



## Review

# Radiological hazard associated with *amang* processing industry in Peninsular Malaysia and its environmental impacts

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## ABSTRACT

Continuous depletion in tin productions has led to a newly emerging industry that is a tin by-product (*amang*) processing industry to harness mega tons of tin by-products produced in the past. *Amang* composed of profitable multi-heavy minerals and rare-earth elements. With poorly established safety and health practices in operating plant, *amang* poses extremely high radioactivity problem associated with high occupational ionizing radiation exposures to workers and continuously impacting the local environment with radioactive contamination from industrial effluent and solid waste into lithosphere and water bodies. The radioactivity level of <sup>238</sup>U and <sup>232</sup>Th series in the mineral varies from few hundreds up to ~200,000 and ~400,000 Bq kg<sup>-1</sup> respectively and are potential to yield more than ~ 30,000 nGy h<sup>-1</sup> of gamma (γ) radiation exposure to plant workers. The study found out that for 8 h of work time, a worker is estimated to receive an average effective dose of 0.1 mSv per day from external γ radiation source with a maximum up to 2 mSv per day for extreme exposure situation. Interferences of different exposure routes for examples inhalation of equivalent equilibrium concentration (ECC) of <sup>222</sup>Rn and <sup>220</sup>Rn progenies and airborne long-lived α particles from the dusty working environment could pose a higher total effective dose as much as 5 mSv per day and 115 mSv per year. The value is 5 times higher than the annual dose limit for designated radiation worker (20 mSv) in Peninsular Malaysia. The study found that 41% of the total received an effective dose received by a worker is contributed by <sup>222</sup>Rn, 32% of airborne particulates and dust, 23% from external γ exposure and 4% from <sup>220</sup>Rn. Based on radioecological risk assessment, the study found out that the aquatic environment is the highly exposed group to ionizing radiation from industrial effluent discharge and sand residues. With the impotent establishment of radiation protection in the industry, plus the country newly introduced long-term plan to revive tin mining as well as its accessory *amang* mineral, it is necessary for the government to harmonize current regulation to improve the worker safety and health as well as sustaining local environment.

## 1. Introduction

*Amang*, locally named for mineral tailings in Malaysia, is a cassiterite-tailing or tin oxide (SnO<sub>2</sub>) secondary product that inevitably derived from mineral separation processes (i.e., gravitational, magnetic and high-tensional separators). It has been classified as a Technically Enhanced Naturally Occurring Radioactive Material (TENORM) after few decades of rapid practice in mineral processing since before 80's and any attempt in mining or processing the mineral is liable for licensing by authorities (Hu et al., 1984; Kandaiya et al., 1987; Hu and Kandaiya, 1985a; AELB, 1991; Udompornwirat, 1991; Tajuddin et al., 1994; Hewson, 1996; Roberts, 1995; Omar et al., 2007).

Produced in mass piles of different matrices; from fine grains of micro-particulates to coarse particulate sizes of sand, *amang* mineral are recognizable in black or dark brownish color; a typical reference to the indigenous stannous-bearing mineral of cassiterite (Penzer, 1921). They are mineralogically distinct to a highly-radioactive black sand beach deposited at Langkawi Island and Batu Feringghi in term of their derivations, textures and mineral compositions (Omar and Hassan, 2002; Omar et al., 2007). Purely-homogenous size of grains of the peninsula black sand-placer deposits are formed due to weathering process of plutonic exposures; eroded by sea and deposited on shore are consisted of primarily 86% of tourmaline and roughly 12% of ilmenite (Omar and Hassan, 2002). Contrarily, *amang* is predominately constituted by great

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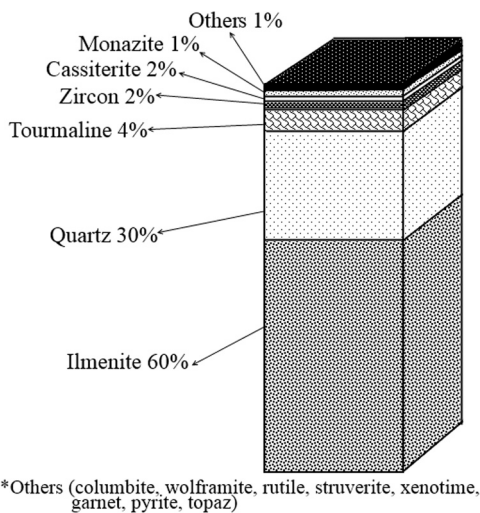


Fig. 1. The distribution % of heavy minerals in *amang* mineral.

the proportion of 60% ilmenite, 30% quartz and multiple trace amounts of other heavy minerals. Udornpornwirat (1991) has reported the typical distributions (%) of multi heavy-minerals in *amang* produced from Peninsular Malaysia (Fig. 1).

Continuing controversy of *amang* processing industries is predominantly engendered by extremely high radioactivity problem associated with high occupational ionizing radiation exposures; e.g., elevated radioactivity concentrations detected for  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay nuclides could span up to about  $\sim 200,000$  and  $\sim 400,000$  Bq kg $^{-1}$ , respectively. Based on dose conversion factor of 0.463 and 0.604 nGy h $^{-1}$  per Bq kg $^{-1}$  (Saito and Jacob, 1995; UNSCEAR, 2008) such amount could possibly yield more than  $\sim 30,000$  nGy h $^{-1}$  of gamma ( $\gamma$ ) radiation exposure (or  $\sim 30,000$  mSv h $^{-1}$  of radiation equivalent dose). Any attempt by *amang* workers for managing and handling *amang* product of monazite bags could pose them to extremely high  $\gamma$  equivalent dose beyond 100 mSv h $^{-1}$ .

In the past few decades, the industrial practices itself and environment layout were quite debatable. In the past, *amang* processing are quite simple (Udornpornwirat, 1991) which some of the tasks in the plant are needed to be manually executed by workers (Dahan, 1990; Omar et al., 2007). Coupled with the poor hygiene environment and messy work (Kandar and Bahari, 1995) assisted by only spades, buckets and wheelbarrows (Hewson, 1996), the problem arises as they are posed by severe radiation exposure (Hu and Kaidaiya, 1985b) as they constantly operated the tasks (Chong et al., 1978; Hu and Koo, 1981). Surrounded by huge stockpiles of *amang* products around the plant (AELB, 1994a), worker such as bulldozer driver is constantly exposed to extreme  $\gamma$  radiation exposure as they executing the machinery task of transporting the mineral (Ramli, 2007; Omar et al., 2008). Prohibited practices of processing thorium cake (yellow-brownish slurry or by product as a result of a chemical leaching process to extract REE) for trade values also were reported in previous time (Hu et al., 1984). Cracking yellow cake or urania also possible at that time considering loosening authority surveillance and monitoring, and lack of comprehension on associated hazards.

Few spots inside of *amang* plant such as monazite storeroom, magnetic separators, tension separators and electrostatic separator have been identified as hotspots (contribute a higher effective dose) owing to substantial mineral dust production which can be indicated by poor visibility inside the plant (Udornpornwirat, 1993). The problem exacerbated by the situation of continuous inhalation and ineluctable ingestion of long-lived alpha emitting particles, EEC of radon ( $^{222}\text{Rn}$ ) and thoron ( $^{220}\text{Rn}$ ) progenies by workers in *amang* processing plants (AELB, 1991; Hewson, 1996). With poorly established radiological

safety and health practices in the plant, usage of film badge, protective mask and cloth by workers during plant operation are not made mandatory by the plant superior. Most of the workers are not made certified as radiation workers by Atomic Energy Licensing Boards Malaysia (AELB) even though the companies are required to possess an operating license under it (Omar et al., 2007, 2008). Meanwhile, for small *amang* plant, they are not made mandatory to secure an operating license from AELB as per excepted on Act 304 (Omar et al., 2007; AELB, 1994a).

Environmental contamination problem associated with radioactive residues from *amang*  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series threaten to further worsen the situation, not to mention other prominent ecological and lithosphere impacts from previous rapid excavating activities of tin-mining. Triggered by unresolved court case of Bukit Merah radioactive *amang* pollution by Asian Rare Earth Sdn Bhd (ARE) in 1982, the *amang* industry has started receiving bad perception and pressure from local and even at international level, which forcing the company to completely cease their operation in 1994. Uncontrolled and rapid processing of *amang* for yttrium value began to provoke the local with releases of unpleasant odor and smoke from the factory which prompted symptom like shortness of breath and probably posing high risk to the inhalation of radioactive airborne dust. The company sustained their old operation as their biggest company share i.e., Beh Mineral Sdn Bhd took over and continued the operation under strict supervision and full guidance by AELB.

The scarcity of tin resources in late 1080s forced the *amang* industry to halt down their productivity and to some extent ceased the operation, from notably 81 processing plants in 1984 to 66 in 1989 (Sulaiman et al., 1994). With few major plants remaining in Bukit Merah, Lembah Kinta (Ramli, 2007), Lembah Klang (Bahari et al., 2007), Kelian Intan (open-pit tin mining) and a few more in Johor and Pahang, *amang* minerals are expected to be continuously produced to some extent in the future. In fact, there is an attempt for tin mining full revival announced by the government in 2019 (New Straits Times, 2019). Motivated by stable tin price ranged US \$18,000–25,000 per tonne for the last 10 years (New Straits Times, 2019), and extremely large uncovered mother lode deposits of 1.2 M per metric tonne as forecasted by US Geological Survey (Yap, 2007), the government are ready to splash the cash for the tin revival in the future.

With the mega reserve of tin lode worth about RM 350 billion deposited along The Main Range Granite batholiths (New Straits Times, 2019) primarily in established protected areas of Royal Belum Reserve Forest, Gopeng and Ulu Kinta Forest Reserves, the cassiterite exploitation and *amang* processing are expected to continuously pose disastrous impact to human and ecology. This review aims on documenting radiological hazard associated with *amang* and their environmental impacts as well as reporting the current status of the industry, which is in high possibility to be revived by the government in the future.

## 2. Method of data collection

### 2.1. Data collection strategies and objectives

In order to systematically and comprehensively collect all data from the previous studies, a conceptual data objective model was developed to sufficiently retrieve all information for a complete review. It also identifies the existence of the data, the availability of that particular data and provides reliability of the finding from this work. The application of the model comprised the following data objectives i.e., (1) the need of  $^{238}\text{U}$  and  $^{232}\text{Th}$  data of local *amang* mineral, final product and intermediate products from local tin mining industries; (2) radiation measurement data in *amang* processing plant; (3) radioactivity levels of industry environment in all medium i.e. air, terrestrial and water; and (4) data of environmental impact assessment and radioactivity pollution in *amang* processing industries.

**Table 1**  
Content of radionuclides and activity concentration related to *amang* mineral at *amang* processing station/mining station in Malaysia.

State/Locale	Amang product/type of sample	Sampling point/information	Radionuclide concentration (Bq kg <sup>-1</sup> )				Reference	
			U <sup>238</sup>	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>		
Klang Valley	sediment	recycle pond	364.46–472.27	427.41–637.61	161.25–230.00	74.80–74.80	Bahari et al. (2007)	
Not stated	sediment	recycle pond	84.14–860.57	105.10–343.32	21.25–198.75	374.10–823.10	Yusof et al. (2001)	
	water	ex-mining pond	0.34–1.05	0.17–0.73	–	–		
Ipoh, Perak	monazite	–	30,628.00	–	–	–	Hamzah and Mahmood (1985)	
	zircon	–	16,178.50	–	–	–	Sharif and Ghazali (1987)	
	xenotime	–	112,385	–	–	–		
	monazite	2 different plant	21,402.55–22,958.65	–	–	–		
	monazite	tin tailing	3087.50	–	–	–	Meor Sulaiman (1988)	
Kinta Valley, Perak	monazite	–	23,218.00	–	–	–	Hamzah and Mahmood (1985)	
	zircon	–	21,118.50	–	–	–	Hu et al. (1981)	
	xenotime	–	67,925.00	–	–	–		
	cassiterite	sand mix	–	666.00	397.75	–		
		cassiterite mix.	palong	–	33.30	33.30	–	Chong et al. (1978)
		ilmenite	stockpile	–	70.30	75.85	–	Sharif and Ghazali (1987)
		zircon	stockpile	–	2519.70	5300.25	–	
		monazite	–	203,034.00	66,751.70	20,757.00	–	
		xenotime	–	–	15,802.70	43,549.00	–	
		monazite	–	26,095.55 ± 1729.00	–	–	–	
		monazite	tin tailing	3050.45	–	–	–	
		zircon	86.45	–	–	–	–	
		ilmenite	tin tailing	49.40	–	–	–	
		<sup>8</sup> Mix ores	tin tailing	2223.00	–	–	–	
		raw <i>amang</i>	stockpile	1025.05–4730.05	706.44–6357.96	–	–	Eng (2004)
	<i>amang</i>	stockpile	1827.8–3445.65	1327.62–5655.58	–	–		
	<i>amang</i>	stockpile	4890.6–18413.85	9171.54–71,167.74	–	–	Ramli (2007); Wagiran et al. (2005)	
	monazite	28,442.05–50,264.5	39,142.46–243,847.66	–	–	–		
	zircon	stockpile	11,893.05–14,721.20	1433.18–2366.98	–	–		
	ilmenite	stockpile	1840.15 – 3581.50	1222.06–2334.50	–	–		
	raw <i>amang</i>	–	1012.70–4680.65	673.96–6130.60	–	–		
Kinta (5 <i>amang</i> processing plants)	monazite	–	28,120.95–49,708.75	37,758.00– 235,272.94	–	–		
	cassiterite	–	494.00–3952.00	263.9–1477.84	–	–		
	waste sand	–	469.30–1197.95	190.82–868.84	–	–		
	wet sand	–	259.35 ± 37.05	166.46 ± 28.42	–	–		
	dry sand	–	629.85 ± 61.75	690.2 ± 44.66	–	–		
	<i>amang</i>	stockpile	14,128.40–18,216.25	40,336.10 ± 68,666.78	–	–		
	<i>amang</i>	stockpile	11,250.85 ± 728.65	24,339.7 ± 389.76	–	–		
	monazite	stockpile	16,474.90 ± 1037.40	45,557.26 ± 641.48	–	–		
	monazite	stockpile	11,769.55 ± 741.00	1421 ± 64.96	–	–		
	zircon	stockpile	12,683.45–14,560.65	1380.4–2277.66	–	–		
	zircon	stockpile	4828.85 ± 333.45	8846.74 ± 182.70	–	–		
	ilmenite	stockpile	1827.80 – 3544.45	1234.24–2241.12	–	–		
	raw <i>amang</i>	stockpile	2087.15–3408.60	2216.76–4612.16	–	–		
	raw ilmenite	stockpile	2235.35 ± 160.55	661.78 ± 36.54	–	–		
	ilmenite (mix)	stockpile	2717.00 ± 197.6	816.06 ± 40.6	–	–		
	leached ilmenite	stockpile	703.95 ± 61.75	300.44 ± 32.48	–	–		
	tourmaline	stockpile	86.45 ± 24.70	77.14 ± 24.36	–	–		
	rutile	stockpile	876.85 ± 74.10	450.66 ± 32.48	–	–		
	monazite	stockpile	35,400.00 ± 2200.00	164,800.00 ± 2000.00	–	–		
	zircon	stockpile	13,000.00 ± 800.00	1700.00 ± 1000.00	–	–		
ilmenite	stockpile	2600.00 ± 100.00	1600.00 ± 100.00	–	–			
monazite	–	9978.80 ± 913.90	–	–	–			

