Radioactivity (Gross-A and Gross-B) Studies of Surface Water Collected from Domiasiat Area, West Khasi Hills District, Meghalaya, India

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Abstract-In this study, the gross activity of the alpha and beta emitting radionuclides present in the naturally occurring water bodies in the Uranium mineralization zone of Domiasiat, West Khasi Hills District, Meghalaya, India was determined. In our study, the gross radioactivity indicates that the water sample under investigation has a low concentration of both alpha and beta emitters and the activity was less than 0.5Bq/l for alpha activity and 1.0Bq/L for beta activity which is lower compared to the WHO prescribed limit. We estimate that the additional equivalent effective dose derived from water consumption is less than 0.3mSv/yr provided the water consumption for the population is on average, 730litres per inhabitant per year. We estimate that the additional equivalent effective dose derived from water consumption is less than 0.3mSv/yr provided the water consumption for the population is on average, 730litres per inhabitant per year (2L per day)

Keywords- Activity; Alpha, Beta; Dose; Intake; Uranium

I. INTRODUCTION

Mother Nature has provided immense natural resources to humanity. Water is one such natural resource which is essential for human life and for health and environment. The first health requirement for any developing nation is abundant supply of clean water. Water, the most indispensable and precious natural resources are expected to be free from pollution. It has two dimension that are closely linked, quantity and quality. In nature, all water contains some impurities. Water quality is one of the highest priority environmental issues. The water resources of a country determine the food production, public health, industrial development and hence its economy. Water in the ocean constitutes up to 97.2%, ice caps of glacier 2.14%, ground water of depth 13,000 ft 0.61% fresh water lakes 0.009%, atmosphere-0.001% and biotic communities- 0.0001%.

Naturally occurring radionuclides of terrestrial origin, also called primordial radionuclides, are present in the environment. Among the primordial radionuclides, the main contributors to external and internal exposure are $^{238}$U and its daughters and $^{232}$Th and its daughters (Yarar et al., 2005). In sense of lives health and environmental pollution, determination of the radioactivity in water (in drinking as well as usage water) is useful (Ozmen et al., 2004). The measurement of radioactivity in drinking water allows the determination of population exposure to radiation by the habitual consumption of water. The occurrence of radionuclides in water and edibles (terrestrial and aquatic) items causes human internal exposure, caused by the decay of radionuclides taken into the body through ingestion. They act when they are incorporated as part of the human food chain. Since the doses from these pathways are strongly related to the amount of radionuclides present, an important objective from the point of view of the radio ecological protection of the population is the accurate evaluation of the amounts received in dietary intake.

Water quality is an important parameter of environmental studies (Karahan et al., 2000). Natural waters contain both alpha- (e.g., $^{238}$U) and beta-($^{40}$K) emitters in widely varying concentrations which are responsible for a generally small fraction of the total dose received from natural and artificial radioactivity (UNSCEAR, 1993). Gross alpha and gross beta radioactivity measurements are of particular interest for routine monitoring purposes.

The Domiasiat area in the West Khasi Hills District of Meghalaya, India has been identified as one having a Uranium ore deposit. Uranium, being radioactive is known to decay to the stable element lead via. a series of steps involving both alpha and beta decay. Because of the decay series, $\alpha$ and $\beta$ emitting radionuclides will be naturally present in the environment which may find their way into streams, springs, rivers etc which inturn can find their way in the diet of man which if left unchecked may pose a serious threat to the health of an individual. Hence, the study was undertaken to determine the $\alpha$ and $\beta$ activity levels in surface waters available in nine selected sampling locations in Domiasiat and its surrounding areas and to assess the dose in which the local populace is being expose through drinking water pathway.

II. EXPERIMENTAL

A. Study Area:

The following are the nine locations in the Domiasiat area in West Khasi Hills District of Meghalaya with their GPS readings where the water samples were collected.

