Radioactivity in the industrial effluent disposed soil

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Abstract

Studies on radiation and radioactivity distribution in the soils of effluent disposed from the sugar industry in India have been conducted. The external gamma dose rates in air and natural radionuclides activities in the soils were measured using an Environmental Radiation Dosimeter and a Gamma-ray Spectrometer respectively. The soil samples were also subject to various physico-chemical analyses. This study revealed some remarkable results that are discussed in the article.

1. Introduction

It is well known that India is one of the countries in the world having the highest background radiation levels\cite{1}. Measurement of natural radioactivity in soil is essential to evaluate any change induced by humans in the future and it is also required to trace the movement of radionuclides from the soil reservoir to environmental and biological systems. The primary

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source of radiation received by human beings is from the natural radioactivity in soils, which derives essentially from $^{40}$K and the radionuclides of the $^{238}$U and $^{232}$Th decay series. The measurement of $^{40}$K is also useful to agronomists for studies connected with plant nutrients [2].

Sethiyathope is one of the places of Tamilnadu in India, has been identified, from the investigations made by Senthilkumar et al. [3, 4], as a polluted area due to the continuous discharge of effluents containing heavy metals with toxic chemicals from the sugar industry. In the present study, an attempt has been made to explore the radiation level and to provide the baseline data for the further assessment of the impact of industrial effluents. Naturally occurring radionuclides, namely, $^{40}$K, $^{232}$Th and $^{238}$U, were measured in soil samples of the region, and the resultant dose in air due to these radionuclides was calculated.

2. Materials and methods

Twenty-two sampling sites have been selected across the polluted area and soil samples were collected according to standard procedures [5]. They were labelled as $S_1$ to $S_{22}$. For the physico-chemical analyses, soil samples were processed according to the standard methods [6, 7] and the heavy metal concentration was determined using ICP-AES. To find the radioactivity levels, the samples were dried in an oven at 110 °C till a constant dry weight was obtained. Then these samples were powdered and sieved through a 150-μm mesh. The processed soil samples were packed in a 250 ml plastic container to its full volume with uniform mass. These containers were sealed hermetically and also sealed externally to ensure that all the daughter products of uranium and thorium and in particular, radon isotopes formed do not escape. A time of 30 days was allowed after packing to attain secular equilibrium between $^{226}$Ra and its short-lived daughter products. The net-weight of each sample was determined before counting. To reduce the contribution from background radiation while recording the spectrum in the laboratory, the samples were kept in a lead shield with a shielding efficiency of 95 percent [8].

The gamma-ray spectrometer was used to determine the activity of the radionuclides, $^{232}$Th, $^{238}$U and $^{40}$K. A sodium iodide [NaI(Tl)] crystal detector of 3” × 3” size combined with an 8k multi-channel analyser (model PCA-II) was used to record the gamma-ray spectra. Standard International Atomic Energy (IAEA) sources were used for calibrating the gamma-ray spectrometer. The counting time for each sample was 20,000 seconds.
Table I: Physico-chemical parameters of soil samples.

| Parameters | Range       | Mean   |
|------------|-------------|--------|
|            | Min         | Max    |
| pH         | 4.60        | 5.90   | 5.30  |
| EC (dSm\(^{-1}\)) | 0.60        | 0.86   | 0.70  |
| Cd (ppm)   | 4.30        | 8.65   | 7.32  |
| Cu (ppm)   | 41.80       | 65.50  | 54.01 |
| Mn (ppm)   | 775.65      | 1215.00| 1015.30|
| Ni (ppm)   | 63.12       | 91.00  | 77.57 |
| Pb (ppm)   | 2.30        | 11.95  | 5.92  |
| Zn (ppm)   | 74.80       | 109.00 | 90.53 |
| Fe (%)     | 7.90        | 17.70  | 12.46 |
| Mg (%)     | 1.70        | 3.43   | 2.66  |

3. Results and discussions

3.1. Physico-chemical analysis of soil

Table I gives the physico-chemical characteristics of the soil samples. All soil samples were clay in texture. This is due to the continuous disposal of effluents. The absorption pattern of heavy metals, pesticides etc. is higher in clay soil [9]. Soil pH plays a vital role on the availability of nutrient, metal elements, existence of microorganisms and maintenance of physical properties. When electrical conductivity (EC) is more than 1 dSm\(^{-1}\), the germination of all crops are affected [10]. The present study shows that the EC values are close to the maximum tolerance limit. The concentration of heavy metals, Cd, Cu, Fe, Mn, Mg, Ni, Pb and Zn are also given in table I. Cd in agricultural soil is mainly due to irrigation with industrial effluents and sewage-sludge in the fields, and it is normally below 1 ppm [11]. Cu at higher concentration is toxic to most plants and the normal amount in soil is 20 ppm [11]. In this study, a high concentration of Cu was observed compared to the normal value. Mn, which is an essential element for plants, shows higher concentrations compared to the normal amount of 850 ppm. One of the highly toxic metals for plants is Ni. The concentration of Ni exceeds the recommended amount of 50 ppm for agricultural soils [12]. The concentration of Pb varied from 2.3 to 11.95 ppm. Though Zn is one of the
Table II: Activities and exposure dose rates of naturally occurring radionuclides in soil samples.

| Radionuclide      | Range               |
|-------------------|---------------------|
|                   | Min | Max  | Mean  |
| 232Th (Bq kg\(^{-1}\)) | 37.3 | 43.2 | 39.75 |
| 238U (Bq kg\(^{-1}\)) | 20.28 | 24.72 | 22.76 |
| 40K (Bq kg\(^{-1}\))  | 220.9 | 270.3 | 253.16 |
| Calculated Dose Rate (nGy h\(^{-1}\)) | 44.7 | 47.8 | 46.1 |
| Observed Dose Rate (nGy h\(^{-1}\)) | 70.0 | 85.0 | 78.3 |

essential elements, it is required only in minute amounts. However, higher concentrations of Zn, which varies from 74.8 to 109 ppm, were found in this study. As the natural abundance of Fe and Mg are more in earth, higher concentrations were noticed.

