ASSESSMENT OF NATURAL RADIOACTIVITY IN WATER AND SEDIMENT FROM AMANG (TIN TAILING) PROCESSING PONDS

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Abstract

Gamma spectroscopy was performed to determine the concentrations of uranium-238 and thorium-232 concentrations in the environment as a consequence of amang processing. In this study 33 water samples and 26 sediment samples were collected from 7 amang processing areas. The concentrations of uranium-238 and thorium-232 were determined by direct counting using a hyper pure germanium (HPGe) detector inter phased with a multi channel analyzer (MCA) . Results showed that the maximum mass and activity concentrations of uranium in water samples were 6.64 ppm and 78.53 Bql^{-1} respectively, while in sediment samples were 69.75 $mgkg^{-1}$ and 860.57 $Bqkg^{-1}$ respectively. The maximum mass and activity concentrations of thorium in water samples were 1.71 ppm and 6.90 Bql^{-1}, while in sediment samples were 157.73 $mgkg^{-1}$ and 637.61 $Bqkg^{-1}$ respectively. Concentrations of uranium-238 and thorium-232 in sediment samples were higher than concentrations of uranium-238 and thorium-232 in water samples, and this may be attributed to insolubility of these radionuclides in water. The concentrations of both radionuclides were higher in sediments collected from ponds involved in the close water recycle system compared to those ponds involved in the open water system. Results also showed that the concentrations of these radionuclides were higher than background indicating that amang processing activity has enhanced the natural radionuclides contents in water and sediment.

Introduction:

Tin mining has been a major activity of Malaysia since 1848. Up till 1980, Malaysia contributed 30.7 % of the world's produce of tin. However her contribution to the world's tin dropped sharply since 1983. By 1996 Malaysia's contributed only 3.9 % and by then there were only 63 mines in operation (Malaysian Department of Mines, 1997). With the drop in tin production and the cost of world's tin, attention shifted toward processing amang (a tin by product) for valuable minerals [1]. Amang is a local (Malaysian) slang word used by the tin mining community to describe tin tailing consisting of a mixture of tin ore, sand and minerals initially discarded by tin miners [5, 16]. Amang or by- product of tin minerals reprocessing, has been found to contain valuable minerals such as ilmenite, zircon, monazite, xenotime, columbite and struvirite that has high demand in production industry [2]. Studies done by the Atomic Energy Licensing Board have shown that the uranium and thorium concentrations vary in monazite, xenotime and ilmenite respectively [3]. Valuable minerals such as monazite ($[Ce,La,Y,Th]PO_4$) are radioactive because they contain naturally occurring thorium. Zircon becomes radioactive when cations, such as Zr^{+4}, are replaced with uranium or thorium [2, 14]. Other minerals may be contaminated with minerals that are radioactive. Amang consists of natural occurring radioactive materials (NORM) such as ^{238}U and ^{232}Th that are technologically enhanced natural occurring radioactive materials (TENORM) during the mining and amang processing activities. Amang which consists of heavy metals is the reason why the mining of tin is blamed for upsetting the ecosystem. Beside the obvious scaring of large and beautiful landscape and turning it into barren lands, tin mining together with amang processing have also been blamed for changing concentration distribution of elements in the ecosystem, namely the distribution of heavy metals as well as NORM in soil and water [15].

In amang processing, separation and concentration of valuable minerals are based on three physical properties, i.e. different specific gravities, magnetic and electrostatic properties. In this process, large volume of water is used in wet gravity separation process and has become a potential source of environmental pollution depending how the water is managed**.** The water may be released directly into the environment (open water management system) or recycle (close water management system). Such activities have been associated with giving rise to radiological environmental problems [16]. The risk of such problem is high due to the fact that legally, amang

plants in Malaysia are categorized as small amang factory and is exempted from licensing by the Atomic Energy Licensing Board (small amang factory) Order 1994 [4].

In Malaysia, there are 113,700 hectares (281,000 acres) of former mining land and 14.4 percent of it is in the form of water pond, used extensively for aqua culture. About four percent has been turned into food production areas, when tin mining collapsed in the 1980s [6].

Using energy dispersive X-ray fluorescence (EDXRF) A.F.Oluwole [10] measured the concentrations of radionuclides and toxic heavy metals in the soil around a lead/tin smelter and also air particulate and mining wastes collected from some tin mines and a tin mill. The concentrations of thorium and uranium reported ranged between 0.01 - 2.94 % and 0.002 - 0.11% in the tailing and between 2.25 - 9.09% and 0.25 - 5.65% in the monazites respectively. Studies by Hu [12] and Kandaiya have also shown the presence of naturally occurring radionuclides in the valuable minerals of amang. Ismail B. [15, 16] reported that amang processing reduces the pH of water and radionuclides contaminates the water and consequently decrease quality of water.