- Phlangdiloin, (N25°31´26.8";E91°24´44.1")
- Wahkaji, (N25°35´93.3";E91°20´48.5")
- Umdohlun, (N25°37´91.1";E91°27´06.1")
- Domiasiat, (N25°33´82.3";E91°22´28.8")
- PhudKylleng, (N25°32´20．5";E91°20´60.8")
- PhudSyngkai, (N25°31´99.8";E91°21´08.2")
- NongbahJynrin, (N25°31´84.9";E91°20´18.9")
- Nongtynger, (N25°20´31.8";E91°12´50.4")
- Mawthabah, (N25°21´11.1";E91°19´60.3")

The location of the Domiasiat area in the West Khasi Hills District of Meghalaya is depicted in Fig-1.
B. Analysis:

1). Sample Collection:
   Water Sample: Samples were collected in a pre-cleaned polythene bottle and acidified with nitric acid to a pH ~ 2-3.

2). Sample Preparation:
   a) Water Sample:
      One (1) liter sample is filtered to remove unwanted impurities. This is then kept for digestion and precipitation is done if high TDS is present. Samples with low TDS were evaporated, deposited in a one inch diameter stainless steel planchet and then ignite at a very high temperature.

3). Sample Digestion:
   a) Gross Alpha:
      The acid extracts (in HNO3 after destruction of any organic matter) are suitably pretreated chemically before the final isolation method. For isolation of transuranic nuclides, 1.0mg Lanthanum carrier is precipitated as LaF3 and the water washed precipitate is deposited on an inch diameter stainless steel planchet by evaporation of an aqueous solution and ignite at a very high temperature and then counted on an Alpha detector probe, manufactured by Nucleonix, India.

   b) Gross Beta:
      For the isolation of precipitated fission product radionuclides, Sulkovich Reagent is added till precipitation. If no precipitate is observed 20mg of Calcium carrier is added and precipitated as Calcium Oxalate carrying Stroncium and rare earth radionuclides and then, evaporating the solution in an aluminium planchet and dried under an infrared lamp. This was then counted using G.M Detector manufactured by Nucleonix, India.

C. Radiation Loss by Self Absorption:
   The radiation from alpha emitters having energy of 8MeV and from beta emitters having energy of 60KeV will not escape from the sample if the emitters are covered by a sample thickness of 5.5mg/cm2. The radiation from a weak alpha emitter will be stopped if covered by only 4mg/cm2 of sample solid. Consequently, for low level counting it is imperative to evaporate all moisture and preferable to destroy organic matter before depositing a thin film of sample solids from which radiation may enter the counter. In counting water samples for gross beta radioactivity, a solid thickness of 10mg/cm2 or less on the bottom area of the counting pan is recommended (APHA, 1986).

D. Calculation:
   The formula used for calculation of gross alpha and gross beta activity is given by
   \[ \text{Activity (Bq/L)} = \frac{Cs+b - Cb}{T \times \eta \times V} \pm \frac{(Cs+b + Cb)^{1/2}}{\eta \times V \times T} \]
   Where,
   \[ Cs+b = \text{Sample counts} \]
   \[ Cb = \text{Background counts} \]
   \[ T = \text{Counting time} \]
   \[ \eta = \text{Efficiency of the detector} \]
   \[ V = \text{Volume of the sample taken in litres} \]

   The minimum detection limit (MDL) is calculated using the following formula:
   \[ \text{MDL} = \frac{3 \sqrt{B}}{T \times \eta \times V} \]
   Where,
   \[ T = \text{Counting time} \]
   \[ \eta = \text{Efficiency of the detector} \]
   \[ V = \text{Volume of the sample taken in litres} \]
   \[ B = \text{Background counts} \]