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Table 1 (continued)

State/Locale	Amang product/type of sample	Sampling point/information	Radionuclide concentration (Bq kg <sup>-1</sup> )				Reference
			U <sup>238</sup>	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>	
Jelapang, Kinta							Sharif and Ghazali (1987)
Bidor	monazite	–	18,117.45 ± 96.33	–	–	–	Meor Sulaiman (1988)
	monazite	tin tailing	3149.25	–	–	–	
Bidor	monazite	not stated	26,429.00 ± 1482.00	–	–	–	Hamzah and Mahmood (1985)
Mambang Diawan	monazite	stockpile	1856.50 ± 9.30	10,287.10 ± 9.30	–	–	Yasir et al. (2007); Majid et al. (2007)
	xenotime	stockpile	6911.10 ± 11.70	3733.1 ± 6.00	–	–	
	ilmenite	stockpile	318.90 ± 2.60	142.7 ± 1.20	–	–	
	sediment	discharge point	1110.50 ± 7.30	1966.6 ± 4.70	–	–	
	sediment	recycle pond	516.8 ± 4.40	292.10 ± 1.90	–	–	
	sediment	recycle pond	262.9 ± 3.90	154.40 ± 1.70	–	–	
	industrial effluent	discharge point	28.98 ± 1.35	29.59 ± 0.89	–	–	
	industrial effluent	recycle pond	35.42 ± 1.63	36.16 ± 1.02	–	–	
	industrial effluent	natural water	28.98 ± 1.35	32.05 ± 0.87	–	–	
	Mambang Diawan	monazite	–	6496.10 ± 1235.00	–	–	–
ilmenite		tin tailing	135.85	–	–	–	Meor Sulaiman (1988)
Gopeng	industrial effluent	tin mining	–	–	0.09–0.10	–	Roberts (1995)
	initial zircon ore	–	N/D	1.02	0.62	–	Hu et al. (1981)
	zircon	ore final product	N/D	0.13	0.49	–	Chong et al. (1978)
Lahat	industrial effluent	processing plant	0.15	0.00	0.32	–	Roberts (1995)
	industrial effluent	processing plant	0.86	0.12	0.21	–	
	industrial effluent	processing plant	–	0.00	0.18	–	
Batu Gajah	industrial effluent	outflow source	0.04	0.08	0.17	–	
Batu Gajah	industrial effluent	small puddle	0.00	0.16	0.36	–	
Kota Baru	industrial effluent	separator	0.00	–	0.08	–	
Kampar	fish contaminated soil	ex-mining pond nearest pond	0.06–0.54 46.60 ± 2.60	0.29–1.23 21.20 ± 1.40	–	7.45–30.60 242 ± 34	Tsyr (2005)
	industrial effluent	ex-mining pond	0.17 ± 0.08	3.20 ± 0.70	–	16.20 ± 3.40	
	industrial effluent	ex-mining pond	–	–	0.06 – 0.05	–	Hamzah et al. (2008)
Kg. Gajah	industrial effluent	ex-mining pond	–	–	0.06 – 0.05	–	Hamzah et al. (2008)
	topsoil (10 cm)	ex-mining area	–	1530.00–20,097.00	5426.00–77,764.00	1241–19,600	Hamzah et al. (2008)
Not stated	monazite	–	24,700.00	284,200.00	–	–	AELB (1994b)
	xenotime	–	61,750.00	20300.00	–	–	
	zircon sand	–	30,875.00	trace amount	–	–	
	ilmenite	–	2470.00	trace amount	–	–	
Not stated	monazite	–	12,350.00–37,050.00	81,200.00–365,400.00	–	–	SEATRAD (1991)
	xenotime	–	37,050.00–86,450.00	40,600.00–203,000.00	–	–	
	zircon	–	24,700.00–49,400.00	40,600.00–57,652.00	–	–	
	cassiterite	stockpile	506.35 ± 49.4	272.02 ± 32.48	–	–	
	rutile	stockpile	901.55 ± 86.45	475.02 ± 48.72	–	–	
	sand residue	stockpile	271.70–1210.30	178.64–905.38	–	–	
	amang sample	processing plant	9125.415	5563.012	–	532.10	Roberts (1995)
	air sampling	separator	889.20	–	–	–	Ghazali et al. (1986)

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Table 1 (continued)

State/Locale	Amang product/type of sample	Sampling point/information	Radionuclide concentration (Bq kg <sup>-1</sup> )				Reference	
			U <sup>238</sup>	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>		
Seri Kembangan	air sampling <i>amang</i>	separator	–	243.60	370.50	–	Hu et al. (1981); Chong et al. (1978)	
		fine particulate	–	–	2752.80	1058.20		
Kg. Manggis	cassiterite mix. clay industrial effluent	separator	–	1261.70	732.60	–	Roberts (1995)	
		pump site tin mining	–	81.40 0.00	85.10 0.00	–		
Not stated	monazite	–	19,142.50	–	–	–	Hamzah and Mahmood (1985)	
Not stated	xenotime	tin tailing	1963.65	–	–	–	Meor Sulaiman (1988)	
Not stated	monazite	–	25,366.90 ± 2013.05	–	–	–	Sharif and Ghazali (1987)	
Air Hitam, Puchong	monazite	–	24,749.40 ± 1235.00	–	–	–	Yasir et al. (2007)	
Puchong (3 stations)	sediment	ex-mining pond	9.37–26.32	30.76–35.34	–	–		
		plant industrial effluent	N/D 36.72	N/D –	0.10 –	–		
Gopeng	<i>amang</i> sample	tin mining	3190.005	7413.56	–	719.9	Roberts (1995)	
		monazite	–	1570 ± 103	–	–	Sharif and Ghazali (1987)	
		monazite	tin tailing	2889.90	–	–	–	Meor Sulaiman (1988)
Kinta Valley	ilmenite monazite	tin tailing	345.80	–	–	–	Hewson (1996)	
		stockpile	–	203,000	–	–		
Dengkil	<i>amang</i> industrial effluent monazite	stockpile	–	243,600	–	–	Roberts (1995)	
		ex-tin mining	–	–	–	0.08–0.14	Yasir et al. (2007)	
Dengkil	monazite	stockpile	832.88 ± 90.56	13,426.68 ± 3307.00	22,011.87 ± 2590.00	3773.15 ± 380.98	Omar and Hassan (2002) Sharif and Ghazali (1987) Bahari et al. (2007)	
		ilmenite	76.31 ± 7.71	97.28 ± 9.88	2099.29 ± 210.65	154.66 ± 16.87		
		zircon	2620.98 ± 250.65	204.83 ± 20.32	8898.21 ± 900.32	179.64 ± 18.05		
		tin	52.83 ± 4.85	24.45 ± 3.0	149.63 ± 20.23	108.04 ± 8.97		
		monazite	–	246.36 ± 20.40	12,452.18 ± 3067	13,573.32 ± 2510		3239.73 ± 150.66
		ilmenite	–	3538.94 ± 250.65	132.44 ± 5.75	822.64 ± 50.85		3219.64 ± 205.75
		contaminated soil	ex-mining area	449.15 ± 25.32	37.52 ± 1.55	105.75 ± 1.75		247.28 ± 8.80
		contaminated soil	plant area	294.46 ± 15.25	103.08 ± 8.25	976.65 ± 5.25		181.74 ± 6.23
		industrial effluent	discharge point	1.52 ± 0.22	1.65 ± 0.35	0.71 ± 0.07		11.75 ± 1.55
		industrial effluent	recycling pond	1.40 ± 0.15	1.57 ± 0.33	0.76 ± 0.07		11.44 ± 2.04
		industrial effluent	ex-mining pond	1.63 ± 0.25	1.57 ± 0.3	0.00		11.75 ± 1.55
		tourmaline	upgrading plant	–	2490.00	5980.00		–
		monazite	–	18,117.45 ± 1272.05	–	–		–
		soil with tin ore	upgrading plant	–	–	857.00		–
		intermediate tin soil/water	raw mineral separator	–	–	268.00 25.00–36.00		–
Air Hitam, Puchong	monazite	–	24,749.40 ± 125.00	–	–	Sharif and Ghazali (1987)		
Selayang Baru	monazite	–	21,563.10 ± 629.85	–	–	Hu et al. (1984);		
Kuantan	monazite	–	18,006.30 ± 642.20	–	–			
West Coast Penin.	monazite	stockpile	43,216.00 ± 21,164.00	264,957.00 ± 3922.00	–		–	

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Table 1 (continued)

State/Locale	Amang product/type of sample	Sampling point/information	Radionuclide concentration (Bq kg <sup>-1</sup> )				Reference
			U <sup>238</sup>	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>	
	thorium cake	stockpile	130,314.00 ± 3108.00	48,729.00 ± 17,242.00	–	–	Hu and Kandaiya (1985a) (1985b)
Gambang	xenotime	stockpile	110,408.00 ± 3367.00	25,641.00 ± 555.00	–	–	Hamzah and Mahmood (1985) Sharif and Ghazali (1987) Meor Sulaiman (1988)
	zircon	stockpile	18,241.00 ± 3108.00	32,967.00 ± 1295.00	–	–	
	ilmenite	stockpile	8214.00 ± 962.00	10,545.00 ± 185.00	–	–	
	monazite	stockpile	31,986.50 ± 2099.50	–	–	–	
	monazite	stockpile	8472.10 ± 568.10	–	–	–	
Sungai Besi	raw amang	stockpile	209.95	–	–	–	
Sungai Way	mix ore	tin tailing	2062.45	–	–	–	Omar et al. (2007)
Not stated	ilmenite	stockpile	–	1397.00	3460.00	–	
	monazite (30%)	stockpile	–	81,906.00	20,975.00	–	
	zircon (65%)	stockpile	–	2021.00	11,303.00	–	
	xenotime (30%)	stockpile	–	41,403.00	93,274.00	–	
	struverite (10%)	stockpile	–	243.00	1061.00	–	
	tourmaline 80%	stockpile	–	2306.00	986.00	–	
	rutile;	stockpile	–	248.00	439.00	–	
	cassiterite	stockpile	–	19.00	36.00	–	
	cassiterite wolframite (74%)	stockpile	–	34.00	230.00	–	
Not Stated	sediment	recycling pond	222.30	251.92	–	–	Mohsen et al. (2007)
	industrial effluent	recycling pond	54.38	1.50	–	–	–
	industrial effluent	discharge point	46.88–66.17	0.92–1.46	–	–	
	industrial effluent	discharge point	63.75–66.10	1.30–1.47	–	–	
	industrial effluent	nearest to pond	61.14–76.48	0.12–0.24	–	–	
	industrial effluent	discharge point	71.55	2.55	–	–	
	industrial effluent	nearest to pond	63.82 –78.53	0.41–1.19	–	–	
	industrial effluent	discharge point	41.56–65.40	0.82–1.97	–	–	
	industrial effluent	nearest to pond	56.53–72.90	1.69–2.81	–	–	
	industrial effluent	discharge point	54.61	6.90	–	–	
Kemaman	zircon	by-product	17,290.00	12,992.00	–	–	Meor Sulaiman and Muslimin (2010)
Puchong	zircon	by-product	19,760.00	2436.00	–	–	
Dengkil	zircon	by-product	19,760.00	3248.00	–	–	
Lahat	zircon	by-product	22,230.00	2436.00	–	–	
Kampar	zircon	by-product	30,875.00	59,682.00	–	–	
Bidor	zircon	by-product	18,525.00	2842.00	–	–	
Not stated	tin slag	–	950.00	40.00	1110.00	–	Lin (2004)
Butterworth	tin smelting plant	tin slag 1	894.59	20,408.82	976.75	1280.27	Lin (2004)
	tin smelting plant (store room)	tin slag 2	1036.43	23,182.77	1153.07	1532.19	
	plant	slag without Ta	601.71 – 1036.43	11,230.45–23,182.77	491.74 – 1153.07	719.64–1532.19	Lin (2004)
		mix of soil-slag	45.89 – 147.92	240.88–1011.83	35.96–148.72	494.71–566.33	
MMC (company)	zircon	by-product	18,401.50 ± 3087.5	–	–	–	Hamzah and Mahmood (1985) Kandaiya et al. (1987)
Not stated	monazite	by-product	–	–	30,600.00 ± 2600.00	–	
Not stated	struverite	by-product	–	–	–	–	

