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Radiological Impact Assessment of Natural Radionuclides and Heavy Metal Contamination in Industrial Tin-Tailing Processing Effluent

(Penilaian Impak Radiologi Radionuklid Tabii dan Pelumusan Logam Berat dalam Efluen Industri Pemprosesan Amang)

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ABSTRACT

This study investigates the radiological hazard and heavy metal contamination of water effluents from Malaysian tin-tailing processing plants. Samples were collected from retention ponds in seven separate tin tailing processing plants scattered throughout the state of Perak, Malaysia. Samples were analysed for radioactivity and heavy metal concentration using Gamma-ray Spectrometry System and Inductively Coupled Plasma-Mass Spectrometry, respectively. The analysis indicates that the concentration of Radium-226 (^{226}Ra), Radium-228 (^{228}Ra), and Potassium-40 (^{40}K) in samples ranged from 2.4 - 34.9 Bq/l, 0.8 - 14.7 Bq/l, and 19.5 - 299.4 Bq/l, respectively. These levels surpassed the control limits (5 Bq/l and 10 Bq/l for ^{226}Ra and ^{228}Ra) set by the regulatory authority. The analysis of the heavy metal contamination showed that the concentration of Arsenic (As) and Lead (Pb) were higher than the Maximum Concentration Level (MCL) of 0.01 and 0.015 mg/L, respectively. Further evaluation of radiological impact showed that the average Annual Effective Doses (AED) by water ingestion and AED for external exposure are 1.43±0.67 mSv/y and 1.71±0.79 mSv/y, respectively. While for non-carcinogenic and carcinogenic risk assessments, the value of hazard index and lifetime cancer risk is 2.1×10-10 and 1.2×10-7, respectively. These research findings suggest that effective treatment of the effluent should be implemented before discharge into the drainage system to prevent the accumulation of radionuclides and heavy metals in the environment, which may pose a risk to public health.

Keywords: Excess lifetime cancer risk; heavy metal; natural radionuclide; radiological impact assessment; tin-tailing

ABSTRAK

Kajian ini bertujuan menentukan bahaya radiologi dan pelumusan logam berat daripada efluen kilang pemprosesan amang. Sampel telah diambil daripada kolam tadahan di tujuh kilang pemprosesan amang di sekitar negeri Perak, Malaysia. Keradioaktifan dan kepekatan logam berat dalam sampel dianalisis masing-masing menggunakan Sistem Spektrometri Sinar-Gama dan Spektrometri Jisim Plasma Gandingan Teraruh. Hasil analisis mendapati kepekatan Radium-226 (^{226}Ra), Radium-228 (^{228}Ra) dan Kalium-40 (^{40}K) dalam sampel masing-masing adalah antara 2.4 – 34.9 Bq/l, 0.8 – 14.7 Bq/l, dan 19.5 – 299.4 Bq/l. Nilai kepekatan ini didapati melepasi nilai had kawalan (5 Bq/l dan 10 Bq/l bagi ^{226}Ra dan ^{228}Ra) seperti yang ditetapkan oleh pihak berkuasa. Analisis terhadap pelumusan logam berat mendapati kepekatan Arsenik (As) dan Plumbum (Pb) adalah lebih tinggi daripada Tahap Kepekatan Maksimum

(MCL) iaitu masing-masing 0.01 dan 0.015 mg/l. Penilaian impak radiologi selanjutnya mendapati bahawa purata Dos Berkesan Tahunan (AED) berpunca daripada pengambilan air efluen dan dedahan luaran masing-masing ialah 1.43±0.67 mSv/tahun dan 1.71±0.79 mSv/tahun. Manakala bagi penilaian risiko bukan karsinogen dan karsinogen nilai pengiraan indeks risiko dan penilaian risiko kanser sepanjang hayat (LCR) masing-masing bernilai 2.1×10-10 dan 1.2×10-7. Hasil kajian ini mencadangkan rawatan berkesan terhadap efluen mesti dilakukan sebelum dibuang ke sistem perparitan bagi mengelakkan pengumpulan radionuklid dan logam berat di alam sekitar yang boleh menjejaskan kesihatan awam.

Kata kunci: Amang; logam berat; penilaian impak radiologi; risiko kanser sepanjang hayat; radionuklid tabii

Introduction

Malaysia has long been known for its abundant natural resources, including fertile land and valuable minerals such as natural rubber and tin, which made it one of the world's leading producers of these materials in the 1970's. However, the 1980's saw a significant decline in the prices of major export raw materials, with tin facing an even more severe situation as export revenues plummeted from US$0.421 billion in 1985 to US$171.05 million in 1986 (Ahmad & Jones 2013). This prompted a shift in focus towards processing the tin by-product, known as tin tailings or amang (Ismail et al. 2003) as a means of recovering valuable rare earth elements (REE) from the minerals present in the tailings. As of 2016, Perak produced about 1,880 tonnes of REE reported by the Department of Mineral and Geoscience Malaysia (JMG 2016). To this end, the amang processing plant in Malaysia was exempt from the Atomic Energy Licencing Act of 1984 between 1994 and 2021, but this order was lifted in 2021 and the plant was relicensed. This move is expected to have significant implications for the extraction of REE and other valuable minerals from tin tailings in Malaysia, as it opens up new opportunities for research and development in this area.

The concentration of naturally occurring radionuclides and heavy metals in water is influenced by the earth's natural processes, however, amang processing activities can increase the concentration of these elements in the environment. Amang processing activities rely on three physical properties, namely, specific gravities, magnetic properties, and electrostatic properties. During the wet gravity separation process, a significant amount of water is required to physically separate the mineral mixture and obtain the desired REE. However, the effluent water generated during the process is usually released into a retention pond or directly into the environment. Although the wet gravity separation process is effective in separating minerals, the use of a large volume of water leads to the accumulation of radionuclides and heavy metals in the effluent water (Zaini, Nor Monica & Ahmad 2009). Therefore, this activity is a matter of concern due to the high contamination of metals (Ismail, Nasirian & Pauzi 2007; Rehman et al. 2018). Over the years, the pollution issue of naturally occurring radionuclides and heavy metals in tin tailings has become increasingly severe in Malaysia (Muhammad Abdullah et al. 2022).

Studies (Alnour et al. 2017; Hamzah, Mardhiansyah & Firdaus 2018; Nasirian et al. 2007) have shown that the concentration of these substances in amang water effluent exceeds the limit set by local regulatory bodies such as the Department of Atomic Energy (ATOM Malaysia), previously known as Atomic Energy Licencing Board (AELB) and the Department of Environment (DOE). Due to this, workers in the amang plant are at risk of external exposure due to the lack of personal protective equipment (PPE) and prolonged exposure to effluent water (Siti Khairunnisaq, Ho & Abd Khani 2018). Typically, the standard method for disposing of water effluent is to release it into a closed-cycle retention pond within each amang plant. However, if the retention pond is poorly constructed, there is a potential for contamination of nearby rivers via surface runoff or groundwater. This may result in an accumulation of dissolved radioactive materials in water sources, leading to potential long-term exposure and health risks for the local community (Dunca 2018; El-Gamal, Sefelnasr & Salaheldin 2019).

Exposure pathways such as external and internal exposure are among the risk to human health that is caused by naturally occurring radionuclides and heavy metals (Kanayochukwu, Ijeoma & Onyenezi 2019; Saha et al. 2017; Siti Khairunnisaq, Ho & Abd Khani 2018). To estimate the health risks posed to humans, many researchers have used the hazard index

and cancer risk indicators for non-carcinogenic and carcinogenic effects, respectively (Mohammadi et al. 2019; Muhammad Abdullah et al. 2021). This study aims to evaluate the concentration of NORM and selected heavy metals (Cu, Cr, Zn, Ni, Pb, As, and Cd) in effluent water from amang plants in Perak, Malaysia, to assess their radiological impact assessment, carcinogenic risk, and non-carcinogenic risk. The findings of this study can provide guidelines on NORM and heavy metal contamination in effluent water in amang plant.

Materials and Methods

Description of Study Area

This study focused on Perak, which is the main location for tin tailings processing in Peninsular Malaysia. Seven amang processing plants in Perak were selected as the study area, as depicted in Figure 1. These plants were chosen based on their processing productivity, with some plants like PP5 and PP7 having ceased processing activity but still storing tin tailings when required. The retention ponds varied across the processing plants as illustrated in Figure 2. Each processing plant had at least one retention pond, except for a few large plants like PP-5 (two ponds) and PP-6 (five ponds). This study collected and analyzed a total of 12 water samples. The study findings were compared to control samples from seven locations located between 1-3 kilometers away from each study site. Publicly accessible places such as police stations and public health clinics were selected as control sites. The results of the study could be used to develop guidelines and take preventive measures to reduce heavy metal contamination in the environment.

