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Investigation of radiogeology and environmental geochemistry of quarry ponds in post-tin mining areas of Phuket Island, southern Thailand

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ABSTRACT

Increasing demand for water supply on Phuket Island for both the local consumption and hotel industry has led to overexploitation of ground- and surface water bodies; hence, it is unremittingly important to monitor the local hydrologic regime. Radiogeological and hydrogeochemical environments are investigated by representative water and surface sediment samples collected from the different quarry ponds located in post-tin mining areas on Phuket Island. The study assessed water quality, analyze the relationship between bedrock weathering and water quality, and identify radiogeological factors and natural radon concentrations, with insights into the local geology of the study areas. The representative quarry ponds of Phuket Island consist of a granite quarry pond (PKW1), a Quaternary sediment quarry pond (PKW2), a granitic-metamorphic quarry pond (PKW3), and the perturbation pond (PKW4), with the formation of a variety of geologic features. All collected water samples show total cation concentrations with $Ca^{2+} > Na^{+} > K^{+} > Mg^{2+}$ and total anion concentrations with $HCO_3^- > Cl^- > NO_3^- > SO_4^{2-}$. However, Gibbs diagrams suggest that rock–water interactions and precipitation are major factors in geochemical processes controlling the water quality. Radon enrichments in the water samples are involved in the local geological structures and hydrogeological environments, as follows: PKW1 > PKW3 > PKW2 > PKW4. No significant correlation between the radon activity concentrations and major/minor ions is shown. Surface sediment samples are reported, and the activity concentrations of radium-228 and thorium-228 (radioactive decay series of thorium-232) are relatively higher than those of radium-226 (radioactive decay series of uranium-238). Therefore, the activity concentrations of natural radionuclides in the surface sediments could be directly influenced by physical weathering, particularly the depositional environments of granitic bedrock around the quarry ponds. Long-term monitoring is necessary to consider the behavior of quarry ponds, which is related to tourism planning and the overall development of Phuket Island.

1. Introduction

Phuket Island, located in Southern Thailand, is one of the world's most popular tourist destinations and with 543 km^2 the largest island in Thailand (Fig. 1; World Tourism Organization, 2021; Wood, 2022), and having a local population of around 418,000 people (reported by Phuket Provincial Administrative Organization, 2022). The economic growth rate of the Phuket Island tourism industry, its key economic factor, has followed Thailand's GDP (World Tourism Organization, 2021), and is expected to bounce back after a decline during the Covid-19 years. In 2022 Phuket welcomed 9 million Thai and foreign visitors, whereas for 2023 around 12 million visitors are expected (reported by Tourism Authority of Thailand, Phuket office, 2022). The number of inbound tourists increases usually during the tourist season from November until

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April, which is parallel to the dry season (December–March). During the eight months long, rainy season supported by the Southwest Monsoon Phuket receives an average of 2000 mm of rainfall (reported by Thai Meteorological Department, 2022). Geographically, Phuket Island has no large rivers, with most water supplies derived from rainfall and subsequently related groundwater resources (Puttiwongrak et al., 2019; Vongtanaboon, 2022). Thus, the water resources of Phuket Island are predominantly surface water ecosystems, which are stored in abandoned tin-mining quarry ponds, and shallow aquifers of mainly weathered granites (Kong, 2017; Vongtanaboon, 2022). The island's three main ponds, Klong Kata (PKW1) in Chalong, Bang Neow Dam (PKW2) in Srisoonthorn, and Bang Wad (PKW3) in Kathu, which are located in the central and southern part of the island, have a combined total capacity of 21.53 million cubic meters (Figs. 1b and 2a - d). During the dry weather-high tourism season the water supplies on Phuket Island are expected to decrease due to increasing demand by both local consumption and the tourism industry (Kong, 2017; Vongtanaboon, 2022). Therefore, providing and developing water sources for tap water production in response to rising water demand is necessary for both hotel businesses and the local population.

From another perspective, Phuket Island is part of the granitoids (granite rocks) of the Southeast Asian Tin Belt, in which outcrops are almost entirely made up of Cretaceous biotite granites, as shown in Fig. 1 (Charusiri et al., 1993; Cobbing et al., 1986; Nakapadungrat et al., 1993; Pollard et al., 1995; Nong et al., 2022). During the late 1980s Phuket was part of a nationwide radiometric survey conducted by the Department of Mineral Resources of Thailand (DMR). Results showed that the granitic plutons on Phuket Island, as part of the Western Fold Belt, have a relatively high level of specific activities of natural radionuclides (e.g., uranium, thorium, and potassium) (Charusiri et al., 1993; Nakapadungrat et al., 1993; Imai et al., 2013; Nieder et al., 2014). Moreover, granitic plutons from Prachuap Khiri Khan along the Gulf of Thailand in the east to the Andaman side with Phuket Island in the west displayed high ratios of thorium and potassium (Charusiri et al., 1993). In contrast to sedimentary and metamorphic rocks, limestones and quartzites, contain rather small amounts of natural radionuclides (Charusiri et al., 1993). The release of high levels of natural radionuclides usually occurs in the environment caused by the weathering of granites and the subsequent erosion and deposition of related minerals (Alnour et al., 2012; Örgün et al., 2005; Tzortzis and Tsertos, 2004). Additionally, open faults and fractures could act as potential pathways for natural radionuclide migration at depth into the hydrological regime (Clamp and Pritchard, 1998; Borchiellini et al., 1991; Durusoy and Yildirim, 2017). As weathered granites contain natural radionuclides related to their origin, their daughter products can escape into the hydrological sphere through the natural weathering and erosion processes escape into the hydrological sphere (e.g., surface water and sediment) (Bottrell, 1993; Durusoy and Yildirim, 2017; Clamp and Pritchard, 1998). In particular, infiltration into fractured bedrock has played a critical role in terms of increased radionuclide activity concentration levels in hydrological regimes (Örgün et al., 2005; Tzortzis et al., 2004). Wind and stormwater runoff are factors that directly influence the movement of radioactive materials (e.g., decomposed granite) into local hydrological regimes (Arogunjo et al., 2009; Fijałkowska-Lichwa, 2014; Odumo et al., 2011). However, the activity concentration can vary from one location to another depending on the rock type (Haroon and Muhammad, 2022; Huang et al., 2023; Beg et al., 2021). For instance, activity concentrations of natural radionuclides are likely to be higher in granites and carbonaceous black shales than in carbonate rocks (Benjami et al., 2017; Huang et al., 2023; Pereira et al., 2015). Consequently, natural radioactivity levels in abandoned quarry ponds of Phuket Island can be assumed to be relatively higher in both the soil and subsurface sediments (Clamp and Pritchard, 1998; Bottrell, 1993). Radon activity concentrations in the water of abandoned quarry ponds can be the result of weathered radioactive materials (e.g., granitic bedrock and soil), which are in direct contact to the water bodies (Haroon and Muhammad, 2022; Kanellopoulos et al., 2018; Benjami et al., 2017).

Radon, a naturally occurring radioactive gas, which can be produced continuously by radioactive materials (e.g., granitic rocks) through alpha decay of radium-226, is highly soluble in water (Clamp and Pritchard, 1998; Borchiellini et al., 1991; Gillmore et al., 2002). In general, the concentration levels of radon in water depend on the isotopic abundance of parent radionuclides (uranium-238 and radium-226) in the bedrock (Arogunjo et al., 2009; Bottrell, 1993; Huang et al., 2023). The recycling of terrestrial rocks has a direct effect on the concentration of radon dissolved in water (Bottrell, 1993; Gillmore et al., 2002). In particular, the hydrogeological setting and source of natural radionuclides might explain differences in radon levels in water samples taken from the various abandoned quarry ponds (Huang et al., 2023; Clamp and Pritchard, 1998; Borchiellini et al., 1991). Hence, radon

