

DISTRIBUTION OF ^{226}Ra RADIONUCLIDE IN UPWELLING EVENT OFF ULSAN, GAMPO AND POHANG, KOREA

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ABSTRACT

DISTRIBUTION OF ^{226}Ra RADIONUCLIDE IN UPWELLING EVENT OFF ULSAN, GAMPO AND POHANG, KOREA. Upwelling is an important event in the sea for it makes the area to become more productive. The objective of this study is to determine the distribution of radium-226 as radiotoxic in the upwelling area in the summer season. Measurements of the horizontal and vertical activities of ^{226}Ra in the Ulsan, Gampo and Pohang waters were conducted in June, August 1999 and June 2000 when the upwelling event was expected to occur. Water temperature, salinity and dissolved oxygen (DO) were also measured concurrently. The thermocline layer or the layer where the temperature decreased drastically occurred at a depth between 30 m to 100 m for samples dated June 1999 and 30 m to more than 100 m for samples dated August 1999 and June 2000. The salinity decreased with depth but the DO concentration increased in this layer. The condition affected the vertical distribution of ^{226}Ra in the study area, where the ^{226}Ra activities showed to be relatively homogeneous vertically in each station. This indicates that the upwelling which occurred in the study area was capable to distribute the ^{226}Ra activities from the bottom to the surface. Thus, the ^{226}Ra distribution at that time did not increase with depth, opposite to what usually occurs in the ocean at normal condition. The ^{226}Ra activities also did not decrease after elapsed time of one year, in fact, it increased. However, horizontally the ^{226}Ra activities decreased with increasing distance from the coastal zone. Based on these results, it can be concluded that the upwelling event in the Ulsan, Gampo and Pohang was effective for increasing the ^{226}Ra activities and was capable to create homogeneous ^{226}Ra activities from the bottom to the surface in the water column, and the source of ^{226}Ra was likely to originate from outside area (i.e. coastal zone) and bottom area.

Keywords: ^{226}Ra activity, upwelling event, Ulsan, Gampo, Pohang.

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INTRODUCTION

Radium is one of the important radionuclides in the sea and one of the most radiotoxic of the naturally occurring radioisotopes that is concentrated in several kinds of marine organisms [1]. The sources of radium in the open ocean mainly come from estuarine, coastal and shelf sediments, from salt marshes, or from fine-grained sediments of the inner shelf via diffusion through pore-water, and the radium is transported to the open ocean by

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current and horizontal mixing [2,3,4] or wind-driven circulation of the surface waters [5]. The ^{226}Ra has higher activities in estuaries and coastal waters than in open and river waters [6]. Ulsan, Gampo and Pohang waters are located in the Japan Sea (East Sea). Over the past decade there has been a growing concern over dumping of radioactive waste in the East Sea Proper Water and adjacent coastal waters [7].

Nozaki *et al* [8] found that ^{226}Ra concentration ($t_{1/2} = 1622$ years) in the surface water increases in the northern part of the North Pacific due to upwelling. In addition, Craig *et al* [9] also discovered that the in-situ source of ^{226}Ra in the vertical diffusion of the deep-water profiles was a function of the parametric upwelling velocity. Thus ^{226}Ra is currently the most important isotope after radiocarbon for the study of rates from circulation and mixing process and Ra is a useful tracer for the studies of oceanic mixing and circulation [10].

Upwelling system is considered to be among the most productive areas of the world ocean. Coastal upwelling regions occupy only 0.1% of the world ocean's surface area, but they account for approximately half of the fish harvest in the world [11], such as Peruvian anchovy which is the largest single fishery developed in an upwelling region. Upwelling system is able to promote upward movement of nutrient-laden, subsurface water into the euphotic zone that helps to provide suitable condition for primary production, which in turn supplies food for the fisheries [12]. Also, typically upwelling areas can be identified by horizontal anomaly in their physical (e.g. temperature, salinity, density, water color), chemical (e.g. oxygen, nutrients), and biological (e.g. chlorophyll, plankton) characteristics. Relative to their adjacent waters, these areas have lower temperatures, higher nutrients, lower oxygen and higher chlorophyll concentration as well as high particulate organic carbon and nitrogen (POC and PON) in the surface water [13]. However, the phenomena of this area have not been much studied effectively yet using the distribution of radionuclide especially with ^{226}Ra (half life 1,600 yr) that is a progeny of ^{238}U [14].