Materials and Methods

Sampling location:

Seven different amang processing plants employing three kinds of water management systems (i.e. open water system, close water natural and close water man made systems) were chosen for this study. Thirty three water samples and 26 sediment samples were taken from seven amang plants. All water and sediment samples were taken from Selangor and Perak State in Malaysia. Hand made water sampler was used for taking water sample from surface level (top), mid and bottom levels of the lakes and ponds. If the depth of pond was less than three meters, only one water sample from top was taken, if the depth was more than three meter and less than four meter, two water samples from top and bottom were taken. If the depth of the lake was more than four meters, three water samples (top, middle and bottom levels) were taken. Water samples were collected and stored in extra clean polyethylene bottle. Water samples collected were labeled as S_XL_Y, where S indicates sampling station and L indicates depth at which the water samples were collected. X represents station number from 1-18, and y represents depth of the water samples from 1-3. For example water sample S_1L_1 means station number 1 and top level of water.

Twenty six sediment samples were collected from two different amang processing plants employing close water natural system. Sediment samples were collected in special PVC container. Sediment sampler model Ejkelkamp with PVC transparent tubes (60,100,150 cm length and 63 mm diameter) was used for taking the sediment samples. Sediment samples collected were labeled as S_XL_Y . Where S indicates sampling station and L indicates depth at which the sediment samples were collected.

Treatment of samples:

The determination of uranium-238 and thorium-232 concentrations in water samples were based on 2000 ml of water samples collected and subsequently evaporated to 200 ml and stored, capped and sealed in Merinelli containers. In sediment samples, large stones and other objects were removed, then were dried in oven at 105ºC for 24 hours to constant mass, then sieved through mesh 500 μm. All sediment samples were weighed and sealed in Merinelli containers. All water and sediment samples were kept for at least four weeks before counting in order to allow the in-growth of uranium and thorium decay products and achievement of secular equilibrium for ^{238}U and ^{232}Th with their respective progenies.

Gamma spectroscopy:

A stand-alone high-resolution gamma spectrometric system was used for the measurement of the energy spectrum of the emitted gamma rays in the energy range between 50 keV and 3000 keV [12]. The gamma spectroscopy system consists of the high purity germanium (HPGe) detector from Oxford Company with an efficiency of 15%. Detector model number is CNVDS30 with crystal characteristics of diameter 45.3 MM, length 47.3 mm, active volume 75 and germanium dead layer thickness 0.3 microns and detector to window distance less than or equal to 5 mm. The end cap outside diameter is 76-mm aluminum 1 mm thick. The spectra were fed through the Amplifier Canbera Model 2020 to the multi channel analyzer with two analog to digital converters and the memory containing 8192 channels. The multi channel analyzer was directly connected to a personal computer where the spectra were processed and stored. In this system bias supply is from Ortec Company. The detector was mounted on a cryostat which was dipped in to a 30 liters dewar filled with liquid

nitrogen. The detector was surrounded by a cylindrical shield consisting of lead with thickness of 5 cm, which provides an efficient suppression of background gamma radiation present at laboratory site. Soil-IAEA-375 was used as standard reference for sediment samples and uranium and thorium mix stock standard solutions were used as standard reference for water samples.

Analysis and Instrumentation:
Gamma spectroscopy was used to determine the concentrations of uranium-238 and thorium-232 in water and sediment samples. Water sample was put into the shielded HPGe detector and the activity concentration present was counted for 86400 seconds (24 hours), while sediment samples were counted for 43200 seconds (12 hours). Prior to the sample measurement, the environmental gamma background at the laboratory site was determined using a blank Merinelli under identical measurement conditions. The laboratory background reading was averaged from four readings taken.

Based on the measured gamma ray photo peaks, emitted by specific radio nuclides in the thorium-232 and uranium-238 decay series, their radiological concentrations in samples collected can be determined. Calculations relied on establishment of secular equilibrium in the samples, due to the much smaller lifetime of daughter radionuclides in the decay series of thorium-232 and uranium-238. More specifically, the thorium-232 concentration was determined from the concentrations of Tl -208 in the samples, and the concentration of U-238 was determined from concentrations of the Bi-214 decay products.

Energy 1120.3 keV belonging to radionuclide Bi-214 was used for measuring mass concentration and activity of uranium-238 in water samples. Energy 2614.4 keV belonging to radionuclide Tl-208 was used for measuring concentration and activity thorium-232 in water samples. Energy 609.3 keV belong to radionuclide Bi-214 was used for measuring mass concentration and activity of uranium-238 in sediment samples. Energy 2614.4 keV belonging to radionuclide Tl-208 was used for measuring mass concentration and activity concentrations of Th-232 in sediment samples.The mass and activity concentrations of radionuclides were obtained using related formula [3].

