NATURAL RADIOACTIVITY IN GROUNDWATER AND SOILS IN JOHOR, MALAYSIA

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ABSTRACT
A study of natural radioactivity in samples of groundwater and soil were collected from nine districts in Johor and it was carried out on years 2008 until years 2010. The totals of seventy locations were selected as sampling point by referred to the reconnaissance maps and have been considered to the certain factor such as groundwater sources, soil types and the surrounding activities such as industrial or agricultur es. At sampling point, the portable gamma-ray survey meter, Model 19 Micro R meter manufactured by Ludlum is used to measure the gamma dose rate at the surrounding areas. In the laboratory, the sample preparation process is made according to the recommended steps and the preparation process of groundwater and soil are different from each other. The technology of HPGe spectrometer is used to counting all samples within 3 hours detection time and the collections of data on radionuclides is in the form of gamma spectrum. Further analysis is depends on the measurement of the natural radioactivity of the elements 238U, 232Th and 40K in order to get the estimation results of the concentration radionuclides contained in groundwater and soil samples. At last, the graphs were plotted for the relationship between the concentration of radionuclides in the samples and gamma dose. At last, graphs were plotted for the relationship between the concentration of radionuclides in the samples and gamma dose.

Keywords: groundwater, soil, concentration.

INTRODUCTION
The Radioactivity is a spontaneous emission of particles or electromagnetic radiation, or both from an unstable nucleus. There are three kinds of radiation which is alpha particle, beta particle and gamma rays with consisting of high energy of photon energy. The natural radioactivity element is commonly can be found in the nature such as air, soil and water. Their characteristic is typically long lived, with half-lives often on the order of hundreds of millions of years.

The presences of natural radioactivity in groundwater and soil need to be studied in order to ensure the level of radioactivity is not high or exceeds the limit as recommended by International Commission on Radiation Protection (ICRP). The average values of environmental background radiation over the world as recommended by the ICRP is 80 nGy/hr (ICRP, 1990).

MATERIAL AND METHODS
This study was conducted to determine the natural radioactivity in groundwater and soil in Johor State. To achieve the objective, there are several methods need to be considered such as location identification, sample preparation, radioactivity counting and dose measurement as well as data analysis.

Specifically, a total of 70 point of locations was selected from the areas of Johore state which is Johor Bahru, Mersing, Kulai, Muar, Segamat, Pontian, Kota Tinggi, Kluang and Batu Pahat. The sampling locations were described through the reconnaissance map which provides the details of soil types, geographical structure and infrastructure available in Johor region.

Besides that, the measurement of dose rate at the sampling point also need to be carried out using the portable gamma survey meter NaI(Tl) detector which is the most widely used device for all kinds of environmental gamma ray surveys due to its efficiency (IAEA, 1979).

All samples were then brought to the laboratory for further analysis which includes filtration, evaporation, drying and storing. In laboratory, the calibrated HPGe gamma-ray spectrometer was used to identify and quantify the existing of radionuclides 238U, 232Th and 40K content in samples of groundwater and soil.

The operational detection is 3 hours in order to get the accurate and precise result during the counting time. In additional, the HPGe gamma-ray spectrometer is direct counting techniques and it also widely used by the previous researchers such as Mohd Zaidi (1989), Sharifuddin (1991), Mireles (2003) in Mexico, Goddard (2002) in Oman, Jibbirri (2001) in Nigeria, Y. Narayana (2001) on India, Quindoset.al. (1994) in Spain and Malanca (1993) in Brazil.

The process of preparation sample is an essential stage in this study. All samples of groundwater and soils are prepared in the laboratory using the appropriate technique.

In the preparation of groundwater sample, it takes place between filtering the samples and stored the sample for 28 days in reasons to achieve the radionuclides secular equilibrium. The groundwater samples are filtered through approximately of 0.45 μm pore size using filters paper. Then following by acidify the water sample with 11 M Nitric acid (HN03) until it reach the PH ~ 2. The reason of this acidified technique is to avoid the growth of microorganism and to minimise water-wall interactions before the analysis for radionuclides (F A Khalil et.al, 2009). Next is evaporation process, where the original size of 2.5L groundwater was subsequently evaporated to size of 0.5Litre with temperature 60-70 °C. After evaporated, then the water were poured into a cleaned Marinelli
beaker. Lastly, sealed the beaker to ensure that 222Rn is not lost and stored to reach the radionuclides secular equilibrium.