The objective of this study is to determine the distribution of ^{226}Ra in the Ulsan, Gampo and Pohang waters, Korea as part of the Japan Sea (East Sea) which is effective to become an upwelling area in the summer season, but with a potential of accumulation of pollution from the surrounding area.

EXPERIMENTAL METHODS

Sampling

During summer seasons the research vessel of "R/V Tamyang" was usually dispatched to carry out multi-purpose ocean research. In June, August 1999 and June 2000 the vessel was launched in the East Sea (Japan Sea),

with the sampling stations as shown in Figure 1. During that time, most universities in Korea which have an oceanography department joined this research. The seawater samples for this study were collected only along station C (Ulsan), D (Gampo) and E (Pohang). The surface seawater samples were collected along stations from coastal to offshore; those were C1-C5, D1-D5 and E1-E5 as horizontal measurements, whereas the vertical measurements were performed only in station C2, D2 and E2 at some different depths.

Temperature and salinity data were obtained using a CTD (Conductivity Temperature and Depth) (SBE 911 plus) attached to a Rosette sampler that was equipped with a sensor to measure the dissolved oxygen (DO) at all stations. The surface seawaters were collected with a submerged water pump, and the deep water samples were collected with Niskin bottles at each sampling station [15].

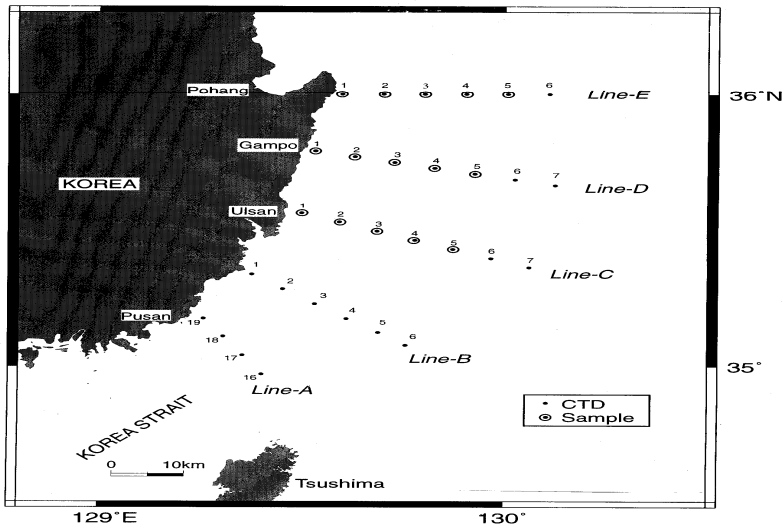


Figure 1. Sampling location of 'R/V Tamyang' during summer season in 1999 and 2000.

Radium Analysis

Seawater samples for analysis of ^{226}Ra were collected at a volume of about 100 liters in acid washed 20-liter polyethylene bottles that had been rinsed at least twice with seawater collected from the location before the final samples were taken. The analysis procedure was conducted according to Yamada and Nozaki [16]. All of the seawater samples were filtered through a

filter paper (Whatman no.2) and subsequently passed to the manganese-coated acrylic fibers (MnO_2 fiber) column at a flow rate $<1 \text{ L min}^{-1}$. Filtering was done directly in the shipboard with special construction of the apparatus as shown in Figure 2.