Sample Preparation

The water samples were prepared according to the standard sample preparation procedure mentioned in the Technical Report Series No. 295. At the study site, water samples were collected from the surface of the water at a depth of 10 cm using a water pump and stored in pre-cleaned plastic containers with a capacity of 1 liter. To prevent the adsorption of radionuclides to the container's wall, the effluent samples collected from the processing plants were immediately treated with 10 mL of hydrochloric acid per liter in sample bottles to a pH of 2 (Eli Syafiqah et al. 2021; IAEA 1989). The sample containers were tightly sealed with covers and transported to the laboratory for further preparation and analysis.

Gamma Spectrometry Analysis

Before gamma spectrometry analysis, each water sample is filtered and measured to 200 mL, then sealed in a 250 mL air-tight Marinelli counting bottle in triplicate. The use of triplicates is an internal standard plan to ensure the validity of the analytical results (IAEA 1989). The samples were stored for one month to attain secular equilibrium between ^{226}Ra and ^{228}Ra and their progenies.

Gamma-ray spectrometry with a high-purity germanium (HPGe) detector was used to determine the activity concentration of radionuclides in the samples. The detector was placed inside a cylindrical lead shield with a thickness of approximately 100 cm and coupled with a multichannel analyzer for data acquisition and gamma spectra analysis. The resulting spectra data were analyzed using CANBERRA's Genie software 2000. To ensure the accuracy of the gamma spectrometry system, a multinuclide standard source of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{137}Cs, ^{88}Y, ^{60}Co, and ^{88}Y (Eckert & Ziegler) was used for energy calibration. The absolute full-energy peak efficiency, ε, was calculated using Equation (1). The detection limit (DL) and minimum detectable activity (MDA) were calculated using Curie's derivation in in Equations (2) and (3), respectively, as reported by Abdullahi, Aznan Fazli and Supian (2019).

$$\varepsilon = \frac{N}{TAY} \tag{1}$$

$$DL = 2.71 + 4.66\sqrt{N_B} \tag{2}$$

$$MDA = \frac{DL}{T\ \varepsilon\ Y\ M} \tag{3}$$

where N is the net area count (count per second); T is the live time in seconds; A is the activity (Bq); Y is the branching ratio fraction; N_B is the background counts (count per second); and M is the mass of the sample (kg).

A certified reference material (CRM) IARMA-004 which contained the radionuclides of ^{60}Co, ^{57}Co, ^{133}Ba, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{210}Pb, and ^{241}Am of known concentration is used to determine the activity content of

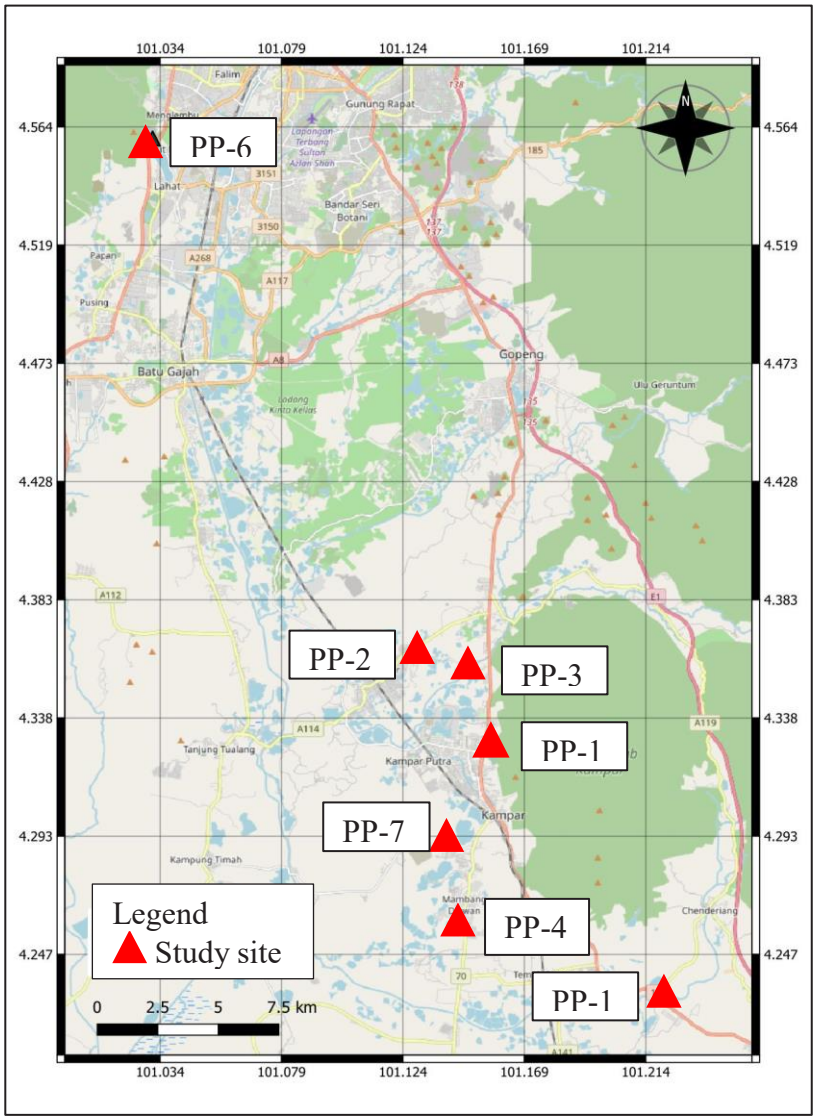

FIGURE 1. Study location of 7 amang processing plant in Perak

radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K in water samples. To calculate the efficiency of desired radionuclides in water samples, an efficiency against energy curve is generated using IARMA-004 (Khoirul Solehah et al. 2020). The efficiency of 1764 keV, 2615 keV, and 1461 keV energy peaks obtained are 0.003, 0.001, and 0.001, respectively. While, DL is within the range of 58.56 ± 9.1, 74.20 ± 11.68, and 92.29 ± 12.68 Bq/kg, respectively, and MDA was found to be within the range of 2.85 ± 0.45, 2.63 ± 0.37 and 25.10 ± 3.45 Bq/kg, respectively, for 1764 keV, 2615 keV and 1461 keV energy peaks.

After the efficiency is determined, energy peak of 351.83 keV (^{214}Pb), 911.32 keV (^{228}Ac), 1460.76 keV (^{40}K), 1764.56 keV (^{214}Bi), and 2614.4 (^{208}Tl) is used to calculate activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K using equation (4). Where N refers to the net count rate per second of -rays for 43200s, is the absolute efficiency of the detector; is the probability of gamma-ray emission and V is the sample volume of 200 mL.

$$A = \frac{N}{\varepsilon \times \theta \times V} \tag{4}$$

Heavy metals analysis

Inductively coupled plasma-mass spectrometry (ICP-MS, model ELAN 9000; PerkinElmer SCIEX) is used to

