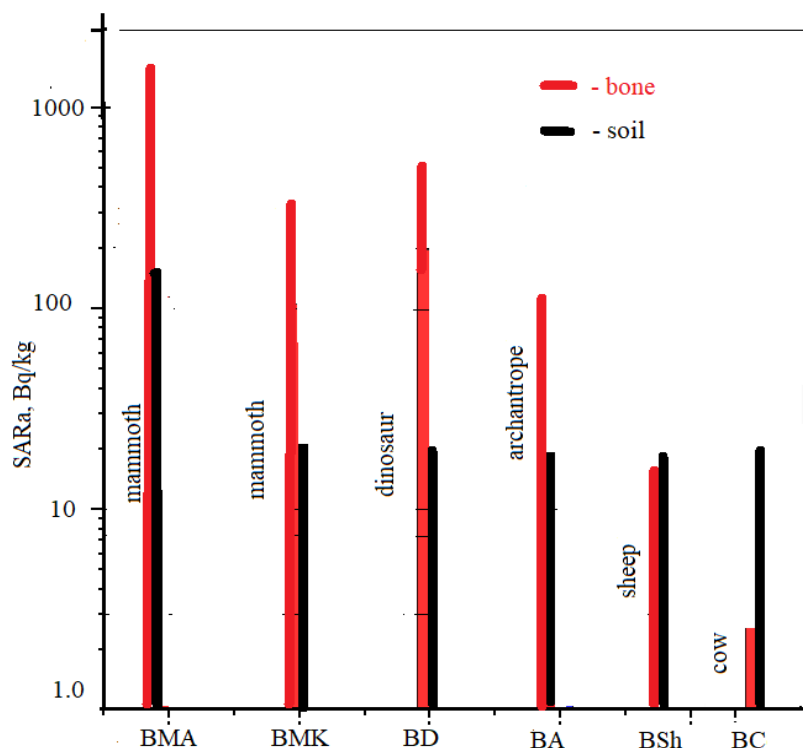


Figure 5. The histogram of S_{Ra} in samples of bones and its soils.

As one can see in Table 1, the values of S_{Ra} of the bone (BMA) and its soil (BMA_s) has been reached very higher values 1543.6 Bq kg⁻¹ and 133.4 Bq kg⁻¹, respectively. In Figure 5 presented a histogram of the S_{Ra} in samples of bones and its soils.

From Figure 5, it is clear that values of S_{Ra} in Angren soil (133 Bq kg⁻¹) is anomalous in comparison with other soils (that in a 17.7 - 25.7 Bq kg⁻¹) with average mean 22.0 Bq kg⁻¹. This showed, the S_{Ra} in Angren soils is 5.2–7.5 times more, than the S_{Ra} in other soils. This reality is explained by the fact that in Angren soils the contents of ²³⁸U very higher 7.8 mg kg⁻¹ [7]. It can be said that the soil is the main donor of ²²⁶Ra for bone remains.

4. DISCUSSION

Some many factors can be effected the penetration of Ra into bone remains in ground. These include the breeding ground of mammals, natural disasters associated with meteorites or unknown sources, as well as natural migration from the soil during the long bedding. There, we assumed the radium-226 is distributed uniformly throughout of the skeletons. Therefore to account for the quantitative distribution of ²²⁶Ra in bone-soil system the age coefficient (K_{age}) was introduced, which that determined by the following ratio (3):

$$K_{age} = S_{Rab}/S_{Ras} \quad (3)$$

where S_{Rab} and S_{Ras} are the specific activity of ²²⁶Ra in bones and soils, respectively, Bq kg⁻¹. In our previous work [23], the age coefficient was not taken into account. The ages of cow (I) and dinosaur (VI) bones were taken as a standard, due to its approximate ages 0.1 Y and 100 mln Y old, which corresponded to values of K_{age} 0.29 and 22.23. Although, the age of sheep skull was known in advance, it was taken as a control (that buried in a 2006-2007). By using these values of cow (I) and dinosaur (VI) calibration curve was obtained, and that presented in Figure 6.