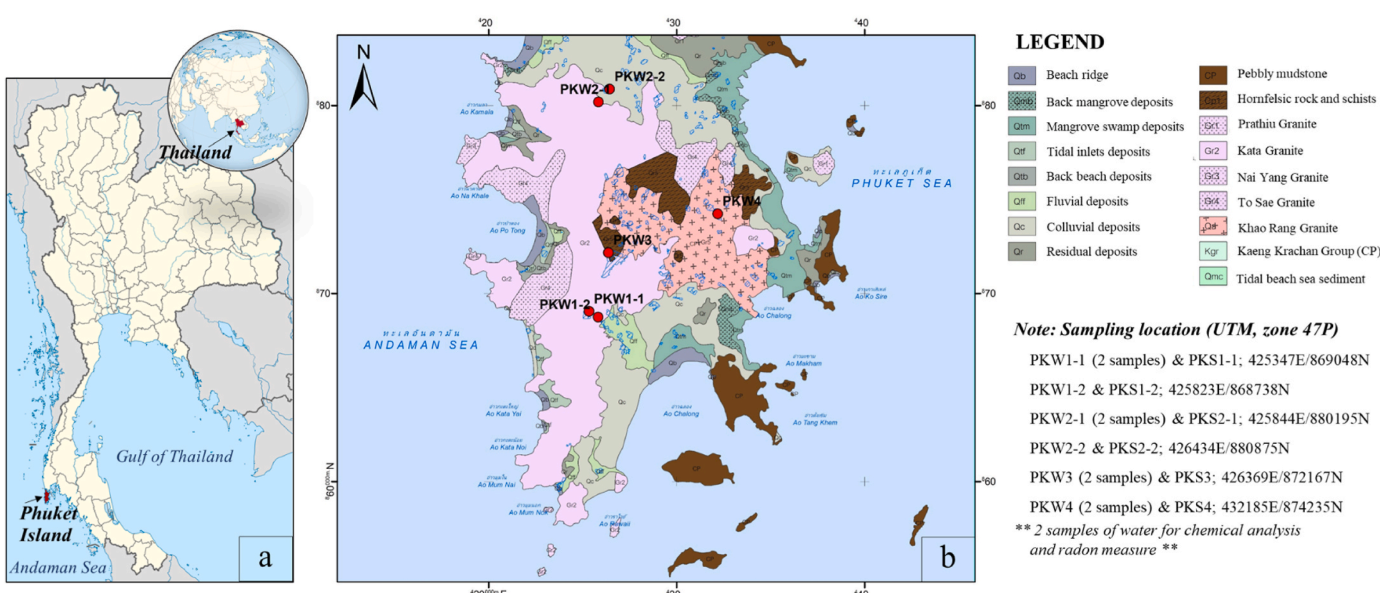

Fig. 1. Simplified world and geological map of the representative abandoned quarry pond on Phuket Island; (a) the location of Phuket Island, and (b) geologic map shown the sampling sites in the post-tin mining areas of Phuket Island.

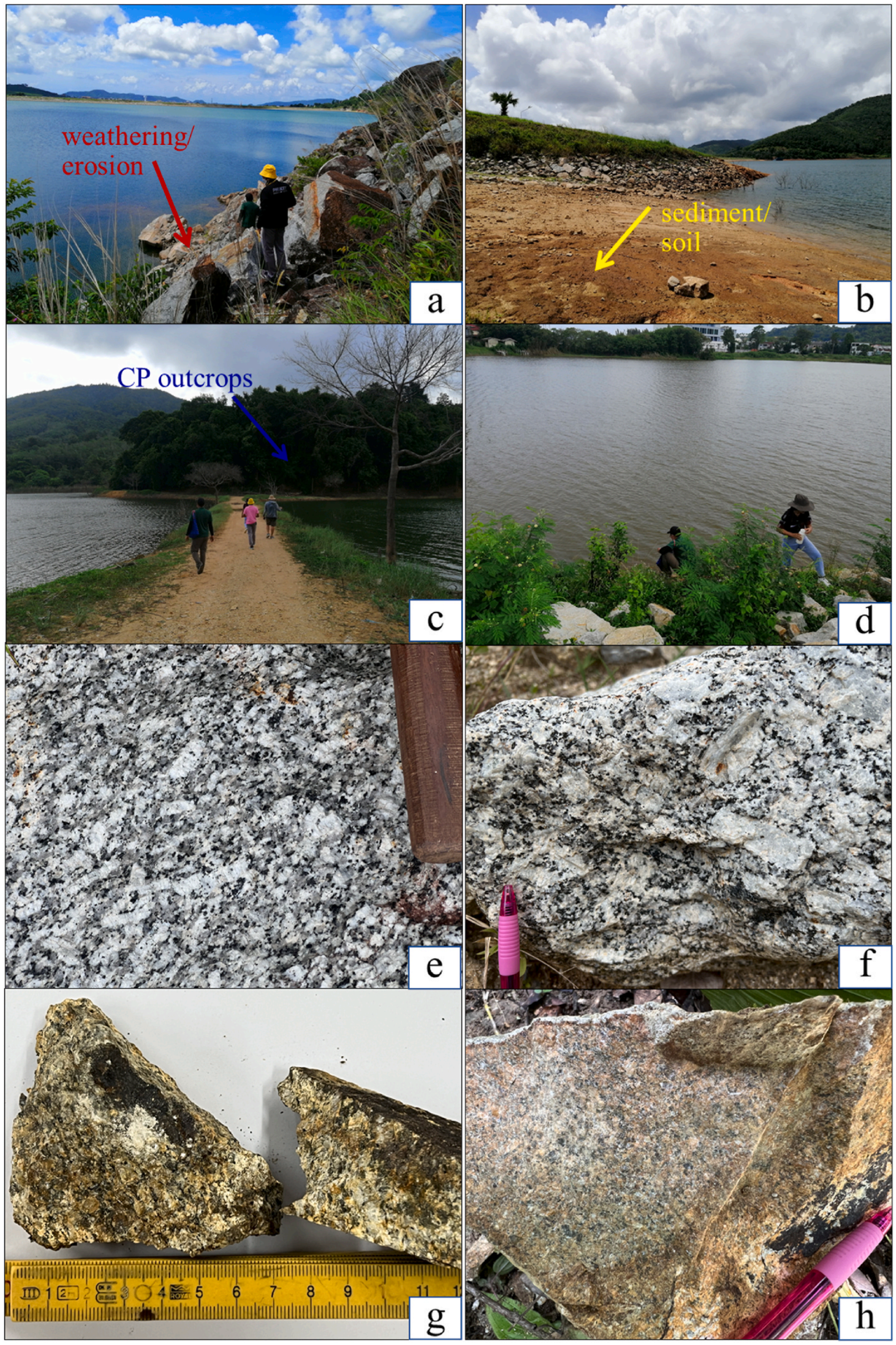

Fig. 2. Phuket islands four main ponds and granite rock samples; (a) PKW1; a granite quarry pond, (b) PKW2; a Quaternary sediment quarry pond, (c) PKW3; a granitic metamorphic quarry pond, and (d) PKW4; a mine perturbation pond, (e) PKS1 outcrop sample; Porphyritic granite with large feldspar phenocrysts, which can be few cm large, grey quartz, and black biotite and hornblende minerals, (f) PKS2 outcrop sample; Porphyritic granite with large feldspar phenocrysts, which can be few cm large, grey quartz, and black biotite and hornblende minerals, (g) PKS3 outcrop samples; Granite with quartz, feldspar and likely some black amphibole and biotite, grain size not larger than about 5 mm, here, somehow weathered already, (h) PKS4 outcrop sample; Granite, relatively fine grained with somehow larger quartz minerals.

activity concentration measurements in abandoned quarry ponds of Phuket Island are important for evaluating the level of natural radionuclides in the hydrological regime.

On the other hand, geochemical monitoring is an essential component of controlling water qualities that are influenced by weathering and erosion of subterranean formations (Eyankware et al., 2020; Kanellopoulos et al., 2018; Nieder et al., 2014; Wang et al., 2023). A quarry pond can contain groundwater, surface water, and rainwater. These waters promote chemical weathering (sulfide oxidation) of bedrock minerals, which leads to an increase in the concentration level of geochemical background values in natural hydrological systems (Abhay et al., 2012; Atanacković et al., 2013; Eyankware et al., 2020; Tran et al., 2020). Moreover, the quality of the waters in a post mining area can be explained by leaching processes, with the origin of major ions from bedrock weathering and ion exchange processes (Abhay et al., 2012; Atanacković et al., 2013; Bucher et al., 2017). Hence, a Piper diagram is often used to identify types of water (e.g., Na–Cl, mixed Ca–Mg–Cl, and Mg–HCO3). In addition, stable isotopes (e.g., $\delta^{18}O$ and $\delta^{2}H$) are typically used to understand the main source of water and the geochemical behavior of elements during rock weathering (Salifu et al., 2020; Musiba and Rwiza, 2021; Sun and Eadington, 1987). Therefore, an evaluation of major ions and trace elements of the waters taken from several post mining areas has identified weathering factors that controlled the water composition (Atanacković et al., 2013; Eyankware et al., 2020; Nieder et al., 2014). However, differences in the geochemical compositions of the waters in abandoned quarry ponds of the post mining area are usually evaluated by the factors of water–rock interaction (incl. Rock weathering), precipitation dominance, and evaporation (Atanacković et al., 2013; Bucher et al., 2017; Bucher et al., 2017, 2017). Und er the mechanism of water−rock interactions, the local hydrogeological environment may influence the chemical properties of water. Thus, water quality might not differ considerably at sites with similar geological formations. In addition, the activity of evaporation–sedimentation in the quarry, particularly during the dry season, might also be the main factor controlling the geochemical background value in the water (Akpanowo et al., 2021; Eyankware et al., 2020; Kanellopoulos et al., 2018; Wang et al., 2023).

Based on the current condition, Phuket Island is at risk of droughts as water demand outstrips supply, especially during the dry high tourism season. Water supplies from quarry ponds in post-tin mining areas might be a solution. However, natural weathering processes in granitic environments need to be monitored for public health purposes, both in terms of radiogeological effects and low-level radiation as well as environmental geochemistry. For these reasons, the information of natural radioactivity and geochemical patterns are extremely important for understanding the status of water quality, as these data can help regional development planning and water management in post-tin mining areas of Phuket Island. In particular, the natural radionuclide concentrations, radium-226, radium-228, thorium-228 and potassium-40, in surface sediments and radon activity concentrations of the waters need to be considered. Additionally, geochemical properties of the surface water need be evaluated in terms of local geology (site effect). However, weathering and erosion of bedrock within the abandoned quarry ponds could have a major impact on the water quality. Such a study, combining all important water related parameter, has not been carried out in Phuket until this day, and even not in Thailand, to our best knowledge. Therefore, this study aims to 1) combine radiogeological and geochemical survey data from selected post-tin mine ponds of Phuket Island, 2) evaluate the data within the context of local geology, and 3) provide a basis for the study of hydrogeological environments, hydrogeochemical characteristics, bedrock weathering, and water quality in abandoned quarry ponds. From there water management decisions might be drawn.

