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^{226}Ra and ^{228}Ra in Water Column of Peninsular Malaysia: Southern South China Sea

CHE ABD RAHIM MOHAMED*, NIOO SIEW YEW and ZAHARUDDIN AHMAD†
Marine Ecosystem Research Center, Faculty of Science and Technology
Universiti Kebangsaan Malaysia, 43000 Bangi, Selangor, Malaysia
E-mail: carmohd@pkrisc.cc.ukm.my; mohamed6566@yahoo.com

The concentration activities of ^{226}Ra and ^{228}Ra were determined at ten stations around Pulau Redang, Malaysia, where high dissolved activity of ^{228}Ra was found at the coastal stations. Meanwhile, activities of ^{226}Ra and ^{228}Ra isotopes in the suspended materials were slightly similar with ^{228}Ra/^{226}Ra ratio ranged from 0.88-1.86. High distribution coefficient (K_d) of radium isotopes found around Pulau Redang indicating that radium are strongly absorbed onto particulate phase. The K_d values of ^{226}Ra and ^{228}Ra were ranged from 0.78×10^5 L g^{-1} to 5.56×10^5 L g^{-1} and from 0.21×10^5 L g^{-1} to 1.86×10^5 L g^{-1}, respectively. The impact caused by the amount of suspended particle material (< 10 mg L^{-1}) on the particle concentration was not found during this study.

Key Words: ^{226}Ra, ^{228}Ra, Suspended particulate matter, Dissolved phase, Distribution coefficient.

INTRODUCTION

Radium is one of the particular environmental concern because its possible risk in significantly increasing the internal radiation dose of both individual and whole population groups[1]. There have only four naturally occurring radium isotopes such as ^{223}Ra (11.4 d), ^{224}Ra (3.64 d), ^{226}Ra (1622 years) and ^{228}Ra (5.75 years)[1-3]. Then in marine environments, ^{226}Ra and ^{228}Ra are readily detectable, soluble and used as a tracer for water circulation[4]. They occur in the seawater primarily by diffusion from sediment *via* the interstitial water[5].

Pulau Redang is located at the southern South China Sea has gazette as a marine park by Malaysian government. This area is reached with various types of coral and attracting for local and foreign tourists. During monsoon season, this area has received large land input from the northeast and southwest monsoons. Therefore the information on radium before and after monsoons in Malaysian water is not well published yet. Then, the purpose of this study is to determine the concentration, distribution and behaviour of ^{226}Ra and ^{228}Ra isotopes in Malaysia waters especially at the Southern South China Sea.

†Malaysia Institute of Nuclear Technology, Bangi, Kajang-43000, Malaysia.

EXPERIMENTAL

About 15 L of surface seawater samples were collected at 10 stations around Pulau Rexdang on July 2003 (Fig. 1). The *in situ* parameters such as salinity, specific conductivity, pH and dissolved oxygen (DO) were measured using calibrated portable meter (Model: YSI-SCT 6810).