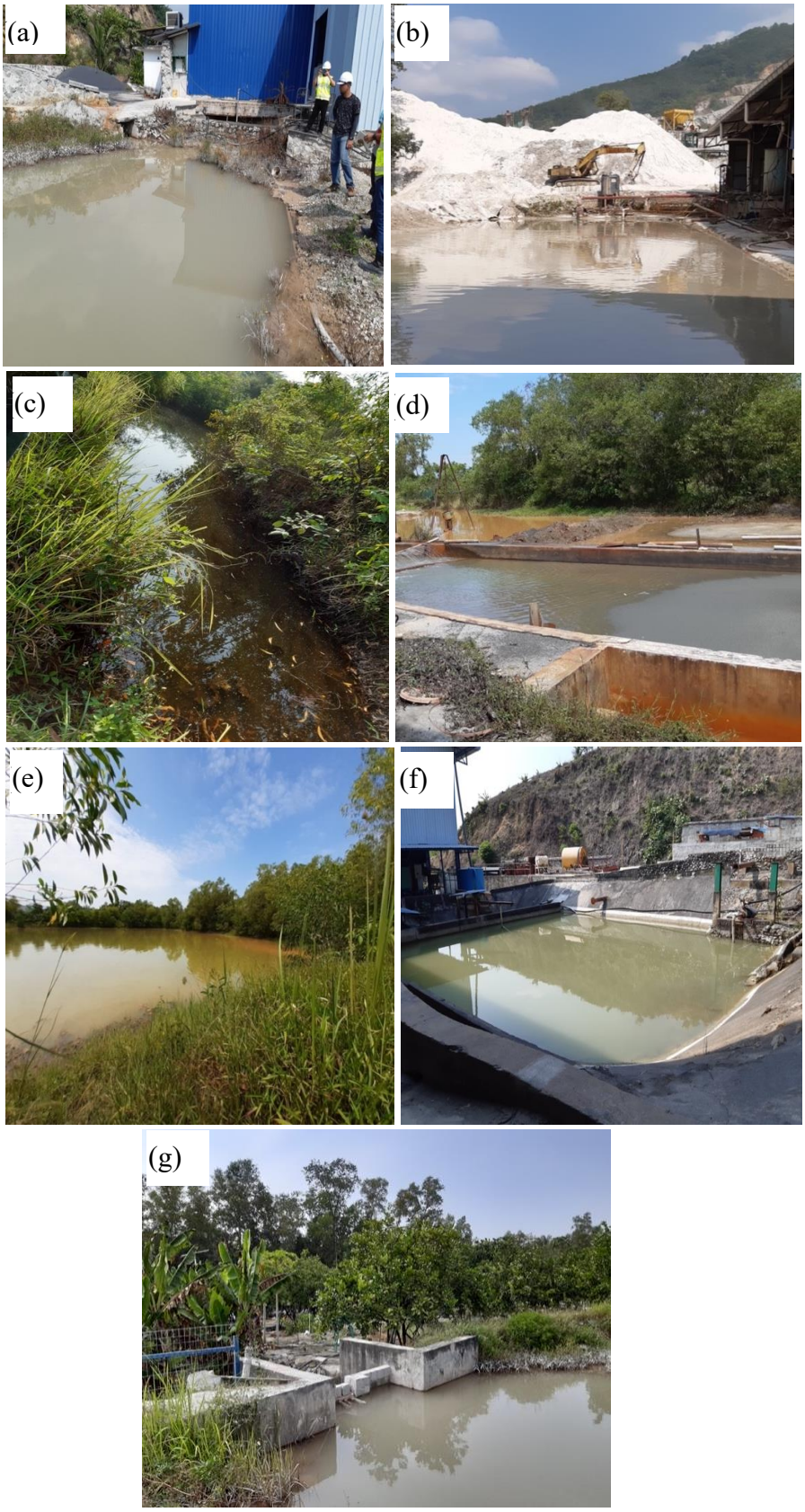

FIGURE 2. Retention pond in each amang plant vicinity. (a) PP-1, (b) PP-2, (c) PP-3, (d) PP-4, (e) PP-5, (f) PP-6 and (g) PP-7

quantify the concentrations of Cu, Cd, Cr, Zn, Ni, Pb, and As contained in the water samples. The samples were filtered using Whatman filter paper, No.4, and were analysed in triplicates to ensure precision and accuracy in the procedure of analyses (Liang et al. 2017). The water samples are filled into a high-density polyethylene bottle for 50 mL each before the subjection of ICPMS. Before this process, the bottle is rinsed with distilled water to remove any contaminants. The calibration was achieved using the multi-element calibration standard (Perkin Elmer Pure Plus) with a concentration of 10 mg/L, and the present elements were; Ag, Al, As, Ba, Be, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, In, K, Li, Mg, Mn, Na, Ni, Pb, Rb, Se, Sr, Tl, U, V, and Zn.

RADIOLOGICAL IMPACT ASSESSMENT

To assess the radiological impacts, the annual effective dose due to water ingestion, gamma radiation absorbed dose rate (D), and annual effective dose (AED) are determined (Ajekiigbe et al. 2017; Al-Harmali 2020; El-Gamal, Sefelnasr & Salaheldin 2019; Gregory, Emmanuel & Ezekiel 2013).

Annual effective dose due to ingestion of water (AED_{ing})
The effective dose arising from the ingestion of radionuclides in water is estimated using a dose coefficient stated by the International Atomic Energy Agency in Table III. 2A (IAEA 2014) and average annual consumption of water. To assess the annual effective dose to the public due to accidental ingestion of effluent water from amang plant is calculated using the Equation (5).

$$\mathrm{AED_{ing}} = \sum \mathrm{C_i} \times \mathrm{C_w} \times \mathrm{EDC} \quad (5)$$

where C_i is the concentration of radionuclide (^{226}Ra, ^{228}Ra, and ^{40}K) in water; C_w is the estimated water consumption (60 liters) (Abdu Nasiru, Aznan Fazli & Nuraddeen Nasiru 2022; Shu'aibu et al. 2021) and EDC is the effective dose coefficient. IAEA has given the dose conversion factor for ^{226}Ra, ^{228}Ra, and ^{40}K as 2.8×10-7, 6.7×10-7, and 6.2×10-9, respectively..

Estimation of Gamma Radiation Absorbed Dose Rate (D)
The absorbed dose rate in the air at 1 metre above the ground has a direct relationship between activity concentrations of natural radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K. UNSCEAR (2000) has given the dose conversion factors for converting the activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K into doses (nGy/h per Bq/l) as 0.462, 0.604, and 0.0417, respectively. The gamma radiation population doses of those living in the area are calculated using Equations (6) and (7).

$$D_{in}\left(\frac{nGy}{h}\right) = 0.92C_{226Ra} + 1.1C_{228Ra} + 0.081C_K \quad (6)$$

$$D_{out}\left(\frac{nGy}{h}\right) = 0.462C_{226Ra} + 0.604C_{228Ra} + 0.0417C_K \quad (7)$$

where D is the dose rate in nGy/h and C^{226}Ra, C^{228}Ra and C^{40}K are the concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K, respectively.

Annual Effective Dose (AED)
Several researchers (Ajekiigbe et al. 2017; Al-Harmali 2020; Gregory, Emmanuel & Ezekiel 2013; Khoirul Solehah et al. 2020) have reported on the assessment of annual effective dose (AED) for irrigation, effluent, and water resources. AED estimation requires consideration of the conversion coefficient from the absorbed dose and the occupancy factor. The activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K are used to compute AED, following the method proposed by UNSCEAR (2000) in Equations (8)-(10) as shown herewith:

$$AED_{in}\left(\frac{mSv}{year}\right) = 4.91 \times D_{in}\left(\frac{nGy}{h}\right) \times 10^{-3} \quad (8)$$

$$AED_{out}\left(\frac{mSv}{year}\right) = 1.23 \times D_{out}\left(\frac{nGy}{h}\right) \times 10^{-3} \quad (9)$$

$$AED_{tot}\left(\frac{mSv}{year}\right) = AED_{in} + AED_{out} \quad (10)$$

where D_{in} and D_{out} are the indoor and outdoor air absorbed dose rates, respectively. AED_{in}, AED_{out}, and AED_{tot} are the indoor, outdoor, and total annual effective doses, respectively. In general terms, the Committee has considered 0.7 Sv/Gy to be the most appropriate average value of the quotient of effective dose rate to absorbed dose rate in air for males and females for environmental exposures to gamma rays (UNSCEAR 2000). To calculate the annual effective dose of workers who are exposed to effluent water during processing activities, conversion coefficients were employed. The assessment took into account the fraction of time the workers spent outdoors and indoors, with the former being 0.2 and the latter being 0.8.

NON CARCINOGENIC AND CARCINOGENIC RISK ASSESSMENT

To assess health risks, the hazard quotient (HQ) and lifetime cancer risk (LCR) were used to categorize them as non-carcinogenic or carcinogenic, respectively. Previous studies (Ahmed et al. 2019; Muhammad Abdullah et al. 2021; Siti Khairunnisaq, Ho & Abd Khani 2018) were consulted for this purpose. It should be noted that the maximum values of H_{ex} and H_{in} were found to be less than or equal to one, indicating that radiation hazards were negligible.

Non-Carcinogenic Analysis

The hazard index (HI) was determined by summing up the hazard quotients (HQs) for each heavy metal to assess the risk associated with exposure to these contaminants. The hazard quotient represents the ratio of dermal absorbed dose (DAD) to dermal reference dose (RfD) for each heavy metal, including Cu, Cd, Cr, Zn, Ni, Pb, and As. The values of DAD, HQ, and HI were calculated using Equations (11) to (14), respectively (USEPA 2004)risk assessment reviewers, remedial project managers (RPMs). The values used in the calculations are presented in Table 1.

$$DAD\ (\frac{\frac{mg}{kg}}{day}) = \frac{DA_{event} \times EV \times ED \times EF \times SA}{BW \times AT} \quad (11)$$

$$DA_{event} = K_p \times C_W \times T_{event} \quad (12)$$

$$HQ = \frac{DAD}{RfD} \quad (13)$$

$$HI = \Sigma HQ \quad (14)$$

where K_p is the dermal permeability coefficient of compound (Cr:1.14×10^{-3} cm/hr; Cd: 3.29×10^{-4} cm/hr; Pb: 1.0×10^{-4} cm/hr Zn: 3.19×10^{-4} cm/hr and As:1.62×10-3 cm/hr;) (Siti Khairunnisaq, Ho & Abd Khani 2018). While C_w is the concentration of heavy metals in water (mg/cm^3), t_{event} is hour of contact with the heavy metals in effluent water per event (average of 8 working hours). Meanwhile, the values for RfD were Cd: 5.0×10^{-6} mg/kg/day; Cr: 7.5×10^{-3} mg/kg/day; As: 1.2×10^{-4} mg/kg/day, Pb:0.424×10^{-3} mg/kg/day Ni: 5.4×10^{-3} mg/kg/day and Zn: 6.0×10^{-2} as stated in the Risk Assessment Information System (RAIS) (Siti Khairunnisaq, Ho & Abd Khani 2018).