2. Geological setting

Phuket Island is located off the west coast of Thailand's southern peninsula as shown in Fig. 1. The geological setting of the island is related to the Western Granitoid Belts of the Southeast Asian Tin Belt, which extends from northern Thailand to western Indonesia through eastern Myanmar and Peninsular Malaysia (Cobbing et al., 1986; Nakapadungrat et al., 1993; Nong et al., 2022; Ridd and Watkinson, 2013). The Shan-Thai block underwent at least three deformation events during the late Carboniferous to early Permian, the late Permian to Early Jurassic, and the Late Cretaceous to early Tertiary, which were marked by igneous intrusions as shown in Fig. 1b (Ridd, 2009; Ridd and Watkinson, 2013; Watkinson et al., 2008). In particular, metallic and nonmetallic minerals with hydrothermal origins, including tin (cassiterite, SnO2) and tungsten (wolframite, (Fe, Mn) WO4), are closely related to granitoid rocks and are found on Phuket Island (Imai et al., 2013; Pollard et al., 1995; Sanematsu et al., 2015; Suwimonprecha et al., 1993). The granite suites can be divided into a number of major groups, including the Prathiu Granite (gr1), Kata Granite (gr2), Nai Yang Granite (gr3), To Sae Granite (gr4), and Khao Rang Granite (gr5) (Charusiri et al., 1993; Nong et al., 2022; Pollard et al., 1995), as shown in Figs. 1b and 2e-h. Although a sequence of granites on Phuket Island has corresponding intrusion ages, the lithological structures of the gr3 and gr4 group overlap closely (Charusiri et al., 1993; Pollard et al., 1995). For instance, the gr1 group consists of a biotite–hornblende granite with medium-to coarse-grained, equigranular to porphyritic, pink feldspar with allanite. The gr2 group, which is the bedrock of the quarry ponds (e.g., PKW1 and PKW3) of Phuket Island, comprises a biotite–hornblende granite that is coarse-to very coarse-grained and porphyritic, with sphene accessories (Figs. 1b, and Fig. 2e–h). Moreover, the gr2 group is intruded by a biotite–muscovite granite. The gr3 group is a muscovite–biotite granite that is coarse-grained and equigranular to porphyritic. The gr4 group is composed of biotite–muscovite granite that is fine-to medium-grained and equigranular to porphyritic. While the gr5 group is muscovite granite that is fine-to medium-grained and grayish white.

On the other hand, Carboniferous to Permian sedimentary rocks of pebbly mudstone, laminated mudstone, sandstone, and turbidite can be found within the granitoid belt with no evidence of contemporaneous volcanism (Ridd and Watkinson, 2013; Suwimonprecha et al., 1993), as shown in Fig. 1b. These rocks are found in the middle portion and along the headlands of the east coast of Phuket Island, including several islets lying just offshore, which are covered mainly by sedimentary and metasedimentary rocks of mudstone, shale, conglomerate shale, siltstone and sandstone. Low-topographic terrain (e.g., intermontane valleys, coastal beaches, flood plains, and swampy areas) is usually mantled by Quaternary unconsolidated sediments of colluvium and alluvium, as shown in Figs. 1b and 2b-c.

3. Methods

3.1. Sampling locations

Sampling locations covered four quarry ponds from various locations in the post-tin mining areas of Phuket Island as shown in Fig. 1. They provided representative values for radionuclide distribution and geochemical data: A granite quarry pond (Figs. 1b and 2a), a Quaternary sediment quarry pond (Figs. 1b and 2b), a granitic-metamorphic quarry pond (Figs. 1b and 2c), and a mine perturbation pond (Figs. 1b and 2d), as shown in Table 3. All surface sediment samples and water samples were collected in March and April 2022 from these four abandoned quarry ponds. Geographical coordinates of the sampling points were recorded using a Garmin GPS device (eTrex Venture® HC, USA). Additional information about the hydrogeological environments (e.g., topography, local geology, and location) of the sampling points can be found in Figs. 1b and 2a-d.

3.2. Radiogeological sampling and measure

Radiogeological sampling methodology and collection followed the International Atomic Energy Agency (IAEA) guidelines. Sediment samples were collected from weathered surface layers of abandoned quarries. Foreign materials (e.g., gravel and grains) were removed from the samples. While water samples were collected in 250 ml bottles at a depth of approximately 0.5 m below the water surface and a distance from the shore of the pond about 5 m in such a way that there was no bubbling. The representative sediment samples were analyzed at the Radioanalytical Laboratory, Office of Atoms for Peace in Thailand, Science, Research and Innovation, Bangkok, Thailand. All samples were dried to a constant weight (24 h in total) and then pulverized and sieved through a 1-mm sieve. After drying and homogenization, sediment samples were kept airtight and sealed in plastic containers with a cylindrical shape (6.5 cm in diameter and 8 cm in height) to prevent any radon exchange between the parent nuclides and their short-lived daughter nuclides. However, sediment samples were measured for the activity concentration of natural radionuclides after being left for 21 days. Secular radioactive equilibrium between the parent nuclides and their short-lived daughter nuclides, such as radium-226 and radon-222 progenies, as well as thorium-228 and radium-224 (and its progenies), was achieved. Essentially, the activity concentration was counted for 80,000 s using a gamma-ray spectrometry system with a high-purity germanium detector (35% relative efficiency of HP-Ge, BSI). The activity concentrations of possible radionuclides include radium-226 (609.31 keV), radium-228 (911.20 keV), thorium-228 (583.79 keV), and potassium-40 (1460.82 keV). A RAD7 electronic radon detector from DURRIDGE Company Inc., Billerica, MA, equipped with a RAD-H2O water accessory device was used to measure the radon concentration in the water samples. The measurement sensitivity of the RAD7 device to radon concentrations, i.e., its lower limit of detection was <0.37 Bq/l. However, the RAD-H_2O required a desiccant to be used at all times to dry the air stream before it entered the device. The relative humidity readings during the measurement remained below 10%.

3.3. Geochemical sampling and analysis

Six representative water samples were collected from the chosen abandoned quarry ponds in post-tin mining areas of Phuket Island to analyze their physical–chemical properties; with two samples from the granite quarry pond (PKW1-1 and 2; Figs. 1b and 2a), two from the Quaternary sediment quarry pond (PKW2-1 and 2; Figs. 1b and 2b), one from the granitic-metamorphic quarry pond (PKW3; Figs. 1b and 2c), and one from the mine perturbation pond (PKW4; Figs. 1b and 2d), as shown in Table 3. Water sampling was conducted during the summer months and low-rainfall period between March and April 2022, making the water samples representative for minimal flow conditions. At the sampling sites, physical properties such as temperature, pH, HCO_3^-, and electrical conductivity (EC) were measured in situ (quarry pond) by a portable pH meter, an ion meter (PXS-HCO3), and a conductivity meter (HI2003), respectively.

The filtration of water samples was performed in the field using 0.45-μm-diameter disposable filters to ensure the removal of suspended solids before storage in prepared bottles. A high-density polyethylene bottle that was narrow-mouthed was presterilized and washed with the sampled water. The water samples were stored as 2-L solutions for qualitative analysis (cations, anions, and heavy elements) and quarter-liter solutions for analysis of the stable water isotopes (δ^2H and $\delta^{18}O$). Nitric acid (HNO_3) (pH values less than 1) was used to acidify the water samples for cation analysis to prevent sedimentation of metals. No chemical reagents were added to the water samples used for anion, heavy element and stable water isotope analyses. Chemical water parameters of environmental geochemistry were determined at the Laboratory of Water Analysis Co., Ltd. (ISO/IEC 17025:2017), Phra Nakhon Si Ayutthaya, Thailand. The stable water isotopes (δ^2H and $\delta^{18}O$) were measured by laser spectrometry (Picarro L2130-I system) at the Thailand Institute of Nuclear Technology (TINT), Nakhon Nayok, Thailand and expressed in δ notations relative to VSMOW, in ‰. The methods of geochemical analysis and detection limits of these parameters are presented in Table 1. The accuracy of hydrogeochemical analysis was determined by calculating the ionic balance, which was within 1:1 ± 0.01% as shown in Table 1. However, the concentration data especially of heavy metals (As, Cd, Cu, Pb, Hg, Mn, and Zn) that were below the limit of detection (LoD) should be removed from the overall data (Table 3). Therefore, the results of qualitative analysis could be used to investigate the geochemistry of the water, including the water properties, hydrochemical characteristics, and bedrock weathering, in the abandoned tin mine ponds on Phuket Island.

