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RADIOACTIVITY IN COMMERCIAL PACKAGED DRINKING AND NATURAL MINERAL WATER IN MALAYSIA

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ABSTRACT

Radiochemistry and Environment Laboratory (RAS) had received bottled drinking water and natural mineral water samples throughout Malaysia from the Ministry of Health Malaysia (MOH). These samples need to undergo radioactivity analysis of gross alpha and gross beta which is part of the drinking water licensing requirements in Malaysia. A total of 83 samples were received in year 2020 and analysed using low background gross alpha gross beta counting system. The results of the analysis found that all samples complied with the mandatory standard requirements set by the MOH, which is not more than 0.1 Bq/L for gross alpha activity and 1.0 Bq/L for gross beta activity.

Keywords: Drinking water, gross alpha, gross beta, natural mineral water

INTRODUCTION

Water is one of the basic necessities in human daily life. Due to the risk of exposure to chemical contamination, microorganisms and radioactive materials, water resource management is important to ensure the safety of a country's water supply. Monitoring program of drinking water is one of the necessary actions in order to ensure the safety of the water supply received. Microorganisms, heavy metals and radioactive substances are the most commonly monitored parameters in drinking water. Among these parameters, radioactive substances found in natural mineral water and drinking water pose the least risk to human health as compared to chemical contamination and microorganisms. This lower risk is attributed to the fact that the radiation dose resulting from the consumption of radionuclides in drinking water is significantly lower than doses from other sources of radiation, as highlighted by the World Health Organization (WHO) in 2017.

The presence of radionuclides in drinking water supplies can occur naturally as well as from human activities that may release radionuclides into the environment such as nuclear facilities, nuclear weapons testing and mining activities. Natural alpha-emitting radionuclides that are the main contributors to gross alpha radioactivity come from the natural decay series of U-238 and Th-232 and their progenies U-234, Th-230, Ra-226, Po-210 and Th-228. The concentrations of natural gross beta activity are from the presence of radionuclides K-40, Ra-228 and Pb-210 (Pintilie et. al., 2016).

The Food Safety and Quality Division (BKKM) under the Ministry of Health Malaysia (MOH) is the governing authority who is responsible to ensure the safety of commercially available bottled drinking water (AMB) and natural mineral water (AMS) that are marketed for public consumption. The Twenty-Five Schedule (Sub-regulation 360B(3)) and the Twenty-Six Schedule (Regulation 360A(7)) of the Food Regulations 1985, Food Act 1983 (Act 281), specify that the standard mandatory limit for gross alpha radioactivity is 0.1 Bq/L and 1.0 Bq/L for gross beta. WHO on the other hand states that the maximum limit for gross alpha is 0.5 Bq/L and 1.0 Bq/L for gross beta to ensure that the dose received by an adult does not exceed 0.1mSv per year, as suggested by the International Commission on Radiological Protection (ICRP, 2008).

The primary objective of this paper is to present the findings of the radioactivity analysis conducted by Radiochemistry and Environment Laboratory (RAS) on bottled drinking water and natural mineral water samples received from across Malaysia. The study aims to assess compliance with the drinking water licensing requirements in Malaysia, specifically focusing on the mandatory standards set by the MOH, which stipulate that gross alpha activity should not exceed 0.1 Bq/L and gross beta activity should not exceed 1.0 Bq/L.

MATERIALS AND METHODS

Study Area and Sampling

Malaysia, located in Southeast Asia, is comprised of two distinct regions, 13 states and three federal territories. The two regions, Peninsular Malaysia, which shares a border with Thailand and East Malaysia, situated on the island of Borneo alongside Indonesia and Brunei are separated by the South China Sea.

The sampling of AMB and AMS throughout Malaysia was conducted by BKKM. The number of samples per state in Malaysia is shown in Table 1. The sampling was conducted for the purpose of license application by companies that wish to market AMB and AMS locally. Apart from that, this sampling was also done to monitor licensed AMB and AMS that are currently marketed. The samples were then sent to RAS Laboratory for analysis.

Table 1. Number of samples per state in Malaysia

State	Number of Samples
Johor	9
Kedah	2
Kelantan	4
Melaka	5
Negeri Sembilan	5
Pahang	5
Perak	5
Perlis	5
Pulau Pinang	2
Sabah	5
Sarawak	14
Selangor	18
Terengganu	1
Federal Territory Labuan	1
Federal Territory Putrajaya	2

Sample Pre-treatment and Preparation

A total of two liter (L) of water was used for analysis. Water was measured using a measuring cylinder and then poured into a 2 L beaker. The samples were evaporated without boiling at temperature less than 85°C (Ho et.al., 2020, Borrego-Alonso et.al., 2023). After the sample volume was reduced to approximately 5-10 ml, the sample was then transferred onto a 2 inch diameter stainless steel planchet with 1/8-inch depth using a dropper. The sample in the planchet was dried under an infrared lamp (Figure 1). The sample was continuously added onto the planchet until all samples were completely transferred and dried.