Results and Discussion:

Before detail discussion was made in this study, the overall finding of this study was prepared first. Figures 1 and 2 show the mass and activity concentrations of uranium-238 and thorium-232 in water samples. S1- S16 are water sampling stations in different amang ponds, S17 and S18 are water sampling stations along a river (S17 being upstream and S18 down stream). L1, L2 and L3 are different depth where water samples were taken. L1 being near the surface and L3 being near the bottom. Figures 3 and 4 show the mass and activity concentrations of uranium and thorium respectively in sediment samples in amang plant number 1. Figures 5 and 6 show the mass and activity concentrations of uranium and thorium in sediment samples in amang plant number 2. Amang plants number 1 and 2 represent two different amang plants. S1-S4 are sediment sampling stations around the ponds, and L1-L4 are the sediment layer (L1 means top sediment layer and L4 means bottom sediment layer).

Figures 1 and 2 show the mass and activity concentrations of uranium-238 and thorium-232 in water samples respectively. Maximum mass concentration of uranium in water samples was 6.64 ppm and maximum activity concentration was 78.53 Bql^{-1} belonging to sample taken at station 8 (ie. S8L1). Maximum mass and activity concentrations of thorium-232 in water samples were 1.71 ppm and 6.90 Bql^{-1} respectively. These readings were recorded in station 15 (ie. S15-L1).

Figures 3 and 4 show the mean mass and activity concentrations of uranium-238 and thorium-232 in sediment samples in amang plant number 1 respectively. Maximum mass concentration of uranium-238 in sediment samples was 69.75 mg/kg and maximum activity concentration was 860.57 $Bqkg^{-1}$. These readings were recorded at station 3 (ie. S3L1). Maximum mass concentration of thorium-232 in sediment samples in amang plant 1 was 157.73 $mgkg^{-1}$ and maximum activity concentration was 637.61 $Bqkg^{-1}$, recorded at station 2 (S2L1).

Figures 5 and 6 show the mass and activity concentrations of uranium-238 and thorium-232 in sediment samples sampled at amang plant number 2. Maximum mass concentration of uranium-238 in sediment samples was 27.59 $mgkg^{-1}$ and maximum activity concentration was 340.40 $Bqkg^{-1}$, these values were observed in station 1 (S1L1). The maximum of mean mass concentration of thorium in sediment samples in amang plant 2 was 150.8 $mgkg^{-1}$ and maximum activity concentration was 609.60 $Bqk^{-1}g$. These readings were recorded at station 1 (S1L1).

Table 1 shows the summary and statistical calculations of data collected from all water and sediment samples. Results from Table 1 shows that the mean concentration of uranium-238 in water samples was 4.34 ± 1.58 ppm and with a range 0.12 - 6.64 ppm. The results also shows that the mean concentration of thorium-232 in water samples was 0.37 ± 0.37 ppm with a range between 0.01 – 1.71 ppm. Mean maximum concentrations of uranium-238 and also thorium-232 were observed in stations 8 (S8-L1) and station 15 (S15-L1) respectively. Both stations were close to water discharge point of the plant and involved with the amang plant using the close water management system

Results from Table 1 shows that the mean concentration of uranium-238 in the sediment samples in amang plant 1 was 18.00 ± 17.55 $mgkg^{-1}$ and the range was between 6.82 - 69.75 $mgkg^{-1}$. The mean concentration of thorium-232 in sediment samples taken from amang plant 1 was 62.05 ± 39.34 $mgkg^{-1}$ and the range was between 26.00 – 157.73 $mgkg^{-1}$. Maximum mean concentrations of uranium-238 and also thorium-232 were observed in stations 3 (S3-L1) and 2 (S2-L1) respectively. These two stations were close to water discharge point, and where the water management in these amang plants is close water system type.

Table 1 also shows the statistical calculations of uranium-238 and thorium-232 in the sediment samples taken from amang plant 2. The mean concentration of uranium in sediment samples in amang plant 2 was 9.62 ± 6.47 $mgkg^{-1}$ and was in the range 4.96-27.95 $mg\text{-}1^{kg}$. The mean concentration of thorium-232 in sediment samples in amang plant 2 was 40.49 ± 39.41 $mgkg^{-1}$ and was in the range 11.92 - 150.80 $mgkg^{-1}$. Maximum mean concentrations of uranium-238 and also thorium-232 were observed in station 1 (S1-L1), i.e. discharge point in this amang plant, where the management system in this amang plant is close water system type.

Table 2 shows statistical calculations and activity concentrations of uranium-238 and thorium-232 in water collected from seven amang plants highlighting the different water samples collected at point of discharge and those collected elsewhere. This table shows their mean, ± standard error mean, median, range and standard deviation. Based on these results the highest uranium-238 and thorium-232 in all amang plants were recorded near or at the point of water discharge (except amang plant 5). The median concentrations of uranium-238 in discharge points were 56.53, 64.93, 71.55, 20.40, 64.00, 54.61and 7.62 Bql^{-1} respectively and for thorium-232 were 1.19, 1.39, 2.55, 1.33, 1.46, 6.9 and 0.12 Bql^{-1} respectively. It should be mentioned that near station S3 in amang plant 5, there were several mounds and valuable minerals next to the point where rainfall could have washed down these minerals and carry them into the pond.

