Determination of Trace Concentration of Uranium in Soils by the Nuclear Track Technique

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Abstract

Solid state nuclear track detector CR-39 has been used to estimate trace concentration of uranium in soil and sand samples from various places of Bangladesh. Uranium contents in soil samples have been found to vary from \(-3.79\) to \(-8.63\) ppm and in sand samples from \(-2.39\) to \(-6.53\) ppm. The mean concentration in soil and in sand samples were found to be \(-4.52\) and \(-2.96\) ppm respectively. The maximum uranium concentration in soil samples was observed in Sylhet while the uranium concentration of sand was found to be maximum in the sea beach of Cox's Bazar. The implication of results is briefly discussed in the paper.

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INTRODUCTION

The whole biotic community of our biosphere cannot avoid the radiation effects. Amongst the different radiation isotopes, uranium is the heaviest element which may be consumed by living species as a result of food chains and environmental conditions. Furthermore, the emanation of α-radioactive radon (222Rn) is associated with the presence in the ground of radium and its ultimate precursor uranium. Although these elements occur in virtually all types of rocks and soils, their amounts vary with the specific site and geological material. As an inert gaseous element, radon can move through the soil freely from its source, the distances being determined by factors such as rate of diffusion, effective permeability of the soil and by its own long half life. The health hazard that it presents in nature has been widely studied in the last decade (Bodansky, 1987; Fleischer, 1988; Nero and Lowder, 1983). Under normal circumstances the dominant contributor to indoor or environmental radon concentrations is the emanation from the soil (Nero, 1985). If uranium rich material lies close to the surface of the earth there can be high radon emanation rates. In such locations, the hazards from radon exposures can be much greater than average. Hence, a knowledge of uranium contents in soils will be useful from the point of view of health physics.

Solid state track detection technique has been widely used for the determination of uranium contents in a variety of solid materials by placing samples in contact with the detector and irradiating with neutrons and measuring the tracks from induced fission (Ginrich, 1975; Chakarvarti and Nagpaul, 1979; Khan et al., 1980). However, Gamboa et al., (1984) have shown that by detection of alpha particles produced by the disintegration’s of uranium and its daughters it is possible to estimate the concentration of uranium in mineral rocks, without the requirement of extra activation or nuclear reactor.

In this paper uranium concentration has been determined from the measurement of alpha particle tracks on CR-39 plastic detectors placed in contact with the samples over a period of time and chemically etched in NaOH solution.
THEORY

Let us consider a radioactive alpha decay chain such as:

\[ A \rightarrow B \rightarrow C \rightarrow \ldots \]  

(1)

Assuming that the decay rate of A is fairly slow compared with the time interval being considered then at equilibrium:

\[ \lambda_A N_A = \lambda_B N_B = \lambda_C N_C \ldots \]  

(2)

where, \( \lambda_A N_A \) etc. are the number of atoms of A etc. decaying per unit time. After \( n \) alpha decays in the series the total number of alphas per unit time will be given by:

\[ N_\alpha = n\lambda N \]  

(3)

In the decay chains \( {}^{238}\text{U} \) transforms into \( {}^{206}\text{Pb} \) and \( {}^{235}\text{U} \) into \( {}^{207}\text{Pb} \):

\[ {}^{238}\text{U} \rightarrow {}^{206}\text{Pb}, \quad n=8 \]

\[ {}^{235}\text{U} \rightarrow {}^{207}\text{Pb}, \quad n=7 \]

In natural uranium the isotopic abundance's of \( {}^{235}\text{U} \) and \( {}^{238}\text{U} \) is given by the ratio:

\[ {}^{235}\text{U} : {}^{238}\text{U} = 1 : 140 \]  

(4)

Let the sample (soil or sand) contain \( X_u \) ppm by weight of uranium. If the sample has a density \( \rho \) then 1 cm\(^3\) (pg) of material contains:

\[ \rho X_u A \frac{10^{-6}}{238} \] atoms of \( {}^{238}\text{U} \) and

\[ \rho X_u A \frac{10^{-6}}{238 \times 140} \] atoms of \( {}^{235}\text{U} \), where \( A \) is Avogadro's number.

Number of alphas emitted from 1 cm\(^3\) of sample due to \( {}^{238}\text{U} \) decay is:

\[ {}^{238}\text{U}_\alpha = 8\lambda_238 \rho X_u A \frac{10^{-6}}{238} \]  

(5)

Number of alphas emitted from 1 cm\(^3\) of sample due to \( {}^{235}\text{U} \) decay is:

\[ {}^{235}\text{U}_\alpha = 7\lambda_235 \rho X_u A \frac{10^{-6}}{238} \frac{1}{140} \]  

(6)

Total number of alphas due to uranium (both \( {}^{238}\text{U} \) and \( {}^{235}\text{U} \)) decay in the sample is:

\[ \alpha_{\text{total}} = {}^{238}\text{U}_\alpha + {}^{235}\text{U}_\alpha \]  

(7)
Let the distance between the plastic detector and the sample be $x$ and let alphas hit the plastic from the sample at an angle $\theta$ (Fig 1).