Carcinogenic Analysis

The carcinogenic risk is a unitless incremental probability of an individual developing cancer over a lifetime due to carcinogenic exposure (Saha et al. 2017). Carcinogenic risk in this study was evaluated as dermal cancer life risks (LCR) using the formula (15).

$$LCR = DAD \times SF \quad (15)$$

where DAD is the dermal absorbed dose (mg/kg/day) and SF_d is the dermal cancer slope factor (mg/kg/day) and it approximates the cancer risk per unit intake dose of an agent to cause cancer over an average lifetime. The cancer risk was evaluated for As, the only element for which SF_d values are available (3.66 mg/kg/day for dermal) (Li et al. 2018; Saha et al. 2017; Siti Khairunnisaq, Ho & Abd Khani 2018).

TABLE 1. Parameters used in DAD calculation

Parameters	Average	Reference
Event frequency, EF (events/day)	0.33*	
Exposure duration, ED (years)	30 years	
Exposure frequency, EF (days/year)	312 days/year**	
Body weight, BW (kg)	77[a]	(USEPA 2011)
Averaging time, AT (days)	6240	
Skin surface area, SA (hands) for adult male (cm^2)	1310[b]	(USEPA 2011)

*8 hours per day, **5.5 days per week, [a]Average body weight of male adult ranging from 25 to 54, [b] Recommended values for surface area of body parts for 21+ years male

Results and Discussion

NORM concentrations in water samples

The natural radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K are common radioactive constituents in groundwater and are often detectable in groundwater supplies. The results of NORM and heavy metal analysis of water samples collected from the amang plant are presented in Tables 2 and 3. The NORM concentrations (Bq/L) in the water samples ranged from 2.4-34.9 Bq/L, 0.8-14.7 Bq/L, and 19.5-299.4 Bq/L for ^{226}Ra, ^{228}Ra, and ^{40}K, respectively (Table 2). The mean concentrations of radionuclides ^{226}Ra and ^{228}Ra in the water samples exceeded the control limits of 10 Bq/L and 5 Bq/L (AELB 2010), respectively, as depicted in Figure 3. The control limits were derived based on the committed effective dose per unit intake through ingestion for members of the public (AELB 2010; UNSCEAR 2000). However, no control limit was issued for radionuclide ^{40}K, as it occurs in a fixed ratio to stable potassium and does not pose any health risks to humans (ANZECC & ARMCANZ 2000).

The values obtained in this study fall within the range reported in previous studies of the closed water system in the amang plant, which was 1.48-71.55 Bq/l and 0.03-36.16 Bq/l for ^{226}Ra and ^{228}Ra, respectively (Mohsen, Ismail & Pauzi 2008; Muhamad Samudi et al. 2007). However, the NORM concentrations in this study were higher than those in Malaysian water samples, which were 2.86±0.79 Bq/l and 3.78±1.73 Bq/l for ^{226}Ra and ^{228}Ra, respectively (Almayahi, Tajuddin & Jaafar 2012). Thus, processing activities involving amang minerals contribute to the increased levels of these radionuclides in the effluent water. Since the NORM content in this study exceeds the values of 10 Bq/l for ^{226}Ra and 5 Bq/l for ^{228}Ra, respectively, treatment of the effluent is necessary before it can be released safely into water resources.

Heavy metals concentrations in water samples

According to the Substance Priority List (SPL), Arsenic (As) poses the most significant potential threat to human health due to its known or suspected toxicity and likelihood of human exposure, followed by Plumbum (Pb), Cadmium (Cd), Chromium (Cr), and Nickel (Ni) (ATSDR 2020).

In this study, the concentration of heavy metals Cd, Cr, As, Pb, and Ni are within the range of 0-0.004 mg/L, 0-0.06 mg/L, 0.01-0.30 mg/L, 0.01-0.09 mg/L, and 0.002-0.07 mg/L respectively, in decreasing order of: As (0.21 mg/kg) > Ni(0.025 mg/kg) > Pb(0.03 mg/kg) > Cr(0.01 mg/kg) > Cd(0.001 mg/kg) (Table 3). These values are in line with those found in the effluent from the industrial zone in Dhaka and water samples in a closed-cycle system in amang plants (Muhamad Samudi et al. 2007; Saha et al. 2017). However, these values still fall within the class IV industrial effluent value stated by DOE Malaysia, for Cd (0.01 mg/kg), Pb (5.00 mg/kg), and Ni (0.20 mg/kg). Meanwhile, the value for heavy metal As, exceeds the class IV industrial effluent value of 0.10 mg/kg. Additionally, all heavy metals except Cr (0.05) in this study exceed the guideline values for chemicals from industrial sources stated by World Health Organization (WHO 2006) which are Cd (0.003), As (0.02), Pb (0.01), and Ni (0.07).

Heavy metals such as As, Cd, and Pb are highly toxic even at low concentrations, with no known benefit to human health. When these metals enter the environment, they can disrupt the aquatic ecosystem (Saha et al. 2017). For example, lead (Pb) is a carcinogenic substance in humans, and its poisoning could lead to disturbances in haemoglobin synthesis. However, dermal exposure to Pb is not considered a significant pathway compared to ingestion and inhalation. Arsenic is one of the agents that can cause skin cancer due to the percutaneous absorption of arsenic into the skin (Post 2003). According to the Agency for Toxic Substances and Disease Registry (ATSDR), skin contact with inorganic arsenic may cause redness and swelling (ATSDR 2015). Recently, the contamination of heavy metals and estimation of potential human health risk analysis has become a highly researched field in other industries as well (Arshad et al. 2020; Kanayochukwu, Ijeoma & Onyenezi 2019; Muhammad Abdullah et al. 2021; Renu et al. 2021).

Pearson's correlation coefficients

Table 4 shows a statistical analysis of the measured radionuclides and heavy metals calculated using Pearson's correlation coefficient. The correlation calculated is used to understand the degree of association that may exist between radionuclides and heavy metals. Positive values approaching 1 indicate a strong positive correlation, which can be observed between radionuclides ^{226}Ra, ^{228}Ra, and ^{40}K. This condition is due to the radionuclides naturally present together in groundwater and also due to the amang extraction processes (Shu'aibu et al. 2021).

TABLE 2. Activity concentrations of radionuclides (Bq/l)

	^{226}Ra (Bq/l)		^{228}Ra (Bq/l)		^{40}K (Bq/l)		References
PP1	10.8-23.0	16.6±6.9	4.3-5.3	4.8±0.7	53.2-119.3	86.2±42.2	This study
PP2	6.0-6.8	6.4±0.4	2.7-6.6	4.1±2.2	27.1-80.3	42.3 ±33.1	
PP3	20.3- 30.2	25.3 ±7.0	7.3-12.9	10 ±2.81	110.6-169.2	139.9±41.4	
PP4	17.8-34.9	26.6±4.8	7.9-14.7	12.6±1.6	156.2-299.4	203.4±43.2	
PP5	11.3-24.9	16.6±2.2	2.1-6.9	3.3±0.8	56.4-125.8	86.8±13.3	
PP6	2.4-31.8	12.0±10.0	0.8-12.9	6.5±4.2	19.5-202.8	81.2±63.0	
PP7	10.8-11.9	11.3±0.7	4.1-6.8	5.7±1.4	70.5-104.1	87.3±23.8	
Radionuclide control limit in water	10.00		5.00		NS		(AELB 2010)
Water samples in Northern Malaysia Peninsula	2.86±0.79		3.78±1.73		152±12.0		(Almayahi, Tajuddin & Jaafar 2012)
Irrigation water in Malaysia	1.51±0.30		0.17±0.09		7.96±3.07		(Khoirul Solehah et al. 2020)
Irrigation water standard (Australia-NZ)	5.00		2.00		NS		(ANZECC & ARMCANZ 2000)
Water resources in Egypt	0.20±0.03		0.11±0.02		0.69±0.08		(El-Gamal, Sefelnasr & Salaheldin 2019)
Water in the closed water system in tin tailing plant	28.98-35.42		29.59-36.16		NS		(Muhamad Samudi et al. 2007)
Amang processing pond	1.48-71.55		0.03-6.90		NS		(Nasirian et al. 2007)
Water resources in Egypt	0.20±0.03		0.08±0.01		0.69±0.07		(El-Gamal, Sefelnasr & Salaheldin 2019)
Effluent from the dump site, in Nigeria	0.40 ± 0.40		0.80 ± 0.20		0.80 ± 0.30		(Ogungbemi et al. 2023)
Rare earth processing plant (LAMP)	0.33 – 0.56		0.18 – 0.68		4.72-4.99		(Matthew Tikpangi et al. 2015)
The mining area in Nigeria	4.22±1.52		9.13±3.80		83.79±21.39		(Ajekiigbe et al. 2017)