(continued on next page)

Table 1 (continued)

State/Locale	Amang product/type of sample	Sampling point/information	Radionuclide concentration (Bq kg <sup>-1</sup> )				Reference
			U <sup>238</sup>	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>	
Penang	first tin slag	by-product	–	6956.00	163,800.00 – 170,400.00 888.00	–	Hu et al. (1981); Chong et al. (1978)
	final tin slag	by-product	–	2738.00	3182.00	–	
Kota Bharu	cassiterite amang sample	by-product processing plant	675.545	1228.962	1480 –	148 125.2	Roberts (1995)
Kaki Bukit, Perlis	(sand mix)	by-product	–	1.67	1.85	–	Hu et al. (1981); Chong et al. (1978)
Selangor	sediment	recycling pond	61.20–340.40	48.10–609.60	23.75–151.25	399.10–848.00	Bahari et al. (2007)
Selangor	water	ex-mining pond	1364.7 ± 11.15	297.29 ± 32.0	–	–	Yusof et al. (2001)
Johor	water	ex-mining pond	554.39 ± 71.01	387.28 ± 43.04	–	–	Yusof et al. (2001)
	water	ex-mining pond	–	–	135.42 ± 15.91	–	
Kota Tinggi	sediment	ex-mining pond	33.22–55.35	70.3–89.3	33.26–55.41	169.46–237.17	Mohd Nazri (2001)
Perak, Selangor	contaminated soil	13 former plants	37.17–1819.4	20.34–4519.79	–	37.56–716.77	Roberts (1995)
Kota Bharu	amang sample	processing plant	85,382.0955	30,046.03	–	2410.10	–

2.2. Data collection and extraction process

In total 58 references were successfully collected in this study and about more than 63% of the sources or works are published at the national level, in local publisher, seminars, and conference proceedings. The most of important data are retrieved from internal and unpublished reports from government department and regulatory body libraries i.e, Department of Geological Survey, Southeast Asia Tin Research and Development Centre, Malaysia Nuclear Agency and Atomic Energy Licensing Board of Malaysia. Most of sources were reported in olden decades as the industry glory dated back in the 1980s–1990s. Specifically 43 collected sources (about 69%) were in time frame from 1978 to 2002, and the rest of the source was reported beyond the time frame and less than 1% were recorded following 2010.

A literature search of published work under scientific citation indexed publication were performed using Web of Sciences, Scopus, PubMed, Google Scholar, ResearchGate and national online journal databases. However, only few sources were found based on topic and title search. Few thesis dissertations of undergraduate and master's research also were collected in this study to optimize data size collection.

2.3. Data treatment and quality control

Since the data are limited, only few specific criteria for source selection are considered. The first criterion is the scope of study which is only in Peninsular Malaysia, subjected only for amang or tin-tailing mineral, not an associate product of cassiterite-magnetite mineral and rare-earth mineral processing. The main requirement considered for reliability of data collected in this study is, it must an original work data not a citation data from another. Other requirements are clear and precise method used, tabulation of raw data and include at least statistical information of central tendency of measurement to verify the precision.

The raw data of radioactivity levels of <sup>238</sup>U and <sup>232</sup>Th were extracted from each of collected sources and tabulated in an Excel file for further analyses. A complete table of raw data for radioactivity levels of <sup>238</sup>U and <sup>232</sup>Th (as well as including <sup>226</sup>Ra and <sup>40</sup>K) is tabulated in Table 1. A

Table 2

Descriptive statistic of radioactivity concentration of <sup>238</sup>U in various samples.

Statistical parameters	<sup>238</sup> U radioactivity content (Bq kg <sup>-1</sup> )		
	Amang concentrated mineral	Residues, sediment and sands	Pond, surface runoff and puddle
n	113	34	39
Mean ± standard error	16,263 ± 2885	325 ± 70	29 ± 6
Confidence level (95%)	10,545–21,981	468	19–39
Standard deviation	30,675	409	30
Kurtosis	18	5	2
Skewness	4	2	1
Range	36–203,034	2–1819	MDL–79

Table 3

Descriptive statistic of radioactivity concentration of <sup>232</sup>Th in various samples.

Statistical parameters	<sup>232</sup> Th radioactivity content (Bq kg <sup>-1</sup> )		
	Amang concentrated mineral	Residues, sediment and sands	Pond, surface runoff and puddle
n	113	34	39
Mean ± standard error	22,075 ± 5010	269 ± 70	4 ± 2
Confidence level (95%)	12,144–32,006	132–406	1–7
Standard deviation	52,064	67	9
Kurtosis	15	6	8
Skewness	4	2	3
Range	19–365,400	2–20,097	MDL–36

statistical verification of distribution using IBM's SPSS Q-Q plot (Statistical Package for Social Science) was used to check the normal distribution of extracting data. Any extreme value was stripped from the list to comply normal distribution of data sampling. Then a descriptive

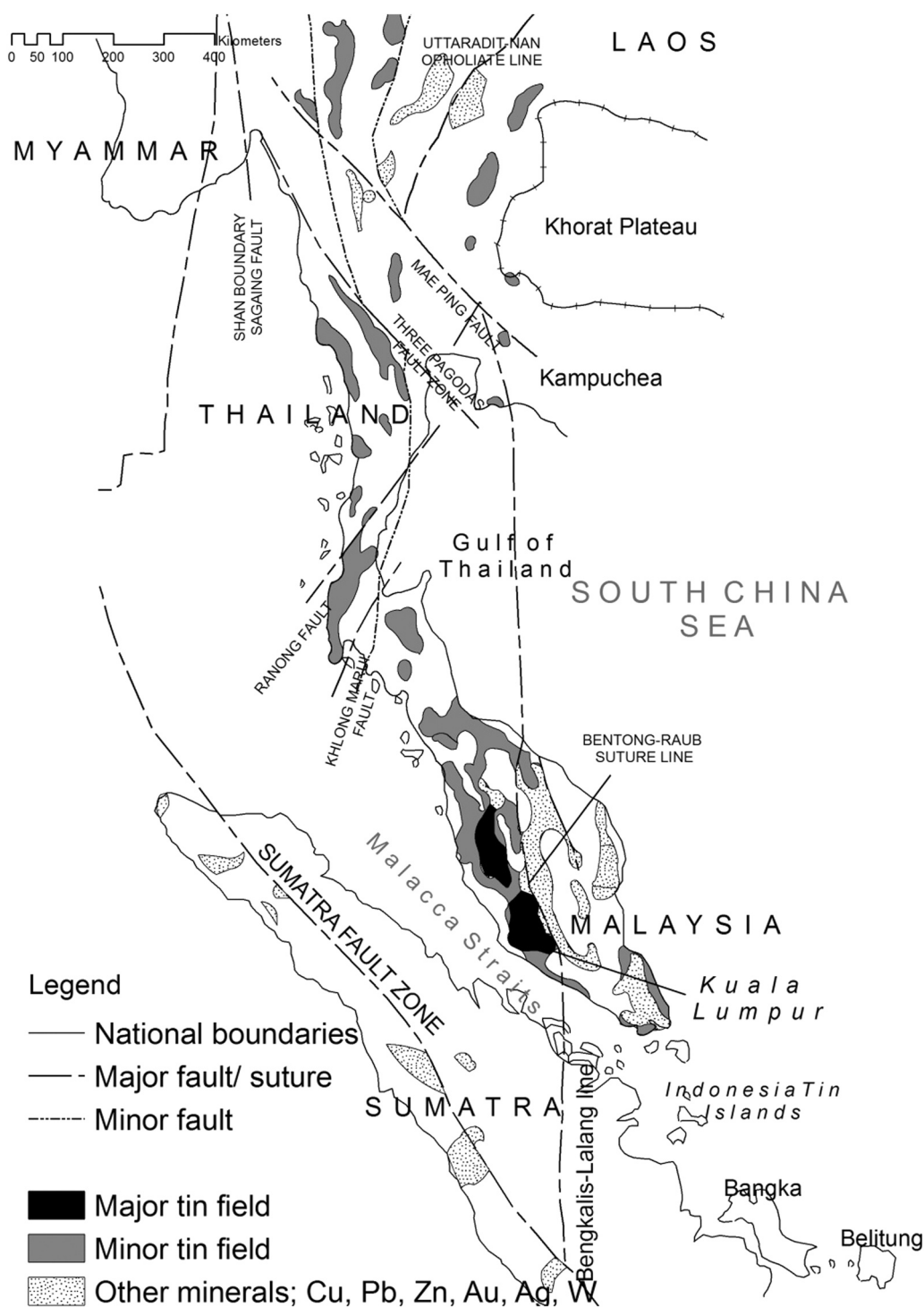


Fig. 2. The cassiterite belts of South East Asian. Re-digitized and compiled data from Schwartz (1995); (Department of Agriculture Peninsular Malaysia, 2002)

statistic was performed to provide information on reliability and precision of collecting data (as shown in Tables 2 and 3). The following steps were also performed for other main data i.e., ECC <sup>222</sup>Rn and <sup>220</sup>Rn progenies, airborne long-lived alpha emitter, and  $\gamma$  radiation dose rate.

### 3. Origin and geological setting

Past history has explained the dominance of Malaysia as the biggest tin producing country in the world's tin history (AELB, 1991; Udompornwirat, 1993; Schwartz et al., 1995) since before World War I; with

30–50% of world tin production were dominated by the country around 1900–1985 (Hails, 1976). The industry emerged to be the biggest economic driving commodity in Malaysia's history since the tin capitalization by foreign miner in 1820s (Lau, 1999). The ideal geographical setting of the Peninsular Malaysia along the rich-stanniferous region of The Southeast Asian Cassiterite Belt favors the enduring million tons of tin reserves for over 2 centuries (Schwartz et al., 1995).

As shown in Fig. 2, The Cassiterite Belt of Southeast Asian stretched out over 2800 km long from Northern Burma across Western and Peninsular Thailand, Peninsular Malaysia and ended at Belitung–Bangka

Islands of Indonesia with the estimated width of the belt over 400 km (Schwartz et al., 1995). Tectonostratigraphically, owing to tectonic evolutions of terranes and continental fragments, The Cassiterite Belt of Southeast Asian across Peninsular Malaysia can be distinguished by two distinct Sn-ore belts; one associated with I-type granitic plutons which scattered all over East Malaya Block and the other associated with large S-type granitic batholiths of Sibumasu block (Agocs and Paton, 1958; Metcalfe, 2013).

During their utmost occasions, most of the ore were derived from the latter Sn-ore belts that being flanked by the main range batholiths; with occasional occurrences in form of placer deposits (alluvium) with the very limited unearthing of vein deposits (lode); (Agocs and Paton, 1958). The cassiterite deposits are developed over igneous bodies under Triassic-complex orogenic processes; which involves folding, intruding and eroding the north-south batholith extension (Roberts, 1995). Alternately 6 other types of minor occurrences also have been identified; replacement of granite associated with other low-Fe aluminosilicate rocks, replacement of basic rocks and other high-Fe aluminosilicate rocks, replacement of carbonate rocks, hydrothermal vein deposits, breccia deposits and pegmatite deposits (Schwartz et al., 1995).

Deep weathering process due to extremely hot and humid equatorial climate also escalated rapid Sn-ore development in the country. Among 99 prominent tin field occurrences in the country as listed out by Schwartz et al. (1995), the most notable occurrences are in Kinta Valley (Ipoh district) and Klang Valley (nearby Kuala Lumpur territory); with both located on Sn-ore belts at Sibumasu block (Hails, 1976) and produces hundred thousand tons of tin per year. Based on compiling statistics from Economic History Malaysia (2020), in total over 10 million tons of tin ore were produced from the country within 1900–2004.

#### 4. Emergence of new industry (*amang*)

For more than a century the tin industry looks promising, but not until 1980 when the industry gradually stagnated, bit by bit showed lack of enthusiasm in mining (Meor Sulaiman, 1991) principally after sustained rising production costs for over 100 years, as well as other factors; multiple global recessions (Ahmad and Jones, 2013), shortfall in global tin market price (from 8000 Sterling pounds  $\text{ton}^{-1}$  to less than half of it) and foreign demands, exhausted resources and competitive domination of alternate metals, aluminum, plastics (Wong and Goh, 1996) and alloys which is more cheaper and environmentally friendly.