Figure 1. Sample drying under an infrared lamp

Sample Counting

Low Background Gross Alpha Gross Beta Counting System (Canberra Tennelec Series 5 XLB, Mirion Technologies, USA) was used to obtain gross alpha and gross beta activity as shown in Figure 2. The alpha and beta efficiency of the instrument was determined by using Am-241 and Sr-90 standard source, respectively. For samples with residual weight of more than 5mg/cm^2, the efficiency was determined using attenuation standards prepared at different weights of U_3O_8 for gross alpha and KCl for gross beta. The counting was conducted for 100 minutes of 3 cycles.

Figure 2: Low Background Gross Alpha Gross Beta Counting System

Calculation of Gross Alpha and Gross Beta Activity

The raw data in count per minute (cpm) obtained from the counting system was used to calculate gross alpha and gross beta activity in Bq/L. The calculation of gross alpha and gross beta activity is performed using Equation (1) (Norfaizal et. al., 2016) and uncertainty is estimated using Equation (2) (Zal U'yun et. al.(a), 2016). The minimum detectable activity (MDA) is calculated based on Equation (3) (Zal U'yun et. al. (b), 2016).

$$A_{GAB} = \frac{cpm_s - cpm_b}{E \times V \times 60} \quad (1)$$

Where:
A_{GAB} : specific activity of gross alpha or gross beta (Bq/L)
cpm_s : gross alpha or gross beta counts per minute in sample
cpm_b : gross alpha or gross beta counts per minute in background
E : counting efficiency
V : sample volume (L)

$U_c(A)/A = [\sqrt{}[(U(n)/n)2 + (U(t)/t)2 + (U(\mathcal{E})/\mathcal{E})2 + (U(V)/V)2 + \ldots.]$ (2)

Where:
$U_c(A)$: uncertainty of gross alpha or gross beta (Bq/L)
A : activity of gross alpha or gross beta (Bq/L)
U(n) : uncertainty of net count for gross alpha or gross beta (cpm)
n : net count for gross alpha or gross beta (cpm)
U(t) : uncertainty of counting time (second)
t : counting time (second)
U(Ɛ) : uncertainty of counting efficiency
Ɛ : counting efficiency
U(V) : uncertainty of sample volume (L)
V : sample volume (L)

$MDA = (1.645)(\sqrt{(C_{bg}/t_c)})$ (3)

Where:
1.645 : confidence level 95%
C_{bg} : background count (cps)
t_c : counting time (second)

RESULTS AND DISCUSSION

Table 2 shows the results of gross alpha and gross beta activity in the AMB sample. Gross alpha activity in the AMB sample was found to be between <0.0011 – 0.0551 Bq/L. Only 6 (9.7%) out of 62 samples received indicated an activity reading above MDA which is between 0.0061 - 0.0551 Bq/L. On the other hand, gross beta activity was in the range of <0.0036 – 0.2087 Bq/L. A total of 25 (40.3%) out of 62 samples had a result that was higher than the MDA which specified between 0.0105 to 0.2087 Bq/L. MDA for gross alpha ranged 0.0011 – 0.0135 Bq/L, and 0.0036 – 0.0060 Bq/L for gross beta. MDA for both gross alpha and gross beta revealed no significant variations between samples.

Table 2. Gross alpha and gross beta activity in bottled drinking water (AMB)

Sample No.	Gross Alpha Activity (Bq/L)	Uncertainty (Bq/L)	MDA (Bq/L)	Gross Beta Activity (Bq/L)	Uncertainty (Bq/L)	MDA (Bq/L)
AMB1	0.0266	0.0087	0.0049	0.1969	0.0116	0.0055
AMB2	<0.0015	-	0.0015	<0.0055	-	0.0055

