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^{226}Ra and ^{228}Ra tracer study on nutrient transport in east coastal waters of Hainan Island, China

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Abstract: Material fluxes (e.g., nutrients) from coastal waters to offshore areas play an important role in controlling the water quality of the adjacent sea areas not only by increasing nutrient concentration but also by changing nutrient structures. In this study, naturally occurring isotopes, ^{226}Ra and ^{228}Ra, were measured with the alpha spectrometry in the Wenjiao-Wenchang and Wanquan estuaries and adjacent sea areas along the east coast of Hainan Island. The excess ^{226}Ra and ^{228}Ra activities were observed by comparison with the values derived from the conservative mixing of freshwater and seawater end-members in both estuaries. Using a one-dimensional diffusion model, the horizontal eddy diffusion coefficient of 3.16×10^5 cm^2/s, for nutrients diffusing from their sources, was derived from ^{228}Ra activities. Consequently, the corresponding nutrient fluxes flowing into the coastal waters were assessed. The results can provide useful information for the study of the mixing and exchange processes of coastal waters as well as dissoluble pollutant transport in this sea area.

Key words: *Ra isotope; ^{226}Ra and ^{228}Ra tracers; horizontal eddy diffusion coefficient; nutrient flux; east coastal waters of Hainan Island*

1 Introduction

Radium (Ra) is an alkaline earth element that is strongly absorbed onto particles and sediments in rivers. With the increase of the ionic strength and the decrease of the particle concentration in estuary regions, most Ra exists as dissolved Ra^{2+} in saline, low turbidity seawater due to desorption from suspended particles and diffusion from sediments (Key et al. 1985; Yang et al. 2002; Lee et al. 2005; Gonneea et al. 2008). Submarine groundwater discharge with high Ra concentration also seeps into the estuarine and nearshore waters (Burnett et al. 1990; Krest et al. 1999; Moore 1997, 1999; Yang et al. 2002; Peterson et al. 2008; Nakada et al. 2011).

The coastal ocean exchanges large amounts of nutrient and energy with the open ocean

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because virtually all terrigenous materials, including water, sediments, dissolved particulate nutrients, and trace elements, enter the coastal ocean with surface or sub-surface runoff. However, the transport of these chemicals from the coast to the open ocean is difficult to quantify because of the complex temporal and spatial variability within these systems (Moore 2000). Ra is a useful tool for tracing the water movement rate in the ocean and, consequently, for investigating the characteristics of material (e.g., nutrient and pollutant) transport to adjacent sea areas (Moore 2000; Charette et al. 2007; Colbert and Hammond 2007).

Okubo (1971) investigated the relationship between two parameters of diffusion, the time and length scales, using the examined data from instantaneous dye release experiments in the upper mixed layer of the North Sea. The study provided a practical means for predicting the horizontal diffusion coefficient of substances from an instantaneous source. Alternatively, Ra isotopes are considered good tracers to study biogeochemical processes on different time scales in the coastal and adjacent sea areas (Kaufman et al. 1973; Yamada and Nozaki 1986; Schmidt and Reyss 1996; Moore 2000; Charette et al. 2007; Colbert and Hammond 2007; van Beek et al. 2008). Mixing parameters like the horizontal eddy diffusion coefficient in coastal regions were derived from the ^{228}Ra activity measured in surface layers (Somayajulu et al. 1996; Huh and Ku 1997; Rengarajan et al. 2002; Men et al. 2006). In recent years, there have been few studies on the horizontal eddy diffusion coefficient in the South China Sea and the corresponding nutrient transport from the coast to adjacent sea areas. Huang et al. (1997) studied the distribution of ^{228}Ra in the surface water of the northeastern South China Sea, and estimated the horizontal eddy diffusion coefficient based on the exponential relationship between the ^{228}Ra activity and the distance offshore.

There are diverse ecosystems in the east coastal waters of Hainan Island. According to the surveys in recent years, the coral reef ecosystem in some regions is being degraded and the living space is shrinking due to the transport of terrigenous pollutants to the local waters. This work aimed to study the distribution characteristics of ^{226}Ra and ^{228}Ra in estuaries and adjacent sea areas along the east coast of Hainan Island, China. The behavior of the two Ra isotopes was investigated during estuarine mixing. A one-dimensional diffusion model was applied to fit the ^{228}Ra data for estimating the horizontal eddy diffusion coefficient in the coastal area.