The soil samples preparations, samples were needs to dry in an oven at temperature 110 °C and followed by pulverized or sieved the samples through mesh 250 microns before keep in the Marinelli beaker.

RESULT AND DISCUSSIONS

The sampling points were identified by referring to the reconnaissance maps which it has been provided by Ministry of Agriculture, Malaysia.

In Figure-1, it shows the scattered points of sampling and were collected from some areas in Johor.

Table-1. The details of measurement dose rate at the selected sampling point in the Johor State.

<table>
<thead>
<tr>
<th>District</th>
<th>Total point of sampling</th>
<th>Gamma dose rate (nGy/h) at the sampling point</th>
</tr>
</thead>
<tbody>
<tr>
<td>Johor Bahru</td>
<td>10</td>
<td>252 ± 34</td>
</tr>
<tr>
<td>Segamat</td>
<td>7</td>
<td>289 ± 83</td>
</tr>
<tr>
<td>Batu Pahat</td>
<td>6</td>
<td>324 ± 88</td>
</tr>
<tr>
<td>Kota Tinggi</td>
<td>7</td>
<td>268 ± 70</td>
</tr>
<tr>
<td>Kulai</td>
<td>8</td>
<td>295 ± 85</td>
</tr>
<tr>
<td>Kluang</td>
<td>9</td>
<td>309 ± 51</td>
</tr>
<tr>
<td>Pontian</td>
<td>7</td>
<td>257 ± 68</td>
</tr>
<tr>
<td>Muar</td>
<td>9</td>
<td>231 ± 55</td>
</tr>
<tr>
<td>Mersing</td>
<td>7</td>
<td>293 ± 81</td>
</tr>
</tbody>
</table>

As show in Table-1, the highest average dose rate is recorded at Batu Pahat with values 324 ± 88 nGy/h and the lowest is recorded at Muar with values 231 ± 55 nGy/h. The estimation average dose rate for the entire of Johore State is 277 ± 44 nGy/h. Most of the recorded values for average dose rate in Johor state areas were considered higher when compared to the World and Malaysia average.

In soils and groundwater samples, the analysis to determine the radioactivity of elements 238U, 232Th and 40K were carried out. The analysis found that the activity concentrations of 238U, 232Th and 40K in soil are 10.6 ± 0.28, 16.3 ± 0.44 and 93.4 ± 10.9 Bq/kg, respectively. The details of radioactivity 238U, 232Th and 40K originated in soil is shown in Table-2.

Table-2. The activity concentrations of element 238U, 232Th and 40K in soil samples.

<table>
<thead>
<tr>
<th>Samples of soil</th>
<th>Activity concentrations (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Range</td>
</tr>
<tr>
<td>238U</td>
<td>7.7 – 25.9</td>
</tr>
<tr>
<td>232Th</td>
<td>13.6 – 30.8</td>
</tr>
<tr>
<td>40K</td>
<td>30.4 - 3299</td>
</tr>
</tbody>
</table>

The radioactivity contents in soil is actually depends on soil types, geological structure and some aspects that should be considered such as soil capillary, soil porosity, soil moistures and human activities which have potential to give a side effects to the soil conditions (Fazali Hashim, 2007).

The Raeq values were also determined from the activity concentrations of 238U, 232Th and 40K contents in soil samples. The values of Raeq in range 29.5 to 76.7 Bq/kg and the average value is 41.1 ± 1.69 Bq/kg (0.17 ± 0.01 mSv/y), which it can be considered less when
compared to the World average (89 Bq/kg) and ICRP (370 Bq/kg or 1.5 mSv/y).

The relationship between gamma dose rate vs Raeq for the analyst of soil samples can be predicted using a satisfactorily graph shown in a Figure-2.