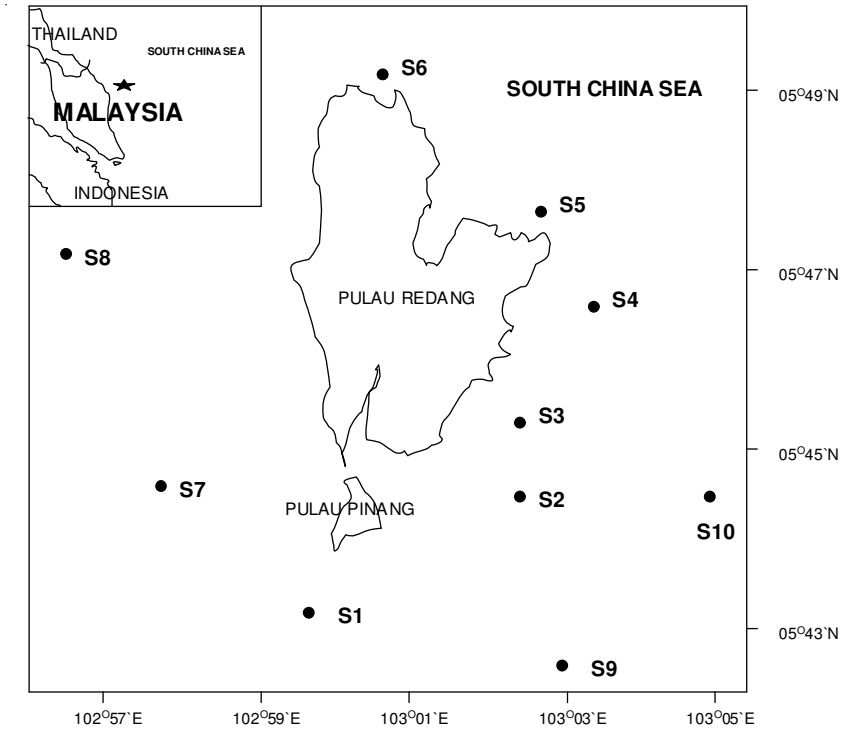

Fig. 1. Sampling stations were conducted around Pulau Redang, Malaysia

In the laboratory, water samples were filtered through the 0.45 μm pore size of pre-weighed membrane filter paper with a flow rate less than 10 mL min^{-1}. The filtrate was acidified with conc. HNO_3 to pH less than 2 and continued spike with 25 and 20 mg mL^{-1} of iron carrier and barium carrier, respectively, as analytical recovery. Sample stirred vigorously before stand 1 h and continue increasing pH to10 with ammonia solution and Na_2CO_3. Then siphon out the supernatant from the filtrate and dissolve the carbonate precipitate with HNO_3 and $HClO_4$. The carbonate aqueous was heating on the hotplate at 70ºC to remove carbon dioxide. After removed carbon dioxide from the aqueous, increasing the pH to 8 with ammonia solution and collect the precipitated material. Purification and counting radium nuclides were performed using the cation exchange resin

and Gross α/β Spectrometer (Model: LB 5100-W; Tennelec), respectively[6]. The same analytical procedure also analyzed for suspended particular matter samples and standard reference material as analyses quality control.

RESULTS AND DISCUSSION

Behaviour of radium in dissolved and particulate phases

The activities of dissolved ^{226}Ra and ^{228}Ra obtained during the cruise at Pulau Redang are varied from station to station (Fig. 1) with ranged from 2.08 to 12.44 mBq L^{-1} for ^{226}Ra. But the concentration activity of ^{226}Ra was about 2.52 and 1.30 mBq L^{-1} at Straits of Malacca and South China Sea, respectively[7]. Meanwhile, ^{226}Ra activities at Bay of Bengal[5,8] varied from 2.0-193.0 and Chao Phraya[9] in ranged of 2.1-4.3 mBq L^{-1}.

Dissolved ^{228}Ra obtained in this study are fluctuated in range of 6.95-33.53 mBq L^{-1} and slightly higher than other studies because most of ^{226}Ra are derived from the continental shelves[7]. Whereas ^{228}Ra activities at Bay of Bengal, Chao Phraya, Straits of Malacca and South China Sea are 3.0-7.6, 2.4-18.4, 8.97 and 2.98 mBq L^{-1}, respectively[5,7,9].

In this study, there is no significant statistical trend between radium isotopes and salinity, where a maximum activity at salinity 31.05 psu was due to the behaviour of radium tends to desorb at mid-salinity range (Fig. 2a). Then from the previous studies[10,11], radium isotopes also have tendency to desorb from the particulate phase at high salinity and slightly increased in statistical proportional with salinity value ranged from 3 to 32 psu. In other case especially found in this study, there are no significant correlation between particulate radium with salinity because radium isotopes might be desorbed as Ra^{2+} when the particulate radium was attacked by high ionic water (high salinity). These Ra^{2+} ions would be attach with anions, such as sulphate, hydroxide or associated with earth alkaline which were causing the concentration of particulate radium were not consistently related with salinity (Fig. 2b).