AMB3	<0.0015	-	0.0015	<0.0055	-	0.0055
AMB4	<0.0015	-	0.0015	<0.0055	-	0.0055
AMB5	<0.0017	-	0.0017	<0.0060	-	0.0060
AMB6	<0.0017	-	0.0017	<0.0060	-	0.0060
AMB7	<0.0017	-	0.0017	<0.0060	-	0.0060
AMB8	<0.0014	-	0.0014	<0.0051	-	0.0051
AMB9	<0.0014	-	0.0014	<0.0051	-	0.0051
AMB10	<0.0019	-	0.0019	0.0135	0.0057	0.0051
AMB11	<0.0045	-	0.0045	0.0406	0.0074	0.0052
AMB12	<0.0016	-	0.0016	<0.0056	-	0.0056
AMB13	<0.0016	-	0.0016	0.0114	0.0057	0.0052
AMB14	<0.0016	-	0.0016	<0.0048	-	0.0048
AMB15	<0.0016	-	0.0016	<0.0048	-	0.0048
AMB16	<0.0014	-	0.0014	<0.0047	-	0.0047
AMB17	<0.0011	-	0.0011	<0.0045	-	0.0045
AMB18	<0.0015	-	0.0015	<0.0045	-	0.0045
AMB19	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB20	<0.0023	-	0.0023	0.0115	0.0051	0.0046
AMB21	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB22	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB23	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB24	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB25	<0.0014	-	0.0014	<0.0046	-	0.0046
AMB26	<0.0025	-	0.0025	0.0705	0.0082	0.0044
AMB27	<0.0014	-	0.0014	0.0146	0.0052	0.0044
AMB28	<0.0020	-	0.002	0.0275	0.0063	0.0048
AMB29	<0.0019	-	0.0019	<0.0044	-	0.0044
AMB30	<0.0015	-	0.0015	<0.0047	-	0.0047
AMB31	<0.0015	-	0.0015	<0.0047	-	0.0047
AMB32	<0.0015	-	0.0015	0.0257	0.0062	0.0048
AMB33	<0.0015	-	0.0015	<0.0048	-	0.0048
AMB34	<0.0015	-	0.0015	<0.0048	-	0.0048
AMB35	<0.0018	-	0.0018	0.0232	0.0058	0.0046
AMB36	<0.0015	-	0.0016	0.0255	0.0059	0.0046
AMB37	<0.0015	-	0.0015	0.0486	0.0073	0.0046
AMB38	0.0484	0.0071	0.0018	0.1431	0.0111	0.0045
AMB39	0.0551	0.0088	0.0025	0.2087	0.0132	0.0045
AMB40	<0.0015	-	0.0015	<0.0045	-	0.0045
AMB41	<0.0015	-	0.0015	<0.0048	-	0.0048
AMB42	<0.0013	-	0.0013	<0.0051	-	0.0051
AMB43	<0.0135	-	0.0135	0.0105	0.0053	0.0048
AMB44	<0.0025	-	0.0025	0.0145	0.0044	0.0036
AMB45	<0.0013	-	0.0013	<0.0051	-	0.0051
AMB46	<0.0019	-	0.0019	0.0131	0.0053	0.0046
AMB47	<0.0015	-	0.0015	0.0479	0.0079	0.0053
AMB48	<0.0015	-	0.0015	<0.0053	-	0.0053

AMB49	0.0099	0.0049	0.0035	0.1384	0.0111	0.0048
AMB50	0.0063	0.0038	0.0033	0.1205	0.0104	0.0048
AMB51	<0.0015	-	0.0015	<0.0046	-	0.0046
AMB52	<0.0012	-	0.0012	<0.005	-	0.005
AMB53	<0.0012	-	0.0012	<0.005	-	0.005
AMB54	<0.0013	-	0.0013	<0.0036	-	0.0036
AMB55	<0.0015	-	0.0015	<0.005	-	0.005
AMB56	<0.0015	-	0.0015	0.0199	0.0063	0.005
AMB57	<0.0018	-	0.0018	<0.0052	-	0.0052
AMB58	0.0061	0.0027	0.0019	0.0141	0.0056	0.0042
AMB59	<0.0017	-	0.0017	0.0302	0.0063	0.0044
AMB60	<0.0017	-	0.0017	<0.0044	-	0.0044
AMB61	<0.0019	-	0.0019	0.0405	0.0064	0.0042
AMB62	<0.0017	-	0.0017	0.0765	0.0085	0.0044
Range	<0.0011 – 0.0551		0.0011 - 0.0135	<0.0036 – 0.2087		0.0036 – 0.0060
Mean	0.0254		0.0020	0.0555		0.0048

The results of the analysis of gross alpha and gross beta in AMS are shown in Table 3. The results revealed that the activity of gross alpha and gross beta ranged between <0.0013 - 0.0469 Bq/L and <0.0048 - 0.2245, respectively. A total of 21 samples were received, with 6 (28.6%) values above the MDA for gross alpha and 15 (71.4%) exceeding the MDA for gross beta.