*NS=Not stated in the study

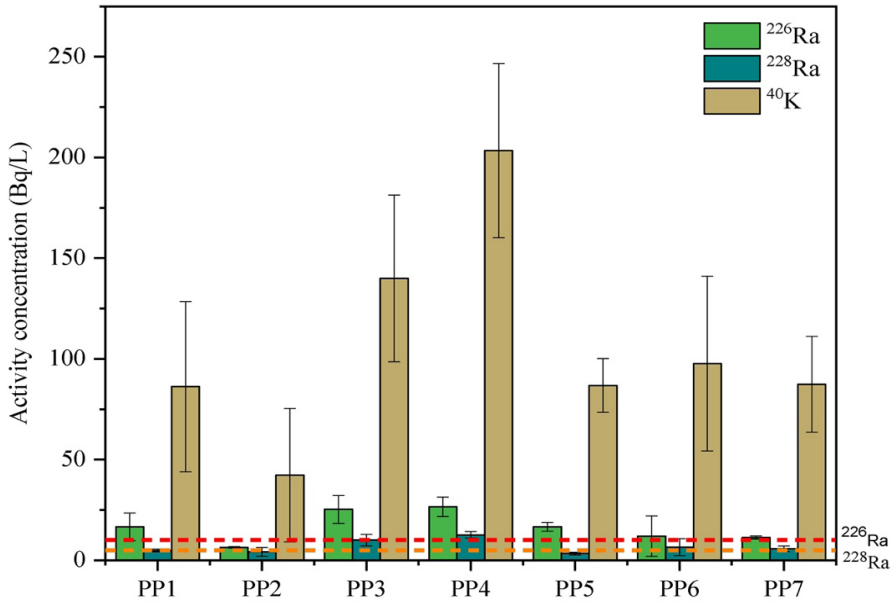

FIGURE 3. NORM activity concentration in water samples

TABLE 3. Activity concentrations of heavy metals (mg/kg)

Location	Cd	Cr	As	Pb	Ni	References
PP1	0.001±0.001	0.001±0.001	0.15±0.198	0.011±0.013	0.015±0.021	
PP2	0.0003±0	0.0021±0	0.0754±0.0005	0.032±0.0004	0.0036±0.0002	
PP3	0.0014 ± 0	0.0010 ± 0.0002	0.0609 ± 0.0012	0.0628 ± 0.0002	0.0340 ± 0.0004	
PP4	0.002±0.002	0.001±0	0.013±0.005	0.015±0.011	0.002±0.001	This study
PP5	0±0	0.001±0	0.628±0.088	0.005±0.004	0.066±0	
PP6	0.002±0.001	0.06±0.07	0.29±0.41	0.09±0.1	0.04±0.04	
PP7	0.002±0.002	0.001±0	0.218±0.007	0.015±0	0.017±0.001	
Industrial effluent (class IV)	0.010	NS	0.100	5.00	0.200	(DOE Malaysia 2009)
Guideline values for chemicals from industrial sources	0.003	0.050	0.02	0.010	0.070	(WHO 2006)
Irrigation water standard	0.05	1.00	2.00	5.00	2.00	(ANZECC & ARMCANZ 2000)
Water in a closed water system in tin tailing plant	NS	NS	NS	0.047–0.196	NS	(Muhamad Samudi et al. 2007)
Industrial zone in Dhaka	0.00034	0.0044	0.00064	0.00521	NS	(Saha et al. 2017)

RADIOLOGICAL IMPACT ASSESSMENT

Radiological impact assessment is determined in this study by calculating several parameters namely; annual effective dose due to ingestion (AED_{ing}), absorbed dose (D), and annual effective dose (AED), as shown in Table 5. The estimated AED_{ing} from ingestion of effluent water from amang plant in this study ranged from 0.73 to 2.61 mSv/year. The AED_{ing} from all the water samples in this study is higher than the WHO's reference level of 0.1 mSv/year (WHO 2006). While all the mean value from the samples is 1.43±0.67 mSv/year which is higher than the water resources in Egypt which is 0.06 mSv/year (El-Gamal, Sefelnasr & Salaheldin 2019), and the effluent samples from the dump site in Nigeria (0.15 mSv/year) (Ogungbemi et al. 2023). These results indicate that the effluent water's NORM concentrations have increased due to processing at the amang facility. Although it is unlikely that employees or members of the public will directly ingest effluent water, the situation could get worse if the effluent is released into water sources. Thus, it became essential to this study to do additional research on effluent as a source of exposure to the environment.

According to Table 5, the absorbed dose rates D_{in} (indoor) and D_{out} (outdoor) for the effluent water in the amang plant in the present study ranged from 13.80 to 54.81 nGy/h and 7.19 to 28.38 nGy/h, respectively. While the mean value of D_{in} (indoor) and D_{out} (outside) are 30.76±14.22 (nGy/h) and 15.91±7.35 (nGy/h), respectively. The mean value of D_{out} (outside) in this study is in range with the study from the mining area in Nigeria which is 11±3.90 nGy/h (Ajekiigbe et al. 2017). However, this value is higher than the water samples from a rare earth processing plant (Lynas Advanced Materials Plant) in Malaysia (0.48-0.79 nGy/h) (Matthew Tikpangi et al. 2015). These values, however, are under the world's average level of 55 nGyh as set by UNSCEAR 2000 (Eli Syafiqah et al. 2021; UNSCEAR 2000).

The calculated annual effective dose (AED) using the concentration of ^{226}Ra, ^{228}Ra, and ^{40}K in effluent water samples in this study are varying from 0.77 to 3.04 mSv/year. The mean value from this study is 1.71±0.79 (mSv/year) which is higher than the recommended limit of external exposure dose for the general public of 1 mSv/year and within the range of irrigation water study in Malaysia which is 1.39 mSv/year (Rahim et al. 2020). Matthew Tikpangi et al. (2015) reported a considerably lower AED value than our study, which is 0.014 mSv/year obtained from Lynas Advanced Material Plant (LAMP) an Australian rare earths processing plant located in Malaysia. However, the value from this study is lower than the study in the Nigerian mining area which has an AED value of 20.25±7.17 mSv/year.

It is worth noting that NORM concentrations in the water samples from the amang plant's retention pond exceeded their permissible values as set by JTA. Moreover, nearly all concentrations of heavy metals exceeded guideline values for chemicals from industrial sources and class IV industrial effluent. Water quality is an essential aspect of any case study that provides valuable information for identifying future contamination of the environment. Various indices have been developed by authorities to summarize water quality for this purpose. The issue of water pollution poses a threat to public health, as heavy metals and other toxic substances from industries can pollute our natural resources (Mohammadi et al. 2019).

NON-CARCINOGENIC RISK AND CARCINOGENIC RISK

Table 6 shows non-carcinogenic and carcinogenic risk values of the hazard index and LCR. The health risk posed by exposure to heavy metals or toxicants is characterized by the hazard index, which is calculated by summing the hazard quotient (HQ) of Cd, Cr, As, Pb, and Ni (Song et al. 2015). A value of <1 for the index is considered safe over a lifetime, and in this study, dermal health risks for workers at amang processing plants were assessed using the index. The potential for adverse health effects related to non-carcinogenic risks associated with overexposure is considered when the HI value is >1 (USEPA 2004). The total hazard index values for Cd, Cr, As, Pb, Ni, and Zn in this study were found to be far lower than <1 (Figure 4), indicating that potential health risks for workers in amang processing plants are under control. Cr appeared to be the main exposure source among the heavy metals, with the non-carcinogenic risk HQ ranked in the order of Cr>As>Ni>Pb>Cd. However, a study by Muhammad Abdullah et al. (2021) showed that HI values for sediment and soil samples in amang plant were close to 1 via dermal contact exposure, while ingestion exposure resulted in values above 1, indicating potential adverse health effects.

Due to the unavailability of carcinogenicity slope factor for heavy metals such as Cd, Cr, Ni, and Pb, only As was used to calculate the carcinogenic risk using the DAD and SF for dermal exposure. Although these metals are also identified as potential carcinogens (Alidadi et al. 2019; Li et al. 2018; Siti Khairunnisaq,

Ho & Abd Khani 2018), it was found that the range of acceptable or tolerable carcinogenic risks via dermal exposure according to USEPA is within the range of 1×10^{-6} to 1×10^{-4} (Liang et al. 2017). The LCR values in this study ranged from 7.2×10^{-9} to 3.6×10^{-7}, which is far lower than the range stated, indicating acceptable values for carcinogenic risks as shown in Figure 5. These values are similar to the LCR calculated for the occupational exposure of farmers in the agricultural field, which is 4.27×10^{-6} (Siti Khairunnisaq, Ho & Abd Khani 2018). However, a study conducted in China on the possible exposure models in the vicinity of the tailing pond showed a high value of 3.4×10^{-3} for the carcinogenic risk, suggesting that the surrounding sites of the tailings pond are highly polluted (Liang et al. 2017). In addition, the average LCR value for dermal contact with soil and sediments in amang plants was found to be 2.01×10^{-4} and 7.69×10^{-4}, respectively (Muhammad Abdullah et al. 2021).

