Mini Review Uranium-Thorium Decay Series in the Marine Environment of the Southern South China Sea

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Abstract

The South China Sea (SCS) is divided into two parts namely northern SCS (nSCS) and southern SCS (sSCS). The sSCS is a semi-closed system that receives rapid large water flushing from the Western Pacific Ocean and the Java Sea during the northeast and southwest monsoon events. Major natural radionuclides in the sSCS are expected to come from river water and terrestrial sediment discharge i.e., Mekong River, Chao Phraya River, Pahang River and Rajang River which contain high lithogenic and biogenic materials. A box model was developed to estimate the amount of 232Th discharge from rivers to the sSCS basin. The result shows that the total flux of 232Th entering into the sSCS was 140.3 × 10^12 Bq/km²/yr, with the highest contribution from the Pahang River followed by the Rajang River, Mekong River and Chao Phraya River. The activity concentrations of natural radionuclides presented herein should be considered useful in order to understand the geochemical behavior of natural radionuclides in marginal sea areas. The review shows that publications on natural radionuclides are still limited; therefore further research needs to be done.

Keywords: Radionuclides; Southern South China Sea; Northeast monsoon; Mekong river; Marginal sea

Introduction

Components of the uranium-thorium U-Th decay series have played an important role in the study of chemical oceanography in the southern South China Sea (sSCS) since the early 1990s. Natural radionuclides are widely used as a tool to investigate the oceanographic process that occurs in the sSCS region such as in the Gulf of Thailand [1,2], the Johor Straits [3], coastal Peninsular Malaysia [4-7], the Vietnam coast [8], the North coast of Java, Indonesia [9], Manila Bay [10,11] and the Sulu Sea [12].

The South China Sea (SCS) is divided into two parts; the northern South China Sea (nSCS) and the southern South China Sea (sSCS). The nSCS includes Taiwan, Hong Kong and the Republic of China, while countries in the SCS basin comprise Malaysia, Singapore, Thailand, Indonesia, Brunei, Indonesia, Cambodia, Vietnam and the Philippines. While four rivers feed into the sSCS; the Mekong River, the Chao Phraya River, the Pahang River and the Rajang River (Figure 1). The Mekong River is the 12th longest river in the world and drains a catchment of 790,000 km² and about 15,000 m³/s as the 8th largest water discharge [13]. This river flows for 4909 km passing through six countries; China, Myanmar, Thailand, Laos, Cambodia, and Vietnam [14]. The Pahang River is located in the Pahang basin, and at 459 km is the longest river in Peninsular Malaysia. The source of the river is the Titiwangsa main range and is a main channel of water drainage to the SCS region [15]. The longest river in East Malaysia is the Rajang River, located in northwest Borneo at about 563 km. In addition, sSCS has a unique geographical structure. Being a semi-enclosed ocean basin, there is a constant exchange of water between the sSCS and the surrounding ocean through the straits. Water is flushed from the western Pacific during northeast (NE) monsoon events and the basin receives a high input of natural radionuclide from land during southwest (SW) monsoon events [6,16]. Furthermore, the geological characteristic of the straits around the sSCS shows a unique structure (Figure 2). Mekong and Chao Phraya River which are located at the Indochina Peninsula consist mainly of Paleozoic-Mesozoic sedimentary rocks with minor intrusive and extrusive igneous rocks. While, the Pahang River which is located at the Malaysian Peninsula consists mainly of Paleozoic-Mesozoic granite and granodiorite rock [17]. The different geological characteristic surrounding the sSCS might contribute to the different behavior of radionuclides in the sSCS. Therefore, the aim of this study is to review published reports of potential sources of natural radionuclide and develop a suitable simple model to estimate the flux of natural radionuclides entering into the sSCS. This review also reports on current levels of natural radionuclides in seawater, sediment and organisms in the sSCS area.