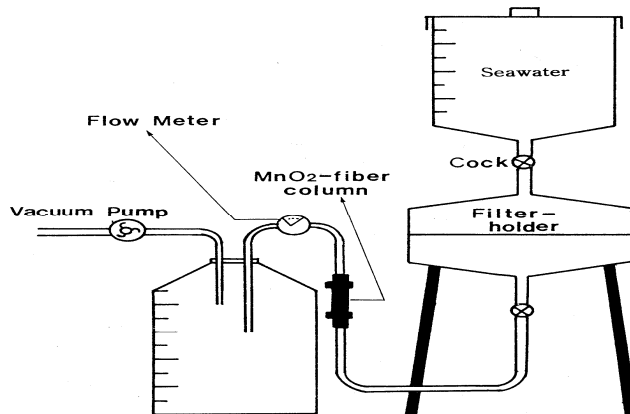


Figure 2. A shipboard apparatus for extraction of dissolved radium.

The MnO_2 fibers were removed and placed in a plastic bag that had been rinsed with deionized water, and immediately were stored in frozen awaiting laboratory analysis. In the laboratory the MnO_2 -fibers were soaked in 300 mL of hot 6N HCl + 1% NH_2OHHCl solution and subsequently the leaching process was carried out. The solution was filtered by squeezing and washing through a membrane filter paper (Whatman no.4). The solution was spiked with 125 mg of Pb^{2+} carrier. After addition of 10 mL of 10N H_2SO_4 , the leached was evaporated to a small volume (10-20 mL). Ra was co-precipitated with PbSO_4 by adding ~150 mL of H_2O to the solution and adjusting the solution to a pH of about 1.8 with 5N NaOH solution. The Pb(Ra)SO_4 precipitate was filtered and ignited at 450 °C for 10 hours. The precipitate was transferred into a plastic counting vial and weighed for determination of chemical recovery (chemical yield). Then the vial was tightly capped and stored for more than 3 weeks to let the short-lived daughter nuclide of Ra grow. The activities of the Ra isotopes were measured by a γ -spectrometer. The ^{226}Ra was calculated from the sum of the count rates of ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 keV). The counting time was typically 1-2 days.

Background and blanks were counted for more than a week. The Pb(Ra)SO_4 counting method was calibrated by following the procedure against NBS (NIST) ^{226}Ra standard.

RESULT AND DISCUSSION

Vertical and horizontal activities of ^{226}Ra in samples dated June, August 1999 and June 2000 are given in Figure 3 and 4. The complete data of ^{226}Ra activities together with water temperature, salinity and dissolved oxygen can be seen in Appendix 1, 2 and 3.

The temperature rapidly decreased vertically with depth (thermocline) which occurred in the layer between 30 m to 100 m for samples dated June 1999 (thick thermocline layer of around 70 m), but for samples dated August 1999 and June 2000 the thermocline layer occurred at 30 m to more than 100 m (thick thermocline layer of more than 70m). In this layer the salinity decreased vertically with depth even though in the surface layer it increased with depth. Conversely, the DO increased with depth in the thermocline layer. According to Lim and Chang [17], Lee [18], Park [19], Kim and Kim [20] and NFRDA [21], a cold water mass is known to occur along the southeastern coast of Korea Peninsula every summer (between June to September). In addition, Yang *et al.* [22] found that at a depth between 75 m and 150 m the water masses of NKCW (North Korea Cold Water) flow in this layer with a temperature range of 1 – 7 °C. This condition pulled the water column upward from the bottom to the up layer (surface), and it is called an upwelling event. It will undoubtedly affect the distribution of some elements. Figure 3 and 4 show the vertical and horizontal distribution of ^{226}Ra . The activities of ^{226}Ra in the surface layer (non thermocline layer) were relative more constant than those in the middle layer. The range activities in the overall study in June, August 1999 and June 2000 were 58.00 – 110.46 (mean=84.15, n=30), 56.30 – 107.69 (mean=85.5, n=29) and 69.9 – 131.61 (mean=100.07, n=30) dpm/1000L respectively, while in surface seawater the values were 58.00 – 110.46 (mean=78.13, n=14), 56.30 – 98.85 (mean=81.39, n=15) and 79.00-131.61 (mean=103.29, n=15) dpm/1000L respectively (Table 1). While the mean activities for the overall study and surface water from June 1999 – June 2000 were 89.91 and 87.60 dpm/1000L correspondingly (Table 1). These values are comparable with the ^{226}Ra concentration in the Eastern Indian Ocean and the China Sea at a similar depth, which has been studied by Nozaki and Yamamoto [23] where the sample collection was done in December 1996, January 1997 and February 1997. They found the surface activities of ^{226}Ra in open water ranging from 59 – 87 dpm/1000L and increasing with depth. The range of activities at a depth of more than 1000 m was 174.7 – 253.9 dpm/1000L. However, Figure 3 show that the vertical distribution of ^{226}Ra was relatively constant from the bottom to the surface. It was due to the upwelling event that could transport ^{226}Ra from the bottom to the surface [8].