Severe decline in the tin mining industry during that period has led to the emergence of tin by-product (*amang*) processing industries (Bahari et al., 2000). *Amang* composed of profitable multi-heavy minerals and rare-earth elements (REE) (Hewson, 1996; Dahan, 1990) and was an economic-importance (Ramli, 2007) during the booming period of the 3rd Industrial Revolution (1980s and 1990s), particularly for semi-conductors and electronic parts. The *amang* processing industry successfully sustained for a long period of time owing to continuous and considerable *amang* production from the past until the present days (Bahari, 2007; Ramli, 2007; Sanusi et al., 2017) and also due to high global demands for REE which dominating few % of total country gross domestic products (GDP) nowadays (Department of Statistics, 2010; Department of Mineral and Geosciences, 2014).

Conventionally in tin mining, for sustainable commercialization and trade purposes, an excellent grade as slightest 0.35% of cassiterite deposit is necessary (Delgado and Capilla, 2014). However, Hewson (1996) reported that in most cases of tin mining in Peninsular Malaysia, only 0.01% of purely Sn (poor grade) are associated in cassiterites. Thus, to be practical, huge piles of *amang* were consistently yielded in that time as a result of highly concentrated Sn ore extraction (35–70%).

In most cases, *amang* were often treated and pre-processed at local mining site before making their ways to upgrading or smelting plant (Meor Sulaiman, 1988) and exported to other countries (Abdul Rahman, 1990). During magnetic segregation processes, ilmenite is first to be differentiated thus leaves behind radioactive, non-conducting and

non-magnetic (non-ferrous) compound (e.g. monazite, xenotime and zircon) (Hewson, 1996). Ilmenite from *amang* were recovered for high-priced titanium oxide whereas the rest were sold to the upgrading plant in Lembah Kinta and Lembah Klang for concentrating monazite, zircon, ilmenite, struverite, columbite, and xenotime as well as their accessory minerals (rutile, leucoxene, anatase, magnetite, limonite, hematite, and corundum) from unproductive gangue of sand and clays. The concentrated minerals then were sold to ore smelting plants for REE purely ore extraction (Delgado and Capilla, 2014; Wong and Goh, 1996).

In fact, purchases of the cassiterite mineral gangue from the mining fields were mainly motivated by unrecovered cassiterite compound, however due to escalating demand after sometimes, switched it to rapid mineral processing industry (Dahan, 1990; Udompornwirat, 1993). During earlier period of monazite recovery, monazite was exploited for thoria (Th oxide) in industrial manufacturing of incandescent thoria lamp (Meor Sulaiman and Muslimin, 2010) before the industry evolved into a rare-earth element processing industry for their highly-favored cerium (25%  $\text{Ce}_2\text{O}_3$ ), samarium and yttrium (2%  $\text{Sm}_3\text{O}_2$  and  $\text{Y}_3\text{O}_2$ ) as well as zirconium, lanthanum, neodymium (Meor Sulaiman, 1988; Jais, 2002). Comparably, xenotime and zircon minerals were also extracted from *amang* for their yttrium and zirconium and some zircon mineral are sold for ceramics industry importance in the manufacturing of glaze and opacifier.

During their prime period 1984–1989, total productions of ilmenite, zircon, monazite and xenotime within five years amounted to approximately 2300,000 metric tonnes, 93,200 metric tonnes, 25,200 metric tonnes and 1900 metric tonnes, respectively. However, after some time, the productions of ilmenite, zircon and monazite in 2010 plunged drastically by 93% (16,947 t), 89% (1124 t) and 66% (609 t) respectively compared to the 1989 productions (Department of Statistics, 2010). Ilmenite suffered the most (99%) due to dramatic depletion of tin production (Dahan, 1990). The productivity continually dropped in 2013 with the total production of ilmenite, zircon, monazite and xenotime declined to 16,947 tonnes, 379 tonnes, 261 tonnes and 97 tonnes, respectively (Department of Mineral and Geosciences, 2014).

Approximately 60 fully-operational *amang* processing plants have been reported in Peninsular Malaysia in 1990 which vary in sizes; from shelter-sized to large compound (AELB, 1991). To date less than half of the figure survived and the rest has ceased their operations after dramatically suffered resource depletion. With estimated 900,000 tonnes of Peninsula's unexplored tin ore reserves (Department of Mineral and Geosciences, 2014), and 16 tin-mining stations still operating in Peninsular Malaysia with steady trend of few tenth to hundred tonnes  $\text{y}^{-1}$  of production (Department of Statistics, 2010; Department of Mineral and Geosciences, 2014), the tin and their accessory mineral processing industries in Malaysia seem to be operated for longer period (Sanusi et al., 2017).

#### 5. Radioactivity of *amang*

The negative impact of the radioactivity associated with *amang* on workers and the environment is well documented by local and international studies. In fact, tin mining itself has attracted public attention due to dredging and excavating activity (Subramanian, 1988) that costs substantial environmental degradation (SEATRAD, 1991). Continuous practices of unearthing tin ores and concentrating their by-products not only posing adverse radiological threat to the public due to highly radioactivity materials but also engendered problem associated with heavy and toxic element contaminations i.e., Cd, Pb, Hg, Ar, Se, and Zn (SEATRAD, 1991; Kandar and Bahari, 1995).

Numerous studies conducted in Peninsula found that *amang* are naturally radioactive due to the association of primordial radionuclides from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series and single gamma emitter of  $^{40}\text{K}$  (Hu and Kandaiya, 1985a, 1985b; Mohd Nazri, 2001; Yasir et al., 2007; Omar, 1991). Classification of radioactive mineral is well defined by the IAEA (1983); which indicated that any mineral containing  $^{238}\text{U}$  and

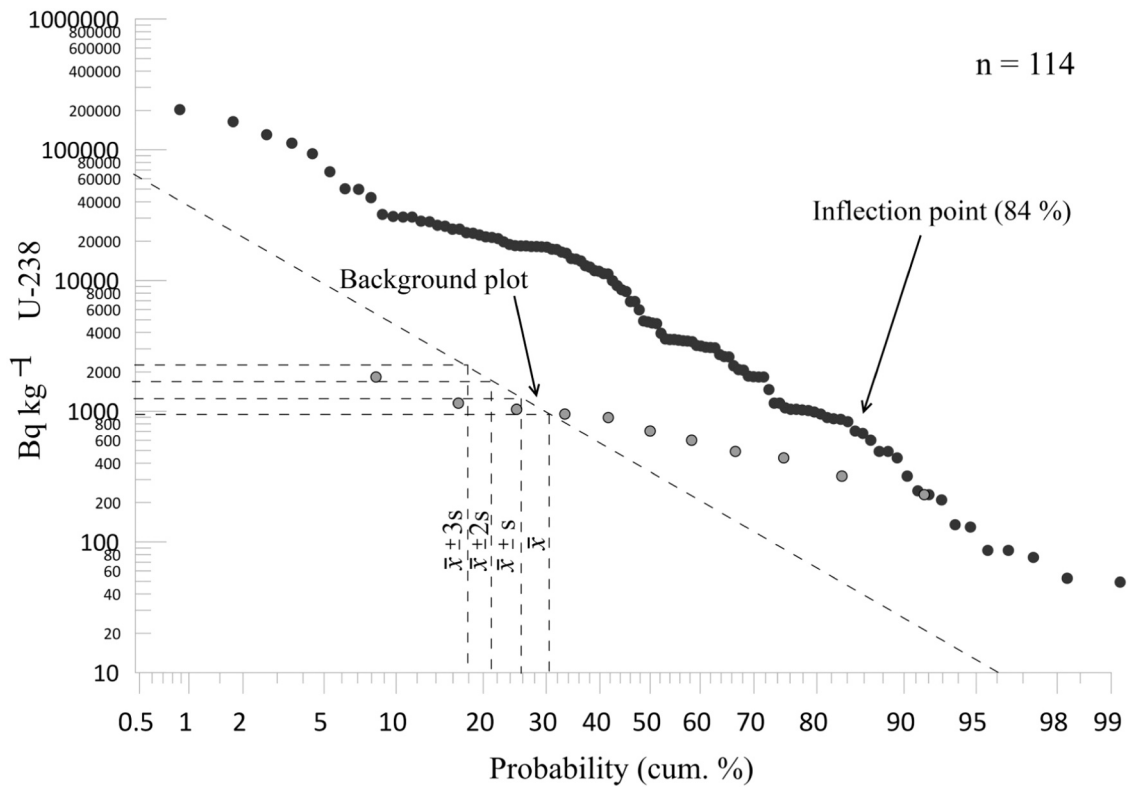


Fig. 3. Threshold value of <sup>238</sup>U radioactivity in *amang* samples.

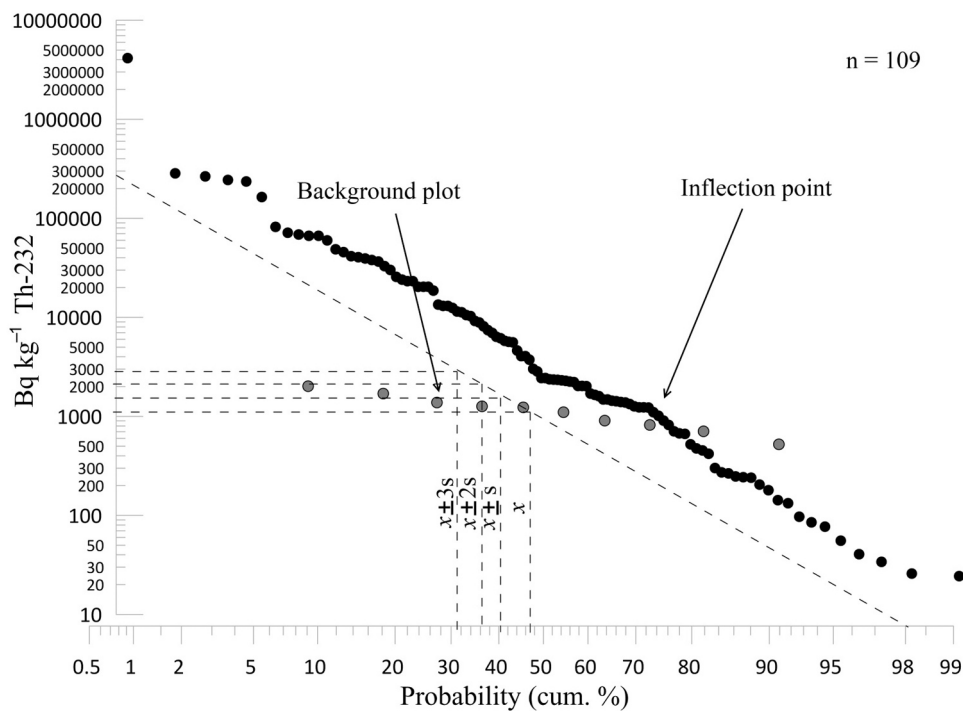


Fig. 4. Threshold value of <sup>232</sup>Th radioactivity in *amang* samples.

<sup>232</sup>Th levels elevated than 2500 Bq kg<sup>-1</sup> and 2030 Bq kg<sup>-1</sup> respectively can be considered as TENORM.

Table 1 shows the tabulation of the radioactivity content of <sup>238</sup>U and <sup>232</sup>Th as well as <sup>40</sup>K in *amang* concentrated products in Peninsular Malaysia based on numerous studies conducted by local and foreign investigators. To assess the central tendency value of the associated

radionuclides and other statistical parameters, Tables 2 and 3 presents the descriptive statistical results from SPSS analysis. Regardless of the type of *amang* final product the mean radioactivity levels of <sup>238</sup>U and <sup>232</sup>Th in the mineral amounted to 16,000 Bq kg<sup>-1</sup> and 22,000 Bq kg<sup>-1</sup>, respectively. The values are 150 times higher than the mean background radioactivity levels of <sup>238</sup>U (~ 100 Bq kg<sup>-1</sup>) and <sup>232</sup>Th (200 Bq kg<sup>-1</sup>) in