Table 1
Methods used to analyze hydrogeochemical parameters of the representative water samples collected from an abandoned quarry pond in the post-tin mining area of Phuket Island.

Parameters	Method used	Detection limits (mg/L)
Physical parameters/ions analysis		
pH	In-house method: TM 001	–
Total dissolved solids, TDS	In-house method: TM 017	25
Fluoride, F^-	Distillation, SPADNS	0.1
Chloride, Cl^-	In-house method: TM 008	6
Iron, Fe^{2+}	Phenanthroline	0.005
Strontium, Sr^{2+}	EDTA Tritrimetric	0.01
Calcium, Ca^{2+}	EDTA Tritrimetric	0.01
Magnesium, Mg^{2+}	EDTA Tritrimetric	0.01
Potassium, K^+	Direct Air-Acetylene Flame	0.01
Sodium, Na^+	Direct Air-Acetylene Flame	0.005
Sulfate, SO_4^{2-}	Turbidimetric	1
Nitrate, NO_3^-	Turbidimetric	1
Bicarbonate, HCO_3^-	Titration	1
Heavy elements		
Arsenic, As	Continuous Hydride Generation/AAs	0.005
Cadmium, Cd	In-house method: TM 040	0.02
Copper, Cu	In-house method: TM 040	0.05
Total Iron, Fe	Phenanthroline	0.1
Lead, Pb	In-house method: TM 040	0.1
Mercury, Hg	Cold-Vapor/AAS	0.0005
Manganese, Mn	In-house method: TM 040	0.05
Stable water isotopes		
Hydrogen isotope, δ^2H	GasBench II-IRMS	±1(‰)
Oxygen isotope, $\delta^{18}O$	GasBench II-IRMS	±0.1(‰)

Remark.
In-house method: TM 008 based on Standard Methods for the Examination of Water and Wastewater, APHA, AWWA&WEE, 23rd ed., 2017, part 4500-Cl-B.
In-house method: TM 030 based on Standard Methods for the Examination of Water and Wastewater, APHA, AWWA&WEE, 23rd ed., 2017, part 4500-SiO_2C.
In-house method: TM 001 based on Standard Methods for the Examination of Water and Wastewater, APHA, AWWA&WEE, 23rd ed., 2017, part 4500-H + B.
In-house method: TM 017 based on Standard Methods for the Examination of Water and Wastewater, APHA, AWWA&WEE, 23rd ed., 2017, part 2540 C.

4. Results and discussion

4.1. Radiogeological characteristics

4.1.1. Radon activity concentration of waters

Radon concentrations of the representative water samples from the four different abandoned quarry ponds in post-tin mining areas of Phuket Island are shown in Figs. 1a–Fig. 2a–d and Table 2. The highest level was found in a granite quarry pond (PKW1; Figs. 2a and 3734 ± 120 Bq/m^3), followed by a granitic-metamorphic quarry pond (PKW3; Figs. 2c and 427 ± 28 Bq/m^3), with an average value of 1191 ± 50 Bq/m^3 (Table 2). Although the PKW3 quarry pond is located on hornfels rock and schists at the contact zone (Figs. 1b and 2c), the water sample also showed relatively high radon concentrations. In contrast, PKW2; Fig. 2b and PKW4; Fig. 2d, located in the Quaternary sediments and mine perturbation ponds as shown in Fig. 1a, exhibited low activities of approximately 379 ± 35 Bq/m^3 and 222 ± 16 Bq/m^3, respectively. Hence, radon levels of water samples may be influenced by the geological environments as shown in Table 2. Therefore, the radon activity concentration followed the order of PKW1 > PKW3 > PKW2 > PKW4.

Previous studies have suggested that radon concentrations measured in groundwater granitic aquifers are usually much higher than those in sedimentary aquifers (Bottrell, 1993; Huang et al., 2023; Örgün et al., 2005). Basically, granitic rocks contain various amounts of uranium (Alnour et al., 2012; Arogunjo et al., 2009; Odumo et al., 2011; Tzortzis

Table 2
Activity concentration of natural radionuclides in the surface sediment samples, and radon activity concentration of the water samples collected from representative abandoned quarry ponds on Phuket Island.

Surface sediment sample	UTM (Zone 47P)		Activity concentration (Bq/kg)				Water sample	Radon concentration (Bq/m^3)
	E (m)	N (m)	Ra-226 (609 KeV)	Ra-228 (911.20 KeV)	Th-228 (583.79 KeV)	K-40 (1460.8 KeV)		
PKS1-1	425347	869048	265 ± 14	314 ± 20	338 ± 18	1120 ± 57	PKW1	3734 ± 120
PKS1-2	425823	868738	182 ± 10	215 ± 14	237 ± 13	904 ± 47		
PKS2-1	425844	880195	149 ± 8	397 ± 24	341 ± 18	714 ± 40	PKW2	379 ± 35
PKS2-2	426434	880875	171 ± 9	271 ± 16	216 ± 11	376 ± 19		
PKS3	426369	872167	109 ± 6	118 ± 7	128 ± 7	266 ± 14	PKW3	427 ± 28
PKS4	432185	874235	50 ± 4	76 ± 6	90 ± 6	130 ± 17	PKW4	222 ± 16

et al., 2004). Thus, radon activity levels increase with increasing uranium concentration in environmental geology. Therefore, it is reasonable to assume that higher radon activity concentrations in the PKW1 and PKW3 samples can be explained by physical weathering processes. The granitic bedrock along the quarry ponds contains dominant radioactive minerals that causes an increase in water radon activity. Nevertheless, decomposition of the Quaternary sediments (from granite weathering) might also be a minor factor for radon concentration increases in the water samples. For instance, the PKW2 sample had a slightly higher radon level than the PKW4 sample that was taken from a mine perturbation pond as shown in Table 2. Preliminary results indicate that radon enrichments in the waters are due to local lithologies and hydrogeological environments. No significant correlation between the radon activity concentrations and major/minor ions has been observed in the water samples of the abandoned quarry ponds in the post-tin mining areas of Phuket Island as shown in Table 2.

4.1.2. *Activity concentration of surface sediments*

Activity concentrations measurements of natural radionuclides (e.g., radium-226, radium-228, thorium-228 and potassium-40) in the surface sediment samples collected from four abandoned quarry ponds provide following results as shown in Table 2 and Fig. 3. The highest activity concentrations were found in the PKS1 samples, except radium-228 and thorium-228, which were found to be slightly higher in the PKS2-1 sample as shown in Fig. 3. In contrast, the PKS4 sample represented the lowest levels for all specific activities, as shown in Table 2 and Fig. 3.

The activity concentration of potassium-40 was higher than those of other natural radionuclides, ranging from 130 ± 17 Bq/kg to 1120 ± 57 Bq/kg, with an average value of 578 ± 32 Bq/kg as shown in Table 2 and Fig. 3. In particular, the PKS1-1 and PKS1-2 samples were taken from areas of porphyritic granites with large feldspar phenocrysts (Figs. 1b, and Fig. 2e and f) showed the highest activity, with values ranging between 1120 ± 57 Bq/kg and 904 ± 47 Bq/kg (Table 2 and Fig. 3). However, the surface sediments were enriched in potassium, which could be partly attributed to feldspar minerals (K-feldspars) and fertilizers that were applied to soil in farming activities (Fares, 2017; Nguyen et al., 2022; Odumo et al., 2011).

In contrast, the PKS3 and PKS4 samples had reported concentrations of potassium-40 that were less than the average value (578 ± 32 Bq/kg) as shown in Table 2 and Fig. 3. The different concentrations of potassium-40 in the PKW2 quarry pond between the two sampling points of PKS2-1 (714 ± 40 Bq/kg) and PKS2-2 (376 ± 19 Bq/kg) are still questionable. However, the weathering and erosion of the Cretaceous biotite granites in the western part of the PKW2 quarry pond (Figs. 1b and 2b) could be one possible effect on the different levels of potassium-40, due to possible variations in the content of K-feldspars. Preliminarily, the order of concentrations of potassium-40 in the surface sediment samples was as follows, PKW1 > PKW2 > PKW3 > PKW4.

