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AUTORADIOGRAPHY FOR U, Th, AND ISOTOPIC DISEQUILIBRIUM STUDY OF SIWALIK FOSSIL BONES

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The relation between the concentration of uranium and thorium atoms in geological samples and the alpha-track production rate in the plastic track detector, LR-115 type II, was used for the analysis of fossil bones collected from the Siwalik Himalayas of India. The alpha-track measurements from the samples under secular equilibrium define the total uranium and thorium concentrations, while fission track measurements yield uranium concentration alone. By combining the results of both measurements, uranium and thorium concentrations can be determined. The results of uranium and thorium concentration determined by alpha-autoradiography are distinctly different from those of fission track analysis, indicating the existence of radioactive disequilibrium in Siwalik fossil bones.

INTRODUCTION

Uranium (U) and thorium (Th) have been present in traces in geological samples since their solidification. An emulsion detector, when placed against the surface of a geological sample, should record the disintegration of alpha-particles. Coppens (1949) gave the U and Th contents of a sample as a function of the alpha-track rate (N) as

$$10^6\,N = 8.58\,k\,U \tag{1}$$

if there is only uranium without thorium, or

$$10^6\,N = 1.92\,k\,Th \tag{2}$$

if there is only thorium without uranium, and

$$10^6\,N = (8.58\,U + 1.92\,Th)\,k \tag{3}$$

if both U and Th are present.

The constant k depends on the composition of the sample and is known as the coefficient of absorption. Coppens et al. (1977) further remarked that the above expressions (Eqs. 1-3) are not valid unless there is radioactive equilibrium. Coppens et al. (1977) improved Eq. 3 to

$$10^6\,N = (9.20\,U + 2.80\,Th)\,k \tag{4}$$

if both uranium and thorium are present and if the decay products are in equilibrium.

Equation 4 was modified by Singh et al. (1986) for use with solid-state nuclear track detectors which can record alpha-particle tracks more precisely. With a cellulose nitrate plastic detector (LR-115 type II), the modified equation is

$$10^6\,N = (6.77\,U + 1.60\,Th)\,k \tag{5}$$

where

$$k = 0.85/\sum(CS/A). \tag{6}$$

In Eq. 6, C is the concentration of an element of atomic weight A, and S is the stopping power of an

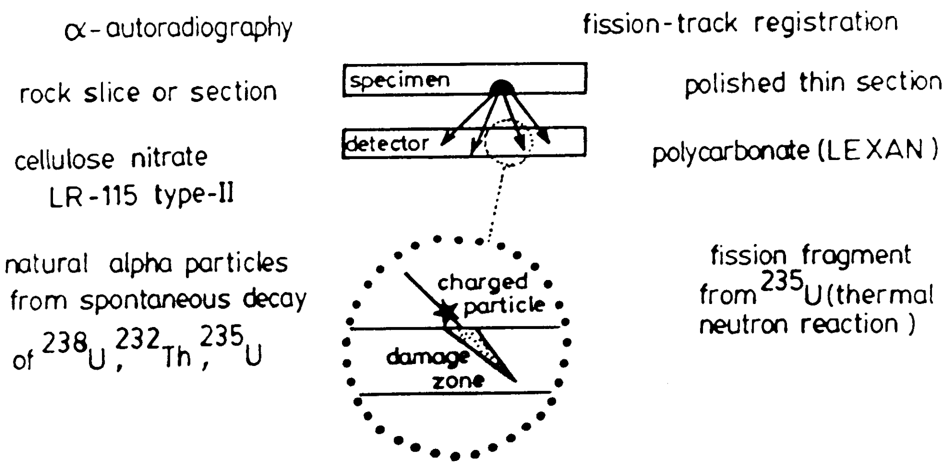

Fig. 1. Mechanism of F/α Technique.

alpha-particle for that element. Thus, knowing the value of k and estimating the uranium concentration by the homogenized fission track technique, the thorium concentration can be easily determined from Eq. 5. A comparison between uranium concentrations determined by alpha-autoradiography and those by other methods, e.g., fission track technique or fluorimetry, allows the detection of radioactive equilibrium/disequilibrium.

In this investigation, the modified relation was used for uranium and isotopic equilibrium/disequilibrium studies of some vertebrate fossil bones collected from the Siwalik Himalayas (Khetpuraly area, Haryana State).