**Table 4**  
External gamma absorbed dose measured in *amang* processing plant.

State/location	Mineral/site/size	Instrument/technique	Gamma ( $\gamma$ ) dose rate nGy h <sup>-1</sup>	Reference
Bukit Merah	<i>amang</i> piles (along the road)	car-borne NaI (TI) scintillation det.	1000–2000	Tajuddin et al. (1994)
N/A (various plant located at central part of west coast of peninsular)	monazite (stockpile in plant)	TLD dosimeter (1 year exposure)	3881 ± 342	Hu and Kandaiya (1985a)
	monazite (storeroom)	Bircon NaI (TI) dose rate meter	>10,000	Hu et al. (1984)
	xenotime (storeroom)		40,000	
	zircon (storeroom)		26,000	
	tin ore (storeroom)		15,000	
	ilmenite (piling in workspace)		10,000	
	tin tailing (outside plant)		5000	
	struvilite and rutile (outside)		3000	
	pyrite (stockpile outside plant)		1000	
	8 locations in Peninsular Malaysia	office areas	Bircon NaI (TI) dose rate meter	300 – 3000
monazite			>400,000	
wet concentrated <i>amang</i> general plant			600–5000	
<i>amang</i> stockpiles			500– 20,000	
ilmenite stockpiles			1200–8000	
zircon stockpiles			1800–7000	
monazite stockpiles			4–15,000	
cassiterite stockpiles			35,000– 180,000	
struverite stockpiles			1600–14,000	
Access tracks stockpiles			8000–35000	
site boundary stockpiles			200–10,000	
tin tailing stockpiles			200–4000	
ex-mining pond bed			300–2000	
plant ambient dose rate		NaI (TI) scintillation detector TLD 100 (LiF:Dy)/ 200 (CaF <sub>2</sub> :Dy)	1400–1600 1566	Mohd Nazri (2001) Bahari et al. (2001)
Kinta Valley	ambient dose rate in quarters		617	
	external $\gamma$ dose rate	NaI(TI) Scintrex BGS-4	100–10,700	Omar et al. (2008)
	external $\gamma$ dose rate		700–18,700	
	external $\gamma$ dose rate		400–16,500	
	external $\gamma$ dose rate		1200–10,200	
	external $\gamma$ dose rate		600–56,000	
	external $\gamma$ dose rate		400–3800	
16 <i>amang</i> plants in Peninsular Malaysia	external radiation	TLD (LiF:Dy) 1 month exposure	420–3500	Omar et al. (2007)
	external radiation	TLD(LiF:Dy) 1 month exposure	514–5600	
	tin smelting plant	NaI (TI) scintillation detector	440–800	Omar et al. (2006)
West coast peninsular	zircon, monazite and ilmenite	car-borne NaI (TI) detection (inside)	250–1560	
	huge dump alongside of road	car-borne NaI (TI) detection (outside)	1980	
	mineral stockpiles	car-borne NaI (TI) detection (inside)	2670	
	tin slag stockpiles	car-borne NaI (TI) detection (inside)	323–960	
	contaminated soil	car-borne NaI (TI) detection (inside)	240–312	
Kinta Valley	17 <i>amang</i> plants (stockpiles)	Ludlum 19 NaI (TI) scintillator	260–50,000	Ramli (2007)
Bukit Merah	$\gamma$ dose rate in <i>amang</i> plant	N/A	8571	Thoste (1994)
	$\gamma$ dose rate in <i>amang</i> plant	radioactive waste storage building	260–457	Ramli (2007)
	$\gamma$ dose rate in <i>amang</i> plant	outside of radioactive waste building	195	
29 <i>amang</i> plants	magnetic separators	TLD (LiF:Dy) 1 month exposure	1143–41,257 (9086)	AELB (1991)
	high tension separators		1371–53,314 (11,657)	
	air table		269–28,063 (8514)	
	dryer		1714–39,503 (7886)	
	conveyor		1526–27,429	
	shaking tables		954–27,429 (6971)	
	lanchute		52,286–4229 (2000)	
	ilmenite dump		1371	

(continued on next page)

Table 4 (continued)

State/location	Mineral/site/size	Instrument/technique	Gamma ( $\gamma$ ) dose rate nGy h <sup>-1</sup>	Reference
	amang dump		1177	
	mineral dump		2577–3411	
	monazite dump		33,017	
	zircon dump		17,526	
	mineral storeroom		1086–12,400 (6869)	
	monazite store		28,971–69943 (55,143)	
	zirconium store		13,714	
	ilmenite store		1903	
	office inside of plant		886–27,429 (7097)	
	quarters		1331	
	guard house		2857	
	plant entrance		1846	
	monazite plant		4800	
	zircon plant		27,429	
	Tin smelting plant		4114	
Pulau Pinang	tin slag ex-dumping (12 sites)	NaI (TI) scintillation detector	914–3714	Lin (2004)
Dengkil, Selangor	12 <i>amang</i> upgrading plants	TLD 100 H (5 month)	549–617	Jais (2002)
		TLD 100 H (3 month)	279–1502	
			564	
Dengkil, Selangor	<i>amang</i> upgrading plant	TLD 100 H 3 month, 3050 hr y <sup>-1</sup>	2960 ± 691	
Not stated	monazite, xenotimes, zircon	Portables dose rate meter	25,000	Hewson (1996)
12 <i>amang</i> plants in Perak	stockpiles of raw <i>amang</i>	GM survey meter Victoreen 491	877–6139	Choong and Hong (1985)
	inside office		526–6578	
	inside laboratory		701–7893	
	plant $\gamma$ dose rate in <i>amang</i> plant		526–6578	
	monazite/xenotime areas		4385–6578	
	zircon working areas		2192–10,524	
	cassiterite working areas		789–2631	
	monazite storage (95% conc.)		35,080–140,320	
	monazite storage (95% conc.)		61,390–228,020	
	monazite (30 tonnes)		157,860	
	monazite (1000 tonnes)		263,100	
	xenotime (20 tonnes)		118,395	
	xenotime (20 tonnes)		219,250	
	zircon (2000 tonnes)		13,155	
	struverite (10 tonnes)		5262	
	ilmenite (500)		2193	
	cassiterite (10)		877	
	(30% of monazite/xenotime)		122,780	
	(30% of monazite/xenotime)		68,406	
	raw zircon (		29,818	
	raw <i>amang</i> (400 – 6000 tonnes)		10,524	
	pyrite (120 tonnes)		1316	
	tantalum-rutile (100 tonnes)		3947	
West Peninsular	<i>amang</i> mineral	Bircon NaI (TI) dose rate meter	1000–8000	Udompornwirat (1993)
	<i>amang</i> mineral		35,000	
	<i>amang</i> plant		7500	
	wet separation area		1000–2000	
	dry section		700–5000	
N/A	outside <i>amang</i> plant		10,000	
	monazite store		70,000	
	processing areas		25,000	
Gopeng	<i>amang</i> pile at tin mining	Exploranium GR – 101 Scintillator	200–6080	Udompornwirat (1991) Roberts (1995)
	layered banks in tailing area		240	
	track near <i>amang</i> piles		160	
	old tin tailing		120–240	
	clay near pond		280	
	track near pond		120	
	tin tailing		40–400	
	collecting drums		160	
	various soil samples		40–240	

(continued on next page)

Table 4 (continued)

State/location	Mineral/site/size	Instrument/technique	Gamma ( $\gamma$ ) dose rate nGy h <sup>-1</sup>	Reference
Kampar	coarse tin tailing		120	
	fine particulate tin tailing		120	
Bidor	old tin tailing		200	
	by-product waste		240	
Simpang Pulai	track near above waste		200	
	old tin tailing		160	
Gunung Sepah	track of tailing		120	
	margin of <i>amang</i> piles		17,920	
Kota Baru	top of <i>amang</i> piles		1840	
	tin tailing		120	
	concentrate bottom of <i>palong</i>		920	
	old <i>amang</i> storage area		680	
Batu Gajah	tin tailing, beyond <i>palong</i>		80	
	gravel over soil		2600	
	weathered/ coarse tin tailing		80–120	
	rotted lateritic soil		160	
Lahat	verge outside plant		8480	
	across road outside plant		240	
	outside plant gate		2080–2320	
	runnel across road		2640	
	plant gate		12,240	
	<i>amang</i> piles		5640	
Jalan Pengkalan	<i>amang</i> pond drainage		2360	
	tin tailing		160	
	clay		280	
	<i>palong</i>		80–200	
Pantai, Selangor	tin tailing		160	
Air Hitam	tin tailing		40–80	
Puchong	tin tailing		40–200	

Parenthesis value = average; *Palong* = wet separator.

superficial soil in Peninsula Malaysia. The mean isotopic abundance of <sup>238</sup>U and <sup>232</sup>Th in *amang* products is ~16,000 Bq kg<sup>-1</sup> and ~22,000 Bq kg<sup>-1</sup>, respectively.

In uranium and thorium exploration, a conventional geotechnics plot of the element of interest based on cumulative probability plots known as Sinclair's plot (Sinclair, 1974) is commonly used to find the threshold levels between background contents and content of highly mineral bearing resources. Specifically, the aim of determining the threshold values is to find a lowest activity level that representing the *amang* mineral. Figs. 3 and 4 shows a cumulative probability plot of <sup>238</sup>U and <sup>232</sup>Th activities in *amang* products. The *black* color plots indicated all 114 data collected from this study whereas *gray* plots indicating a reduced plot of data from the point of inflection to the lowest point. The motivation of representing the *gray* plots is to make graphical comparison between <sup>238</sup>U and <sup>232</sup>Th background plots with all collected data. The determination of the threshold levels was based on the inflection points or skewed trend of *black* plots which showing a more disperse and less connected plot trend. For <sup>238</sup>U, the plots indicated an average threshold value ( $\bar{x}$ ) equivalently to 950 Bq kg<sup>-1</sup> with a range ( $\bar{x} \pm s$ ) of 1250–650 Bq kg<sup>-1</sup>, whereas for <sup>232</sup>Th the  $\bar{x}$  value is 1100 Bq kg<sup>-1</sup> with a range ( $\bar{x} \pm s$ ) of 1500–500 Bq kg<sup>-1</sup>. In other word, about 68.2% of radioactivity measurements taken in any *amang* samples will give an average value between 650 and 1250 Bq kg<sup>-1</sup> and 1500–500 Bq kg<sup>-1</sup> for <sup>238</sup>U and <sup>232</sup>Th, respectively.

Up to date, the IAEA widely employed a reference of 2500 Bq kg<sup>-1</sup> (<sup>238</sup>U) and 2030 Bq kg<sup>-1</sup> (<sup>232</sup>Th) levels for worldwide TENORM classification. This value could be a slightly higher reference for other countries with lower radioactivity levels associated with their mineral industries. This will be a loophole and inadequacy in law for regulatory body and enforcement in Malaysia to govern and supervise uncontrolled *amang* processing activities as the calculated values in Malaysia (650–1250 Bq kg<sup>-1</sup> and 1500–500 Bq kg<sup>-1</sup>) is half of IAEA reference values. Therefore, the threshold values estimated in this study is important for guideline revision or for enacting a new act related to

TENORM, owing to large assessed data that representing heavy mineral processing industries in Peninsular Malaysia.

Depending on the type of end product, some of the mineral compound could associate with the extremely high content of <sup>238</sup>U and <sup>232</sup>Th decay nuclides up to ~ 60,000 Bq kg<sup>-1</sup>, for instance monazite. One of the notable finding was recorded by Hewson (1996), where the <sup>232</sup>Th content of 50,000 ppm was spotted in local monazite piles. With the highest relative abundance (few tenth %) in the earth's crust and identical ionic size of the impurity ion of the, lanthanum (La) and cerium phosphate (CePO<sub>4</sub>) in the monazite lattice molecule can easily blend and consolidate with the ion (AELB, 1991).

In contrast to monazite, ilmenite is scarcely produced as a radioactive mineral. In some case in the Peninsula, the <sup>238</sup>U content in the mineral could be as low as 4 ppm (Meor Sulaiman, 1988). However, contaminations of U and Th impurities due to their internal binding with the host ions could prompt ilmenite to a highly radioactive mineral (Omar et al., 2007; Hewson, 1996; Hu et al., 1984) with a substantial content of <sup>238</sup>U and <sup>232</sup>Th decay nuclides up to 290 ppm and 575 ppm, respectively (Eng, 2004). Another possibility of radionuclides association in *amang* final products is through adsorptions of U and Th into the defect lattice surfaces of host and direct cation exchanges between them (Meor Sulaiman and Muslimin, 2010). For instance, the content of <sup>238</sup>U and <sup>232</sup>Th in some sample of zircon stockpiles could be as low as 6 ppm (Meor Sulaiman, 1988) spanning to maximum 5,000 ppm (SEATRAD, 1991) probably due to defect lattice surfaces of host and direct cation exchanges.

Unperfected conventional magnetic and physical separation process of the *amang* also results in a high degree of radioactive contamination of *amang* product (Udompornwirat, 1993) along with the factor of trade demands (Omar et al., 2007) since some of the industrial purchaser favored impurified or low grade of *amang* products (contaminated mineral) to cut off the processing rates.

**Table 5**  
Measurements of total airborne alpha activity, equilibrium equivalent concentration (EEC) of <sup>222</sup>Rn and <sup>220</sup>Rn progenies in *amang* processing plants in Malaysia.