III. RESULTS AND DISCUSSION
   The maximum water radioactivity concentrations for gross alpha and gross beta were 0.33 ± 0.02 and 0.56 ± 0.06 Bq/L obtained in Nongbahjynrin and Kylleng sampling sites respectively. World Health Organisation (WHO, 2004) advises 0.1Bq/L for gross alpha and 1.0Bq/L for gross beta activity as limit values for drinking water. Gross beta activities of all water samples from all the locations are seriously under the reference value of 1.0Bq/L. In the case of gross alpha an activity, the values varies with location and are lower or slightly higher than the recommended WHO values of 0.1Bq/l (Yarar et al., 2005). The higher values of alpha activity observed are however lower than the maximum contamination limit (0.5Bq/l for gross alpha) as prescribe by WHO. Even though, most of the beta activities were below the minimum detection limit, but for some months where beta activity is observed, the gross beta activity is higher than gross alpha activity. This phenomenon is in accordance with that observed by Karahan et al., in various surface waters in Istanbul, Turkey. The opposite however, was observed by Ozmen et al., in limestone and igneous based areas. From Table: I, we can see that all the sampling stations exhibit gross alpha activity but the concentration differs from one station to the other. The gross alpha activity in Phlangdiloin ranges from 0.03 – 0.21Bq/L with an arithmetic mean of 0.08 ± 0.02 Bq/L. Wahkaji and Undohlun alpha activity ranges from 0.03 – 0.15Bq/L and 0.03 – 0.14Bq/L respectively with a mean of 0.09 ± 0.03Bq/L each. Domiasiat and Nongtynger share a
mean of 0.07 ± 0.02Bq/L but with different ranges. Domiasiat alpha activity ranges form 0.03 – 0.14Bq/L while Nongtynger varies from 0.03 – 0.15Bq/L. The alpha activity in Kylleng ranges from 0.03 – 0.21Bq/L with a mean of 0.09 ± 0.03Bq/L. Syngkai, Nongbahjynrin and Mawthabah alpha activities ranges from 0.07 – 0.18Bq/L, 0.05 – 0.33Bq/L and 0.04 – 0.28Bq/L with a mean value of 0.11 ± 0.03, 0.13 ± 0.03 and 0.12 ± 0.03 Bq/L respectively. Only in one or two months the beta activity exceeds the minimum detection limit of 0.23Bq/L in all the selected locations.

It can be seen that the alpha activity in all the sampling locations does not exceed 0.35Bq/L which is well below the prescribe maximum contamination limit (MCL) of 0.5Bq/L. The high α-activity observed in water samples from Kylleng, Syngkai, Mawthabah, Nongtynger and Nongbahjynrin can be attributed to the presence of the uranium ore deposit and the leaching process that occurs in the area. The geological formation of these selected locations is mainly based on sandstone and igneous. These formations have an ability to retain the radioactivity element (Cowart & Burnett, 1994). The radioactivity concentration in around the uranium ore deposit site is higher than the other sites (i.e., Wahkaji, Umdohlun etc.).

According to the TSE standard, the activity concentrations in drinking water should be 0.035Bq/L for gross alpha and 0.37Bq/L for gross beta activities. (TSE, 1997) (Ozmen et al., 2004). As seen from Fig:2, the gross alpha activity concentration is higher than the TSE guidelines limit in all the sampling sites but lower than the WHO maximum contamination prescribe limit (0.5Bq/L) (WHO, 2004). The gross beta radioactivity concentration in all locations is however lower than the TSE guidelines limit except for one or two months. The beta radioactivity of most of the water samples can be readily attributed to K-40 and Ra-228 concentration where as the alpha emissions are due to a mixture of radioisotopes with different chemical behaviors and all belonging to the natural radioactive series (Ozmen et al., 2004). Gross alpha activity in natural water is mainly due to uranium and radium isotopes because thorium solubility is low (Osmond & Ivoich, 1992). Principally, Ra-226 and occasionally Th-232, Po-210 or Ra-224 are the main contributors to the total alpha particle activity in water samples (Damla et al., 2006).