EXPERIMENTAL TECHNIQUE

The homogenized fission track technique (Fisher 1970) was used for U estimations in fossil bone samples. In this method, the sample was powdered and pressed into a pellet which was sandwiched between Lexan plastic disks and irradiated with a known dose of thermal neutrons from a reactor along with a suitable U-rich standard glass dosimeter pellet. After irradiation, the plastic detectors were removed and etched in 6.25 N NaOH at 70°C for 25 min to obtain the fission track radiographs of the samples. These were then scanned under a microscope to determine the induced track densities. A comparison of the induced fission track densities recorded in the Lexan covering the unknown and standard pellets gave the U content in the sample, obtained by the following equation (Fleischer et al. 1975; Fisher 1977):

$$U_x = U_s \, \rho_x / \rho_s \tag{7}$$

where x and s refer to unknown and standard, respectively, and ρ is the induced fission track density.

To obtain alpha-autoradiographs, a sheet of cellulose nitrate plastic detector (LR-115 type II) was placed against the surface of the polished sections of the samples which were prepared by grinding with emery powder of mesh size varying from 100 to 600 μm and polished with cerium oxide. The whole assembly was then stored undisturbed for two months to allow the tracks to accumulate. The surface area of ≈1 cm^2 was appropriate for track scanning. After the accumulation period, the plastic sheet was removed and etched for 2 h in 2.5 N NaOH at 60°C. The alpha-tracks were then counted using an Olympus binocular microscope with a magnification of 600 X. The mechanism of the technique is shown in Fig. 1.

RESULTS AND DISCUSSION

Uranium concentration by fission track radiography

The induced fission track densities recorded in the Lexan plastic detectors, covering the unknown and standard pellets, were counted. Then, using Eq. 7, the uranium content of the unknown samples was calculated, which is reported in Table 2.

Uranium concentration by alpha-autoradiography

To derive the U and Th concentrations in fossil bones, through alpha-autoradiography, it is sufficient to determine the number (N) of optically visible alpha-particle tracks emitted per cm^2/s and to utilize Eq. 5. However, the value of the coefficient of absorption of alpha particles (k), which depends upon the composition of

Table 1. Calculation of k for fossil bones.

Element	A	C	S/A	CS/A
Ca	40	0.3829	0.047	0.017996
P	31	0.1773	0.049	0.008687
F	19	0.0726	0.053	0.003920
O	16	0.3668	0.066	0.024208

$\Sigma CS/A = 0.0548$
$k = 0.85/\Sigma(CS/A) = 15.51$

Table 2. U concentration determined by alpha-autoradiography and fission track analysis in fossil bones.

Sample	Sample	Specimen	10^6N	U_α(mg/kg)	U_f(mg/kg)
	Kh4	Bovid joint bone	3751	36	43
	Kh5	Elephant jaw	4336	41	52
Khetpuraly (Haryana)	Kh6	Bovid joint bone	1038	10	63
	Kh7	Elephant molar	3751	36	109
	Kh8	Bovid limb	2327	22	64
Nariangarh (Haryana)	FN3	Tortoise bone	7998	76	165
	FN4	Unidentified	5799	55	119
	FS1	Elephant tusk	6100	58	118
Saharanpur (U.P.)	FS2	Elephant tusk	2951	28	94
	FS3	Molar teeth	4976	48	102
	FS4	Unidentified	6620	63	159

U_f - U concentration determined by fission-track analysis.
U_α - U concentration determined by α-autoradiography.

the sample and is given by

$$k = 0.85/\Sigma(CS/A) \quad (8)$$

is not known. The constant k is calculated using the ratio S/A as already reported (Singh et al. 1986) and was found to be 15.51 for fossil bones (Table 1).

Since thorium is reported (Udas and Mahadevan 1974) to be absent in Siwalik fossil bones, Eq. 5 gets modified as

$$U = 10^6\, N / 6.77\, k. \quad (9)$$

As for the fossil bones, k was found to be 15.51. Hence, knowing N, the value of U can be calculated. Uranium concentrations, determined by alpha-autoradiography and fission track radiography, are reported in Table 2. Since it is known (Oakley 1963) that bones pick up uranium from ground water (as is used to infer relative ages of bones that are buried at a given site), the effect demonstrated in Table 2 is reasonable.

Study of radioactive equilibrium/disequilibrium

A comparison of the results of uranium content calculated by alpha-autoradiography and fission track analysis (which gives the true uranium content without taking the daughters into account) shows that these two values are distinctly different, confirming the presence of radioactive disequilibrium in the fossil bone samples. This corroborates the results already reported by some authors (Fisher 1977; Sharma et al. 1983; Singh et al. 1992).

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