Location	Type of sample/ Site	Technique/ detector	Specific activity concentration (Bq m <sup>-3</sup> )			Reference
			Alpha (α) emitter	EEC <sup>222</sup> Rn progenies	EEC <sup>220</sup> Rn progenies	
West Peninsular	mineral separator	Alpha dosemeters silicon surface barrier det.	–	12.13	4.58	Omar et al. (2007)
			15.59	2.42		
			34.65	2.29		
			41.58	3.95		
			55.44	3.31		
			31.19	5.35		
			25.99	5.86		
			20.79	3.82		
			32.92	3.69		
			43.32	10.06		
			24.26	4.58		
			31.19	5.09		
			25.99	3.31		
			25.99	3.06		
24.26	6.36					
45.05	10.18					
12.13	4.58					
N/A	monazite	Solid state nuclear track detector (on the mineral stockpile)	86,580.00 ± 9583.00	–	–	Hu and Kandaiya (1985a, 198b)
			177,600.00 ± 21,238.00	–	–	
N/A	thorium waste xenotime zircon	.	57,350.00 ± 9768.00	–	–	Hu and Kandaiya (1985a)
			4440.00 ± 1443.00	–	–	
N/A	monazite work area storeroom	two-filter methods (grab sampling)	–	1147.00 ± 814.00	4810.00 ± 2960.00	Hu and Koo (1983)
		two-filter methods (grab sampling)	–	1073.00 ± 518.00	3108.00 ± 2368.00	
		modified Kusnetz and Tsivoglou method	–	60.64	–	Hu and Koo (1983)
		modified Kusnetz and Tsivoglou method	0.18	–	–	
N/A	<i>amang</i> plant	glass fibers filters papers	0.10	–	–	Hewson (1996)
		airborne dust sampling	0.25	–	–	
		gravimetric mass determination	0.27	–	–	
Bukit Merah	monazite store area		1.52	–	–	Hu and Koo (1981)
			3.74	–	–	
N/A	monazite storeroom		–	–	–	Hu and Koo (1983)
			–	–	–	
Ipoh	struvilite		N/D	–	–	Hu and Koo (1983)
			2.33	–	–	
Tronoh	monazite storeroom	air sampling (port. alpha scintillation)	4.40	–	–	Hu and Koo (1983)
			–	–	–	
Upgrading plant	monazite		N/D	–	–	Hu and Koo (1983)
			1.52–0.30	–	–	
Upgrading plant	<i>amang</i>		N/D–5.55	–	–	Hu and Koo (1983)
			–	–	–	
Upgrading plant	struvilite		1.04	–	–	Hu and Koo (1983)
			–	–	–	
Kg. Gajah	ex-mining site	diffused-junction photodiode sensor (on soil)	116.92 – 20.35	–	–	Hamzah et al. (2008)
			–	–	–	
Kinta Valley	monazite zircon	gross alpha 2π geometry gas flow proportional counter Tennelec Series 5 LB5500	956,746.00	–	–	Ramli (2007)
			80,159.80	–	–	
Kinta Valley	ilmenite	proportional counter Tennelec Series 5 LB5500	15,514.80	–	–	Ramli (2007)
			–	–	–	
Kinta Valley	cassiterite rutile	(measurement on stockpile)	5171.60	–	–	Ramli (2007)
			646,450.00	–	–	
Kinta Valley	tourmaline		1292.90	–	–	Ramli (2007)
			–	–	–	
Amang plant	work areas		0.23 ± 0.04	–	–	Hewson (1996)
			0.02 ± 0.02	–	–	
Amang plant	work areas		0.21 ± 0.03	–	–	Hewson (1996)
			0.13 ± 0.02	–	–	
Amang plant	monazite plant		–	–	–	Hewson (1996)
			–	–	–	
Amang plant	work areas	Tennelec TC257 alpha spectrometer	0.03 ± 0.01	–	–	Hewson (1996)
		Gilian HFS 513 A dust sampling pump	0.34 ± 0.05	–	–	
Amang plant	work areas		0.12 ± 0.02	–	–	Hewson (1996)
			6.80 ± 0.60	–	–	
Amang plant	work areas		0.12 ± 0.02	–	–	Hewson (1996)
			–	–	–	

(continued on next page)

Table 5 (continued)

Location	Type of sample/ Site	Technique/ detector	Specific activity concentration (Bq m <sup>-3</sup> )			Reference
			Alpha ( $\alpha$ ) emitter	EEC <sup>222</sup> Rn progenies	EEC <sup>220</sup> Rn progenies	
(12 stations)	work areas tin slag ex dump site	Radon Thoron Daughter Monitor (Tracerlab)	0.29 ± 0.04	–	–	Lin (2004)
			–	10.40 – 310.14	0.36–13.47	
Dengkil, Selangor	12 <i>amang</i> plants	WLX (RTD), Pylon Electronic Inc. Canada	–	17.33 – 296.28	–	Jais (2002)
2 <i>amang</i> plants		WLX (RTD), Pylon Electronic Inc. Canada	–	40.30 – 583.90	–	Jais (2002)
West Peninsular	processing areas	Tennelec TC257 alpha spectrometer	240,000.00	69.31	3.82	Udompornwirat (1991)
	monazite store	Gilian HFS 513 A dust sampling pump	–	51.98	3.82	Udompornwirat (1993)
			8000.00–1900,000.00	155.94	1.02–11.46	
<i>Amang</i> plant at	monazite store		–	277.22	89.11	Roberts (1995)
Gopeng, Perak Mining areas	old tailing	Pylon AB-5 radon emanator	–	27,400–3 8100	12500.00–12700.00	
	old tailing	Hammer spike system	–	141,800.00–199,700.00	4900.00–15,100.00	
	old tailing	15 – 50 cm depth in soil	–	8900.00	2900.00	
	gravelly old tailing	direct measurement on sample	–	47,300.00	17,400.00	
	<i>amang</i>			8500.00	76,500.00	
	<i>amang</i> piles			35,900.00	161,700.00	
Gunung Sepah Kota Baru	old tailing		–	36,500.00	1300.00	
	<i>amang</i> piles		–	49,900.00–106,400.00	89,800.00–12,000.00	
	old tailing			21,700.00	5200.00	
Batu Gajah	gravel over soil			2014.00	58,500.00	
Lahat	verge outside plant			9600.00	34,200.00	
	across road			21,600.00	15,800.00	
Pantai Puchong	old tailing	Pylon AB-5 radon emanator	–	2300.00	29,00–41.00	Roberts (1995)
29 <i>amang</i> plants	old tailing			16,500.00	700.00	
					–	
located in West Peninsular	magnetic separators	Roken TR personal air sampleR PS-4	0.02–2.89	58.04	5.17	AELB (1991); Sulaiman et al. (1994)
	high tension	Tennelec LB 5100 low alpha-beta counter	0.03–0.78	52.67	5.02	
	office	RDA-200 monitoring system	0.09–0.21	30.94	8.68	
	air table	Alpha dosimeters model 550/560	0.01–0.24	65.04	4.26	
	shaking table	(in-situ air sampling)	0.02–3.79	27.03	2.35	
	dryer		0.10–0.92	38.81	8.20	
	lanchute		0.18	34.65	3.42	
	monazite store		0.18–0.26	103.61	32.54	
	general dump		0.02–0.18	34.31	2.43	
	<i>amang</i> dump		0.13	51.46	1.35	
	zirconium store		0.10	104.30	1.35	
	general store		–	24.08	2.28	
	quarters		0.02	17.19	2.43	
	monazite plant		4.88	34.36	34.34	
	zirconium plant		0.21	17.18	3.79	
	zirconium dump2960		0.19	87.13	6.49	
	conveyor		0.17	34.36	1.35	
	tin plant		0.99	17.19	1.35	
			–	–	6.49	
			–	34.36	5.14	

## 6. Ionizing radiation exposures from *amang*

In radioactive decay, in order to achieve isobar stability, naturally occurring radionuclides in <sup>238</sup>U and <sup>232</sup>Th decay series including other radionuclides i.e. K<sup>40</sup>, <sup>87</sup>Rb, <sup>138</sup>La, <sup>147</sup>Sm dan <sup>176</sup>Lu spontaneously emitted an amount of energy in forms of radiation or particle i.e., gamma radiation ( $\gamma$ ) and alpha ( $\alpha$ ) and beta ( $\beta$ ) particles. Depend on energy magnitude and radioactivity strength of the source, indirect or direct interaction between these radiations and matter (living tissue) results in energy deposition which would cause a detrimental effect on

human tissues by chemical and biological changes on human cell structure, beside of increases productivity of free ion and radical via excitation or sub-excitation.

Workers in the industry are the most affected group of radiation exposures. Few conducted investigations by Hu and Koo (1983), Hu and Kandaiya (1985a), Choong and Hong (1985), AELB, 1991; Bahari et al., 2001, 2007; Hewson, 1996; Kandaiya et al., 1987; Mohsen et al., 2007; Omar et al., 2006, 2007, 2008; SEATRAD, 1991; Ramli, 2007; Udompornwirat, 1991; Udompornwirat, 1993 have identified 3 predominant routes and pathways of radiation exposures namely; (1) external  $\gamma$

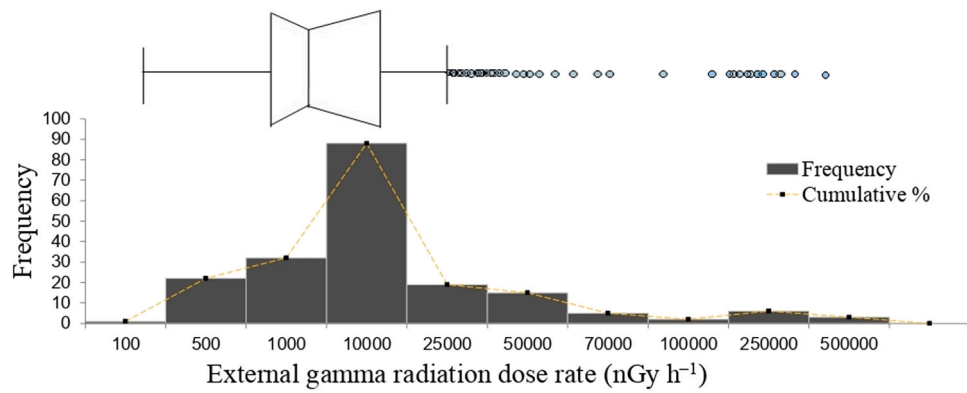


Fig. 5. Notched boxplot and histogram of absorbed gamma dose rate measured in *amang* plant.

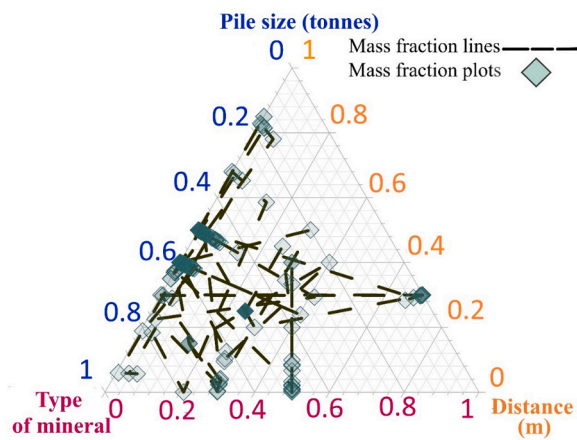


Fig. 6. Ternary plot of normalized absorbed gamma dose rate in *amang* plant.

radiation from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series, (2) internal radiation exposure to  $\alpha$  and  $\beta$  particles from inhalation of radon ( $\text{Rn}^{222}$ ) and thoron ( $\text{Rn}^{220}$ ) and their progenies, and (3) internal exposures to  $\gamma$  radiation,  $\alpha$  and  $\beta$  particles as a result of unavoidable ingestion and inhalation of airborne radioactive dust particularly from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series. The second and latter route howbeit only considered internal exposure from inhalation of  $\alpha$  emitting nuclides due to impracticable measurement of internal  $\gamma$  radiation and complex intake rate for ingested nuclides as well as for taking consideration of insignificant contributions of  $\beta$  particles compared to highly destructive  $\alpha$  particles.

As tabulated in Tables 4 and 5 numerous radiation surveys of external  $\gamma$  radiation and radiometric monitoring of airborne  $\alpha$  emitters, and EEC of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies were conducted by local and foreign researchers in Peninsular Malaysia. Fig. 5 graphically illustrated the distribution of measured absorbed  $\gamma$  dose rate in nearly 200 spots in the *amang* processing plants in Peninsular Malaysia. In most of the plant,

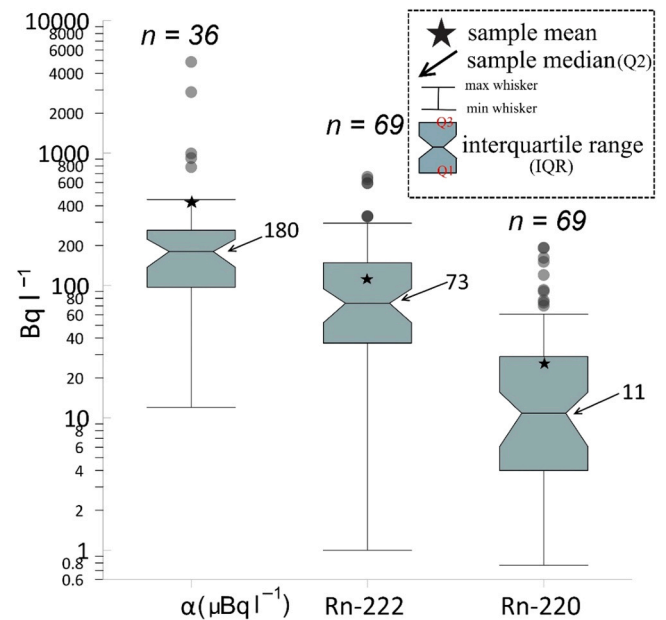


Fig. 7. Boxplot of concentration (given in  $\mu\text{Bq l}^{-1}$ ) for airborne alpha emitters and EEC of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  in air in *amang* plant (in  $\text{Bq l}^{-1}$ ).

high dose range between 5000–17,500  $\text{nGy h}^{-1}$  is a prevalent range in the *amang* industry. This is 50–175 times higher than the mean background level. Statistically, based on the interquartile range (IQR) plots in Fig. 5 approximately 100  $\gamma$  measurements (50% of the data) in the plants indicated high dose range of 900 (25th percentile) to 13,000  $\text{nGy h}^{-1}$  (75th percentile). The outlier points show anomalous extreme cases of high  $\gamma$  dose beyond 25000  $\text{nGy h}^{-1}$  probably due to highly concentrated  $^{238}\text{U}$  and  $^{232}\text{Th}$  in final products or nearby monazite storeroom and surrounded by huge stacks of the mineral. The external  $\gamma$

Table 6

Descriptive statistic of total airborne alpha activity, equilibrium equivalent concentration (EEC) of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies in *amang* processing plants in Malaysia.