2 Sampling and methods

2.1 Study site

Hainan Island is located in the southern part of China, across the Qiongzhou Strait from the Leizhou Peninsula of Guangdong Province. On its eastern side, it borders the South China Sea (Fig. 1). Our study area was focused on the east coastal area of Hainan Island, including two estuaries, the Wenjiao-Wenchang and Wanquan estuaries. The Wenjiao and Wenchang rivers, with total lengths of 56 km and 37 km, respectively, directly connect with

the Bamen Bay, which has a surface area of 40 km^2 and an average water depth of 1 to 2 m. The two rivers have a total runoff of 8×10^8 m^3/year and a mean suspended sediment load of 1.0×10^5 t/year (Wang et al. 2006). The Wanquan River is the third largest river in Hainan Province, with a total length of about 160 km and a drainage area of about 3.6×10^3 km^2. The runoff of the Wanquan River shows a significant seasonal variation, with an annual average runoff of 5.2×10^9 m^3/year and a mean suspended sediment load of 3.9×10^6 t/year (Wang 2002; Wang et al. 2006). In our study area, a significant feature is the notable diversity of habitats, such as rocky shores, sandy beaches, mangroves, sea grass beds, and especially, coral reefs. Frequent tropical cyclones strike the island in August and September every year (Mao et al. 2006).

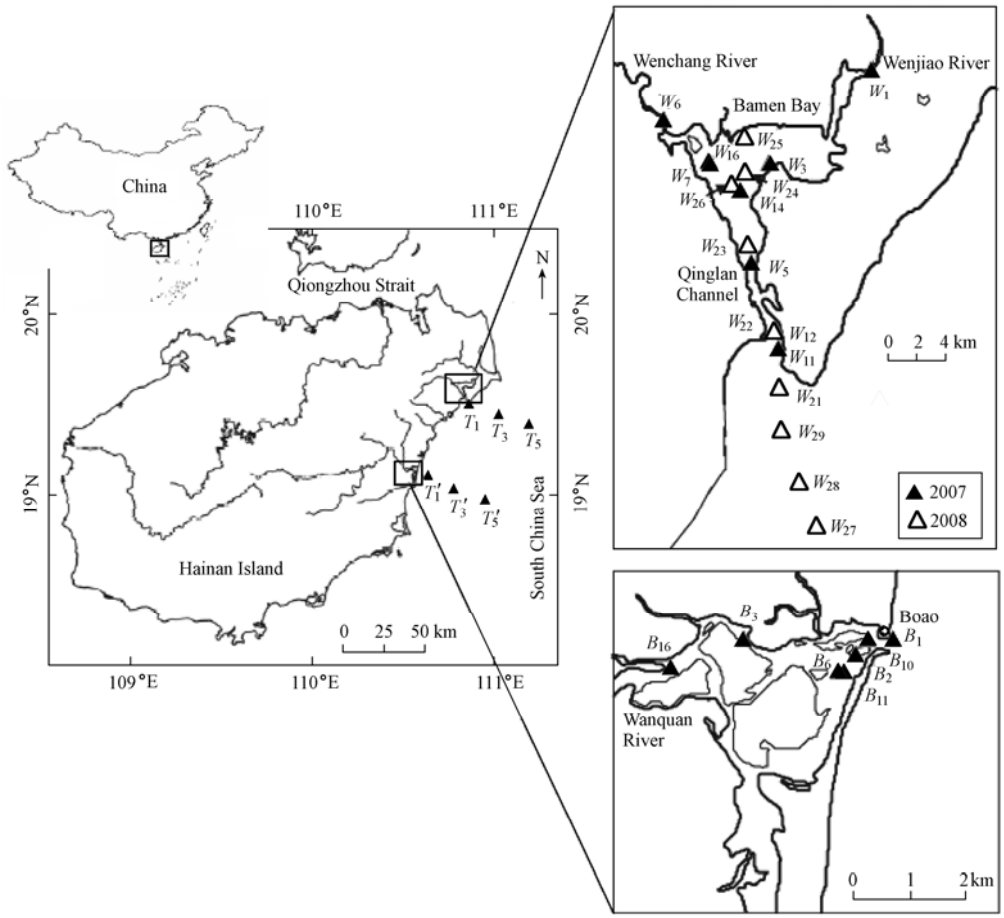

Fig. 1 Sampling sites in study area along east coast of Hainan Island

2.2 Methods

Most of the coastal and estuarine water samples in this study were collected in August 2007. We also collected samples at the sampling sites W_{21}-W_{29} in the Wenjiao-Wenchang Estuary in July 2008 after a typhoon event.