The probability that the direction vector lies in any angle interval between $\theta$ and $\theta + d\theta$ is

$$P(\theta)d\theta = \frac{1}{2} \cos \theta d\theta$$

(8)

The probability at depth $x$ of reaching the surface is given by:

$$P(x) = \int_{\theta_1}^{\theta_2} \frac{1}{2} \cos \theta d\theta = \frac{1}{2} (1 - \sin \theta_2)$$

(9)

The number of alphas coming from a layer $dx$ in the sample is given by the equation (Andam, 1985):

$$\alpha_{\text{total}} \, dx \, \frac{1}{2} (1 - \sin \theta_2) = \frac{1}{2} \alpha_{\text{total}} \left(1 - \frac{x}{R_{\alpha}}\right) \, dx$$

(10)

where, $R_{\alpha}$ is the alpha particle range in the sample. This represents the track density $\rho_t$ in the plastic:

$$\rho_t = \frac{1}{2} \alpha_{\text{total}} \int_0^{R_{\alpha}} \left(1 - \frac{x}{R_{\alpha}}\right) dx = \frac{1}{4} \frac{\alpha_{\text{total}} R_{\alpha}}{\alpha_{\text{total}}}$$

(11)

Using (5) - (7), (11) can be expressed as:

$$\alpha_{\text{total}} = \frac{4 \rho_t}{R_{\alpha}}$$

(12)

Hence, $X_u$ can be estimated from the relation:

$$X_u = \frac{4 \times 238 \rho_t}{R_{\alpha} \rho A \left[8 \lambda_{238} + \frac{7 \lambda_{235}}{140}\right] \times 10^{-6}}$$

(13)
EXPERIMENTAL

In order to use in the present study, 250μm thick CR-39 plastic sheet procured from Pershore Mouldings Ltd., England was cut into small pieces of the size 2.0 cm x 1.5 cm. Each of these small pieces was used as a detector.

A total of 73 samples (49 soil samples and 24 sand samples) have been collected from various places spanning over several regions of Bangladesh (Fig.2). The samples were generally collected from the slopes of ponds and rivers at a depth of about 1 metre from the ground level. Such a level was chosen in order to ensure an old age of the samples which may be the representative of the particular place. At first the samples were dried in the laboratory and then ground into fine powder. With the help of a wooden sieve, the samples were cleaned of coarse grains and bits of straw, glass etc. About 150 grams of each sample was taken into a plastic can of height 7.5 cm and diameter 6.5 cm. Four detectors were exposed in direct contact with the powdered sample as shown in Fig. 3. The exposures were made for 15 days with the detector held in $2\pi$ geometry.

After irradiation, each of the detectors was etched in 6N NaOH solution at a constant temperature of 70 ± 1 °C for 4 hours. The central portion of the detector strips were scanned using a binocular research microscope at a magnification of 450x (45x objective and 10x eyepiece). The track density was determined by counting the alpha particle tracks in 60 fields of view. For each sample, an average track density of the four detectors placed in a can was taken. Five unexposed detectors were etched and studied under the microscope in the beginning of the experiments in order to determine the common background. All subsequent counts were corrected for this background value. The alpha track densities were transformed into uranium concentrations using Eqn. (13). The ranges of alpha particle were taken to be 107 μm and 71 μm in soil and sand respectively (Gamboa et al., 1984).

RESULTS AND DISCUSSIONS

Table 1 gives the average alpha track densities (tracks cm$^{-2}$d$^{-1}$) for soil samples of various places of Bangladesh registered by each set of detectors. The corresponding uranium concentrations in ppm (parts per million) are also presented in the same table and the results obtained are displayed in Fig. 4. The highest concentration has been found in Sylhet and the lowest in Rajshahi. The significantly higher concentration of uranium in Naogaon soil sample is in agreement with earlier measurement (Islam et al., 1991) of high radon activity in soil and dwellings of the same locality.
For sand samples the average alpha track densities and corresponding uranium concentrations are presented in Table 2 and the results are displayed in Fig 5. The samples of Cox's Bazar sea beach show highest uranium concentration while those of Naogaon show lowest uranium concentration.

The comparison of Tables 1 and 2 shows that except for in the samples of greater Chittagong, the uranium concentrations in the soil samples from all other places are consistently higher than those of sand samples. The mean uranium concentrations in soil and sand samples are $4.52 \pm 0.35$ and $2.96 \pm 0.28$ ppm respectively.