Statistical parameters	Absorbed $\gamma$ dose rate ( $\text{nGy h}^{-1}$ )	Airborne $\alpha$ -emitters ( $\text{Bq m}^{-3}$ )	EEC $^{222}\text{Rn}$ progenies	EEC $^{220}\text{Rn}$ progenies
Total data	218	36	69	69
<sup>a</sup> n	193	46	46	46
Mean $\pm$ standard error	18,892 $\pm$ 3619	1.00 $\pm$ 0.25	116.88 $\pm$ 35.06	179.60 $\pm$ 123.01
Confidence level (95%)	11,755–26,029	0.50–1.50	46.26–187.50	68.15–427.36
Median	2430	0.21	36.73	4.42
Standard deviation	50,271	1.67	237.82	834.30
Kurtosis	26	3.55	12.78	24.738
Skewness	5	2.09	3.57	4.967
Range	100–228,020	0.01–6.80	10.40–1147.00	0.36–4810.00

<sup>a</sup> Data of direct measurement on mineral sample/ stockpiles were excluded from statistical analysis.

**Table 7**

Estimation of effective doses for different cases and situation due to external  $\gamma$  radiation and internal  $\alpha$  exposure of airborne  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series, and equilibrium equivalent concentration (EEC) of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies in *amang* processing industries in Malaysia.

Case/situation of exposure by hour (h)	Total effective dose ( $\mu\text{Sv}$ )				
	External $\gamma$ radiation	$\alpha$ emitting radionuclides <sup>a</sup>	EEC $^{222}\text{Rn}$ progenies	EEC $^{220}\text{Rn}$ progenies	Total
Full-time 8 h	105.79	150.48	186.74	17.45	460.46
Half-time 4 h	52.89	75.24	93.37	8.73	230.23
Minimum exposure 1 h	13.22	18.81	23.34	2.18	57.55
One-minute standing	0.22	0.31	0.39	0.04	0.96
Few second walking across	~0.02	~0.01	~0.01	~0	0.04
<sup>b</sup> Full-time 8 h (range)	0.56–2240.00	3.80–1546.93	1.47–970.22	0.58–116.15	6.41–4873.30

Effective dose conversion factor  $\alpha$  emitting radionuclides for  $^{238}\text{U}$  is  $5.7 \times 10^{-6}$  Sv Bq $^{-1}$  and  $^{232}\text{Th}$  is  $1.2 \times 10^{-5}$  Sv Bq $^{-1}$  (ICRP 137, 2017); for radon 11.23 mSv per WLM whereas 1 WLM =  $6.37 \times 10^5$  Bq h m $^{-3}$ ; for thoron, 3.23 mSv per WLM whereas 1 WLM =  $4.68 \times 10^4$  Bq h m $^{-3}$ ; considering 2000 h y $^{-1}$  at work (ICRP 115, 2010).

<sup>a</sup> Averaged value of dose contributions from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series.

<sup>b</sup> Based on tabulated result in Table 6.

radiation exposure received by workers varies from one spot to another depends on size of mineral stockpile, a type of mineral product and distance from the source. Based on ternary plot of normalized measured  $\gamma$  dose rates in Fig. 6, type of final products and pile size dominated the chart compared to the distance of exposure. Distance of source to the exposed person is no longer significant as the source of exposure comes from everywhere inside the plants.

Table 6 shows the detailed values of descriptive statistics of absorbed  $\gamma$  dose rate in air, radioactivity levels of airborne  $\alpha$  emitters, EEC  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies measured in *amang* plant. The highest recorded  $\gamma$  absorbed dose rate in air in *amang* plant is  $\sim 200,000$  nGy h $^{-1}$ . The maximum value is attributed to highly radioactive from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series in high grade of monazite stockpiles. In some *amang* plants, there are few indications of low  $\gamma$  absorbed dose rate  $\sim 100$  nGy h $^{-1}$  which is comparable to the average background count of metamorphic and sedimentary geological background. The value probably measured in the absence of mineral piles or away off from the plant. In terms of radioactivity levels compared to other exposure route i.e., airborne  $\alpha$  emitters,  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies, it is fair to said that the radioactivity levels of  $^{238}\text{U}$  and  $^{232}\text{Th}$  that contributed to external  $\gamma$  radiation are 10 times higher compared other sources. Based on notched boxplots in Fig. 7., the exposure routes of airborne  $\alpha$  radionuclide emitters and  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies only averagely indicated moderate radioactivity levels in air with the range levels show almost compatible values in air. This is probably due to the open air layout of most *amang* plants and temperate wind speed of hot humid climate of the country. However, for extreme cases, in a closed room i.e, monazite storeroom, extremely high radioactivity levels of nearly 5000 Bq l $^{-1}$  for airborne long-lived radionuclide were detected, with few hundred Bq l $^{-1}$  of radon and thoron levels were measured in the air.

It is useful to interpret the radioactivity and  $\gamma$  absorbed dose in air to the dosimetric unit of effective dose for comparing risks and impact of different type of radiation exposures to different tissues in the biological system of humans. Table 7 shows estimated values of effective doses for different cases and situation due to multi routes of exposure from different radiation sources. All estimated values in Table 4 are calculated based on effective dose conversion factor from ICRP 74 (1996), ICRP 137 (2017) and ICRP 115 (2010). For exposure to airborne  $\alpha$  emitters;  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies; the calculations considered averaged updated effective dose conversion factors from (ICRP 115, 2010), for a Reference Worker with an average breathing rate of 1.2 m $^3$  h $^{-1}$  during an 8-h working day (ICRP 66, 1994) whereas for airborne  $\alpha$  radionuclide emitters, 5-mm AMAD aerosols of dry particulate and Type S deposited material were considered for an annual intake rate of 2400 m $^3$  y $^{-1}$  (ICRP 66, 1994). The effective dose conversion factors for all exposure routes are listed as footnotes in Table 7.

Averagely, the *amang* industry could consistently pose considerable  $\gamma$  radiation exposure of about nearly  $\sim 20,000$  nGy h $^{-1}$  to workers. This is 100 times higher than the average dose rate received by peoples' lives in

a granitic region with substantial levels of natural U and Th. As shown in Table 4, by assuming 8 h of worktime, a worker could possibly receive an average of 0.1 mSv per day and for extreme cases of highly concentrated NORM in stockpiles of xenotime and monazite, a worker could experience effective dose up to 2 mSv per day from external gamma radiation source alone. Even though the value is 1 per 500 of the ICRP 103 newly introduced 50 mSv of occupational dose limit for planned exposure situation, interferences of different exposure routes i.e., airborne long-lived  $\alpha$  particles and inhalation of  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies could promote higher total effective dose of  $\sim 5$  mSv for 8 h of work time per day, and 115 mSv per year (assuming ICRP annual working hour 2000 h y $^{-1}$ ). In fact, in Malaysia the worker in the *amang* processing plant is not a designated radiation worker like medical industries. Beside that the estimated values of effective dose for external  $\gamma$  radiation (Table 4) is based on conversion coefficient of 0.7 Sv Gy $^{-1}$  which is not the best estimate for conversion of air Kerma to effective dose, and only applicable for  $\gamma$  irradiation from the soil and not practicable for a truncated solid cone of mineral stockpile geometry. A huge stockpile of  $\gamma$  irradiation could possibly promote dose greater than the factor of 0.7 Sv Gy $^{-1}$ .

In addition, the total effective dose received by workers could be higher than the calculated values in Table 4, as internal exposure routes from ingested airborne of long-lived  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay nuclides are disregarded owing to unavailable data. It required complex quantification of intake rate (Bq m $^{-3}$ ) using bio-assay and sampling or screening sweats and fecal matters. With the substantial deliberation rate of 1572–21,664  $\mu\text{g m}^{-3}$  of large suspended particulates of 92–230  $\mu\text{m}$  from mineral separator machineries (Suki and Abidin, 1990), daily ingested radionuclides by worker are possible to give an increment of a few mSv to the total estimated effective dose.

The annual occupational effective dose estimated from this study (115 mSv y $^{-1}$ ) indicated compatible agreement with an effective dose estimated in tin mining industries in Nigeria that is 99.9 mSv y $^{-1}$  (Ibeanu, 2003). In fact the effective dose contribution from *amang* or tin-tailing product from Peninsular Malaysia is relatively low compared to other highly concentrated products like high-grade heavy minerals, for example Egyptian high-grade monazite and thorium cake which contributed about 633 and 498 mSv y $^{-1}$  of total effective dose (El-Afifi et al., 2017). India's monazite also indicated compatible range of annual effective dose with the study finding that is ranging from 2 to 230 mSv y $^{-1}$  (Pillai, 2007).

Based on estimated effective doses in Table 4, 41% of total effective dose received by a worker is dominated by internal exposure from radon inhalation, then followed by 32% of long-lived  $\alpha$  emitting radionuclide inhalation, 23% from external  $\gamma$  exposure and the lowest contribution is from thoron inhalation which is less than 1%. Even though the radioactivity level of external  $\gamma$  radiation is considerably higher than others, due to destructive ionizing energy of  $\alpha$  particles to internal biological tissues ( $\sim 1$ –9 MeV),  $\alpha$  exposures from inhalation of radon and long-

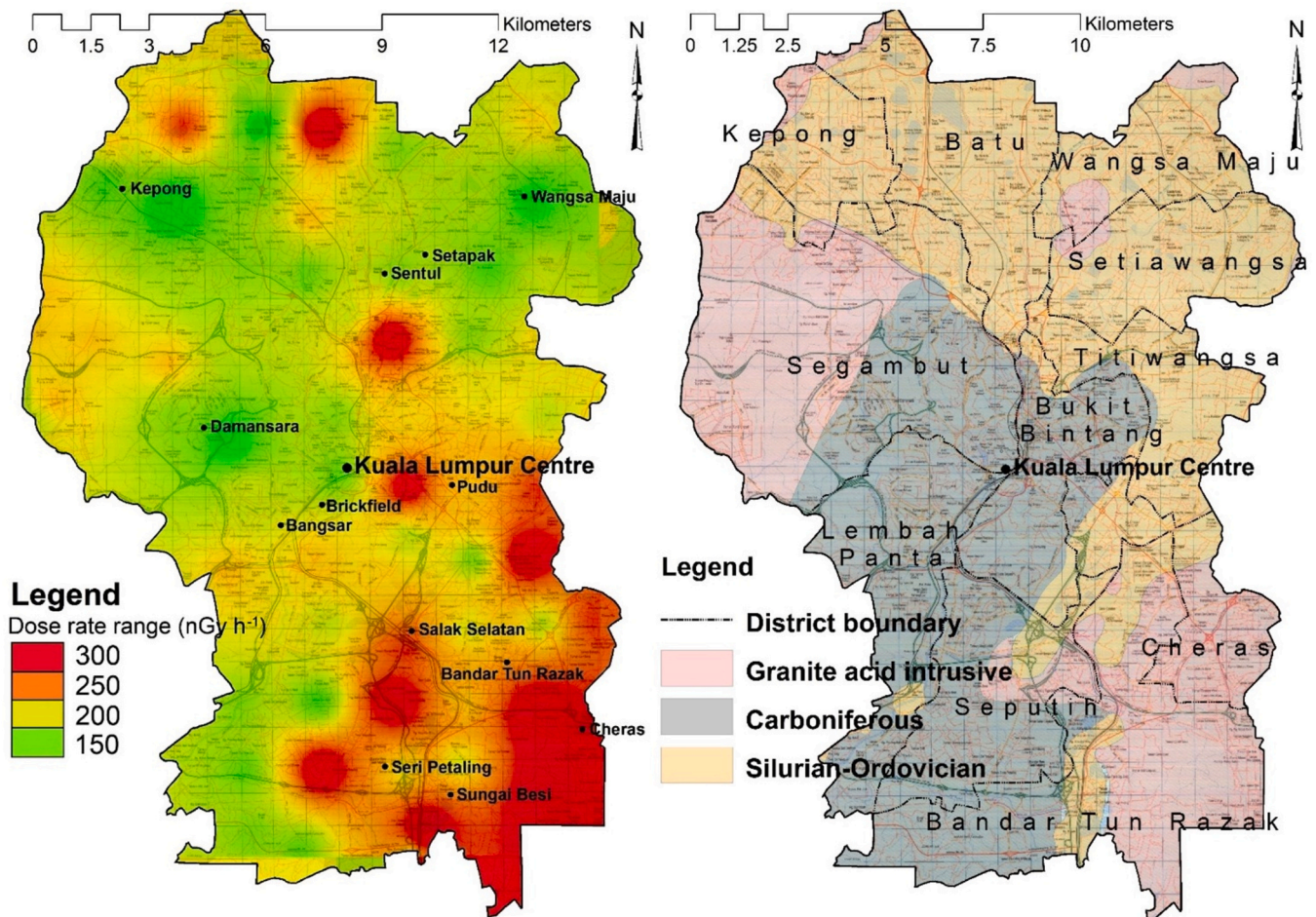


Fig. 8. Isodose map of measured terrestrial gamma radiation in Kuala Lumpur (formerly known as Lembah Klang) and geological map of the place.

lived radionuclides from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series are considered to be the main source of hazardous radiation exposures to workers (Sulaiman et al., 1994). The low magnitude of effective dose due to inhalation of thoron  $^{220}\text{Rn}$  progenies is mainly due to low  $\alpha$  particle energy of 0.8 MeV and extremely short exposure time  $\sim 55$  s compared to radon  $^{220}\text{Rn}$   $\sim 3.8$  days, with considerable  $\alpha$  energy of 5.5 MeV.