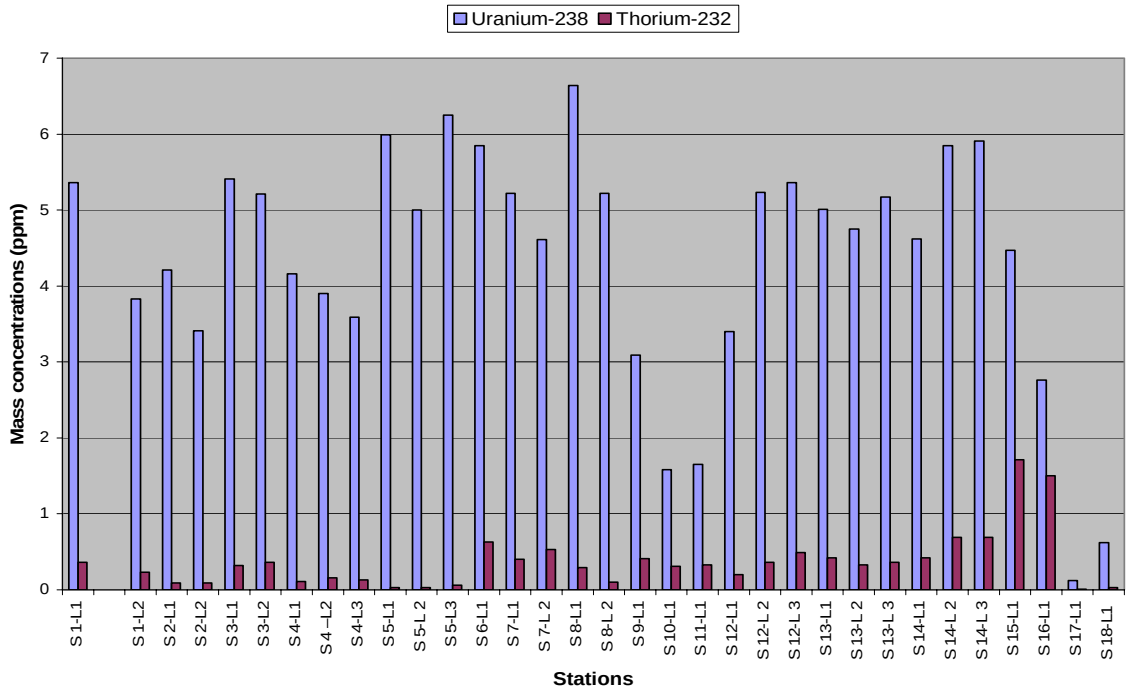

Figure 1. Mass concentrations of U-238 and Th-232 in water samples

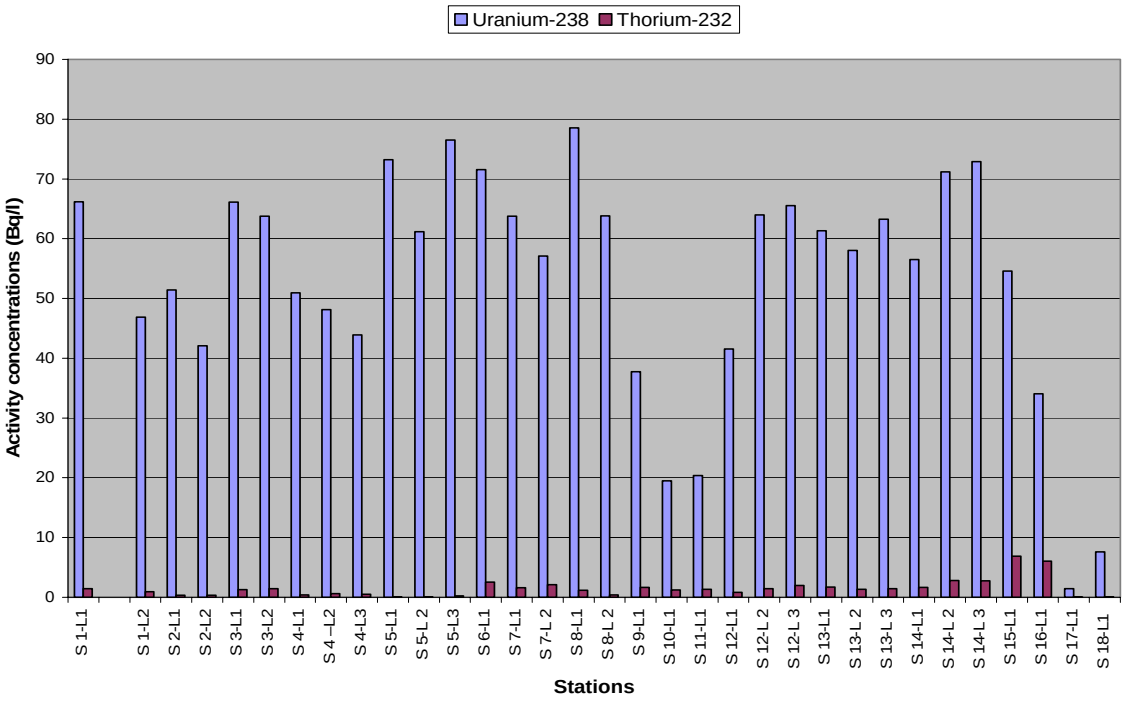