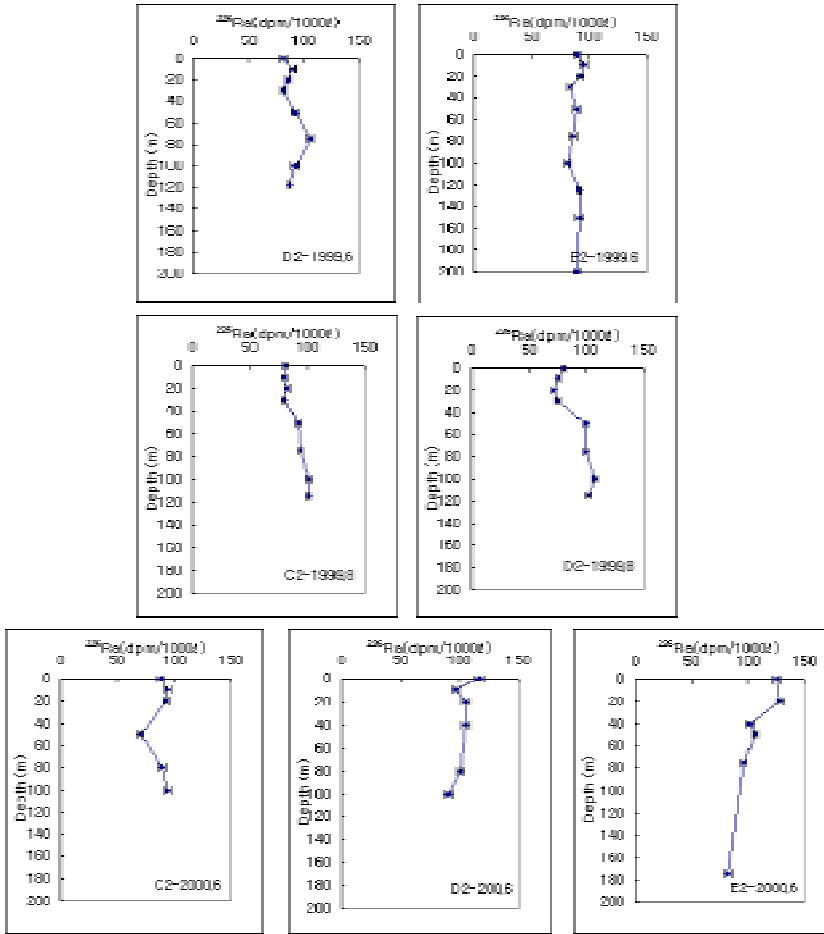


Figure 3. Vertical distribution of ^{226}Ra in Ulsan (C) in August 1999 and June 2000; Gampo (D) in June 1999 and August and June 2000; Pohang (E) in June 1999 and June 2000.