Samples were collected and enriched as follows: (1) About 20 L of water was collected using a submerged pump installed at a water depth of 0.5 m and filtered immediately through cellulose filters (pore size: 0.45 μm). It was then stored in pre-cleaned polyethylene containers. (2) 4.00 dpm of ^{229}Th-^{225}Ra solution (Eckert and Ziegler Isotope Products, 7229) was added as an internal tracer while the sample solution was stirred and acidified to reach a pH value of 2, and then allowed to stand for about 6 h to equilibrate with seawater. (3) NH_4OH, $KMnO_4$ and $MnCl_2$ solutions were added to form an amorphous dark brown suspension of MnO_2 at a pH value of 9. (4) The suspension was stirred for 0.5 to 1 h, and allowed to settle for more than 12h. (5) The precipitate was separated from the supernatant and dissolved in HNO_3 and H_2O_2 solutions (Dimova et al. 2007).

The following separation and purification procedures were described by Hancock and Martin (1991). Briefly, the $Pb(NO_3)_2$ solution, dilute H_2SO_4, and solid K_2SO_4 were added to the acidic solution mentioned above to form the $Pb(Ra)SO_4$ co-precipitate. The co-precipitate was centrifuged and redissolved in the ethylene diamine tetraacetic acid (EDTA) solution. Then, this solution was transferred through an anion exchange column (DOWEX 1X8-200, 100-200 meshes, chloride form, 50 mm in height, and 7 mm in diameter) for desulfidation, and this column was washed in 13-mL EDTA solution with a concentration of 0.01 mol/L and a pH value of 10. After that, thorium (Th) and actinium (Ac) were retained on the column (Th separation). Later, the solution was transferred from the anion column onto a cation exchange column (DOWEX 50WX8-200, 200-400 meshes, 80 mm in height, and 7 mm in diameter) to elute plumbum (Pb) and residual Th and Ac. Finally, Ra isotopes were electrodeposited onto a stainless-steel disc and determined by the alpha spectrometry (Canberra 7200-08). ^{226}Ra is an alpha-emitting isotope, while ^{228}Ra is a beta-emitting isotope. Hence, the ^{226}Ra activity was calculated immediately after the counting, but the ^{228}Ra activity could only be obtained indirectly through the activity of its daughters ^{228}Th and ^{224}Ra after the disc was stored for more than six months.

3 Results and discussion

We present the ^{226}Ra and ^{228}Ra data and $^{228}\mathrm{Ra}/^{226}\mathrm{Ra}$ activity ratio (henceforth denoted as [228/226]) in two estuaries (sampling sites W_1-W_{16} in the Wenjiao-Wenchang Estuary and B_1-B_{23} in the Wanquan Estuary) and offshore sea waters (sampling sites T_1, T_3, and T_5 on transect T, and T_1', T_3', and T_5' on transect T') in August 2007 and in the Wenjiao-Wenchang Estuary (sampling sites W_{21}-W_{29}) in July 2008 after a typhoon event. The sampling site B_{23} is on upper reach of the Wanquan River and not shown in Fig. 1. The detailed sampling information and the Ra isotope activities are shown in Table 1.

Table 1 Sampling sites, water depth, salinity, and analytical results of Ra isotopes in estuaries and adjacent sea areas along east coast of Hainan Island, China