### A. Determination of the Effective Dose Equivalent

The following equation is used to calculate the doses (USE-EPA, 1988)

\[ DR = A \times IR \times ID \]

TABLE: I MEAN, RANGE WITH STANDARD DEVIATION OF GROSS ALPHA ACTIVITY CONCENTRATION IN WATER FROM DIFFERENT SAMPLING LOCATIONS

<table>
<thead>
<tr>
<th>Location</th>
<th>Phlangdiloin</th>
<th>Wahkaji</th>
<th>Umdohlun</th>
<th>Domiasiat</th>
<th>Kylleng</th>
<th>Syngkai</th>
<th>Nongbahjynrin</th>
<th>Mawthabah</th>
<th>Nongtynger</th>
</tr>
</thead>
<tbody>
<tr>
<td>n</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
<td>19</td>
</tr>
<tr>
<td>Range</td>
<td>0.03–0.21</td>
<td>0.03–0.15</td>
<td>0.03–0.14</td>
<td>0.03–0.12</td>
<td>0.03–0.21</td>
<td>0.07–0.18</td>
<td>0.05–0.33</td>
<td>0.04–0.28</td>
<td>0.03–0.15</td>
</tr>
<tr>
<td>Mean</td>
<td>0.08</td>
<td>0.09</td>
<td>0.09</td>
<td>0.07</td>
<td>0.09</td>
<td>0.11</td>
<td>0.13</td>
<td>0.12</td>
<td>0.07</td>
</tr>
<tr>
<td>S.D</td>
<td>0.02</td>
<td>0.03</td>
<td>0.03</td>
<td>0.02</td>
<td>0.03</td>
<td>0.03</td>
<td>0.04</td>
<td>0.04</td>
<td>0.02</td>
</tr>
</tbody>
</table>

n: Number of sampling months; S.D: Standard Deviation

![Fig. 2 Comparison of Mean alpha activity from different samples with TSE and WHO standard limits](image)

Where,

- \( DR \) = Effective Equivalent Dose (mSv)
- \( A \) = Activity (Bq/L)
- \( IR \) = Intake of water for a person in one year (2L/day) = 730Litres.
- \( ID \) = Ingestion dose equivalent factor = 3.58 x 10^-7 mSv/Bq x 2

In the calculation, following the procedure of Fernandez et al., (1992), it is considered that more than 50% of the annual dose corresponds to radium (gross alpha radium). Therefore, in water with a concentration of, say, 0.08Bq/L, the effective equivalent dose is:

\[ DR = 0.08\text{Bq/L} \times 730\text{Litre/yr} \times 3.58 \times 10^{-7} \text{ Sv/Bq} \times 2 \]

\[ DR = 0.042\text{mSv/yr}. \]

Assuming that Ra-226 is the main contributor to the alpha activity, from Table: II, the calculated radium effective dose value was seen to be minimum in Domiasiat
with 0.037mSv/yr and maximum in Nongbahjynrin with 0.068mSv/yr.

TABLE: II EFFECTIVE DOSE RA-226 EQUIVALENT IN DIFFERENT LOCATION WITH MEAN ACTIVITY CONCENTRATION

<table>
<thead>
<tr>
<th>Sl.No</th>
<th>Location</th>
<th>Mean Activity (Bq/L)</th>
<th>Annual Effective Equivalent Dose (mSv/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Phlangdiloin</td>
<td>0.08</td>
<td>0.042</td>
</tr>
<tr>
<td>2</td>
<td>Wahkaji</td>
<td>0.09</td>
<td>0.047</td>
</tr>
<tr>
<td>3</td>
<td>Umdohlun</td>
<td>0.09</td>
<td>0.047</td>
</tr>
<tr>
<td>4</td>
<td>Domiasiat</td>
<td>0.07</td>
<td>0.037</td>
</tr>
<tr>
<td>5</td>
<td>Kylleng</td>
<td>0.09</td>
<td>0.047</td>
</tr>
<tr>
<td>6</td>
<td>Syngkai</td>
<td>0.11</td>
<td>0.057</td>
</tr>
<tr>
<td>7</td>
<td>Nongbahjynrin</td>
<td>0.13</td>
<td>0.068</td>
</tr>
<tr>
<td>8</td>
<td>Mawthabah</td>
<td>0.12</td>
<td>0.063</td>
</tr>
<tr>
<td>9</td>
<td>Nongtynger</td>
<td>0.07</td>
<td>0.037</td>
</tr>
</tbody>
</table>