Figure 2**.** Activity concentrations of U-238 and Th-232 in water samples

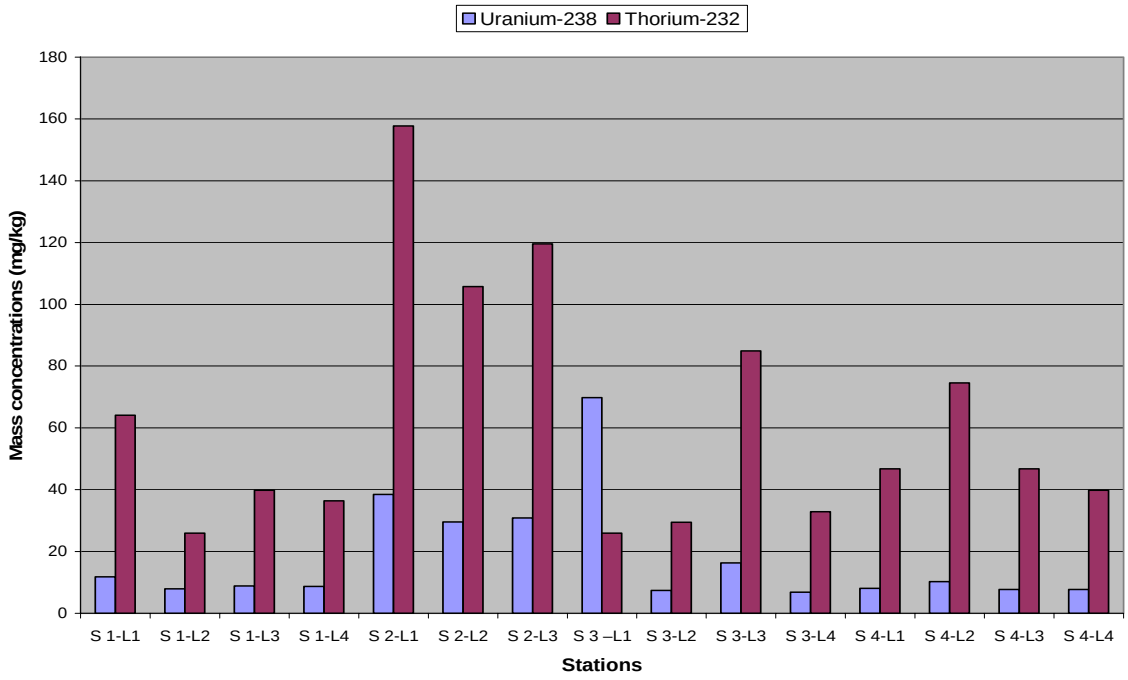

Figure 3. Mass concentrations of U-238 and Th-232 in sediment sample (amang plant 1)