Uranium thorium (U-Th) decay series in marine environments

There are three major actinide nuclides with a very long half-life; 238U (t½ 4.5 × 10⁹ years), 232Th (t½ 1.4 × 10⁶ years) and 226Ra (t½ 7.0 × 10⁴ years) as well as others described in other studies such as Cheng et al. and Loeff [18]. The decay process of these radionuclides produce three decay series as shown in Figure 3. The U-Th decay series begins with 238U, 232Th and 226Ra, and ends with stable isotopes of lead [18-20]. In a closed system where the growth of the parent is balanced with the decay of the daughter, the ratio between parent and daughter is equal to unity (1). However, in a natural environment such as in marine, freshwater and terrestrial ecosystems disequilibrium will occur between parent and daughter [21-23].

Generally, the members of a U-Th decay series enter marine environments through four main pathways; in-situ production of a daughter nuclide, river transport, the diffusion process through sediment and from the atmosphere (Figure 4) [20].

Most natural radionuclides are commonly absorbed onto particles, deposited on seafloors and have their own parents. For example, 234U, 236Ra and 210Po are produced from the decay of 238U, 232Th and 210Pb, respectively. Meanwhile, natural radionuclides of riverine input such as 238U, 232Th and 230Th are carried in detrital particles from source during...
The characteristics of each radionuclide in a uranium-decay series are unique and thus suitable for use as indicators of marine environmental conditions e.g., $^{210}\text{Pb}$, $^{210}\text{Po}$ and $^{234}\text{Th}$ are used as tools to estimate biological productivity the water column [25-27]. The ratio

river flow to the ocean [24]. In addition, the input of $^{218}\text{Pb}$ resulted from emissions of $^{222}\text{Rn}$ from rock and soil, and is also scavenged from the atmosphere by precipitation processes before being deposited onto the ocean surface [16].
### Uranium-238 series

| Element | Decay Product | Half-life |
|---------|---------------|-----------|
| U-238   | Pa-234        | 1.2 min   |
| Th-234  | Pa-231        | 32800 y   |
| Th-230  | Ra-226        | 1600 y    |
| Ra-228  | Ra-222        | 5.75 y    |
| Ra-224  | Ra-223        | 3.7 d     |

### Uranium-234 series

| Element | Decay Product | Half-life |
|---------|---------------|-----------|
| U-234   | Th-230        | 75400 y   |
| Th-231  | Th-228        | 1.9 y     |
| Th-223  | Ac-225        | 6.1 hr    |
| Ac-227  | Ra-223        | 11.4 d    |

### Uranium-235 series

| Element | Decay Product | Half-life |
|---------|---------------|-----------|
| U-235   | Th-232        | 3.64*10^10 y |
| Th-231  | Th-228        | 1.9 y     |
| Th-227  | Ac-225        | 6.1 hr    |
| Ac-227  | Ra-223        | 11.4 d    |

**Figure 3:** Natural uranium-thorium decay series, colored according to particle reactivity. The arrows represent decay with changes in atomic number (Z) and number of nucleons (N) indicated. All three series end with a stable lead isotope [18].

**Figure 4:** Schematic diagram showing oceanic cycles of selected members of the U and Th decay series. Horizontal arrows indicate radioactive decay characterized by a constant rate. Vertical arrows denote fluxes across the sediment-water interface and also removal from atmosphere (for 210Pb) through chemical scavenging and particle settling [18,20].
of radionuclides such as $^{230}$Th/$^{232}$Th and $^{231}$Pa/$^{230}$Th has been used to determine pollution levels and biological productivity in open oceans [28], where both $^{231}$Pa and $^{235}$U are also produced throughout the water column at a constant production ratio rate value of $^{231}$Pa/$^{230}$Th = 0.093, especially in marginal seas.