Radium-226 concentrations in the surface sediments ranged from 50 ± 4 to 265 ± 14 Bq/kg, with an average value of 154 ± 8 Bq/kg as shown in Table 2 and Fig. 3. The highest radium-226 concentration was recorded in the granite quarry pond (PKW1; Figs. 1b and 2a), especially in the PKS1-1 sample (265 ± 14 Bq/kg), followed by the PKS2-2 sample (171 ± 9 Bq/kg) collected from the Quaternary sediment quarry pond (PKW2; Figs. 1b and 2b). A slightly higher activity concentration in the PKW1 quarry pond could be explained by physical weathering of granitic rocks (Figs. 1b and 2e-h). The concentrations in the sediment from the granitic-metamorphic quarry pond (PKW3; Figs. 1b and 2c) were still significantly higher than the concentrations in the sediments from the mine perturbation pond (PKW4; Figs. 1a and 2d), as shown in Table 2 and Fig. 3. Note that one side of the PKW3 quarry is Cretaceous biotite granites (Kata Granite), and the other side is

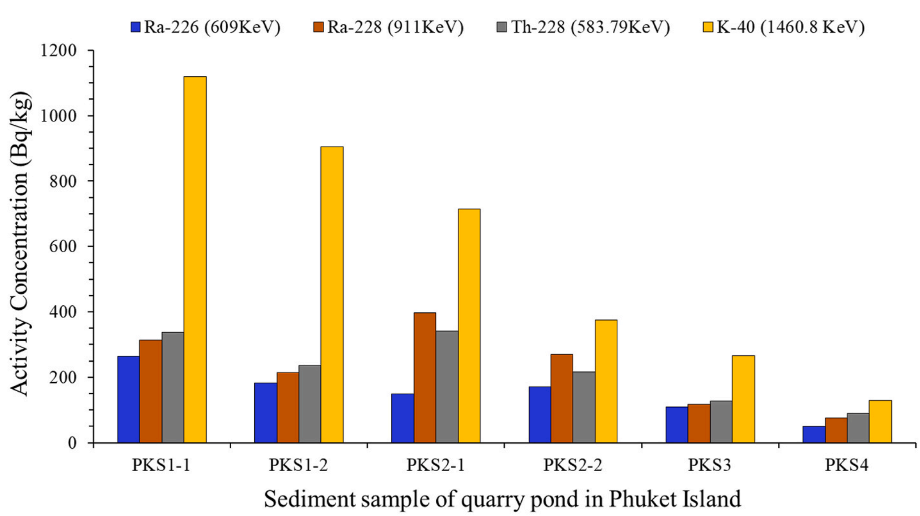

Fig. 3. Activity concentrations of natural radionuclides in the representative surface sediment samples collected from different abandoned quarry ponds of Phuket Island.

Permian–Carboniferous hornfels rocks (Phuket Group) as shown in Fig. 1b. The outcrop might potentially have increased the amount of surface sediments, which may partly account for the reduced radiation levels (Table 2 and Fig. 3). Noticeable differences in the activity levels of radium-226 in the sediment samples could be explained by the hydrogeological behavior and geochemical features of each quarry pond. Therefore, the order of radium-226 average activity concentrations in the abandoned quarry pond in the post-tin mining area of Phuket Island was as follows: PKW1 > PKW2 > PKW3 > PKW4.

The activity concentrations of radium-228 and thorium-228 were calculated using a 911.20 keV and 583.79 keV gamma photopeak, and values ranged from 76 ± 6 Bq/kg to 397 ± 24 Bq/kg (avg., 232 ± 14 Bq/kg) and 90 ± 6 Bq/kg to 341 ± 18 Bq/kg (avg., 226 ± 12 Bq/kg), respectively, as shown in Table 2 and Fig. 3. Unusually, the sediment samples from the PKW2 quarry pond had higher levels of radium-228 and thorium-228 than the samples from the PKW1 quarry pond. The activity concentrations of the PKS2-1 and PKS2-2 samples collected from the same pond were not similar, whereas a higher radiation content was shown in the PKS2-1 sample (Table 2 and Fig. 3). Hence, the radionuclide distribution might differ greatly, even within the same quarry pond; however, the sediment properties depend on the geological environments of the sampling point (Figs. 1b and 3). Excess radium-228 and thorium-228 in the PKS2-1 sample could be directly related to natural thorium from weathered granitic bedrock. Therefore, the weathering and denudation of granitic bedrock in the western part of the PKW2 quarry pond will probably become the leading cause of enrichment of the thorium series in surface sediments. Moreover, a higher fixation of the radionuclides in surface sediments is possibly due to organic enrichment and shifts the pH of the water to slightly less alkaline conditions (Amanjeet, Kumar et al., 2017; Benjami et al., 2017).

Remarkably, all of the surface sediment samples collected from the different abandoned quarry ponds in the post-tin mining area of Phuket Island showed that the activity concentrations of radium-228 and thorium-228 (radioactive decay series of thorium-232) were higher than that of radium-226 (radioactive decay series of uranium-238) as shown in Table 2 and Fig. 3. Therefore, the activity concentrations of natural radionuclides in the surface sediment samples in the post-tin mining areas of Phuket Island could be directly influenced by physical weathering, particularly the depositional environments of granitic bedrock in the area of the quarry ponds.

4.2. Hydrogeochemical characteristics

4.2.1. water properties

Physical properties, hydrogeochemical data, and sampling points of the water samples collected in the different abandoned quarry ponds in the post-tin mining area of Phuket Island are represented in Table 3. The water samples had slightly alkaline pH values up to 7.9 that were likely impacted by former tin mining activities and local geological environments. The water was taken from the Quaternary sediment quarry pond (PKW2; Figs. 1b and 2b), and it showed a slightly less alkaline tendency (pH = 7.4). The TDS contents varied from 25 mg/L (PKW2 pond) to 72 mg/L (PKW4 pond), with an average value of 43 mg/L as exhibited in Table 3.

Most of the water samples had low contents of HCO_3^-, with values below 10 mg/L, except for the mine perturbation pond (PKW4; Figs. 1b and 2d), which showed a high value of 68 mg/L, as shown in Table 3. The concentrations of Ca^{2+} were lower in PKW2 (1–2.6 mg/L) and higher in the PKW4 (22 mg/L) quarry ponds. The total ion concentrations were as follows: (a) the cation concentrations were $Ca^{2+} > Na^+ > K^+ > Mg^{2+}$, and (b) the anion concentrations were $HCO_3^- > Cl^- > NO_3^- > SO_4^{2-}$ as exhibited in Table 3. While the concentration of major ions in precipitation on Phuket Island has followed the order $SO_4^{2-} > Cl^- > Na^+ > NO_3^- > Ca^{2+} > Mg^{2+} > K^+$ as shown in Table 3. Hence, the variation in solutes of the water samples in the post-tin mining area of Phuket Island was mainly controlled by the local geological environment (Fig. 1b and Table 3). On the other hand, heavy metals, such as As, Cd, Cu, Fe, Pb, Hg, Mn and Zn, were present in extremely low concentrations (LoD; below the limit of detection), as shown in Table 3. Therefore, waters of the abandoned quarry pond in the post-tin mining area of Phuket Island have no detectable amount of heavy metal contamination.

Processing geochemical survey data of anions and cations and total dissolved solids are placed in the triangles of a Piper diagram to evaluate the hydrochemistry of the water samples, as represented in Fig. 4. A preliminary examination can be classified by the hydrochemical compositions into three types: the PKW1-1, PKW1-2, PKW2-1 and PKW2-2 samples are the Na–Cl type, the PKW3 sample is the mixed Ca–Mg–Cl type, and the PKW4 sample is the Mg–HCO3 type, as shown in Fig. 4.

Table 3
Hydrogeochemical data of the representative water samples collected from an abandoned quarry pond in the post-tin mining areas of Phuket Island.

Parameters	PKW1-1	PKW1-2	PKW2-1	PKW2-2	PKW3	PKW4	Rainwater at Phuket	WHO (2022)	EU (2020)
pH	7.7	7.6	7.4	7.4	7.7	7.9	4.2	6.5–8.5	6.5–9.5
TDS	38	42	25	25	54	72	28	600–1000	500
Cation; mg/L									
Ca^{2+}	5.1	4.5	2.6	1.0	8.3	22	0.32	75	100
Mg^{2+}	1.5	1.5	1.2	1.7	1.7	4.1	0.08	30	–
K^+	3.5	3.3	4.9	2.4	3.9	5.1	0.28	12	–
Na^+	6.9	6.5	5.0	5.0	7.4	7.1	0.75	200	–
Fe^{3+}	0.07	0.08	0.17	0.09	0.15	0.40	–	0.3	0.2
Anion; mg/L									
HCO_3^-	6	6	4	6	8	68	–	–	–
Cl^-	8	7	6	6	10	12	1.2	250	250
NO_3^-	0.18	0.01	0.74	0.98	0.01	0.01	0.17	3	3
SO_4^{2-}	0.2	0.2	0.31	0.2	0.23	0.52	0.25	250	250
Heavy element; mg/L									
As	LoD	LoD	LoD	LoD	LoD	LoD	LoD	0.01	0.01
Cd	LoD	LoD	LoD	LoD	LoD	LoD	LoD	0.003	0.005
Cu	LoD	LoD	LoD	LoD	LoD	LoD	LoD	2	2
Pb	LoD	LoD	LoD	LoD	LoD	LoD	LoD	0.01	0.01
Hg	LoD	LoD	LoD	LoD	LoD	LoD	LoD	0.001	0.001
Mn	LoD	LoD	LoD	LoD	LoD	LoD	LoD	0.5	-
Zn	LoD	LoD	LoD	LoD	LoD	LoD	LoD	4	-

Remark: *LoD, below the limit of detection; As (0.005 mg/L), Cd (0.02 mg/L), Cu (0.05 mg/L).*
Fe (0.1 mg/L), Pb (0.1 mg/L), Hg (0.0005 mg/L), Mn (0.05 mg/L), and Zn (0.05 mg/L).