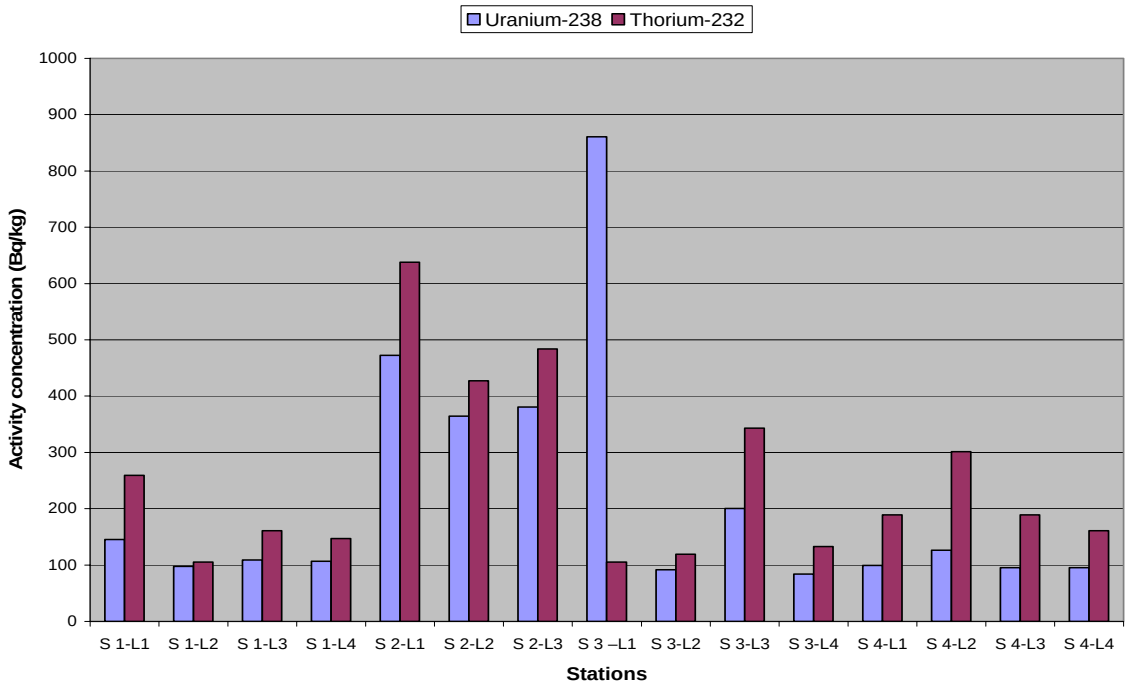

Figure 4. Activity concentrations of U-238 and Th-232 in sediment samples (amang plant 1)

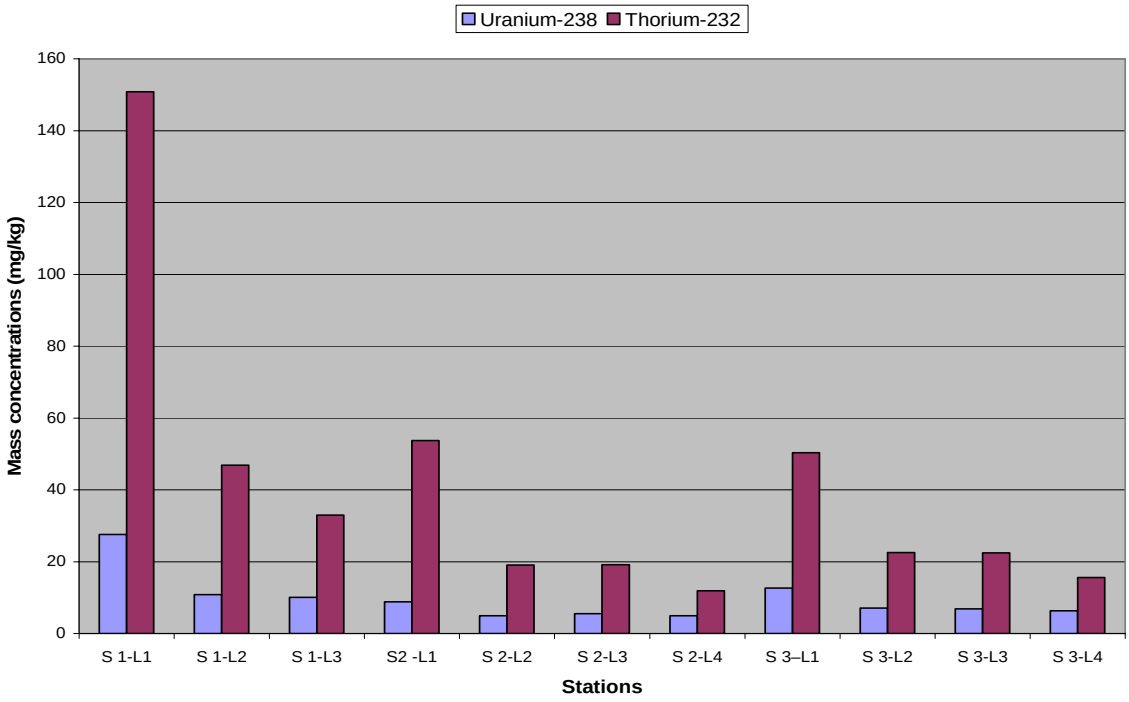

Figure 5. Mass concentrations of U-238 and Th-232 in sediment samples (amang plant 2)