**Potential source and budget of radionuclides in the southern South China Sea**

The sSCS has a unique geographical structure as it is a semi-enclosed ocean basin with water exchanges through the straits and bay. The area is dominated by the unique characteristic of an east Asian monsoon system with northeast winds during winter (November–February) and southwest winds during summer (June–August) [29,30]. Surface water current flows from north to south during the northeast monsoon and in the reverse direction during the southwest monsoon (Figure 5). This results in large amounts of water flushing both artificial and natural radionuclides from the western Pacific Ocean and the Java Sea into the sSCS during northeast and southwest monsoon events, resulting in a high deposition of radionuclides in this region [6]. Several studies have shown that monsoon events contribute significantly to high radionuclide activities in the sSCS basin. High $^{228}$Ra and $^{228}$Ra activity has been observed in surface sediment on the eastern coast of Peninsular Malaysia; the source of which are neighboring countries and the Western Pacific Ocean during northeast monsoon events [6]. Recent studies have also shown high $^{210}$Po activity in Malaysian coastal waters as a result of haze event in dry season during southwest monsoon [16].

Besides monsoon phenomena, the SCS also have experienced with the typhoon events especially at the sSCS region [31]. However, the typhoon event is rare occurred in sSCS compared with nSCS because the SCS located at the Equator region and always immune from the natural disaster [32]. Typhoon Vamei formed in the sSCS was a rare typhoon event that occurred at sSCS in year 2001 with a wind speed more than 36 m/s has significant affecting the atmospheric and oceanic conditions [33].

In addition to the monsoon, the sSCS also receives major radionuclides from the river either in particulate, dissolved or soil forms. The SCS receives about 1600 million tons per year (Mt/yr) of detrital sediments from numerous rivers including the Pearl, the Red, and the Mekong [29]. Terrigenous materials in the SCS are mainly generated through transport from the continent via large rivers (e.g., the Mekong River, the Rajang River) representing 8.4% of estimated global fluvial sediment discharge to the world’s oceans [34]. In the sSCS, the highest sediment discharge is from the Mekong River (160 Mt/yr), while the Chao Phraya, Pahang and Rajang Rivers discharge 11 Mt/yr, 20.4 Mt/yr and 30 Mt/yr, respectively [17] (Table 1). Based on that scenario, the sSCS can be considered the largest sink for fluvial sediments in the SCS basin, and is thus expected to receive more natural radionuclides from its contributing rivers.

The riverine input of radionuclides such as $^{235}$U, $^{232}$Th and $^{228}$Ra from major rivers to the sSCS is listed in Table 1. These radionuclides are carried in the particulate load of rivers where reactions occurring

| River (area)                  | Drainage area $10^3$ km² | Sediment discharge Mt/year | $^{232}$U (Bq/kg) | $^{232}$Th (Bq/kg) | $^{228}$Ra (Bq/kg) |
|------------------------------|--------------------------|----------------------------|------------------|--------------------|-------------------|
| Mekong (Vietnam)             | 790                      | 160                        | n.a              | 50.7               | n.a               |
| Chao Phraya (Thailand)       | 160                      | 11                         | 33.33            | 48                 | n.a               |
| Pahang (Peninsular Malaysia) | 19                       | 20.4                       | n.a              | 102.5              | 56                |
| Rajang (East Malaysia)       | 50                       | 30                         | 58               | 27.8               | 45                |

Note: n.a = not available

Data sources: a) Huy and Luyen [53], b) Srisuksawad et al. [1] c) Mohamed et al. [6] d) Wan Mahmood and Yi [4], e) Yusoff and Mohamed [38], f) Wan Mahmood et al. [44], Yi et al. [43] h) (Milliman and Syvitski [13], l. Liu et al. [55]) j) Staub et al. [54].

Table 1: Drainage area, suspended sediment discharge of major rivers flowing directly into the Southern South China Sea (sSCS) and radionuclide activity.
in the river may modify radionuclide fluxes to the oceans [24]. The drainage area, sediment load and radionuclide activity data were used to estimate radionuclide fluxes from river sediments (Equation 1) to the sSCS basin using the equation below:

\[
F = \frac{C}{A} \times S \times A
\]

Where \(F\) is the fluxes of radionuclides, \(C\) is the radionuclides activity and \(A\) is the drainage area. While an equation 2 was apply to estimate the total fluxes of radionuclides contribute from four rivers into the sSCS. Where \(\Delta F\) is the total fluxes of radionuclide entering into the sSCS, \(C_{mk}\), \(C_{cp}\) and \(C_{ph}\) are the activities of radionuclide in Mekong River, Chao Phraya River, Pahang River and Rajang River, respectively. Then \(S_{mk}\), \(S_{cp}\), \(S_{ph}\) and \(S_{ph}\) are refer to the amount of sediment loading contributed from Mekong, Chao Phraya, Pahang and Rajang Rivers, respectively. Finally, \(A_{mk}\), \(A_{cp}\), \(A_{ph}\) and \(A_{ph}\) are refer to the drainage area of Mekong, Chao Phraya, Pahang and Rajang Rivers, respectively.

