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Uranium Estimation and Isotopic Disequilibrium Study of Siwalik Fossil Bones

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Abstract—Solid-state nuclear track detectors have been used for measuring uranium and thorium concentrations in fossil bone samples collected from the Siwalik Himalayas of India. The fission track measurement yields uranium concentration alone, while α-autoradiography of samples under secular equilibrium yields the total uranium and thorium concentrations. Thus, by combining the results of both measurements, uranium and thorium concentrations are determined. The results of uranium content, as determined by fission track analysis, are distinctly different from those of α-autoradiography, indicating the existence of radioactive disequilibrium in Siwalik fossil bones.

1. INTRODUCTION

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

$$10^6\ N = 8.58\ ku \tag{1}$$

or

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

and

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

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 (1977) further remarked that the above expressions [equations (1–3)] are not valid unless there is radioactive equilibrium. Also, the expressions are derived using a microscope with a minimum detection limit of 3 μm. Using a microscope which can identify trajectories of α-particles up to 2 μm, Coppens (1977) improved equation (3) to

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

Equation (4) was modified by Singh *et al.* (1986a) for use with solid-state nuclear track detectors which can record α-particle tracks more precisely. With cellulose nitrate plastic (LR-115 type 2), the modified equation is

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

where

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

In equation (6), C is the concentration of an element of atomic weight A, and S is the stopping power of an α-particle for that element. Thus, knowing the value of k and estimating the U concentration by the homogenized fission track technique, the Th concentration can be determined easily from equation (5). A comparison between the U concentrations determined by α-autoradiography and those by other methods (e.g. fission track or fluorimetry), allows the detection of radioactive equilibrium/disequilibrium. In the present investigation, the modified relation is used for a U and isotopic equilibrium/disequilibrium study of some Siwalik fossil bones collected from the Nadha Sahib area of Haryana (India).

2. EXPERIMENTAL TECHNIQUE

The homogenized fission track technique (Fisher, 1970), was used for U estimations in fossil bone samples. In this method, the sample is powdered and pressed into a pellet which is 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. The lexan plastic is then etched and scanned. A comparison of the induced fission track densities recorded in the lexan covering the unknown and standard pellets gives the U content in the sample, obtained by the following equation (Fisher, 1977):

$$U_{un} = U_{st} \cdot \rho_{un}/\rho_{st}$$

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

To obtain α-autoradiographs, a sheet of LR-115 type 2 plastic is placed against the surface of the polished sections of the samples which are prepared by grinding with emery powder of mesh size varying from 100 to 600 μm and polished with cerium oxide. The whole assembly is then stored undisturbed for 2 months to allow the tracks to accumulate. A surface area of ≈ 1 cm^2 is appropriate, both for preparation and, later, for scanning. After the accumulation period, the plastic sheet is removed and etched for 2 h in 2.5 N NaOH at 60°C. The α-tracks are then counted using an Olympus binocular microscope with a magnification of 600×.

3. RESULTS AND DISCUSSION

To derive the U and Th concentration in fossil bones, it is sufficient to determine the number (N) of optically visible α-particle tracks emitted per cm^2 per s and to utilize equation (5). However, the value of k, which depends upon the composition of the sample and is given by

$$K = 0.85/\sum(CS/A)$$

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

3.1. Uranium concentration by α-autoradiography

If U and Th represent the concentrations of uranium and thorium, N the number of α-tracks/cm^2/s and k the coefficient of absorption of α-particles, we have

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

if both U and Th are present. Since thorium is reported (Udas and Mahadevan, 1974) to be absent in Siwalik fossil bones,

$$\mathrm{U} = 10^6\ N/6.77\ k.$$

Table 1. Calculation of k for fossil bones

Element	A	C	S/A	CS/A
Ca	40	0.382	0.047	0.018
P	31	0.177	0.049	0.008
F	19	0.072	0.053	0.004
O	16	0.366	0.066	0.024

$\sum CS/A = 0.548$, $k = 0.085/\sum(CS/A) = 15.51$

where C is the concentration of an element of atomic weight A and S is the stopping power of an α-particle for that element.

Table 2. Uranium content determined by α-autoradiography and fission track analysis in fossil bones

Sample number	Specimen	10^6 N	U_α (ppm)	U_f (ppm)
Nadha Sahib				
N 16	Unknown	8,930	85.04	23.95
N 17	Unknown	17,413	165.84	23.48
N 18	Unknown	14,064	133.95	9.29
N 19	Unknown	9,376	89.30	23.01
N 20	Unknown	12,278	116.94	22.69
N 21	Unknown	12,502	119.06	10.97
N 22	Unknown	14,511	138.20	11.97
N 23	Unknown	8,706	82.92	25.75
Naraingarh				
V 1	Unknown	6,251	59.53	36.81
V 2	Molar teeth, piece of stegodon	22,101	210.49	40.66
V 3	Unknown	8,483	80.79	45.04
V 4	Teeth of elephant	2,455	23.38	18.12
V 7	Unknown	7,813	74.41	18.24
V 8	Elephant teeth fragment	6,027	57.41	20.64
V 10	Limb bone	6,697	63.78	21.80
V 11	Unknown	6,929	65.91	22.90

U_α = U content determined by α-autoradiography.
U_f = U content determined by fission track analysis.
Value of k (absorption coefficient) for fossil bone = 15.51.

For fossil bones, k is found to be 15.51. Hence, knowing N, we can calculate the value of U. Uranium concentrations, determined by α-autoradiography and fission track radiography, are reported in Table 2.

3.2. Study of radioactive equilibrium/disequilibrium

A comparison of the results of uranium content calculated by α-autoradiography and fission track analysis (which gives the true U content without taking the daughters into account), shows that these two are distinctly different, which confirms the presence of radioactive disequilibrium in the fossil bone samples. This corroborates the results already reported by some authors (Sharma *et al.*, 1983; Singh *et al.*, 1986b).

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