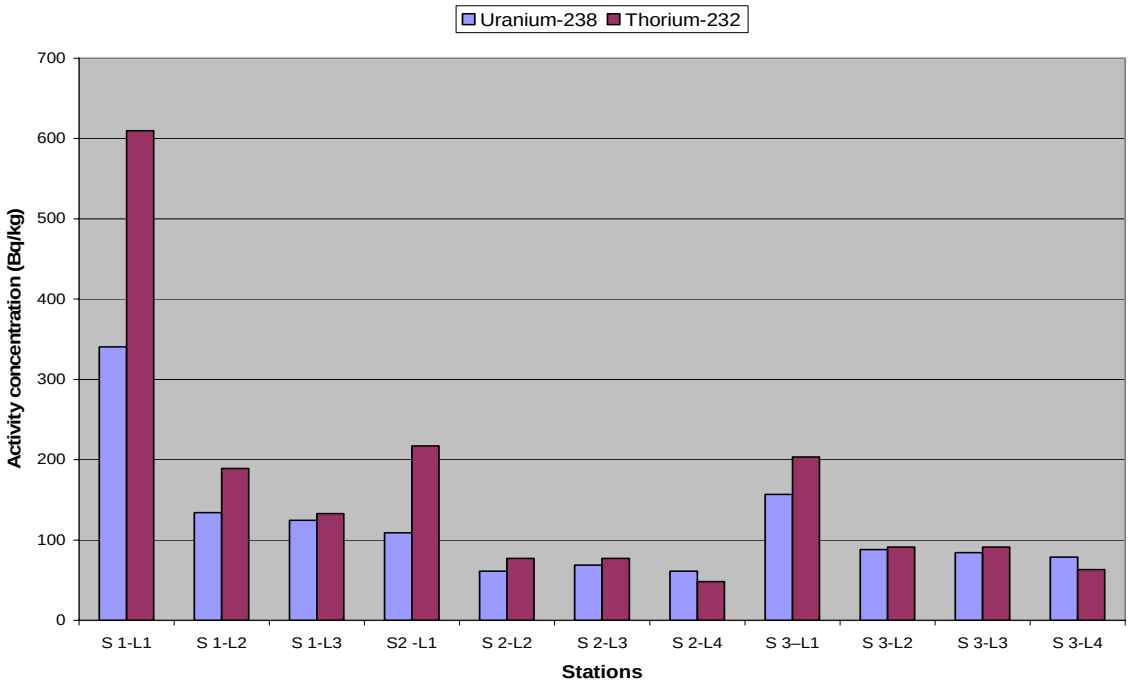

Figure 6. Activity concentrations of U-238 and Th-232 in sediment samples (amang plant 2)

Table 1. Statistical calculations of water and sediment samples

Sample	Sample size	Mean	Std error of mean	Lower 95% Conf limit	Upper 95% conf limit	Minimum	Media (50 percentile)	Maximum
U in Water sample	33	4.34 ppm	0.275	3.785	4.909	0.12 ppm	4.750 ppm	6.64 ppm
Th in water sample	33	0.37 ppm	0.064	0.231	0.501	0.01 ppm	0.331 ppm	1.71 ppm
U in Sediment sample (amang plant 1)	15	18.00 mg/kg	4.533	8.285	27.730	6.82 mg/kg	8.84 mg/kg	69.75 mg/kg
Th in sediment sample (amang plant 1)	15	62.05 mg/kg	10.159	40.261	83.844	26.00 mg/kg	46.80 mg/kg	157.73 mg/kg
U in sediment Sample (amang plant 2)	11	9.62 mg/kg	1.952	5.275	13.975	4.96 mg/kg	7.13 mg/kg	27.59 mg/kg
Th in sediment sample (amang plant 2)	11	40.49 mg/kg	11.882	12.024	66.970	11.920 mg/kg	22.57 mg/kg	150.80 mg/kg

Table 2. Activity concentrations of ^{238}U and ^{232}Th in water collected from 7 amang plant.

Amang Plants/Sample	Uranium-238				Thorium-232			
	Mean ± sem (Bq/L)	**Median (Bq/L)**	**Range (Bq/L)**	**St. dev.**	**Mean ± sem (Bq/L)**	**Median (Bq/L)**	**Range (Bq/L)**	**St. dev.**
Plant 1								
S1-Point of discharge	56.53±9.645	56.53	46.88-66.17	13.64	1.19±0.27	1.19	0.92-1.46	0.38
S2	46.74±4.680	46.74	42.06-51.42	6.62	0.37±0.15	0.37	0.35-0.38	0.21
Plant 2								
S1-Point of discharge	64.93±1.18	64.93	63.75-66.10	1.66	1.39±0.09	1.39	1.3-1.47	0.12
S2	47.64±2.05	48.11	43.88-50.92	3.54	0.532±0.6	0.51	0.43-0.63	0.10
S3	70.28±4.67	73.21	61.14-76.48	8.09	0.16±0.04	0.12	0.12-0.24	0.07
Plant 3 (pond 1)								
S1-Point of discharge	71.55	71.55	71.55	0.00	2.55	2.55	2.55	0.00
S2	60.44±3.33	60.44	57.11-63.77	4.71	1.89±0.28	1.89	1.61-2.16	0.39
S3	71.17±7.36	71.17	63.82-78.53	10.40	0.8±0.39	0.80	0.41-1.19	0.55
Plant 3 (pond-2)	37.75	37.75	37.75	0.00	1.65	1.65	1.65	0.00
Plant 4								
S1-Point of discharge	20.40	20.40	20.40	0.00	1.33	1.33	1.33	0.00
S2	1952	19.52	19.52	0.00	1.27	1.27	1.27	0.00
Plant 5								
S1-Point of discharge	57.03±7.75	64.00	41.56-65.40	13.42	1.42±0.33	1.46	0.82-1.97	0.58
S2	60.866±1.52	61.31	58.04-63.25	2.63	1.50±0.10	1.47	1.34-1.70	0.18
S3	66.87±5.19	5.19	56.53-72.90	9.0	2.42±0.37	2.77	1.69-2.81	0.64
Plant 6								
S1-Point of discharge	54.61	54.61	54.61	0.00	6.9	6.9	6.9	0.00
S2	34.05	34.05	34.05	0.00	6.07	6.07	6.07	0.00
Plant 7								
S1-Down stream	7.62±	7.62	7.62	0.00	0.12	0.12	0.12	0.00
S2-Up stream	1.48±	1.48	1.48	0.00	0.03	0.03	0.03	0.00