A simple box model has been developed to estimate total fluxes of \(^{232}\)Th discharge from the river to the sSCS (Figure 6). Total fluxes of \(^{232}\)Th is \(1.40 \times 10^5 \) Bq/km²/yr with the highest amount contributed by the Pahang River (\(110.05 \times 10^3 \) Bq/km²/yr) followed by the Rajang River (\(16.68 \times 10^3 \) Bq/km²/yr), the Mekong River (\(10.27 \times 10^3 \) Bq/km²/yr) and the Chao Phraya River (\(3.3 \times 10^3 \) Bq/km²/yr).

High inputs from the Pahang River might be related to its geological formation. The Pahang basin’s geographical location in Peninsular Malaysia has extensive granitic rock compared to other areas in Southeast Asia (Figure 7) [35]. Thorium is highly concentrated in granitic rock with a value range of 8-33 ppm [36]. During the weathering process, Th in the tetravalent state can be adsorbed onto surface minerals of granitic rock such as apatite, monazite and zircon then transported from rivers to the oceans [36,37]. The normal ratio of \(\text{Th}/\text{U}\) in most granitic rock ranges from 3.5 to 6.3 [36] while the value of \(\text{Th}/\U\) in marine sediment adjacent to Peninsular Malaysia is influenced by granitic rock.

**Study of natural U-Th decay series in the southern South China Sea: Is there enough information?**

Studies measuring natural radioactivity in various media around the sSCS have been conducted since the early 1990s. A number of expeditions such as Research on the Seas and Island (ROSES) organized by Universiti Sains Malaysia (USM) and the Prime Scientific Sailing Expedition (EPSP’09), which covered the west and east coasts of Peninsular Malaysia, the Exclusive Economic Zone (EEZ) of Sabah Sarawak, the Sulu Sea and the Celebes Sea have been held for Malaysian marine scientists. These expeditions have contributed new knowledge of radionuclide behavior in the sSCS near Malaysia [3,5,7,12,16,23,38,41-45].

Table 2-4 presents the values of natural radionuclide in sediment, sea water and biota respectively as collected in the sSCS and adjacent sea areas. The value of \(^{234}\)U and \(^{238}\)U in sediment ranges from 6.83 Bq/kg to 65.3 Bq/kg and 5 Bq/kg to 91.5 Bq/kg, respectively [1,3,38,39]. Natural thorium isotopes such as \(^{230}\)Th, \(^{232}\)Th and \(^{233}\)Th in sediments were detected in various areas in the sSCS such as the Johor Strait, the Vietnam coast and on the Malaysian coast with values ranging from 1.83 Bq/kg to 262.83 Bq/kg, 9 to 62.83 Bq/kg and nd to 200 Bq/kg, respectively [1,3,6,44,46]. The values of \(^{228}\)Ra and \(^{226}\)Ra in sediment from the sSCS ranged from 2.9 Bq/kg to 64 Bq/kg and 23 to 130 Bq/kg, respectively [1,4,6,43]. In addition, high \(^{232}\)Th activity was detected in sediment in the sSCS region (~262.83 Bq/kg) [3] which might due to marginal sea characteristics where the ocean receives more input of \(^{232}\)Th from the straits surrounding the sea. Furthermore, a high organic matter contents in sediment also reported at the sSCS e.g., western coast of Borneo which can influence the concentration activity of natural radionuclides in sediments. A strong statistical correlation value between organic matter contents with natural radioisotopes of uranium such as \(^{234}\)U (\(r^2=0.959, p<0.01\)) and \(^{238}\U (r^2=0.904, p<0.01\) are well discussed by Yusof and Mohamed [38]. The adsorption of uranium with organic matters will occurred through the process of ion exchange for formation the stable of U(IV) in surface sediments [22,47].
The activity of isotopes in sediments in the sSCS is generally higher compared to other regions such as the Red Sea and the north eastern coast of India, thus further investigation is essential to determine the various sources of radionuclide entering the sSCS region.