Table 7 presents the results of the linear Pearson's correlation coefficient analysis conducted to assess the relationships and degree of association among the measured radiological variables in the water samples studied. The strength of the relationship observed between the radionuclides and radiological variables is classified as very strong. However, overall, water collected from the retention pond at the amang plant poses a low radiological threat to the environment. The contribution of water samples to the overall radiation dose at the plant is negligible when compared to soil, tin tailings, and sediment samples (Muhammad Abdullah et al. 2021; Sanusi et al. 2021).

TABLE 4. Pearson correlation of radionuclides and heavy metals in water samples

	^{226}Ra	^{228}Ra	^{40}K	Cd	Cr	As	Pb	Ni	Zn
^{226}Ra	1.00								
^{228}Ra	**0.82**	1.00							
^{40}K	**0.92**	**0.93**	1.00						
Cd	-0.02	0.25	0.18	1.00					
Cr	-0.24	-0.10	-0.16	0.08	1.00				
As	-0.25	-0.63	-0.37	-0.16	0.19	1.00			
Pb	-0.04	0.16	-0.06	0.06	0.79	-0.16	1.00		
Ni	0.03	-0.39	-0.20	-0.18	0.31	**0.87**	0.19	1.00	
Zn	0.24	0.31	0.08	-0.06	0.38	-0.32	**0.85**	0.15	1.00

TABLE 5. Radiological impact assessment on effluent water samples from amang plant

Location	^{226}Ra (Bq/l)	^{228}Ra (Bq/l)	^{40}K (Bq/l)	AED_{ingest} (mSv/y)	Din (nGy/h)	Dout (nGy/h)	AED (mSv/y)	Reference
PP1	16.6±6.9	4.8±0.7	86.2±42.2	1.15	27.56	14.18	1.53	This study
PP2	6.4±0.4	4.1±2.2	42.3 ±33.1	0.73	13.80	7.19	0.77	
PP3	25.3 ±7.0	10 ±2.81	139.9±41.4	2.09	45.57	23.55	2.53	
PP4	26.6±4.8	12.6±1.6	203.4±43.2	2.61	54.81	28.38	3.04	
PP5	16.6±2.2	3.3±0.8	86.8±13.3	1.08	25.24	13.00	1.40	
PP6	12.0±10.0	6.5±4.2	81.2±63.0	1.18	24.57	12.73	1.36	
PP7	11.3±0.7	5.7±1.4	87.3±23.8	1.15	23.78	12.32	1.32	
Mean	16.1±7.4	6.8±3.22	104.1±52.2	1.43±0.67	30.76±14.22	15.91±7.35	1.71±0.79	
Irrigation water in Malaysia	1.51±0.30	0.17±0.09	7.96±3.07	-	-	-	1.39	(Khoirul Solehah et al. 2020)
Water samples in Malaysia	2.86±0.79	3.78±1.73	152±12	-	-	10	0.01	(Almayahi, Tajuddin & Jaafar 2012)
Water resources in Egypt	0.20±0.03	0.08±0.01	0.69±0.07	0.06	-	-	-	(El-Gamal, Sefelnasr & Salaheldin 2019)
Effluent from the dump site, in Nigeria	0.40 ± 0.40	0.80 ± 0.20	0.80 ± 0.30	0.15	-	-	-	(Ogungbemi et al. 2023)
Rare earth processing plant (LAMP)	0.33 – 0.56	0.18 – 0.68	4.72-4.99	-	-	0.48-0.79	0.006-0.01	(Matthew Tikpangi et al. 2015)
The mining area in Nigeria	4.22±1.52	9.13±3.80	83.79±21.39	-	-	11.00±3.90	20.25±7.17	(Ajekiigbe et al. 2017)

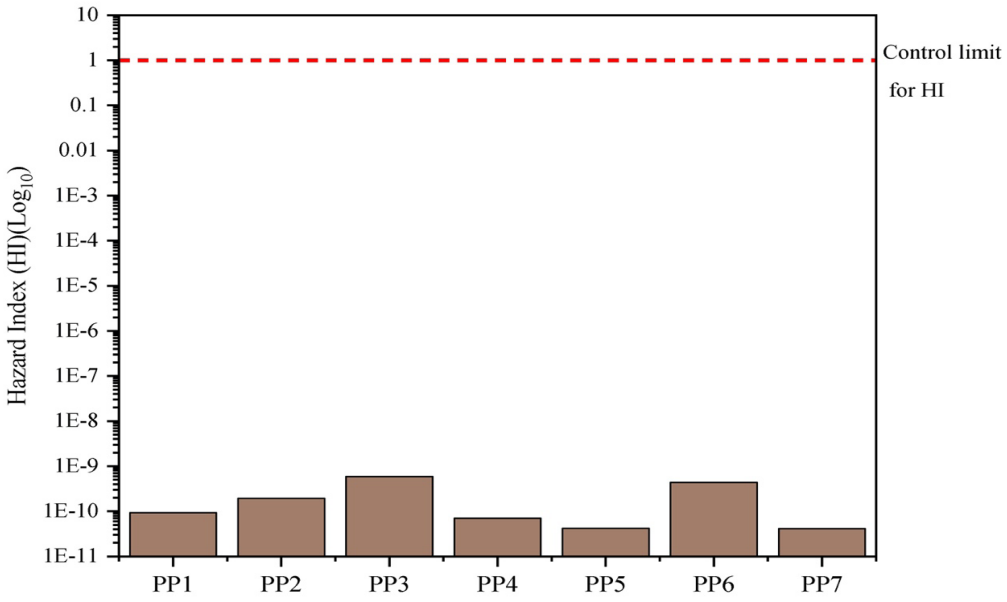

FIGURE 4. Hazard index of water samples in amang plant

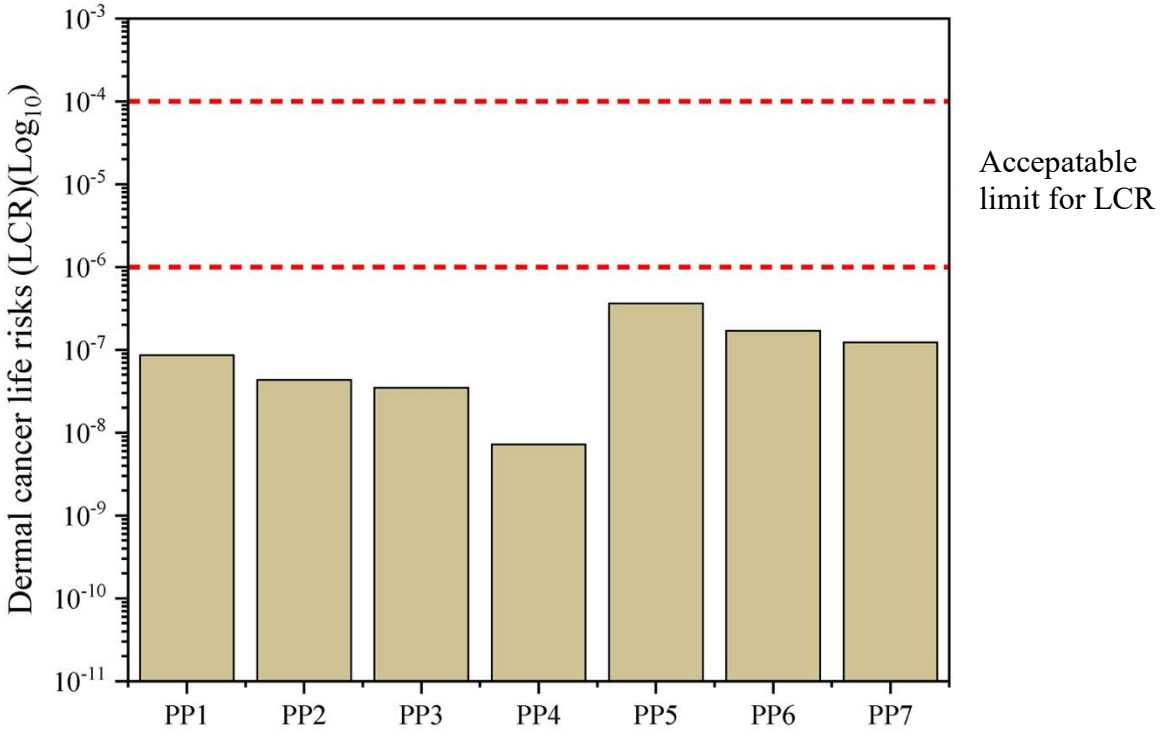

FIGURE 5. Dermal cancer life risks (LCR) of water samples in amang plant

TABLE 6. Carcinogenic and non-carcinogenic risk assessment on effluent water samples from amang plant