## 7. Radioactivity contamination to environment

The NORM contaminations to soil body due to rapid tin mining and *amang* processing in the past are publicly known. The situation is worsened with uncontrolled exploitation of rock aggregates, tin tailing sands and clays from the ex-tin mines and mineral processing for construction materials, road pavement and landfill material for infrastructural development in Kuala Lumpur and the nearby districts of Selangor (Hassan and Hamzah, 1996; Omar et al., 2006; Yap, 2007).

Previous study of isodose mapping by the corresponding author; Sanusi et al. (2017) found that the dose levels of background  $\gamma$  radiation in Kuala Lumpur (formerly also known as Lembah Klang; most productive areas for tin and *amang* processing industry) are technically enhanced by radioactivity contamination of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  from past tin-mining and recent rapid urbanization activities. As shown in Fig. 8, anomalous dose enhancements in certain places are shown by yellow, orange and red contours with  $\gamma$  dose range of 200–400 nGy h<sup>-1</sup>. In general, geological features of metamorphic and sedimentary from the Carboniferous and Silurian-Ordovician ages typically indicated low background radiation i.e., below 150 nGy h<sup>-1</sup> as they composed of low natural radioactivity level compared to acidic igneous granitic

geological regions. Based on the cumulative probability plot, ANOVA and *t*-test, the work statistically verified the enhancement of background radiation exposure in Kuala Lumpur due to TENORM with estimated increased dose rates of  $57 \pm 12$ ,  $74 \pm 8$  and  $134 \pm 11$  nGy h<sup>-1</sup> for granitic, Silurian and Carboniferous geological regions respectively. However, the study is limited to external gamma exposure only. It is believed that indoor radon exposure also prominent source in the study area. This would pose member of public to considerable  $\gamma$  radiation exposures mainly to radon inhalation due to past radioactivity contaminations in soil.

Numerous works of assessing and monitoring radioactivity levels in ambient air, terrestrial and water bodies as well as water, sediments are well documented by Dahan (1990), Abdul Rahman (1990), AELB (1991), SEATRAD (1991), Roberts (1995), Hu et al. (1995, 1996), Hewson (1996), Bahari et al. (2000, 2001, 2007), Yusof et al. (2001), Lim (2004), Eng (2004), Majid et al. (2007), Mohsen et al. (2007), Yasir et al. (2007, 2008) and Hamzah et al. (2008). Most of the work focused on radioactivity contamination levels in soil and industrial discharged pond rather than biota.

*Amang* pond is created as the *amang* processing required huge water supply (Lim, 2004; Majid et al., 2007; Mohsen et al., 2007) to extract interest mineral from host composite through physical separation to eliminate sand and sediment waste (Bahari et al., 2007; Yasir et al., 2007). Usually the water resources are obtained from natural sources such as local river or water spring (Yasir et al., 2007) and the industrial effluent wastes were discharged to man-made water reservoirs or ponds. The water from the pond are recycled (Mohsen et al., 2007; Bahari et al., 2007) for next uses for cost effective operation as well as to contain it

Table 8

Estimation of  $^{238}\text{U}$  and  $^{232}\text{Th}$  activity concentrated in biotas and their absorbed doses using ERICA program in 2 different ecosystems due to TENORM contamination.

Ecosystem/type of biotas	Radiological parameters					Screening value ( $\mu\text{Gy h}^{-1}$ )	Risk quotient
	Activity in organism ( $\text{Bq kg}^{-1}$ )		Absorbed dose rate ( $\mu\text{Gy h}^{-1}$ )				
	$^{238}\text{U}$	$^{232}\text{Th}$	Internal	External	Total		
<i>Terrestrial</i>							
Amphibian	1.778	0.105	0.045	7.67E-05	0.045	40.000	0.003
Bird	0.409	0.105	0.012	2.72E-05	0.012	40.000	0.001
Mollusc-gastropod	10.956	2.468	0.320	2.75E-05	0.320	40.000	0.024
Reptile	1.685	0.584	0.054	5.79E-05	0.054	40.000	0.004
Annelid	10.956	2.468	0.320	7.67E-05	0.320	40.000	0.024
Arthropod – detritivorous	3.374	1.364	0.112	8.26E-05	0.112	40.000	0.008
Flying insects	3.374	1.364	0.112	2.81E-05	0.112	40.000	0.008
Grasses and Herbs	41.561	43.040	1.987	6.21E-05	1.987	400.000	0.015
Lichen and Bryophytes	295.750	102.220	9.501	2.8E-05	9.501	400.000	0.071
Mammal – large	1.778	0.036	0.044	6.75E-06	0.044	40.000	0.003
Mammal – small-burrowing	1.778	0.036	0.044	6.48E-05	0.044	40.000	0.003
Shrub	19.825	16.409	0.853	2.65E-05	0.853	400.000	0.006
Tree	2.145	0.340	0.059	7.89E-06	0.059	400.000	0.000
<i>Aquatic</i>							
Amphibian	7812.547	75,682.441	1110.090	7.85E-06	1110.090	40.000	83.257
Benthic fish	4867.830	16,754.327	260.892	8.79E-05	260.893	400.000	1.957
Bird	3533.384	75,682.441	1089.440	3.31E-06	1089.440	40.000	81.708
Crustacean	13,442.387	14,031.807	263.693	5.42E-04	263.694	400.000	1.978
Insect larvae	13,442.387	58,759.659	897.469	1.05E-03	897.470	400.000	6.731
Mammal	7812.547	75,682.441	1110.090	2.48E-06	1110.090	40.000	83.257
Mollusc – bivalve	37,404.750	244,346.166	3642.787	1.73E-04	3642.787	400.000	27.321
Mollusc – gastropod	37,404.750	244,346.166	3642.787	2.92E-04	3642.787	400.000	27.321
Pelagic fish	4867.830	16,754.327	260.892	3.52E-06	260.892	400.000	1.957
Phytoplankton	4750.203	275,034.378	3929.731	2.39E-05	3929.731	400.000	29.473
Reptile	7812.547	24,168.687	381.255	9.40E-05	381.255	40.000	28.594
Vascular plant	24912.996	2,297,178.717	32,670.292	5.43E-04	32,670.292	400.000	245.027
Zooplankton	13442.387	12,627.451	243.794	2.26E-05	243.794	400.000	1.828

from ecological pollution. The practices technically would accumulate substantial radioactivity from  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay nuclides in the water bodies. As presented in Tables 2 and 3, the mean radioactivity levels of  $^{238}\text{U}$  decay nuclides in stagnant watery bodies discharged from *amang* processing are  $29 \text{ Bq l}^{-1}$  with maximum value could be as high as  $79 \text{ Bq l}^{-1}$ . The value is 14 times higher compared to average U radioactivity in surface water  $\sim 2 \text{ Bq l}^{-1}$  and 3–4 times higher than the underground water in Malaysia.

In contrast to  $^{238}\text{U}$ , radioactivity in  $^{232}\text{Th}$  decay nuclides shows low level with a mean of  $4 \text{ Bq l}^{-1}$  and maximum value of  $36 \text{ Bq l}^{-1}$ . Most of considerable radioactivity levels were detected in solid effluent and pond sediment samples with a mean of 325 and  $269 \text{ Bq kg}^{-1}$  for  $^{238}\text{U}$  and  $^{232}\text{Th}$ , respectively. The accumulation of radionuclides developed in the sediments over time as these heavy isotopes are insoluble to water pond. In the past time before *amang* was known for its favorable rare earth mineral or even now in tin-mining industry, it was reported that most of *amang* waste or gangue effluents mainly from tin processing and cracking treatment were dumped to surrounding environment whilst their wastewaters were channeled to the nearest local river. Not to mentioned large open-air stockpiles of *amang* which is abandoned and stacked outside the plant along the road (Dahan, 1990; Tajuddin et al., 1994; Udornwirat, 1991; Hewson, 1996; Omar et al., 2006, 2008), unsheltered to nature carriers such as wind and rain that inevitably could pollute close environment especially the river.

Hu et al. (1995) reported that long-term *amang* processing practices could acidify water bodies of close environment with the detected acidity of waste effluent up to pH 3. Simultaneously, works by Bahari et al., (2000, 2007) found that the practices have turned normal pond into acidic one (pH level of 3–4) with unappealing black-brownish stagnant water. In fact, local government has started their long-term initiatives to conserve the environment, sustainability and rehabilitation of ex-mining and dumping site by turning it into new townships, housing estates (Lin, 2004; Bahari et al., 2000, 2007), recreational park (Bahari et al., 2000; Mohd Nazri, 2001; Ang and Ho, 2004; Yap, 2007), freshwater fish farm (Roberts, 1995) and vegetable cultivation station

(Yusof et al., 2001; Mohd Nazri, 2001; Tsy, 2005; Yasir et al., 2007).

In order to assess the radiological risk to biota a radioecological assessment tool ERICA. It is a software developed by The ERICA Project (2004–2007) which specialized for environmental risks against ionizing radiation including screening and management). The project was conducted between by a collaboration between 15 organizations in from 7 European countries. The specific task includes doses estimation to organisms, site screening, identification of exposed organism group in 3 main different ecosystems: (1) marine, (2) aquatic and (3) terrestrial. In this study, Tier 2 assessment has been chosen to facilitate multiple series data exposure scenarios with 2 main isotopes selected i.e.,  $^{238}\text{U}$  and  $^{232}\text{Th}$  being released in two ecosystem i.e, aquatic and terrestrial which consisted of two different group biotas. Estimation of Tier 2 is based on comparison of calculated whole-body doses of individual reference organisms with Tier 1 screening dose rate, whereas Tier 1 dose screening formulated on a comparison basis between isotope activity in environmental media against Environmental Media Concentration Limits (EMCL). The risk estimated (quotient) is given by (Oughton et al., 2013):

$$RQ = \sum_{i=1}^n \frac{M_n}{EMCL_n}$$

where: RQ = total estimated risk quotient (dimensionless).  $M_n$  = measured isotope activity in environmental media; Unit for  $M_n$  = water ( $\text{Bq l}^{-1}$ ); soil or sediment ( $\text{Bq kg}^{-1}$ ); water ( $\text{Bq m}^{-3}$ );  $EMCL_n$  = Environmental Media Concentration Limit.

The value of  $EMCL_n$  utilized for radioecological assessment in this work is based on a value of  $40 \mu\text{Gy h}^{-1}$  for terrestrial animal, birds, amphibians and reptiles, and  $40 \mu\text{Gy h}^{-1}$  for plants and aquatic organism. According to the program, the estimated value below this threshold indicated no measurable population effect would occur due to radiation exposure released in reference ecosystem. The derived threshold values are taken from IAEA, USDOE and UNSCEAR. Other radioecological parameters considered in this assessment are an occupancy factor of biotas, media concentration ratio and radiation weighting factor.

The result of estimation of  $^{238}\text{U}$  and  $^{232}\text{Th}$  activity concentrated in biotas, their absorbed doses and risk quotient using ERICA are tabulated in Table 8. The radioecological risk RQ indicated high value ( $\sim 2\text{--}245$ ) for aquatic organisms due to their small mass of their biological system. Besides, their occupancy factor in an aquatic environment increases the exposure rate by increasing the isotopes accumulation factor or transfer rate. The ERICA program indicated “red alarm” for the exposure scenario and suggesting a continuous screening should be taken over the reference organisms. In contrast, the terrestrial ecosystem shows the least exposed environment system with most of RQ value less than 0.1 (ranging from 0 to 0.07). The main factor of low exposure of dose to this group is due to bigger mass and low occupancy factor to the contaminated media.

## 8. Conclusions

This review unveiled the significance potential of radiological threat from *amang* processing industry. The potential risks arise from excessive hazardous radiation exposure associated with highly radioactive levels in *amang* mineral. Few hotspots for example monazite storeroom and mineral separators have been identified to contribute high effective dose to workers. The study found out that averagely a worker is estimated to receive an average total effective dose of 0.5 mSv per day based on 8 h working time exposure and a maximum of 5 mSv per day for extreme exposure situations from all possible exposure routes. Averagely, it is estimated that an occupational exposure to the worker is equivalent to 115 mSv  $\text{y}^{-1}$  and exceeding 50 mSv annual dose limit recommended to designated radiation worker by ICRP. It is noteworthy to conclude that the level of effective dose experienced by the workers is within the high level dose exposure with imminent threat to the experience long term stochastic effects even though too small relatively to cause immediate danger of deterministic effects. In fact, an accumulation of low-level dose as a consequence from long haul of processing practices throughout worker life could triggered long-term effect of carcinogenesis. With current government motivation to revive tin mining and *amang* processing mineral, and loose establishment of regulations on radiological protection and radiation safety in mineral processing in Malaysia, it is necessary for the regulatory bodies to harmonize current acts or enacting new regulation and legislation for the industry. The exacerbating situation arises as *amang* worker is not required to be registered as a radiation worker. Prominent threat to the ecosystem has been spotted based on contaminated industrial effluent discharge to aquatic system as well sand residue from *amang* mineral processing. Radioecological risk assessment from this study revealed that the aquatic biotas are among the highly exposed group to radiation exposure from *amang* processing industries. A newly industrial warrant is required for important industrial operations for examples decommissioning, decontamination, and radioactive waste disposal especially when large volumes of waste are involved in order to sustain local environment form NORM contamination.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2020.111727.

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