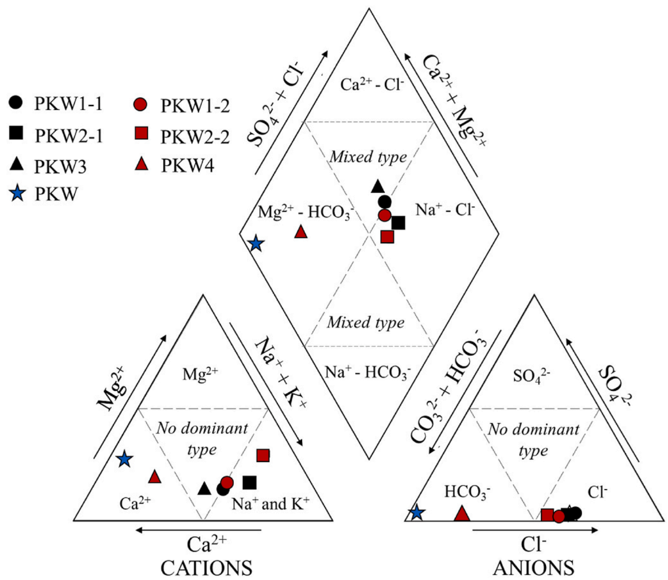

Fig. 4. Piper diagram of the representative water samples collected from different abandoned quarry ponds of Phuket Island.

Hydrogeochemical heterogeneity could be associated with the specific conditions of a local geological setting. In particular, the spatial variability of the geochemical characteristics of the PKW3 and PKW4 quarry ponds is clearly exhibited (Fig. 4). With regard to abundant anions in the water samples, the highest values of HCO_3^- and SO_4^{2-} in the PKW4 pond occurred in water samples that were collected from the areas where sediment perturbation was produced by the post-tin mining area (Table 3 and Fig. 4). Therefore, the PKW4 quarry pond is unmistakably associated with a mine perturbation zone within sedimentary lithologies (Figs. 1b and 2d). However, the total cation concentrations of the PKW1-1 and PKW1-2 samples were very low, which points to a lower effect of chemical weathering of the granitic bedrock (Table 3). Therefore, the quality of water is associated with local circulation conditions, the residence period of surface water, and rock–water interactions.

From another perspective, distributions of ion concentrations in the water samples of the different abandoned quarry ponds in the post-tin mining areas of Phuket Island are represented by a modified Schoellor diagram, as shown in Fig. 5. The concentrations of Ca^{2+} in the water samples taken from the granite quarry pond (PKW1) and the granitic-metamorphic quarry pond (PKW3) were lower than that in the sample collected in the mine perturbation pond (PKW4) as represented in Table 3 and Fig. 5. The PKW2 quarry pond, especially PKW2-2, is located on residual deposits comprising rock fragments and silts (Figs. 1b and 2b) and had lower Ca^{2+} contents (Table 3 and Fig. 5). The Mg^{2+} concentrations of the PKW1 and PKW3 quarry ponds had similar ranges (PKW1: 1.5 mg/L, and PKW3: 1.7 mg/L). Significantly, the water samples in the fractured granitic ponds of PKW1 and PKW3 were characterized by low concentrations of K^+(Table 3 and Fig. 5). Therefore, these waters might be affected by former tin mining activities.

On the other hand, the water samples collected in the PKW2 quarry pond had slightly lower concentrations of Na^+ than the other quarry ponds as shown in Table 3 and Fig. 5. No differences in Cl^- concentrations were observed between fractured granitic ponds (PKW1; Fig. 2a and PKW3; Fig. 2c) and in the mine perturbation pond (PKW4; Fig. 2d), except for the PKW2 sample. SO_4^{2-} shows quite similar contents in most water samples, except for the PKW4 sample, which had a relatively higher concentration (Table 3 and Fig. 5). Noticeably, the SO_4^{2-} concentration of the PKW4 sample might be related to the presence of inorganic tin compounds, indicating the possibility of water contamination. Moreover, the HCO_3^- concentrations showed a similar range between 4 and 8 mg/L, except for the PKW4 pond, which had a value of 68 mg/L (Table 3 and Fig. 5). In the case of the NO_3^- content, water samples from the PKW2 quarry pond had slightly higher concentrations than those from other abandoned tin mines of Phuket Island. In the hills and mountains around the PKW2 pond rubber plantations can be found, where fertilizer use is quite usual. Runoff from these areas into the pond, mainly during the rainy season, can bring NO_3^- into the pond and thus increase its concentrations. However, fertilizer use in rubber plantations might be less than in other agricultural areas, so that the overall nitrate concentrations are comparable lower and algae blooms for this pond are not documented. Moreover, high Fe and Mn concentrations in the PKW4 pond may indicate abundant organic matter in the post-tin mining area (Table 3 and Fig. 5).

Gibbs diagrams consisting of TDS versus Cl/(Cl + HCO_3) and TDS versus (Na + K)/(Na + K + Ca) were used to evaluate the hydrogeochemical signatures of the water samples in the different abandoned quarry ponds in the post-tin mining area of Phuket Island as presented in Fig. 6. Unique properties of water samples can be categorized as rock weathering, precipitation, and evaporation, as shown in Fig. 6. The water samples from the PKW1 and PKW3 quarry ponds were clearly affected by water–rock interactions (rock weathering), indicating that the contribution from chemical weathering dominates (Fig. 6). The PKW2 samples are located in a precipitation-dominant zone, indicating that the major ion constituents are primarily controlled by rainwater (Fig. 6). The PKW4 sample had dominant cations (Na^+ and Ca^{2+}) and

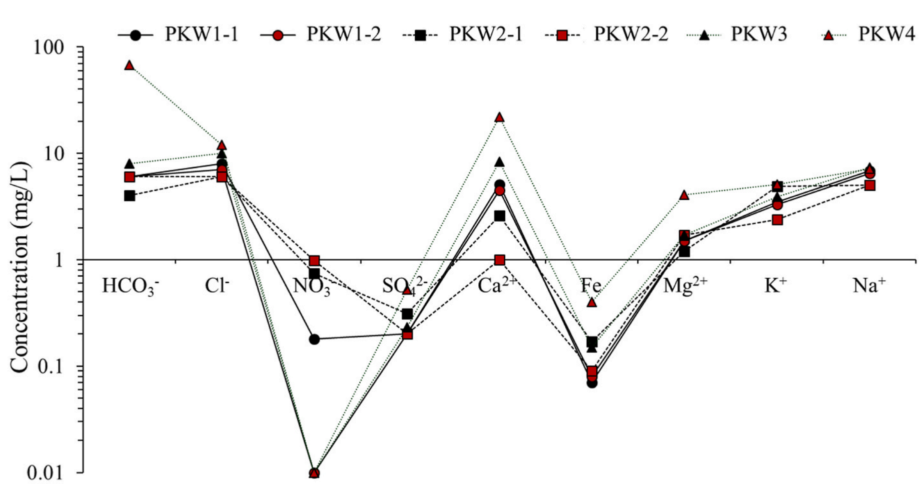

Fig. 5. Modified Schoellor diagram representing concentration of cations and anions from representative water samples collected from different abandoned quarry ponds of Phuket Island.

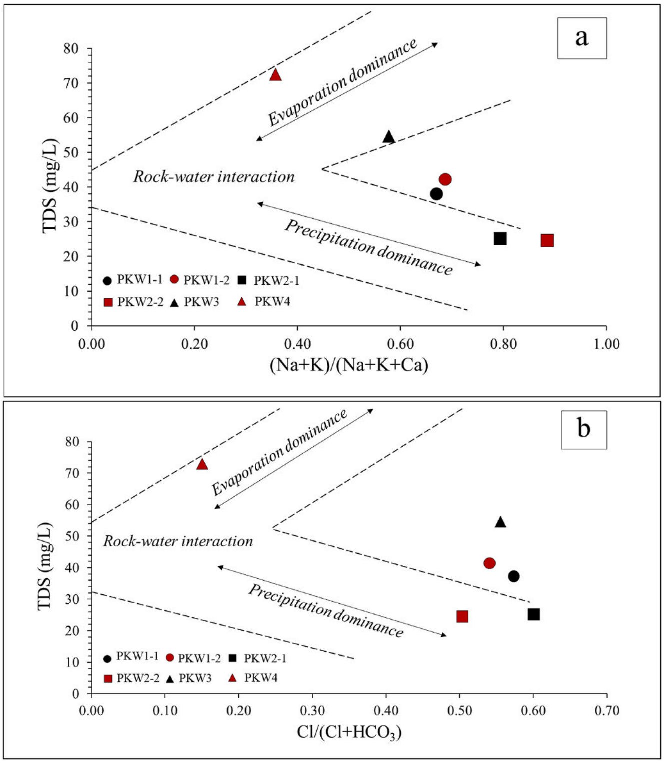

Fig. 6. Gibbs diagram representing the ratio; (a) Na + K/(Na + K + Ca) and (b) Cl/(Cl + HCO_3) as a function of TDS of the representative water samples collected from different abandoned quarry ponds of Phuket Island.

dominant anions (Cl^- and SO_4^{2-}) that clearly indicate evaporation dominance. According to the Piper diagram (Fig. 4), the PKW4 sample clearly indicates magnesium bicarbonate water, which reflects the dominance of carbonate-rich sedimentary rocks and/or weathering products. Furthermore, the small excess Na^+ content could be regulated by cation exchange processes when Ca^{2+} and Mg^{2+} were exchanged with Na^+ in the clay mineral sediment.