Activities of radium in particulate phase obtained during the cruise were ranging from 4.32-10.29 and 5.12-9.82 Bq g^{-1} for ^{226}Ra and ^{228}Ra, respectively. Since the activities of both radium nuclides in particulate phase surrounding Pulau Redang are almost similar, we assumed that was little external inputs of both isotopes came from the neighboring area and not caused by tourism activities.

Activity ratio and distribution coefficient

The activity ratios of ^{228}Ra/^{226}Ra in dissolved and particulate phases from Pulau Redang are varied in range of 1.88 to 7.00 and from 0.88 to 1.86, respectively, where ^{228}Ra are more enriched in dissolved phase and opposite occur for ^{226}Ra (Fig. 3). Then the ^{228}Ra/^{226}Ra ratio in dissolved

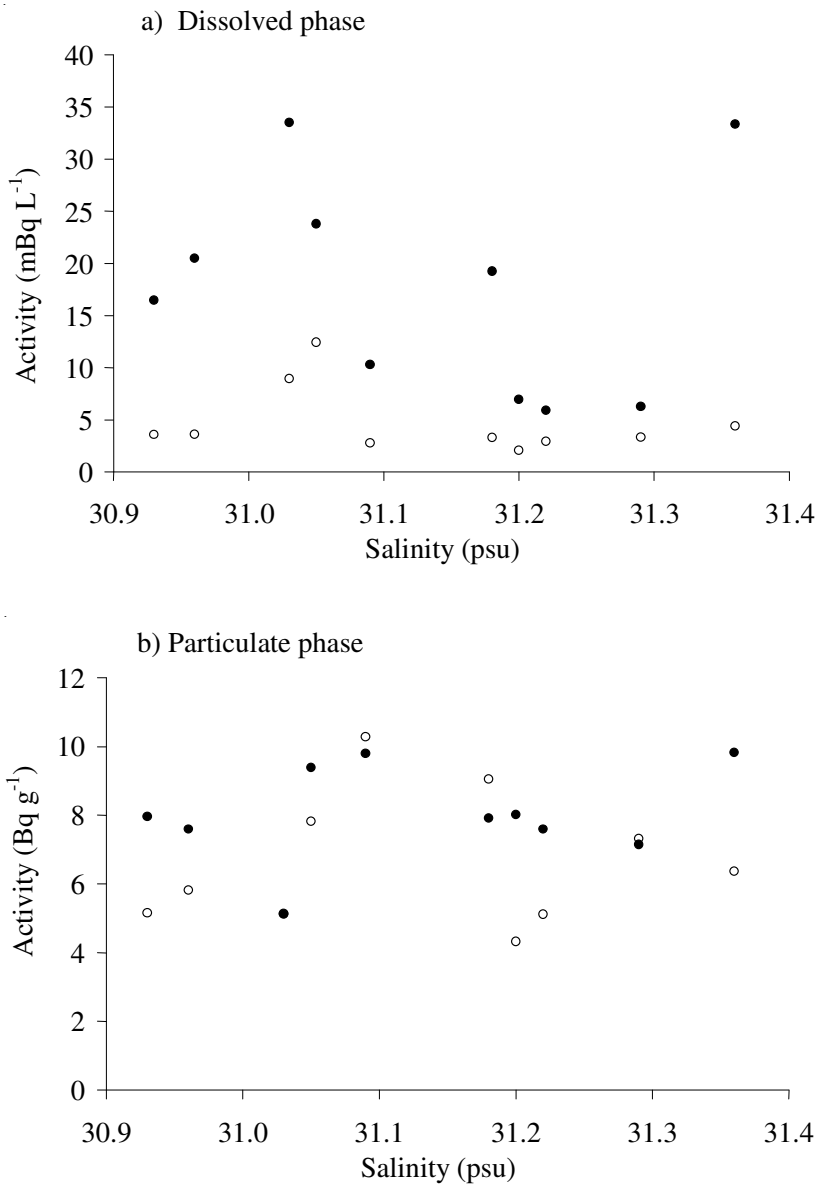

Fig. 2. Distribution of radium in dissolved and particulate phase obtained during sampling, where open and close circles are for ^{226}Ra and ^{228}Ra, respectively

phase at stations S9 and S10 are lower than other stations resulting from the depletion of ^{228}Ra in dissolved phase which is related to the solubility of uranium over than thorium will cause a limited transport of ^{228}Ra to the open sea[12].