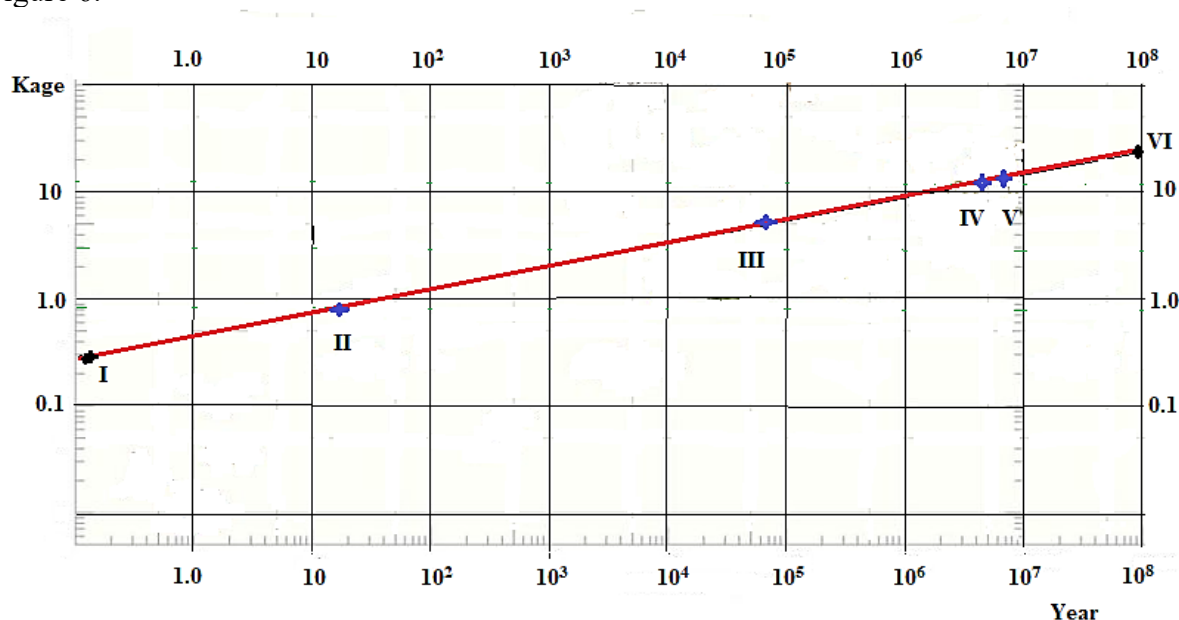


Figure 6. Calibration curve for assessing ages of bone remains.

From calibration curve of Figure 6, we assessed ages of the bones: 15.4 ± 2.1 Y for sheep, ≤ 0.10 mlnY for archanthropus, 8.2 ± 1.6 mln Y and 8.5 ± 1.6 mlnY for Kashkadarya and Angren mammoths at the values of $K_{age} = 0.77$ (II), 3.59 (III), 11.02 (IV) and 11.57 (V), respectively.

The assessed ages of bone remains are listed in the Table 2, where presented values of S_{Ra} , K_{age} and ages of cow-I; sheep-II; archanthropus-III; mammoths-IV,V and dinosaur-VI.

Table 2. The assessing of ages and values of S_{Ra} and K_{age} for bone remains.

Periods	Bone and soils	S_{Ra} , Bq kg ⁻¹	K_{age}	Age, Y
I	BC BC _s	6.45 ± 2.1 22.4 ± 6.0	0.29	0.1 ± 0.01
II	BSh BSh _s	17.4 ± 3.5 22.4 ± 6.0	0.77	15.4 ± 2.1
III	BA BA _s	77.5 ± 15.3 21.6 ± 4.7	3.59	≤ 0.1 mln
IV	BMK BMK _s	283.3 ± 70.8 25.7 ± 5.9	11.02	8.2 ± 1.6 mln
V	BMA BMA _s	1543.6 ± 257.2 133.4 ± 27.1	11.57	8.5 ± 1.6 mln
VI	BD BD _s	506.9 ± 106.2 22.8 ± 4.7	22.23	100 ± 17.0 mln

As shown the Table 2, the using of age coefficient (K_{age}) showed the assessment possibility of the ages of bones. Many years studies a show, the CR-39 track detectors very suitable for determination the specific activity of ²²⁶Ra in bones and led us to fact is being correlation between S_{Ra} and ages of skeleton. The example, the age of control sheep bone found out 15.4 ± 2.1 Y at $K_{age} = 0.77$ that corresponded to 2006-2007 that was buried. Ages of archanthrope made up ≤ 0.1 mln Y, the mammoths of Kashkadarya and Angren were found 8.2 ± 1.6 and 8.5 ± 1.6 mln years, respectively.

5. CONCLUSIONS

In this work, a simple, accessible, highly sensitive and selective method was developed, for assessment of the ages of the ancient bones by measuring specific activity of ²²⁶Ra on the CR-39 track detectors.

The calibration curve has showed the existence of correlation between specific activity of ²²⁶Ra and ages of skeletons. It is established the age of the Selungur archanthrop is ≤ 0.10 mln Y. The old of bones Kashkadarya and Angren mammoths were 8.2 and 8.5 mln years. That claimed, some 10 mln years ago in territory Central Asia had been life.

As is well known, ancient bone remains have being the material cultural heritage of the country, where they are discovered. So, it is difficult to collect them in a center for study due

to being of some barriers. This estimating method of the age of ancient bones is not complex to implement. I would like to see others results by using of this method.

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