S1-S3 are sampling station, **Sem:** Standard error mean and **St.D:** Standard deviation

The higher uranium-238 and thorium-232 concentrations in water samples collected at down stream relative to upstream, suggested that amang processing enhances their concentrations. In the case of plants employing close water system, such enhancement is expected with every recycling process.

The enhancement of NORM in water may also be attributed to the acidity of the recycling water. Such acidity is caused by the acidic nature of amang [16]. Acid conditions caused the radionuclides to dissolved in water.

Another finding from this study is that, the mean mass and activity concentrations of thorium-232 in all sediment samples (amang plants 1 and 2) were higher than the mass and activity concentrations of uranium-238 in sediment samples (Table 1). However this was the opposite in water samples.

Results from this also showed that average concentrations of uranium-238 and thorium-232 in both water and sediment samples were higher than those measured from areas that were not involved in amang processing or tin mining activities. According to R.M.R. Almedia [6], natural uranium-238 concentration in ground water range from 0.1 to 10 ppb, while in this study the mean maximum concentration in water sample reported was 6.64 ppm (in S8-L1), or 6600 times more than the maximum concentration of uranium-238 in natural ground water. Natural uranium is the only radioactive substance for which chemical toxicity is the limiting factor in risk assessment the maximum contaminant level for uranium is 20 μgl^{-1} [6]. As mentioned mean concentration of uranium-238 in amang water samples was 4.34 ppm, it means the average concentration of uranium in amang water samples was around 220 times more than maximum contamination level of uranium. A. Martin Sanchez [18] reported low concentration of uranium series in water samples in Extramadura (Spain), ranging from 0.024 to 2.69 ppb and most of them were below 1.0 ppb. Likewise the uranium concentration of Slovenian spas area ranged from 0.2 to 2.7 ppb [Kobal, 9]. According to Boyle (1982) the mass concentration of thorium in natural water is around 0.005 – 0.5 ppb. In this study the concentration of thorium-232 in water samples ranged from 0.03 – 1.7 ppm. The maximum mean concentration of thorium-232 in water taken at station S15-L1 was 3400 times higher than those reported by Boyle in ground water. I. G. E. Ibeanu [8], showed that the measured concentration levels of uranium and thorium in tin tailing samples and the measured dose rates in Nigeria were found to be elevated with values up to approximately 100 times above background levels of control soils.

Higher concentrations of uranium-238 and thorium-232 in sediment relative to water observed in this study supported other earlier reports [15, 16]. Higher concentrations of both radionuclides in sediment is attributed to the insolubility of minerals bearing radionuclide in this water, such minerals include monazite, zircon and ilmenite.

Ismail *et al*. [15, 16] and Redzuwan *et al.* [2] carried out similar studies in Perak and Selangor in Malaysia respectively. Ismail *et al* reported uranium-238 and torium-232 mass concentrations ranging from 6.93- 11.45 $mgkg^{-1}$ and 27.72-120.88 $mgkg^{-1}$ respectively. Redzuwan reported activity concentrations of uranium-238 and thorium-232 ranging from 6.27-435.95 $Bqkg^{-1}$ and 12.90-301.59 $Bqk^{-1}g$ respectively. Our finding were in correlations with those of Ismail *et al*. and Redzuwan *et al*. in both the mass and activity concentration of both radionuclides and their differences between uranium-238 and thorium-232.

Conclusion:

Gamma ray spectrometry definitely appeared to be a useful and sensitive method for obtaining actual information on radionuclides in the environments. A total of 33 water samples and 26 sediment samples taken from amang processing plants/ river and ponds where analyzed for uranium-238 and thorium-232 concentrations. Results further confirm other earlier limited studies that amang processing enhances NORM into TENORM. Concentrations of uranium-232 were higher in water than thorium-232.However it was the opposite in sediment. Overall uranium-238 and thorium-232 concentrations were higher in sediment than water indicating the insolubility of these NORM in water and suggesting that they remained in mineral form in the sediment.

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