Sampling site	North latitude (°)	East longitude (°)	Salinity	Depth of sampling site (m)	^{226}Ra (dpm/100L)		^{228}Ra (dpm/100L)		[228/226]	
					^{226}Ra activity	Error	^{228}Ra activity	Error	Ratio	Error
W_1	19.648	110.903	0		23.99	0.84	28.23	1.36	1.18	0.07
W_3	19.612	110.836	17.9	1.8	25.94	0.82	139.50	4.19	5.38	0.25
W_5	19.573	110.823	22.6	5.9	23.66	0.72	126.10	3.70	5.33	0.25
W_6	19.573	110.767	0	2.1	23.92	0.92	34.69	1.51	1.45	0.09
W_7	19.613	110.795	9.5	3.8	33.60	1.51	121.40	5.37	3.61	0.21
W_{11}	19.538	110.841	28.5	5.9	18.34	0.69	59.20	2.19	3.23	0.19
W_{12}	19.545	110.840	33.3	5.5	12.98	0.44	38.94	1.39	3.00	0.19
W_{14}	19.602	110.815	27.4	1.9	20.38	0.60	117.10	3.33	5.75	0.27
W_{16}	19.614	110.794	8.9	1.8	34.32	0.99	124.10	3.58	3.62	0.15
W_{21}	19.523	110.842	7.5	4.7	17.26	0.66	71.09	3.25	4.12	0.24
W_{22}	19.545	110.838	2.2	4.8	22.77	0.80	55.47	2.60	2.44	0.14
W_{23}	19.580	110.821	0.7	6.2	15.32	0.54	45.47	2.44	2.97	0.19
W_{24}	19.609	110.818	0.5	1.8	15.39	0.52	28.98	2.43	1.88	0.17
W_{25}	19.624	110.818	0.1	1.9	17.00	0.79	27.49	2.31	1.62	0.16
W_{26}	19.605	110.810	0.4	2.1	23.80	0.97	36.88	3.68	1.55	0.17
W_{27}	19.467	110.867	27.7	25.0	12.20	0.50	33.34	1.77	2.73	0.18
W_{28}	19.485	110.855	25.6	18.0	12.79	0.62	24.58	1.73	1.92	0.16
W_{29}	19.506	110.843	16.1	9.0	14.87	0.60	46.27	2.33	3.11	0.20
B_1	19.156	110.587	33.9	3.0	12.79	0.42	30.93	1.08	2.42	0.15
B_2	19.153	110.581	2.4	5.0	15.30	0.52	28.03	1.14	1.83	0.11
B_3	19.156	110.563	0	1.4	13.57	0.58	17.57	1.02	1.29	0.10
B_6	19.150	110.578	7.0	1.4	18.93	0.57	32.64	1.30	1.72	0.10
B_{10}	19.155	110.583	12.2	3.2	15.37	0.56	23.34	1.11	1.52	0.10
B_{11}	19.150	110.579	6.2	1.2	17.84	0.59	32.61	1.27	1.83	0.11
B_{16}	19.151	110.549	0	1.3	7.74	0.50	8.37	0.70	1.08	0.13
B_{23}	19.252	110.438	0		10.20	0.57	9.97	0.85	0.98	0.11
T_1	19.508	110.865	35.6	18.0	10.60	0.75	18.32	1.48	1.73	0.19
T_3	19.453	111.024	35.6	43.0	9.01	0.55	12.51	0.93	1.39	0.13
T_5	19.396	111.188	35.6	68.0	9.79	0.56	12.18	0.91	1.24	0.12
T_1'	19.115	110.634	35.2	14.0	14.33	1.23	22.14	2.18	1.55	0.20
T_3'	19.044	110.778	35.6	70.0	8.29	0.52	11.77	0.91	1.42	0.14
T_5'	18.977	110.947	35.6	87.0	9.69	1.04	19.09	2.29	1.97	0.32

Note: Quoted uncertainties are ±1σ.

3.1 Distribution of ^{226}Ra and ^{228}Ra in Wenjiao-Wenchang Estuary

Sampling sites in the Wenjiao-Wenchang Estuary are shown in Fig. 1. In August 2007, the ^{226}Ra and ^{228}Ra activities were 13.0-34.3 dpm/100L and 34.7-140 dpm/100L, respectively. In July 2008, as influenced by the typhoon event, the ^{226}Ra and ^{228}Ra activities decreased to 12.2-23.8 dpm/100L and 24.6-71.1 dpm/100L, respectively. The low Ra concentrations may be due to the strong turbulent mixing and seawater dilution caused by the typhoon. As expected, the maximum desorption event in July 2008 occurred in the region with a lower salinity range than that of the region in August 2007 (Figs. 2(a) and 2(c)). ^{226}Ra and ^{228}Ra concentrations were well above the conservative mixing line between the river and seawater end-members, especially in August 2007. This distinct convex curvature indicated the excess addition of Ra into the dissolved phase. Ra sources include the desorption from riverine suspended particles, the diffusion from bottom sediments, and most importantly, the submarine groundwater discharge (Hussain et al. 1999; Krest et al. 1999; Moore 1999; Yang et al. 2002; Peterson et al. 2008; Nakada et al. 2011).