Location	Cd	Cr	As	Pb	Ni	Hin	LCR	Reference
PP1	0.001±0.001	0.001±0.001	0.15±0.198	0.011±0.013	0.015±0.021	0.93×10^{-11}	8.66×10^{-8}	This study
PP2	0±0	0.002±0	0.075±0.001	0.032±0	0.004±0	1.9×10^{-10}	4.33×10^{-8}	
PP3	0.001 ± 0	0.001± 0	0.061±0.001	0.063±0	0.034 ± 0	5.9×10^{-10}	3.47×10^{-8}	
PP4	0.002±0.002	0.001±0	0.013±0.005	0.015±0.011	0.002±0.001	7.0×10^{-11}	7.22×10^{-9}	
PP5	0±0	0.001±0	0.628±0.088	0.005±0.004	0.066±0	4.2×10^{-11}	3.62×10^{-7}	
PP6	0.002±0.001	0.06±0.07	0.290±0.410	0.090±0.100	0.04±0.04	4.4×10^{-10}	1.70×10^{-7}	
PP7	0.002±0.002	0.001±0	0.218±0.007	0.015±0	0.017±0.001	4.1×10^{-11}	1.23×10^{-7}	
Mean	0.001±0.001	0.009±0.021	0.205±0.210	0.033±0.032	0.025±0.023	2.1×10^{-10}	1.2×10^{-7}	
Water in a close water system in tin tailing plant	NS	NS	NS	0.047–0.196	NS	0.14±0.06	0.22±0.1	(Muhamad Samudi et al. 2007)
Industrial effluent (class IV)	0.010	NS	0.100	5.00	0.200			(DOE Malaysia 2009)
Guideline values for chemicals from industrial sources	0.003	0.050	0.02	0.010	0.070			(WHO 2006)
Irrigation water in Malaysia						0.01	-	(Khoirul Solehah et al. 2020)
Water samples in Northern Malaysia Peninsula						0.06	-	(Almayahi, Tajuddin & Jaafar 2012)
Industrial zone in Dhaka	0.00034	0.0044	0.00064	0.00521	NS			(Saha et al. 2017)
Rare earth processing plant (LAMP)						0.003-0.007	-	(Matthew Tikpangi et al. 2015)
Water resources in Egypt						-0.1 – 0.001		(El-Gamal, Sefelnasr & Salaheldin 2019)

TABLE 7. Pearson correlation coefficients among radioactive parameters for water samples

	AED_{ingest} (mSv/yr)	D_{in} (nGy/h)	D_{out} (nGy/h)	AED_{in} (mSv/y)	AED_{out} (mSv/y)	AED_{tot} (mSv/y)	H_{in}	LCR_{tot}
AED_{ingest} (mSv/yr)	1.000							
D_{in} (nGy/h)	0.992	1.000						
D_{out} (nGy/h)	0.993	1.000	1.000					
AED_{in}(mSv/y)	0.993	1.000	1.000	1.000				
AED_{out}(mSv/y)	0.992	1.000	1.000	1.000	1.000			
AED_{tot}(mSv/y)	0.992	1.000	1.000	1.000	1.000	1.000		
H_{in}(mSv/y)	0.205	0.194	0.194	0.194	0.194	0.194	1.000	
H_{ex}(mSv/y)	-0.464	-0.410	-0.413	-0.412	-0.410	-0.410	-0.260	1

CONCLUSIONS

In summary, the concentration of ^{226}Ra and ^{228}Ra exceeded the control limit stated by Atom Malaysia, as well as the standard for irrigation water stated by the Australian and New Zealand Environment and Conservation Council (ANZECC). While all heavy metals concentration in the samples is below the MCL level except for As and Pb. Heavy metal As and Pb exceeds the MCL value of 0.01 and 0.015 mg/L, respectively. However, the dermal health risk and the radiological impact assessment calculated using these values show nearly all assessments are below permissible levels. This indicates that dermal exposure is not a significant pathway for workers, compares to ingestion and inhalation pathways. However, proper treatment of this effluent water must be implemented to protect the environment and reduce human health risks.

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REFERENCES

Abdu Nasiru Muhammad, Aznan Fazli Ismail & Nuraddeen Nasiru Garba. 2022. Annual effective dose associated with radioactivity in drinking water from tin mining areas in north-western Nigeria. *Journal of Radiation Research and Applied Sciences* 15(3): 96-102. https://doi.org/10.1016/j.jrras.2022.06.008

Abdullahi Shittu, Aznan Fazli Ismail & Supian Samat. 2019. Determination of indoor doses and excess lifetime cancer risks caused by building materials containing natural radionuclides in Malaysia. *Nuclear Engineering and Technology* 51(1): 325-336. https://doi.org/10.1016/j.net.2018.09.017

AELB. 2010. *Akta Perlesenan Tenaga Atom 1984*. Peraturan-Peraturan Pelesenan Tenaga Atom (Perlindungan Sinaran Keselamatan Asas) 2010.

Ahmad, S. & Jones, D. 2013. The importance and significance of heritage conservation of the ex-tin mining landscape in Perak, Malaysia, the abode of grace. *The Asian Conference on Asian Studies* 2013: 38-54.

Ahmed, M., Matsumoto, M., Ozaki, A., Van Thinh, N. & Kurosawa, K. 2019. Heavy metal contamination of irrigation water, soil, and vegetables and the difference between dry and wet seasons near a multi-industry zone in Bangladesh. *Water (Switzerland)* 11(3). https://doi.org/10.3390/w11030583

Ajekiigbe, K.M., Olise, F.S., Sejlo, G.T., Yinusa, S.T., Amadi, V.N. & Olaniyi, H.B. 2017. Gamma spectrometric analysis of soil, sediment and water samples of granitic-Type solid mineral mining activities. *Journal of Radiation and Nuclear Applications An International Journal* 2(1): 29. https://doi.org/10.18576/jrna/020105

Al-Harmali, A. 2020. Assessment of natural radioactivity hazards in selected water samples collected from northern regions of Oman. *IOP Conf. Series: Materials Science and Engineering*. No. 757.

Alidadi, H., Belin, S., Sany, T., Zarif, B., Oftadeh, G. & Mohamad, T. 2019. Health risk assessments of arsenic and toxic heavy metal exposure in drinking water in northeast Iran. *Environmental Health and Preventive Medicine* 24(59): 1-17.

Almayahi, B.A., Tajuddin, A.A. & Jaafar, M.S. 2012. Radiation hazard indices of soil and water samples in northern Malaysian Peninsula. *Applied Radiation and Isotopes* 70(11): 2652-2660. https://doi.org/10.1016/j.apradiso.2012.07.021

Alnour, I.A., Wagiran, H., Ibrahim, N., Hamzah, S. & Elias, M.S. 2017. Determination of the elemental concentration of uranium and thorium in the products and by-products of amang tin tailings process. *AIP Conference Proceedings* 1799. https://doi.org/10.1063/1.4972913

ANZECC & ARMCANZ. 2000. *Australian and New Zealand Guidelines for Fresh and Marine Water Quality*.

Arshad, H., Zahid Mehmood, M., Hussain Shah, M. & Abbasi, A.M. 2020. Evaluation of heavy metals in cosmetic products and their health risk assessment. *Saudi Pharmaceutical Journal* 28(7): 779-790. https://doi.org/10.1016/j.jsps.2020.05.006

ATSDR. 2020. Support Document to the 2019 Substance Priority List. April: 1-9. www.atsdr.cdc.gov/SPL

ATSDR. 2015. Arsenic - ToxFAQs™. ToxFAQs. 2015. http://www.atsdr.cdc.gov/toxfaqs/index.asp

DOE Malaysia. 2009. *Environmental Quality (Industrial Effluent) Regulations 2009*. Percetakan Nasional Malaysia Berhad.

Dunca, A.M. 2018. Water pollution and water quality assessment of major transboundary rivers from Banat (Romania). *Journal of Chemistry* 2018: 9073763. https://doi.org/10.1155/2018/9073763

Eli Syafiqah Aziman, Aznan Fazli Ismail, Siti Fatimah Jubri, Muhammad Abdullah Rahmat & Wan Mohd Razi Idris. 2021. Environmental impact assessment of post illegal mining activities in Chini Lake with regards to natural radionuclides and heavy metals in water and sediment. *Journal of Radioanalytical and Nuclear Chemistry* 330(3): 667-683. https://doi.org/10.1007/s10967-021-08049-4

El-Gamal, H., Sefelnasr, A. & Salaheldin, G. 2019. Determination of natural radionuclides for water resources on the west bank of the Nile River, Assiut Governorate, Egypt. *Water* 11(2): 311. https://doi.org/10.3390/w11020311

Gregory, A.O., Emmanuel, E. & Ezekiel, A.O. 2013. Gamma spectroscopy analysis of produced water from selected flow stations in Delta State, Nigeria. *International Journal of Environmental Monitoring and Analysis* 1(5): 167-174. https://doi.org/10.11648/J.IJEMA.20130105.11

Hamzah, Y., Mardhiansyah, M. & Firdaus, L.N. 2018. Characterization of rare earth elements in tailing of ex-tin mining sands from Singkep Island, Indonesia. *Aceh International Journal of Science and Technology* 7(2): 131-137. https://doi.org/10.13170/aijst.7.2.8622

IAEA. 2014. *A Procedure for the Rapid Determination of 226Ra and 228 Ra in Drinking Water by Liquid Scintillation Counting*. International Atomic Energy Agency.