In sea water, high values of $^{226}\text{Ra}$ and $^{238}\text{Ra}$ isotopes were detected in the EEZ of Peninsular Malaysia with values of up to $2.7 \times 10^3 \text{ mBq/L}$ and $3.5 \times 10^3 \text{ mBq/L}$ respectively as published by Amin et al. (Table 3) [48]. A high value of radium isotopes in sea water corresponds to the high activity value of radium isotopes detected in surface sediments from the east coast of Peninsular Malaysia [6]. In addition, oil and industry gas contribute up to almost 560 Bq/kg of $^{226}\text{Ra}$ from sludge discharge to the marine environment [49]. Radium $^{226}\text{Ra}$ isotope activity in sea water was higher in the SCS compared to the entire South China Sea and western Pacific Ocean regions due to high radionuclide depositional fluxes from enhanced dry precipitation caused by haze events [16].

On the other hand in marine organisms, high $^{210}\text{Po}$ and $^{234}\text{Th}$ values were detected in green mussels collected from the Johor Strait with average values of 150 Bq/kg and 388 Bq/kg. It is plausible that the Straits of Johor might be liable to certain levels of radionuclide contamination from exiting natural radionuclides and anthropogenic activity inputs [50]. Furthermore, concentrations of $^{238}\text{Ra}$ in fish (0.80-2.13 Bq/kg) from the eastern coast of Peninsular Malaysia [4] also show high values

| Radionuclide | Samples | Technique | Range values (average) Bq/kg | Country (Area) | References |
|--------------|---------|-----------|-----------------------------|----------------|------------|
| $^{234}\text{U}$ | Sediment core (n=22) Alpha detector | 21.12-65.3 | Thailand gulf | [1] |
| | Sediment core (n=2) Alpha detector | 6.83-62.17 | Malaysia (Borneo port) | [38] |
| | Sediment core (n=25) INAA | 30-91.5 (65.0) | Malaysia (Johor Strait) | [3] |
| | Sediment core (n=22) Alpha detector | 20.67-63.67 (33.33) | Thailand gulf | [1] |
| $^{238}\text{U}$ | Surface sediment (n=51) ICP-MS | 5-50 (23) | Thailand (eastern coast of gulf) | [39] |
| | Sediment core (n=2) Alpha detector | 8.5-58 | Malaysia (Borneo port) | [38] |
| $^{226}\text{Ra}$ | Surface sediment (n=51) HGPe detector | 2.9-53.2 | Thailand (eastern coast of gulf) | [39] |
| | Sediment core (n=16) HPGe detector | 16-46 (30) | Malaysia (EEZ of Peninsular) | [4] |
| | Surface sediment (n=31) HPGe detector | 16-21 | Malaysia (EEZ of Sabah and Sarawak) | [43] |
| | Surface sediment (n=15) Alpha detector | 8-48 | Malaysia (East coast of peninsular) | [6] |
| | Surface sediment (n=18) HPGe detector | 13-64 | Malaysia (West coast of peninsular) | [6] |
| $^{230}\text{Th}$ | Sediment core (n=16) HPGe detector | 28-57 (56) | Malaysia (EEZ of Peninsular) | [4] |
| | Surface sediment (n=31) HPGe detector | 23-45 | Malaysia (EEZ of Sabah and Sarawak) | [43] |
| | Surface sediment (n=15) HPGe detector | 12-130 | Malaysia (East coast of peninsular) | [6] |
| | Surface sediment (n=18) HPGe detector | 36-89 | Malaysia (West coast of peninsular) | [6] |
| $^{232}\text{Th}$ | Sediment core (n=3) Alpha detector | 23.17-164 | Malaysia (marine port of Sabah, Labuan and Klang) | [46] |
| | Surface sediment (n=31) Alpha detector | 9-12 | Malaysia (EEZ of Sabah) and Sarawak | [44] |
| | Sediment core (n=22) Alpha detector | 22.17-62.83 | Thailand gulf | [1] |
| | Sediment core (n=31) Alpha detector | 6.8-27.8 | Malaysia (EEZ of Sabah and Sarawak) | [44] |
| | Sediment core (n=22) Alpha detector | 4-108 (48) | Thailand gulf | [1] |
| | Sediment core (n=25) INAA | 30.67-262.83 (86.67) | Malaysia (Johor Strait) | [3] |
| | Sediment core (n=3) Alpha detector | 1.83-153.67 | Malaysia (marine port of Sabah, Labuan and Klang) | [46] |
| | Surface sediment (n=15) Alpha detector | 65-145 | Malaysia (East coast of peninsular) | [6] |
| | Surface sediment (n=18) Alpha detector | 35-59 | Malaysia (West coast of peninsular) | [6] |
| $^{234}\text{Th}$ | Sediment core (n=33) HPGe detector | n.d-200 | Vietnam (northern coast) | [55] |
| | Sediment core (n=25) HPGe detector | n.a | Indonesia (north coast of Java Sea) | [9] |
| $^{210}\text{Po}$ | Sediment core (n=13) Apha detector | 2.33-141.4 | Malaysia (EEZ of Sabah) | [56] |
| | Sediment core (n=22) Alpha detector | 11.67-143.33 | Thailand gulf | [1] |
| | Sediment core (n=10) Alpha detector | 22-110 | Philippines (area affected by toxic harmful algal bloom (HAB)) | [11] |