Since the disintegration of radionuclides is a random process only an estimate of uranium concentrations in soil and sand samples can be obtained. The exposure time (15 days) followed in this study is long enough to get a large number of tracks in the plastic detectors. Furthermore, in order to reduce the statistical error, tracks over 60 fields of view were counted for each small piece of detector. In spite of these, a negligible error in this study may arise from the diffusion of radon ($^{222}\text{Rn}$) gas which arises in the decay chain of uranium.

CONCLUSIONS

Uranium concentration in the soil of Sylhet has been found to be very high. This necessitates the investigation of uranium mine in that region. The higher uranium concentration in the reddish soil samples of Naogaon is in agreement with the high radon activity (Islam et al., 1991) in that place.

Among sand samples, the sea beach sand of Cox's Bazar shows high uranium concentration. This is consistent with previously reported (Kasim, 1988) high radon - thoron emanation from the sea beach sand of Cox's Bazar. Thus, there seems to be a correlation between uranium content of the soil and the radon emanations rate.

The method provides the best combination of simplicity, accuracy and reliability by utilizing the track detecting solids to record the alpha particles emitted by uranium and its daughters. It goes without saying that this method does not require any expensive instrumentation which makes the technique very useful and attractive for developing countries like Bangladesh.
Acknowledgments

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REFERENCES


Table 1

Mean uranium concentration in soil samples collected from different places in Bangladesh.

<table>
<thead>
<tr>
<th>Source of samples</th>
<th>Average alpha track density per day (cm$^2$ d$^{-1}$)</th>
<th>Average estimated uranium content (ppm by wt.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rajshahi</td>
<td>150.28 ± 1297</td>
<td>3.79 ± 0.33</td>
</tr>
<tr>
<td>Naogaon</td>
<td>196.87 ± 14.69</td>
<td>5.96 ± 0.37</td>
</tr>
<tr>
<td>Dinajpur</td>
<td>159.64 ± 13.20</td>
<td>4.02 ± 0.33</td>
</tr>
<tr>
<td>Rangpur</td>
<td>202.76 ± 14.87</td>
<td>5.11 ± 0.37</td>
</tr>
<tr>
<td>Barishal</td>
<td>207.84 ± 15.08</td>
<td>5.24 ± 0.38</td>
</tr>
<tr>
<td>Meherpur</td>
<td>216.52 ± 15.37</td>
<td>5.45 ± 0.39</td>
</tr>
<tr>
<td>Chittagong</td>
<td>224.06 ± 15.99</td>
<td>5.65 ± 0.39</td>
</tr>
<tr>
<td>Rangamati</td>
<td>178.46 ± 13.68</td>
<td>4.50 ± 0.34</td>
</tr>
<tr>
<td>Sylhet</td>
<td>342.55 ± 19.11</td>
<td>8.63 ± 0.48</td>
</tr>
</tbody>
</table>
Table 2

Mean uranium concentration in sand samples collected from different places in Bangladesh.

<table>
<thead>
<tr>
<th>Source of samples</th>
<th>Average alpha track density per day (cm$^2$ d$^{-1}$)</th>
<th>Average estimated uranium content (ppm by wt.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rajshahi</td>
<td>90.24 ± 10.35</td>
<td>2.39 ± 0.26</td>
</tr>
<tr>
<td>Chapai-Nawabgonj</td>
<td>77.75 ± 9.74</td>
<td>1.97 ± 0.25</td>
</tr>
<tr>
<td>Naogaon</td>
<td>56.32 ± 8.50</td>
<td>1.43 ± 0.22</td>
</tr>
<tr>
<td>Dinajpur</td>
<td>62.62 ± 8.80</td>
<td>1.59 ± 0.22</td>
</tr>
<tr>
<td>Cox's Bazar</td>
<td>257.83 ± 16.67</td>
<td>6.53 ± 0.42</td>
</tr>
<tr>
<td>Rangamati</td>
<td>173.13 ± 13.88</td>
<td>4.39 ± 0.35</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Fig. 1. Plastic in contact with alpha decaying material.

Fig. 2. Map of Bangladesh indicating the sources of samples.

Fig. 3. Arrangement of detectors for the measurement of uranium concentration in soil and sand samples.

Fig. 4. Mean uranium concentrations of soil samples from various places in Bangladesh.

Fig. 5. Mean uranium concentrations of sand samples from various places in Bangladesh.
Plastic

Fig. 1

α - decaying material

$\theta_L$, $\theta$, $R_\alpha$
Bay of Bengal

• Source of sample

Fig. 2

13
Fig. 4
Fig. 5