4.2.2. Stable isotope composition

Stable isotope analysis has become a powerful tool for environmental tracers used to investigate water samples under complex geological processes. In particular, tracing the stable isotopes δ^2H and δ^{18}O is essential for understanding the hydrogeochemical and hydrological characteristics by which water is stored under complex hydrogeological conditions (Salifu et al., 2020; Musiba and Rwiza, 2021). Preliminarily, the δ^{18}O and δ^2H values of water samples taken from the different abandoned quarry ponds in the post-tin mining area of Phuket Island are presented in Table 4 and Fig. 7. Mostly, the water samples are characterized by lighter isotopes, represented by δ^{18}O values ranging from −4.27‰ to −1.24‰ (average value of −3.27‰), and δ^2H values varying between −24.95‰ and 13.13‰ (average value of −20.58‰) (Table 4 and Fig. 7). The stable isotope compositions of the PKW1 and PKW2 samples were close to that of rainwater (Table 4 and Fig. 7). The difference between the stable isotope compositions of the two samples in the PKW2 quarry ponds (Figs. 1b and 2b) could be explained by an evaporation rate that varies between sampling points. The highest isotopes were discovered in the PKW4 sample (Figs. 1b and 2d) from a mine perturbation pond (δ^{18}O value, −1.24‰ and δ^2H value, −13.13‰), followed by the PKW3 sample from a granitic metamorphic quarry pond (δ^{18}O value, −3.03‰ and δ^2H value, −20.08‰). Consequently, the stable isotope compositions can be used to trace hydrological processes at different spatial scales in the post-tin mining area on Phuket Island (Table 4 and Fig. 7).

Table 4
Isotopic signatures of the representative water samples collected from an abandoned quarry pond in the post-tin mining areas of Phuket Island.

Raw water sample	δ^{18}O (±0.05‰)	δ^2H (±0.5‰)
PKR1-1	−4.27	−24.95
PKR1-2	−3.94	−23.89
PKR2-1	−3.53	−20.63
PKR2-2	−3.58	−20.79
PKR3	−3.03	−20.08
PKR4	−1.24	−13.13
Rainwater-PK	−5.80	−30.89
Seawater	−0.78	−7.8

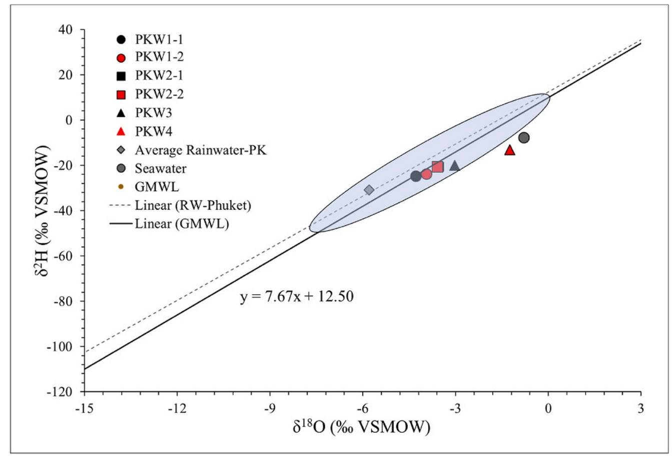

Fig. 7. Relationships between the δ^{18}O and δ^2H for the water samples in Phuket, seawater, and average Phuket's rainwater, 95% of the isotopic composition of the meteoric water in Phuket, the GMWL and LMWL represented the global meteoric water line and the local meteoric water lines, respectively.

On the other hand, the local meteoric water line of Phuket Island (LMWL-PK, δ^2H = 7.66; δ^{18}O + 12.495, Hydro Informatics Institute, 2021, Fig. 7) is slightly higher than the global meteoric water line, indicating that the effects of the different dominant rainfall types are dominantly affected by maritime air masses. The stable isotope composition of the local rainwater on Phuket Island has a δ^2H of −30.89‰ and a δ^{18}O of approximately −5.80, which are significantly different from those of seawater (δ^2H value, −7.8‰ and δ^{18}O value, −0.787.8‰) and the global meteoric water line (GMWL; δ^2H = 8.02; δ^{18}O + 9.47, Craig, 1961), as shown in Fig. 7. While a steeper slope and higher y-intercept of the LMWL-PK compared with the GMWL might result from a tropical monsoon climate and less below-cloud secondary evaporation. Most of the δ^{18}O and δ^2H composition of the representative water samples fall along 95% of the isotopic composition of the meteoric water in Phuket (n = 223, n 95% = 213), indicating that the water has originated from local rainfall. No representative water samples plotted close to the LMWL-PK. However, the enriched isotopic composition of PKW3 and PKW4 stand outside Phuket's meteoric water range and is lower than the isotopic composition of seawater. This likely indicates the evaporative trend from the average isotopic composition of Phuket's rainwater, as shown in Fig. 7.

Noticeably, the stable isotope compositions of the waters in the different abandoned quarry ponds in the post-tin mining areas of Phuket Island are affected not only by large-scale geological features but also by local landform factors (e.g., precipitation, air temperature, and topography). However, Phuket Island is especially affected by monsoon rainfall due to its warm temperate and tropical monsoon climate, which has a dominant rainfall and warm throughout the year (27–34 °C). Therefore, the evaporation might have a significant effect on the water isotope composition of abandoned tin mine areas.

4.3. Sources of natural radioactivity and geochemical patterns

Radiogeological results of the waters and surface sediment samples taken from the representative abandoned quarry ponds in the post-tin mining areas of Phuket Island consist of radon concentrations (water samples) and natural radionuclides (surface sediment samples), as shown in Table 2 and Fig. 3. Preliminarily, the highest activity concentrations of radium-228 and thorium-228 (radioactive decay series of thorium-232), radium-226 (radioactive decay series of uranium-238), and potassium-40 are belong to a biotite–hornblende granitic area (Figs. 1b and 2e-h). Outstanding, the natural radionuclide of radon represented the highest concentration in the representative water samples from the granite quarry pond (PKW1; Fig. 2a), while the lowest level was recorded in the mine perturbation pond (PKW4; Fig. 2d). Remarkably, radon was inactive and easily dissolved gas originating from the surrounding rocks. Thus, the radon diffusion rate in the water from the different abandoned quarry ponds of Phuket Island depends on their porosity and the characteristics of the bedrock (Borchiellini et al., 1991; Bottrell, S.H., 1993; Kanellopoulos et al., 2018). However, weathering processes could have favored further local enrichment of in granitic environment (Figs. 1 and 2).

On the other hand, concentrations of the natural radionuclides (i.e., radium-226, radium-228, thorium-228 and potassium-40) in the surface sediment samples also show the highest average value in the PKS1 samples. These outcrop samples were especially observed porphyritic granite with large feldspar phenocrysts, besides quartz, and biotite and hornblende (Figs.. 1b and 2e-h). Therefore, the highest concentration of natural radionuclides in both the waters and surface sediments has been recorded in the granite quarry pond (PKW1; Figs. 1b and 2a), which is related mainly to Cretaceous biotite granites (Kata Granite). The geological assumption of radium-226 in the surface sediment samples could be attributed to a body of intrusive igneous rock that is rich in uranium (Arogunjo et al., 2009; Beg et al., 2021; Odumo et al., 2011). However, discrepancy in the activity concentrations may result from their decay products during alteration and weathering. This is supported even further by radiological data, especially for the water samples collected in the granite quarry pond (PKW1; Fig. 2a) and mine perturbation pond (PKW4; Fig. 2d), clearly showing that the radon concentration in the PKW1 water sample was significantly higher than that in the PKW4 water sample. In this case, it can be explained that radioelements in the abandoned quarry pond of Phuket Island have been significantly fractioned during weathering of bedrock (Figs. 1 and 2). Moreover, the high feldspar mineral content and high accessory mineral content was especially effective on the radioactivity concentration. Consequently, the existence of radon and radium-226 indicates that the granitic bedrock surrounding the quarry ponds that are enriched with uranium is its source.

The concentrations of radium-228 are higher than those of radium-226 in the surface sediments, indicating that this bedrock consists of thorium that is more abundant than uranium (Table 2 and Fig. 3). Consequently, the activity concentrations of thorium-228 measured in the surface sediment samples clearly emanate from thorium. The PKW2-1 sediment sample presents the highest concentrations of radium-228 and thorium-228 and is located in close proximity to Cretaceous biotite granites (Kata Granite) (Figs. 1b and 2b), with low pH values shown in the waters (Table 3). Thus, the observation suggests a significant influence of natural radionuclides that impact the water quality in the different abandoned quarry ponds of Phuket Island.