[228/226] followed the same pattern as the ^{228}Ra activity in the Wenjiao-Wenchang Estuary and varied greatly with the salinity, as shown in Figs. 2(a) through (d). In August 2007, [228/226] in freshwater was lower than 2, and it increased sharply to the maximum value of 5.8 at moderate salinity, and then decreased through the high-salinity region in the mixing zone. In July 2008, [228/226] increased from the value lower than 2 in freshwater as well to the maximum value of 4.1 and then decreased with the increasing salinity. Although the two isotopes have similar chemical properties, the difference in their half-lives will affect the production rates because of their respective parents. Short half-lives result in higher production rates (Beck et al. 2007). Thus, we know that the growth rate of ^{228}Ra is higher than that of ^{226}Ra, accounting for [228/226] of more than 1 in most cases. Meanwhile, if we assumed that the desorption of ^{226}Ra and ^{228}Ra from suspended particles was similar and the contribution from submarine groundwater discharge was negligible, the diffusion of these two isotopes from bottom sediments have been much different to account for the large difference in [228/226]. However, this may not have been the case, as the diffusion from sediments cannot cause such a large difference in [228/226]. It was reported that in estuaries with salinity ranging from 2 to 20, where Ra isotopes were excessively released into the seawater, the maximum values of [228/226] were within the range of 3 to 5 (Elsinger and Moore 1983; Rengarajan et al. 2002). In this study, the maximum [228/226] observed was close to 6 in 2007 and greater than 4 in 2008 in the Wenjiao-Wenchang Estuary. Such results most likely suggested discharge of submarine groundwater with unusually high [228/226] (Swarzenski 2007; Charette 2007), which should be further investigated.

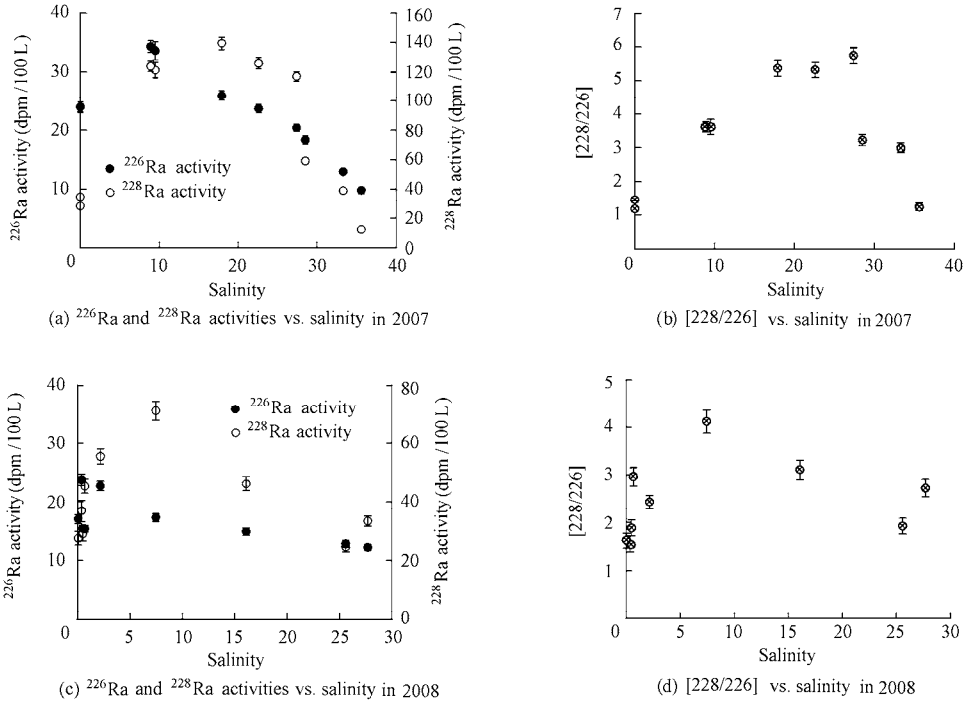

Fig. 2 ^{226}Ra and ^{228}Ra activities and [228/226] vs. salinity for Wenjiao-Wenchang Estuary