In Table: III, we compare our findings with those values in different countries, it is clear that they are generally low in terms of ground water in Arizona (Holbert et al., 1995), Finland (Solanen, 1988) and Newark basin, New Jersey (Szabo and Zapecza, 1987), bottled water in Spain (Duenas et al., 1997) and drinking water in Venezuela (Sajo-Bohus, 1997) and Quebec, Canada (Zikovsky, 2006). However it is interesting to see that the values of the alpha activity concentration observed in surface and ground water of Rio Grande do Norte, Brazil (Malanca et al., 1988) is very much comparable with the activity observed in Domiasiat and its adjacent areas. We can see that the mean alpha activity of Rio Grande do Norte and Domiasiat area are comparable i.e., 0.092Bq/L and 0.099Bq/L respectively. Also the maximum observed value in Rio Grande do Norte, Brazil and Domiasiat area is 0.35Bq/L and 0.33Bq/L respectively. (Malanca et al., 1988)

From Fig: 3, it is worth noting that the mean value of gross alpha activity in all the countries is not that much different even though there is a marked difference in the maximum value. This however signifies that the high alpha activity is confined to certain places only, which may constitute only a small percentage of the total activity recorded which is true for the case of the Domiasiat area where the high activity is confined to the uranium mineralization zone. We can therefore say that the mean gross alpha activity in all these place are similar and lies in the range of 0.09Bq/L to 0.21Bq/L, which is a very small range indeed.

TABLE: III COMPARISON OF MEAN AND MAXIMUM GROSS ALPHA ACTIVITY CONCENTRATION IN DIFFERENT COUNTRIES

<table>
<thead>
<tr>
<th>Place</th>
<th>N</th>
<th>Mean</th>
<th>S.D</th>
<th>Maximum</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Domiasiat area, India</td>
<td>171</td>
<td>0.09</td>
<td>0.01</td>
<td>0.33</td>
<td>This Work</td>
</tr>
<tr>
<td>Arizona</td>
<td>667</td>
<td>0.17</td>
<td>-</td>
<td>5.66</td>
<td>Holbert et al., 1995</td>
</tr>
<tr>
<td>Finland</td>
<td>282</td>
<td>0.17</td>
<td>-</td>
<td>3.10</td>
<td>Solanen, 1988</td>
</tr>
<tr>
<td>New Jersey</td>
<td>259</td>
<td>-</td>
<td>-</td>
<td>4.59</td>
<td>Szabo and Zapecza, 1987</td>
</tr>
<tr>
<td>Spain</td>
<td>84</td>
<td>0.14</td>
<td>0.24</td>
<td>1.36</td>
<td>Duenas et al., 1997</td>
</tr>
<tr>
<td>Venezuela</td>
<td>25</td>
<td>0.12</td>
<td>0.07</td>
<td>0.54</td>
<td>Sajo-Bohus, 1997</td>
</tr>
<tr>
<td>Quebec, Canada</td>
<td>236</td>
<td>0.21</td>
<td>1.25</td>
<td>17.98</td>
<td>Zikovsky, 2006</td>
</tr>
</tbody>
</table>

n ➔ number of samples; S.D ➔ Standard Deviation

Fig: 3 Comparison of Mean and Maximum gross alpha activity concentration in different countries
IV. CONCLUSION

Our survey and study of gross radioactivity indicates that the water sample under investigation has a low concentration of both alpha and beta emitters and the activity was less than 0.5Bq/l for alpha and 1.0Bq/L for beta. We conclude that the radioactivity of water samples from all the selected locations is well below the limit set by the WHO. We estimate that the additional equivalent effective dose derived from water consumption is less than 0.3mSv/yr provided the water consumption for the population is on average, 730 litres per inhabitant per year. To the best of our knowledge, this is the first detailed study of gross alpha and gross beta activity concentration in water samples in Domiasiat and its adjacent areas. However, more work has to be done in order to identify the individual radionuclides contributing to the total gross radioactivity. This is a matter of further research work.

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