Figure-2. The relationship between gamma dose rate vs Raeq for the analyst of soil samples.

This study shows a positive linearity relationship between gamma dose rate vs Raeq with R2 value is 0.2. The R2 value for that graph is very low and it means the relationship between gamma dose rate and Raeq is weak. The natural radioactivity of 238U, 232Th and 40K also found in groundwater and it can be a soluble in water. Therefore, it have a potential to give risk and harm to the human health effects especially when drinking the untreated water. Thus, increase the concern about quality of groundwater is important and it needs to be strictly controlled. For this reason, studies of groundwater for human consumption must be performed in order to guarantee it have a low radioactivity (F A Khalil et al, 2009).

Table-3. The activity concentrations of element 238U, 232Th and 40K in groundwater.

<table>
<thead>
<tr>
<th>Samples of soil</th>
<th>Activity concentrations (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Range</td>
</tr>
<tr>
<td>238U</td>
<td>0.08 – 0.27</td>
</tr>
<tr>
<td>232Th</td>
<td>0.14 – 0.52</td>
</tr>
<tr>
<td>40K</td>
<td>1.17 – 3.99</td>
</tr>
<tr>
<td>Total</td>
<td>1.39 – 4.78</td>
</tr>
</tbody>
</table>

In this study, the average of radioactivity concentrations 238U, 232Th and 40K in groundwater are 0.17 ± 0.04, 0.23 ± 0.04 and 2.14 ± 0.09 Bq/l, respectively. The details of radioactivity 238U, 232Th and 40K originated in groundwater is shown in Table-3.

The total radioactivity (238U + 232Th +40K) in range from 1.48 to 4.49 Bq/l with an average 2.54 ± 0.17 Bq/l, it can be considered two times higher than the value recommended by IAEA. The value of 1 Bq/l is recommended for the total radioactivity of 238U, 232Th and 40K in water (IAEA, 1989). The abundance of 238U, 232Th and 40K in groundwater depends on what kind of mineral derived from aquifer rocks and the soil compounds (Gilkeson and Cowart, 1987; Andreo and Carrasco, 1999).

In groundwater analyst, the ratio of Th/U range from 0.62 to 3.58 with an average 1.42. Specifically, the ratios of Th/U obtained in all samples of groundwater are less than 1 and it means that the radioactivity of uranium is greater than thorium. It happens because of the characteristic 238U which is easily soluble in groundwater (Laina Salonen, 1994). If high solubility of 238U in groundwater, it can caused the abundance of CO2 and give some affects to the water purity (Hiisvirta, 1991).

In this study, the ratio of Th/U revealed that the radioactivity of thorium is dominantly greater than uranium in groundwater. It happened due to the radionuclides of 232Th is actively mobile and absorb from the soil surrounding to the groundwater. Besides that, the relationship between gamma dose rate and total radioactivity (238U + 232Th + 40K) in groundwater is shown in Figure-3. The graph shows a positive linearity between total radioactivity and gamma dose rate. The R2 value is very small which is 0.18.

CONCLUSIONS

The finding in this study shows the natural radioactivity of 238U, 232Th and 40K was contained in the samples of groundwater and soil which have been
collected from the selected point at certain location in Johor areas. The estimations of average activity concentration of 238U, 232Th and 40K in soil are 10.6 ± 0.28, 16.3 ± 0.44 and 93.4 ± 10.9 Bq/kg. While, the average activity concentration of 238U, 232Th and 40K in groundwater are 0.17 ± 0.04 Bq/l for 238U, 0.23 ± 0.04 Bq/l for 232Th and 2.14 ± 0.09 for 40K. Besides that, the values measured for Radium Equivalent Activity, Raeq in soils samples is range 29.5 to 76.7 Bq/kg with average value is 41.1 ± 1.69 Bq/kg (0.17 ± 0.01 mSv/y). The positive linear graph were plotted and the relationship between gamma dose rate vs Raeq in the analyst of soil sample and gamma dose rate vs total radioactivity in the analyst of groundwater is weak because of the R2 value is 0.1 and 0.2, respectively.

REFERENCES