IAEA. 1989. *Technical Report Series No. 295: Measurement of Radionuclides in Food and the Environment*.

Ismail Bahari, Nasirian Mohsen & Pauzi Abdullah. 2007. Radioactivity and radiological risk associated with effluent sediment containing technologically enhanced naturally occurring radioactive materials in amang (tin tailings) processing industry. *Journal of Environmental Radioactivity* 95(2-3): 161-170. https://doi.org/10.1016/j.jenvrad.2007.02.009

Ismail, B., Yasir, M.S., Redzuwan, Y. & Amran, A.M. 2003. Radiological environment risk associated with different water system in amang factory. *Pakistan Journal Of Biological Science* 6(17): 1544-1547.

JMG. 2016. *Malaysian Minerals Yearbook 2016.* Department of Mineral and Geoscience Malaysia.

Kanayochukwu, J., Ijeoma, H. & Onyenezi, J. 2019. Health risk assessment of cadmium, chromium and nickel from car paint dust from used automobiles at auto-panel workshops in Nigeria. *Toxicology Reports* 6: 449-456. https://doi.org/10.1016/j.toxrep.2019.05.007

Khoirul Solehah Abdul Rahim, Zalita Zainuddin, Mohd Idzat Idris, Wahmisari Priharti, Murtadha S.H. Aswood, Solehah Khoirul Abdul Rahim, Zalita Zainuddin, Wahmisari Priharti & Murtadha Aswood Sh. 2020. Determination of the radiological risk from the natural radioactivity in irrigation at selected areas of Peninsular Malaysia. *Sains Malaysiana* 49(6): 1439-1450. https://doi.org/10.17576/jsm-2020-4906-22

Matthew Tikpangi Kolo, Siti Aishah Binti Abdul Aziz, Mayeen Uddin Khandaker, Khandoker Asaduzzaman & Yusoff Mohd Amin. 2015. Evaluation of radiological risks due to natural radioactivity around lynas advanced material plant environment, Kuantan, Pahang, Malaysia. *Environmental Science and Pollution Research* 22(17): 13127-13136. https://doi.org/10.1007/s11356-015-4577-5

Li, R., Kuo, Y.M., Liu, W.W., Jang, C.S., Zhao, E. & Yao, L. 2018. Potential health risk assessment through ingestion and dermal contact arsenic-contaminated groundwater in Jianghan Plain, China. *Environmental Geochemistry and Health* 40(4): 1585-1599. https://doi.org/10.1007/S10653-018-0073-4

Liang, Y., Yi, X., Dang, Z., Wang, Q., Luo, H. & Tang, J. 2017. Heavy metal contamination and health risk assessment in the vicinity of a tailing pond in Guangdong, China. *International Journal of Environmental Research and Public Health* 14(12): 1557. https://doi.org/10.3390/ijerph14121557

Mohammadi, A.A., Zarei, A., Majidi, S., Ghaderpoury, A., Hashempour, Y., Saghi, M.H., Alinejad, A., Yousefi, M., Hosseingholizadeh, N. & Ghaderpoori, M. 2019. Carcinogenic and non-carcinogenic health risk assessment of heavy metals in drinking water of Khorramabad, Iran. *MethodsX* 6: 1642-1651. https://doi.org/10.1016/j.mex.2019.07.017

Mohsen Nasirian, Ismail Bahari & Pauzi Abdullah. 2008. Assessment of natural radioactivity in water and sediment from amang (tin tailing) processing ponds. *The Malaysian Journal of Analytical Sciences* 12(1): 150-159.

Muhamad Samudi Yasir, Amran Ab Majid, Redzuwan Yahaya, Ismail Bahari & Wong Siew Kim. 2007. Impak aktiviti pemprosesan amang sistem tertutup ke atas kualiti air dan sedimen setempat. *The Malaysian Journal of Analytical Sciences* 11(2): 370-378.

Muhammad Abdullah Rahmat, Aznan Fazli, Eli Syafiqah, Nursyamimi Diyana, Faizal Mohamed & Irman Abdul. 2022. The impact of unregulated industrial tin-tailing processing in Malaysia: Past, present and way forward. *Resources Policy* 78: 102864. https://doi.org/10.1016/j.resourpol.2022.102864

Muhammad Abdullah Rahmat, Aznan Fazli Ismail, Nursyamimi Diyana Rodzi, Eli Syafiqah Aziman, Wan Mohd Razi Idris, Tukimat Lihan, Wan Mohd Razi Idris & Tukimat Lihan. 2021. Assessment of natural radionuclides and heavy metals contamination to the environment: Case study of Malaysian unregulated tin-tailing processing industry. *Nuclear Engineering and Technology* 54(6): 2230-2243. https://doi.org/10.1016/j.net.2021.12.013

Nasirian Mohsen, Bahari Ismail, Pauzi Abdullah & Azizah Jaafar. 2007. Gamma hazards and risk associated with norm in sediment from amang processing recycling ponds. *The Malaysian Journal of Analytical Science* 11(1): 314-323.

Ogungbemi, K.I., Adedokun, M.B., Ibitoye, A.Z., Oyebola, O.O. & Owoade, R.L. 2023. Estimation of radiological impact of the activities of olusosun dump site on workers and dwellers of Olusosun, in Lagos Southwest Nigeria. *Journal of Radiation Research* 64(1): 53-62. https://doi.org/10.1093/jrr/rrac067

Post, G. 2003. *Environmental Assessment and Risk Analysis Element: Dermal Absorption of Inorganic Arsenic from Water*. New Jersey Department of Environmental Protection Division of Science, Research and Technology.

Rehman, K., Fatima, F., Waheed, I. & Hamid Akash, M.S. 2018. Prevalence of exposure of heavy metals and their impact on health consequences. *Journal of Cellular Biochemistry* 119(1): 157-184. https://doi.org/10.1002/jcb.26234

Renu, K., Chakraborty, R., Myakala, H., Koti, R., Famurewa, A.C., Madhyastha, H., Vellingiri, B., George, A. & Gopalakrishnan, A.V. 2021. Molecular mechanism of heavy metals (lead, chromium, arsenic, mercury, nickel and cadmium) - induced hepatotoxicity - A review. *Chemosphere* 271: 129735. https://doi.org/10.1016/j.chemosphere.2021.129735

Saha, N., Safiur Rahman, M., Ahmed, M.B., Zhou, J.L., Ngo, H.H. & Guo, W. 2017. Industrial metal pollution in water and probabilistic assessment of human health risk. *Journal of Environmental Management* 185: 70-78. https://doi.org/10.1016/j.jenvman.2016.10.023

Sanusi, M.S.M., Ramli, A.T., Hashim, S. & Lee, M.H. 2021. Radiological hazard associated with amang processing industry in Peninsular Malaysia and its environmental impacts. *Ecotoxicology and Environmental Safety* 208: 111727. https://doi.org/10.1016/j.ecoenv.2020.111727

Shu'aibu, H.K., Khandaker, M.U., Baballe, A., Tata, S. & Adamu, M.A. 2021. Determination of radon concentration in groundwater of Gadau, Bauchi State, Nigeria and estimation of effective dose. *Radiation Physics and Chemistry* 178: 108934. https://doi.org/10.1016/j.radphyschem.2020.108934

Siti Khairunnisaq Rudzi, Ho Yu Bin & Intan Idayu Abd Khani. 2018. Heavy metals contamination in paddy soil and water and associated dermal health risk among farmers. *Malaysian Journal of Medicine and Health Sciences* 14: 2-10.

Song, D., Zhuang, D., Jiang, D., Fu, J. & Wang, Q. 2015. Integrated health risk assessment of heavy metals in Suxian County, South China. *International Journal of Environmental Research and Public Health* 12(7): 7100-7117. https://doi.org/10.3390/ijerph120707100

UNSCEAR. 2000. Sources and effects of ionizing radiation, united nations scientific committee on the effects of atomic radiation UNSCEAR 2000 report to the general assembly. *UNSCEAR Report* I: 1-556.

USEPA. 2004. Risk Assessment Guidance for Superfund (RAGS) Volume I. Human Health Evaluation Manual (HHEM). Part E. Supplemental guidance for dermal risk assessment. *USEPA* 1 (540/R/99/005). https://doi.org/EPA/540/1-89/002

WHO. 2006. *Guidelines for Drinking-Water Quality*. Geneva: World Health Organization.

Zaini Hamzah, Nor Monica Ahmad & Ahmad Saat. 2009. Determination of heavy minerals in 'amang' from Kampung Gajah ex-mining area. *The Malaysian Journal of Analytical Sciences* 13(2): 194-203.

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