The percentage of AMS samples that exceed the MDA value for AMS was much higher as compared to AMB. This demonstrated that the activity of gross alpha and gross beta in AMS, is higher in general compared to AMB. The typical processing of AMB involves filtration and purification procedures, which have the potential to eliminate radioactive-contained minerals and heavy metals, resulting in a reduced presence of radioactivity (Khandaker, et.al., 2017).

However, this finding needs further investigation due to the smaller number of AMS samples as compared to AMB, which also causes the percentage of AMS samples with activity more than MDA to be higher.

Table 3. Gross alpha and gross beta activity in bottled natural mineral water (AMS)

Sample No.	**Gross Alpha Activity (Bq/L)**	**Uncertainty (Bq/L)**	**MDA (Bq/L)**	**Gross Beta Activity (Bq/L)**	**Uncertainty (Bq/L)**	**MDA (Bq/L)**
AMS1	<0.0013	-	0.0013	<0.0053	-	0.0053
AMS2	<0.0036	-	0.0036	0.0107	0.0053	0.0049
AMS3	<0.0021	-	0.0021	<0.0053	-	0.0052
AMS4	<0.0021	-	0.0021	<0.0052	-	0.0052
AMS5	<0.0021	-	0.0021	<0.0052	-	0.0052
AMS6	<0.0123	-	0.0094	0.0123	0.0065	0.006
AMS7	<0.0021	-	0.0021	0.0732	0.007	0.0049
AMS8	<0.0021	-	0.0021	<0.0048	-	0.0048
AMS9	0.0063	0.0036	0.0028	0.0667	0.0082	0.0045
AMS10	0.0039	0.0029	0.0024	0.0474	0.0074	0.0047

AMS11	<0.0014	-	0.0014	0.0129	0.0061	0.0045
AMS12	<0.0013	-	0.0013	<0.0051	-	0.0051
AMS13	0.0102	0.0048	0.0033	0.2245	0.0105	0.0036
AMS14	<0.0038	-	0.0038	0.0243	0.0061	0.0048
AMS15	<0.0034	-	0.0034	0.0144	0.0053	0.0045
AMS16	<0.0042	-	0.0042	0.0139	0.0053	0.0046
AMS17	0.0094	0.005	0.0038	0.0424	0.0105	0.0044
AMS18	0.0469	0.012	0.0058	0.0771	0.0086	0.0046
AMS19	<0.0038	-	0.0038	0.0681	0.0080	0.0043
AMS20	0.0141	0.0053	0.0032	0.0439	0.0073	0.0043
AMS21	<0.0047	-	0.0047	0.0224	0.0058	0.0044
Range	<0.0013 – 0.0469		0.0013 – 0.0094	<0.0048 – 0.2245		0.0036 - 0.0060
Mean	0.0151		0.0020	0.05028		0.0048

The gross beta activity in all samples was found higher than the gross alpha activity. This finding is in agreement with Yussuf, et. al, (2012) who determined the activity of U-238, Th-232 and K-40 in natural mineral and drinking water samples. The activity concentration of U-238 and Th-232 which contributed to gross alpha activity in a sample, was found to be substancially lower than activity of K-40 which correlated to gross beta activity in a sample. Hence, the activity of gross beta was found to be higher compared to gross alpha. Apart from that, secular equilibrium that has been established between U-238 and its progenies, Th-234 and Pa-234m, making both radionuclides prominent β emitters alongside K-40 (Borrego-Alonso et.al., 2023).

The range of MDA for gross alpha and gross beta in AMB is 0.0011 – 0.0135 Bq/L and 0.0036 – 0.0060 Bq/L, respectively. The MDA range for gross alpha in AMS was 0.0013 – 0.0094, which was greater than the MDA range for gross alpha in AMB. On the other hand, the range of MDA for gross beta was found to be consistent in both AMB and AMS samples. This is in line with the findings by Ho et. al., (2020) that MDA for gross alpha was less consistent compared to gross beta. The inconsistent MDA value of gross alpha in the AMS sample was caused by the presence of more residues in the sample compared to AMB resulting in different MDA values. Higher residues level in a sample will contribute to lower counting efficiency, thus elevating the MDA value.

Although there are samples that have radioactivity more than MDA, but the activity concentration is still lower than the standard mandatory limit set by MOH. Thus, it is safe to say that the radiation dose received from consuming AMS and AMB marketed in Malaysia does not exceed the limit suggested by ICRP.

CONCLUSION

The results of gross alpha and gross beta analysis on all AMB and AMS samples received from MOH in the year 2020 shown that the values obtained are below the mandatory standard limit, which is 0.1 Bq/L for gross alpha and 1.0 Bq/L for gross beta. This shows that the radiation exposure received from the consumption of marketed AMB and AMS in Malaysia is at a safe level.

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