Note: EEZ = exclusive economic zone; n.d = not detected (below detection limit); n.a = not available, HPGe= High purity vertical germanium detectors, INAA= Instrumental neutron activation analyses

Table 2: Range values of radionuclides in marine sediment in the southern South China Sea area.
compared to other areas such as fish from Japan (226Ra: 0.008 Bq/kg) and Puget Sound, USA (226Ra: 0.003-0.75 Bq/kg) [4]. In addition, 210Po and 210Pb activity in the west coast of Peninsular Malaysia shows high values compared to 226Ra [23,51]. Furthermore, high activity values of 231Pa and 231Pb detected in zooplankton as published by Mohamed and Kuan (1996) P. Radiometric dating of estuarine sediments from the Rajang River, Mekong River and Chao Phraya River. The activity values of natural radionuclides in organisms, sediment and seawater in the sSCS were also compiled and reviewed. Unfortunately, there is limited data available on the distribution and behavior of some natural radionuclides such as 231Pa, 234Th, 210Bi, 7Be and 10Be in the sSCS. It is strongly recommended that further and more complete research is undertaken to study the behavior of these radionuclides in the sSCS [58].

Conclusion

This review discusses potential sources and budgets of natural radionuclides in the sSCS. The total flux of 232Th discharge to the sSCS was successfully estimated from the box model with a value of 140.3 × 10^3 Bq/km²/yr. The highest flux of 232Th contributed to the sSCS is from the Pahang River with a value of 110.05 × 10^3 Bq/km²/yr followed by the Rajang River, Mekong River and Chao Phraya River. The activity values of natural radionuclides in organisms, sediment and seawater in the sSCS were also compiled and reviewed. Unfortunately, there is limited data available on the distribution and behavior of some natural radionuclides such as 231Pa, 234Th, 210Bi, 7Be and 10Be in the sSCS. It is strongly recommended that further and more complete research is undertaken to study the behavior of these radionuclides in the sSCS.

Acknowledgements

The authors would like to thank the Ministry of Science, Technology and Innovation (MOSTI), for providing the research grant (04-01-02-SF0801). Thanks are also due to all the laboratory members and staff of Pusat Pengajian Sains Sekitaran dan Sumber Alam, Faculty of Science and Technology, UKM.

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