3.2. Gamma-ray spectral analysis of soil

The concentration of primordial radionuclides estimated in the soil samples is presented in table II. The world average concentration of 232Th, 238U and 40K is 30, 35 and 400 Bq kg\(^{-1}\) [13]. When compared to the activity concentration of radionuclides with the world average value, 232Th is higher by a factor of 1.33 whereas both, 238U and 40K, are lower by a factor of 0.62.

The external dose rates have been obtained from the soil activity concentration of the three primordial radionuclides. They are given together with the activity concentration of three primordial radionuclides for the soil samples in table II. A graphic plot [fig. 1] of observed dose rate with that of calculated using UNSCEAR [13] conversion factors yielded a good correlation with a co-efficient of 0.89. Figure 2 shows the frequency distributions of the activities of 232Th, 238U and 40K. The activity distributions of 232-Th and 238-U are practically symmetrical, while the left tail in the activity distribution of 40K is slightly longer than the right as can be seen in the figs. 2a, 2b and 2c, respectively. The activities corresponding to 232Th, 238U and 40K are fitted to a normal curve.
Figure 1: Correlation between calculated and observed dose rates.

Figure 2a: Frequency distribution of $^{232}$Th activities.
Figure 2b: Frequency distribution of $^{238}$U activities.

Figure 2c: Frequency distribution of $^{40}$K activities.
Table III: Gamma dose rates from naturally occurring radionuclides.

| Range                  | 232Th | 238U  | 40K  |
|------------------------|-------|-------|------|
| Dose rate (nGy h⁻¹)    |       |       |      |
| Min                   | 24.84 | 8.7  | 9.28 |
| Max                   | 28.77 | 10.6 | 113.5|
| Median                | 26.41 | 9.94 | 10.73|
| Total Dose (nGy h⁻¹)  |       |       |      |
| 232Th                  | 53.27 | 60.5 |      |
| 238U                   | 45.2  | 48.42|      |
| 40K                    | 46.89 |      |      |
| Percentage contribution|       |       |      |
| 232Th                  | 53.27 | 60.5 |      |
| 238U                   | 18.71 | 22.38|      |
| 40K                    | 20.09 | 24.35|      |

### 3.3. Comparison of radioactivities with other environments

It was found that mean activities of 238U and 232Th were higher by a factor of 1.55 and 2 respectively, when compared to the Indian average values [14]. When compared to the world average values, the activity of 232Th is higher by a factor of 1.33, while 238U and 40K are lower in their activities [13].

### 3.4. Gamma dose rates from naturally occurring radionuclides

From the concentration of 232Th, 238U and 40K, the gamma dose rates $D$ in air were computed using the procedure reported by Beck et al. [38] and the results are given in table III.

$$ D = 0.666S_{\text{Th}} + 0.429S_{\text{U}} + 0.042S_{\text{K}} (\text{nGy h}^{-1}), \quad (1) $$

where, $S_{\text{Th}}$, $S_{\text{U}}$, and $S_{\text{K}}$ is the specific activity concentration of 232Th, 238U and 40K, respectively.

The gamma dose rate due to the presence of 232Th, 238U and 40K varies from 45.2 to 48.42 nGy h⁻¹ with a median of 46.89 nGy h⁻¹, which is lower than the world average value of 59 nGy h⁻¹. Table III also shows the results of percentage contribution of 232Th, 238U and 40K to the external dose rates in air. The median values of percentage contribution of the dose rates are, respectively, 56.3%, 21.5% and 22.84%. Narayana et al. [15] have reported the percentage contributions to be 46.7% and 13%, respectively for 232Th and 40K, which are much lower than those of the present study. For Indian
soils, Mishra and Sadasivam [14] have reported these percentage contributions to be 33.6% and 48.7% due to $^{232}$Th and $^{40}$K, respectively. The world wide average values reported by UNSCEAR [16] for the radioactive contributions to the gamma dose rates in air from $^{232}$Th and $^{40}$K are 40% and 35%, respectively.

3.5. Correlation between heavy metals and radionuclides

An attempt has been made to study the correlation between the heavy metals and radioactive elements present in soil samples. A good correlation was observed between the $^{232}$Th activity and Fe ($r = 0.76$), Mg ($r = 0.66$), Pb ($r = 0.68$), Zn ($r = 0.68$) and Ni ($r = 0.68$). These heavy metals (Fe, Mg, Pb, Ni and Zn) are part of the composition of thorium bearing minerals such as Thorlanite, Yttrocrasite, Zirkelite, Brammerire-thorutite series and Polymignite [17]. So this might be one of the reasons that the activity concentration of $^{232}$Th is higher than the concentration of $^{238}$U in the soil samples.

4. Conclusion

Gamma-ray spectral analysis of soil samples reveals the presence of $^{232}$Th, $^{238}$U and $^{40}$K in all the sites of the studied area. The activity concentration of $^{232}$Th is higher than $^{238}$U. A good correlation was observed between $^{232}$Th and Fe, Mg, Pb, Zn, and Ni, which are in the composition of thorium bearing elements. The median of the gamma dose rate of 46.89 nGy h$^{-1}$, is due to the presence of $^{232}$Th, $^{238}$U and $^{40}$K. It is lower compared to the world average value. The data obtained in this study will serve as a baseline survey for primordial radionuclide concentration in soil.

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