On the other hand, the Klong Marui Fault Zone (KMFZ) has been considered in studies of the local geological setting of Phuket Island as related to granite suites (i.e., Khao Prathiu, Kata Beach, Ao Nai Thon, and Khao Tosae), providing further evidence of the presence of a body of intrusive igneous rock at depth (Charusiri et al., 1993; Ridd and Watkinson, 2013; Watkinson et al., 2008). A large variation in activity concentrations in the surface sediment samples was related to the hydrogeochemical patterns of the water (Figs. 3 and 6). In particular, the concentration of potassium-40 in the surface sediments represents increased values, and the presence of ion concentrations in the waters decreased, as shown in Tables 2 and 3 For instance, the highest potassium-40 concentration was recorded in the sediment samples of the PKW1 quarry pond, but the total ion concentration in the water samples was the lowest. However, the high concentrations of natural radionuclides in the surface sediments are not related to the radionuclides contaminating waters but have been directly affected by the weathering of granitic bedrock (Bucher et al., 2017; Kanellopoulos et al., 2018; Pereira et al., 2015).

From another perspective, an investigation of hydrogeochemical data in the waters taken from the representative abandoned quarry ponds on Phuket Island represents the variation in geochemical compositions that are indicative of discontinuous transitions in their hydrogeological environments (Figs. 4–6). A clear differentiation of the local geological settings leads to the classification of four geological ponds: the granite quarry pond (PKW1; Figs. 1b and 2a), the Quaternary sediment quarry pond (PKW2 Figs. 1b and 2b), the granitic metamorphic quarry pond (PKW3 Figs. 1b and 2c), and the mine perturbation pond (PKW4 Figs. 1b and 2d). Multi-element geochemical surveys of the water samples have shown that PKW1 and PKW2 (Na–Cl type), PKW3 (mixed Ca–Mg–Cl type), and PKW4 (Mg–HCO_3 type) are nevertheless directly linked to local geological environments. Furthermore, the highest SO_4^{2-} value in the PKW4 water sample would indicate sulfate reactions together with associated cations from the contaminations of the mine perturbation zone. Compatible with the local geological setting is Permian–Carboniferous hornfels rock (Phuket Group) outcrops in both the eastern and western PKW4 pond (Figs. 1b and 2d). In contrast, the low-level correlation between cations of Ca^{2+} and Mg^{2+} and HCO_3^- in the PKW1 water samples may reflect the contribution from the dissolution of Ca^{2+} containing minerals, e.g. Ca^{2+} feldspar, in the granite quarry. This indicates that alkali metals contributed to the PKW1 water samples from reactions with Na^+ -and K^+-bearing silicate minerals. Notably, the Gibbs diagrams (Fig. 6) show that the water composition is primarily controlled by the reactions of dominant rocks and precipitated water, especially in the Quaternary sediments of the quarry pond, with the exposed minerals in the bedrock. Thus, the various elemental compositions present in the waters directly reflect the physical properties of the bedrock and geological environments around the abandoned quarry pond of Phuket Island.

Mostly, geological environments control the geochemical distribution of elements in the abandoned quarry pond of Phuket Island. Stable isotopes of δ^2H and $\delta^{18}O$ have suggested that most of the water samples are representative of lighter isotopes and have been identified as surface water bodies (Table 4 and Fig. 7). Except for the PKW4 sample, heavy isotopes are shown. However, the water characteristics could be related to physical weathering in surface bedrocks and probably linked to the residence time of water in the quarry ponds. Interestingly, geochemical variations in the water samples in the different abandoned quarry ponds of Phuket Island are probably related to rainfall that has resulted from a tropical monsoon climate. Consequently, the compatibility geochemical analysis and stable isotope compositions reflect the local geological variety that is expected to be impacted by physical weathering processes, rainfall intensity, and an active tectonic regime (Atanacković et al., 2013; Musiba and Rwiza, 2021; Salifu et al., 2020). Therefore, geochemical characteristics of the waters have been associated with the different geological environments that are used to uniquely identify geochemical patterns for each abandoned quarry pond in the post-tin mining area of Phuket Island.

4.4. Assessment of water quality for human consumption

The water samples collected from the quarry ponds in post-tin mining areas of Phuket Island are assessed using the contamination indicators and comparing them to standards (e.g., World Health Organization; WHO, 2022, European Union; EU, 2020; Din et al., 2023;

Iqbal et al., 2020; Koki et al., 2018; Rahman et al., 2023) as shown in Table 3. However, the water quality parameters are focused on the activity concentrations of natural radionuclides (e.g., radon, radium-226, radium-228, thorium-228 and potassium-40) and the key geochemical features (e.g., TDS, Ca^{2+}, Mg^{2+}, Cl^{-}, NO_3^{-}, As, and Cd) of a contaminant impact of hydrogeological setting (Tables 2 and 3). For instance, the chloride and nitrate content, all the representative water samples were below the most desirable limits both of the WHO, 2022; EU, 2020, indicating good quality water for human consumption purpose (e.g., World Health Organization; WHO, 2022, European Union; EU, 2020; Din et al., 2023; Iqbal et al., 2020; Koki et al., 2018; Rahman et al., 2023). The concentrations of sulfate and magnesium are within the prescribed limit, as well as F and NO_3^{-} contents are found within the permissible limit of WHO, also followed the EU, which is no result in the human system (e.g., gastrointestinal irritation). Moreover, the concentration of heavy elements (e.g., As, Cd, Cu, Pb, and Hg) were present well the desirable levels as shown in Table 3.

Outstanding, the quarry ponds in post-tin mining areas of Phuket Island have not been affected by seawater intrusion and other activities of a former tin mine, except the weathering and denudation of granitic bedrock has a great influence on the water quality. Therefore, an assessment of the existence of activity concentrations of natural radionuclides in water are necessary due to weathering and denudation of granitic bedrock with water-rock interaction. It may be localized recharge could transport activity concentrations of natural radionuclides into the surrounding aquifers. The natural radionuclides exist in groundwater aquifers due to the physical weathering of granitic rocks is, in turn, acting as long-term significant pollution sources to the quarry ponds. Although, in the present study, the fluctuation of radon concentration and natural radioactivity in the water samples collected from the quarry ponds of post-tin mining areas is small and desirable limit for a human consumption that fixed by the WHO (2022). A long-term plan is necessary to put more efforts on the study of water quality and activity concentration in the presented quarry ponds of post-tin mining areas in Phuket Island. Furthermore, planning for sustainable development of the quarry ponds of post-tin mining areas in Phuket Island, the water pollution prevention and control should be strictly in the discharge of urban sewage and tourism industry. As well as the vigorously promote sediment dredging in the quarry ponds of post-tin mining areas to reduce the accumulation of activity concentration in hydrological environment.

5. Conclusions

This study has been conducted to enhance the comprehensive understanding of the hydrogeological environments of different abandoned quarry ponds in post-tin mining areas of Phuket Island to minimize potential negative effects when water is used for human consumption. Such an integrated work has not been done in Phuket before, which is one of the main tourist destinations of Thailand.

Preliminarily, weathering and erosion of Cretaceous biotite granites are regarded as being responsible for the increased radon levels (water samples) and activity concentrations of natural radionuclides (surface sediment samples). However, the variability in hydrogeochemical environments is mainly controlled by water−rock interactions, precipitation reactions, and evaporation.

The activity concentrations of natural radionuclides (radium-226, radium-228, thorium-228 and potassium-40) in surface sediments are strongly dependent on the specific geological setting of each abandoned quarry pond and the type of quarry pond was significantly correlated with the activity concentration of natural radionuclides. Moreover, the radiogeological interpretation of the water and surface sediment samples indicated that the physical weathering of bedrock directly influenced the hydrological systems of the abandoned quarry ponds. Activity concentrations of a granular detrital form of these weathering products were taken from the surface sediment/water. Therefore, the activity concentration of natural radionuclides in surface sediments depends on the amount of radioactive materials in geological environments.

Although the assessment of the activity concentration of natural radionuclides leads to the determination of a general characterization of the abandoned quarry pond, long-term monitoring is still necessary to understand hydrological changes. Activity concentrations of radionuclides in the dry season could be used to determine the surface water quality based on radionuclide contamination that was not previously present in these areas. Geochemical compositions of the water tentatively reflect the richly diverse hydrogeological environments and different bedrock types of the abandoned quarry ponds. However, a variety of weathering and erosion processes continued to break down the bedrock of the quarry pond, which were factors that controlled the water composition. Especially, acid rain (rainwater at Phuket Island, pH = 4.2) slowly dissolves rocks of abandoned quarry ponds due to chemical reactions between the acid and the minerals in the rock. Hence, the proportion of solutes acquired by water–rock interactions depends on bedrock formation dominance and rock weathering. Long-term monitoring is necessary to consider the geochemical behaviors of waters, such as major and trace elements and trace metals, and the potential effects of weathering due to longer dry seasons. However, the current water quality assessment allows the pond waters for human consumption. A long-term monitoring scheme also can and will ensure this for the future.

CRediT authorship contribution statement

Wipada Ngansom: Conceptualization, Investigation, Writing – original draft. **Dumrongsak Rodphothong:** Investigation. **Thawatchai Itthipoonthanakorn:** Formal analysis, Investigation. **Saroh Niyomdecha:** Investigation, Methodology. **Helmut Dürrast:** Supervision, Writing – review & editing. **Weerawat Intaratat:** Resources. **Patchareeya Chanruang:** Resources. **Chakrit Saengkorakot:** Resources. **Monthon Yongprawat:** Investigation, Methodology.

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.

Data availability

No data was used for the research described in the article.

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