3.2 Distribution of ^{226}Ra and ^{228}Ra in Wanquan Estuary

Unlike the natural topography in the Wenjiao-Wenchang Estuary, the topography is quite complex in the Wanquan Estuary. There are small sandbars close to the mouth, separating the river flow into southern and northern branches. Sampling sites in the Wanquan Estuary were located in the southern branch (Fig. 1). The ^{226}Ra and ^{228}Ra activities and [228/226] there were 7.04-18.9 dpm/100L, 8.37-32.6 dpm/100L, and 1.0-2.4, respectively, which were significantly lower than those in the Wenjiao-Wenchang Estuary. The relationship between the ^{226}Ra or ^{228}Ra activity and salinity also demonstrated non-conservative mixing, except for one sampling point where salinity was 33.9 (Figs. 3(a) and (b)). This implies that the impact of groundwater discharge in the Wanquan Estuary on the Ra concentration might not be as significant as that in the Wenjiao-Wenchang Estuary. At the sampling point with the salinity of 33.9, we observed an increase of the ^{228}Ra activity but a decrease of the ^{226}Ra activity, probably indicating the extra input of ^{228}Ra to the mixing zone. A similar phenomenon was found in the Yangtze Estuary by Elsinger and Moore (1984). We suspect that the diffusion from bottom sediments other than groundwater discharge provided ^{228}Ra to the water body, while addition of ^{226}Ra from these sources was masked by water dilution.

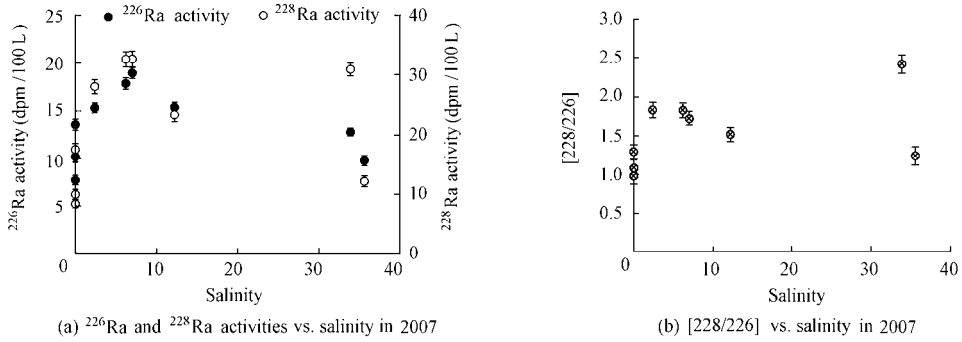

Fig. 3 ^{226}Ra and ^{228}Ra activities and [228/226] vs. salinity for Wanquan Estuary

3.3 Estimation of horizontal eddy diffusion coefficient

^{228}Ra in ocean surface waters is continuously supplied from the continental shelf sediments by the decay of its parent ^{232}Th. Horizontal mixing of the water body promotes the transport of ^{228}Ra from the surface mixing layer offshore into the open ocean. It is possible to use ^{228}Ra as a tracer to estimate the exchange rate of coastal waters. If net advection can be neglected, and the system is in a steady state, the distribution pattern of the ^{228}Ra activity versus distance offshore can be expressed with a simple one-dimensional diffusion model as follows (Moore 2000):

$$K_{\mathrm{h}} \frac{\partial^2 A}{\partial x^2} - \lambda A = 0 \tag{1}$$

where A is the ^{228}Ra activity (dpm/100L); x is the distance offshore (km); λ is the decay constant of ^{228}Ra, and λ = 0.12/year; and K_{h} is the horizontal eddy diffusion coefficient (cm^2/s). Under boundary conditions $A = A_0$ at x = 0 and $A \to 0$ at $x \to \infty$, the solution of Eq. (1) is then expressed as

$$\ln A = \ln A_0 - x\sqrt{\frac{\lambda}{K_{\mathrm{h}}}} \tag{2}$$

Eq. (2) describes the decrease of ^{228}Ra with increasing x as an exponential function. Thus K_{h} can be calculated from the slope S of a plot of $\ln A$ versus the distance offshore ($K_{\mathrm{h}} = \lambda / S^2$). Fig. 1 shows two transects T and T' in the adjacent sea areas. Profiles of the ^{226}Ra and ^{228}Ra activities as a function of the distance offshore for these two transects are illustrated in Fig. 4.

Moore (2000) showed a significant decrease of Ra isotopes with the distance offshore within 50 km from the coast. However, in this study, we saw a slightly decrease of the ^{226}Ra and ^{228}Ra activities at transect T (Fig. 4(a)). It has been reported that ^{228}Ra is known to build up to high concentrations in extended continental shelf areas (van der Loeff et al. 1995; Kim et al. 2005), which would constitute an alternative source of ^{228}Ra from neighboring regions. Fig. 4(b) shows an increase of the ^{228}Ra activity further offshore at transect T' that causes a poor linear regression result (R^2 = 0.036 7). This suggests an additional ^{228}Ra source that contributed to the

offshore areas. In this case, the mixing model assumptions cannot be met, and thus, transect T' was not included for the diffusion coefficient calculation. However, the estimate at transect T seemed more robust, and any subsequent ^{226}Ra or nutrient flux calculations would only be based on this transect.