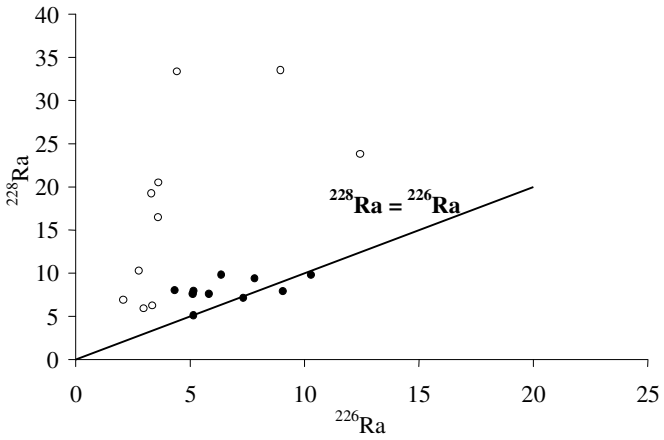

Fig. 3. Correlation between ^{228}Ra and ^{226}Ra in dissolved (circle; mBq L^{-1}) and particulate (closed; Bq g^{-1}) phases obtained during sampling

High solubility of uranium would also contribute a dissolved of ^{226}Ra into coastal seawater, where the contribution concentrations of ^{226}Ra desorbed from suspended sediments are little[9,10,13]. This was due to the long half-life of ^{226}Ra (1600 years) compare to their residence time in sea-water. Thus, dissolved ^{226}Ra will desorbed from the suspended sediments would transport to the open sea before it mixing with the coastal water, but low solubility of ^{232}Th was causing the retention of dissolved ^{228}Ra at coastal water. Besides this, the shorter half-life (5.75 years) of ^{228}Ra would cause the mixing of ^{226}Ra desorbed from suspended sediments with coastal waters. Consequently, highly activity ratio in dissolved phase would be found at coastal stations especially stations S3 and S6. On the other hand, the relationship correlation between ^{228}Ra and ^{226}Ra in the particulates are located closed to the line of ^{228}Ra equal ^{226}Ra was indicating that the source of particulate found at study area are almost same from similar origin (Fig. 3).

The distribution coefficient (K_d) is a measure the tendency of an element to be associated and transported with the particulate phase[14]. It is also widely used to understand and determine the eventual fate of metals and radionuclides released into the aquatic environment. In this study, the distribution coefficient (K_d) is defined as;

$$K_d = \frac{[A]_p}{[A]_d \times SPM}$$

where the $[A]_p$ is the activity of particulate ^{226}Ra or ^{228}Ra (Bq g^{-1}), $[A]_d$ is the activity of dissolved ^{226}Ra or ^{228}Ra (Bq g^{-1}) and SPM is the concentration of total suspended particulate matter (g L^{-1}).

The K_d values of ^{226}Ra and ^{228}Ra are ranged from 0.78×10^5 to 5.56×10^5 L g^{-1} and 0.21×10^5 to 1.86×10^5 L g^{-1}, respectively. High K_d values of both radium isotopes are indicated that radium nuclides are highly reactivity and adsorbed strongly onto the suspended particulate in marine environment.

The plot of K_d values *vs.* contents of suspended particle was insignificant with weak statistical correlation ($R^2 < 0.5$), where less contents of suspended (< 10 mg L^{-1}) might be the factor causing the particle concentrations effect had not seem in this study as well reported by Baskaran and Santschi[15]. The lower concentrations of suspended particles in the aquatic environment might be decrease the adsorption of radionuclide onto the particulate phase.

Conclusions

The activities of ^{228}Ra measured during this cruise are two times higher than others study and most radium isotopes were strongly adsorbed into suspended particles. Particle concentration effect is not found for samples containing a little amount of suspended particles (< 10 mg L^{-1}).

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