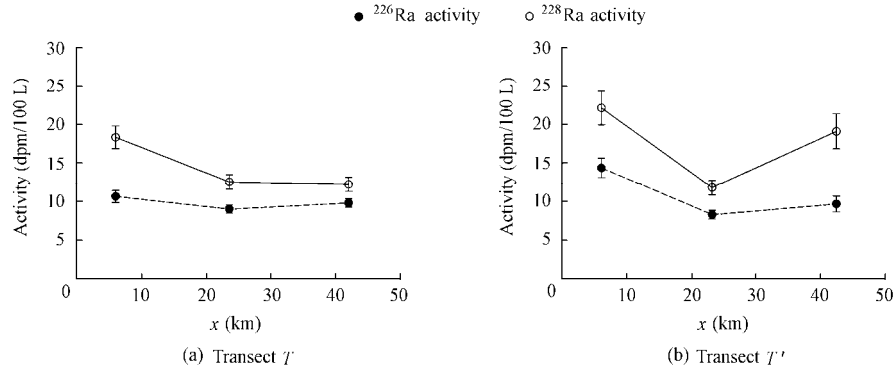

Fig. 4 ^{226}Ra and ^{228}Ra activities as a function of distance offshore for transects T and T'

Fig. 5 shows $\ln A$ as a function of the distance offshore for transects T and T'. The slope of the regression line was –0.011 dpm/(100L·km) at transect T, and the K_h value of 3.16×10^5 cm^2/s was obtained from Eq. (2) based on the mixing model assumptions.

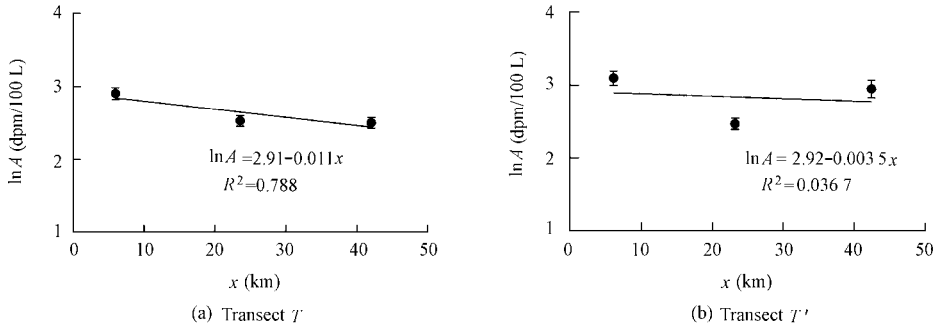

Fig. 5 ln$A$ as a function of distance offshore for transects T and T'

Table 2 summarizes the horizontal eddy diffusion coefficients calculated using the ^{228}Ra activity in some regions. Huang et al. (1997) estimated the horizontal eddy diffusion coefficient of 2.3×10^6 cm^2/s in the northeastern South China Sea. Clearly, the estimated K_h value in this study was an order of magnitude less than that reported by Huang et al. (1997).

For a conservative tracer, its flux can be estimated from K_h and the offshore concentration gradient (Moore 2000). The gradient of ^{226}Ra was 0.096 dpm/(100L·km) in the study area. Thus, the corresponding offshore ^{226}Ra flux for unit cross-sectional area was 2.62×10^9 dpm/(km^2·d). In the study area, there was a better pycnocline at the water depth of about 10 m, which inhibited Ra transport from the bottom to overlying water. Therefore, we assumed that Ra was transported offshore in this 10-m deep surface layer; thus, the offshore flux of ^{226}Ra for unit length was 2.62×10^7 dpm/(km·d). This value can be used to balance the

^{226}Ra flux to the ocean. Moore (2000) estimated the ^{226}Ra flux from the coastline to the ocean and concluded that a substantial volume of groundwater discharge was required to balance the Ra budget. Moreover, application of the ^{228}Ra-derived K_h can also help assess the nutrient fluxes to the open South China Sea.

Table 2 Horizontal eddy diffusion coefficients in some regions, calculated with ^{228}Ra activity

Region	K_h (10^6 cm^2/s)	Reference	Region	K_h (10^6 cm^2/s)	Reference
South Pacific	10-100	Kaufman et al. (1973)	Northeastern South China Sea	5.7-16	Xie et al. (1995)
East Pacific	0.1-10	Knauss et al. (1978)	Northeast Pacific	1-50	Huh and Ku (1997)
Sub-Arctic waters	5-10	Moore et al. (1980); Men et al. (2006)	Western Yellow Sea	29	Men et al. (2006)
Coast of Western North Pacific	0.4-40	Yamada and Nozaki (1986)	Nansha Sea area	13	Huang et al. (1996)
Coast of California-Japan Offshore	40	Cochran (1992); Men et al. (2006)	Northeastern South China Sea	2.3	Huang et al. (1997)
Seto Inland Sea	1.1-1.4	Kasemsupaya et al. (1993)	East coast of Hainan Island	0.32	This paper

3.4 Horizontal diffusion flux of nutrients from coast to offshore area

One important pathway for terrigenous nutrient transport from land to the open ocean is via the diffusion process. In this study, we used the nutrient concentrations at transect T' as an example. Based on the offshore nutrient gradients (NO_3^-, PO_4^{3-}, and SiO_3^{2-}) and the K_h value estimated in section 3.3, the nutrient fluxes Q from the coast to the open South China Sea can be calculated as follows:

$$Q = K_h \iota \tag{3}$$

where ι is the horizontal nutrient gradient, which was 0.46 μmol/(m^3·m) for NO_3^-, 0.03 μmol/(m^3·m) for PO_4^{3-}, and 1.03 μmol/(m^3·m) for SiO_3^{2-} in this study. The horizontal eddy diffusion coefficient K_h was 3.16×10^5 cm^2/s. Using Eq. (3), the nutrient fluxes in our study area were 1.3 mol/(m^2·d) for NO_3^-, 0.082 mol/(m^2·d) for PO_4^{3-}, and 2.8 mol/(m^2·d) for SiO_3^{2-}. The N : P ratio of 16 was the same as the Redfield ratio. The changes of the nutrient concentration could result in the shift in the N : P ratio during the diffusion process. Chen et al. (2001) reported that in the wet season of six months, the Kuroshio current imported $(288\pm26)\times10^9$ mol of N, $(20.6\pm1.9)\times10^9$ mol of P, and $(412\pm37)\times10^9$ mol of Si into the South China Sea through the Bashi Channel. In this study, if the nutrients were transported offshore in the 10-m deep surface layer, the total amounts of nutrients for the study area with a 100-km coastline within six months were 2.3×10^8 mol of N, 1.5×10^7 mol of P, and 5.1×10^8 mol of Si. Based on a comparison with the estimates made by Chen et al. (2001), the horizontal transport in this work supplied a small portion of the nutrients into the South China Sea. However, its potential impact on the east coast of Hainan Island cannot be ignored, and the variation of the N : P

ratio should be further investigated. Moreover, other nutrient fluxes from the coastal to offshore areas can also be estimated by the diffusion coefficient obtained by this study, which could be helpful to understanding the way aquatic environmental pollution along the coast affects offshore areas.

4 Conclusions

The activity of Ra isotopes (^{226}Ra and ^{228}Ra) showed non-conservative behavior in the mixing zone of the Wenjiao-Wenchang and Wanquan estuaries, suggesting an extra input of Ra into the Estuary, and especially the possible contribution from submarine groundwater discharge. It should be mentioned that the potential lateral source of Ra contributing to the offshore area at transect T' meant that the model assumptions could not be met there. Consequently, we could estimate the eddy diffusion coefficient only from the data at transect *T*. From the distribution of the ^{228}Ra activity with the distance offshore at transect *T*, the horizontal eddy diffusion coefficient was obtained, which was 3.16×10^5 cm^2/s, and the associated nutrient fluxes were 1.3 mol/(m^2·d) for NO_3^-, 0.082 mol/(m^2·d) for PO_4^{3-}, and 2.8 mol/(m^2·d) for SiO_3^{2-}. Although these estimates only considered a small part of nutrient inputs into the South China Sea, they can still provide a firm basis for the future research when we are committed to reducing their influences on the water quality by controlling the nutrient structures in the coastal ecosystem of east coast of Hainan Island.

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