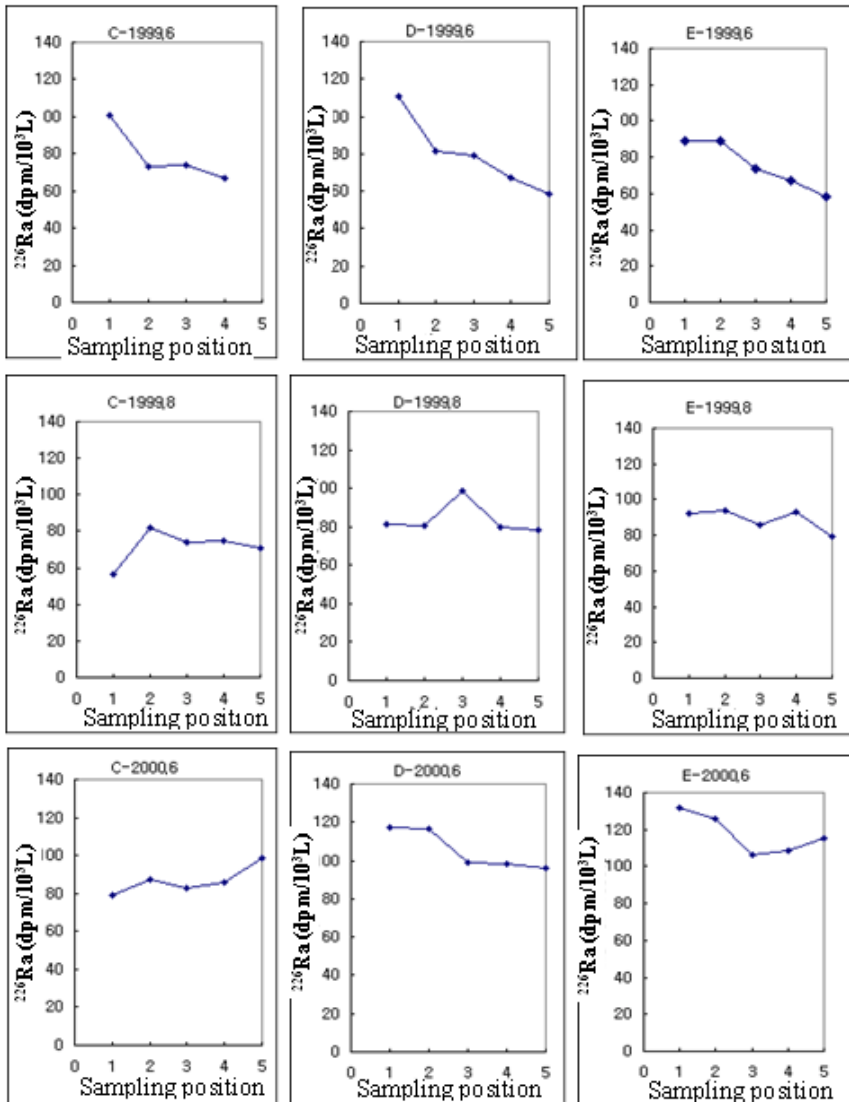


Figure 4. Horizontal distribution of surface ^{226}Ra in Ulsan (C), Gampo (D) and Pohang (E) in June 1999 and August and June 2000.

The ^{226}Ra activity did not show any decrease after elapsed time of one year both in the overall study and surface water, in fact, the activity increased (Table 1). Hence the decay process did not affect the decreasing of ^{226}Ra . Thus mixing process increased the supply of ^{226}Ra higher than the reduction of ^{226}Ra due to decay process. Kumar and Li [24] stated that the regeneration

of ^{226}Ra is mainly from the underlying sediment rather than through the dissolution of particles in the water column. Compared with previous study in Tsushima Surface Water (TSW) by Kim [25], this activity is much lower because the range of activities in TSW for samples dated September 1991 was 109 – 380 (mean=236) dpm/1000L. The activities of the ^{226}Ra between the overall study and surface seawater did not show any significant difference, which could be due to upwelling of deep water affecting the spreading water masses. According to Kennish [26], the radioactive in sea can be diluted and dispersed by current, turbulent diffusion, isotopic dilution and biological transport. Thus, upwelling that occurred during that time was effective to dilute and disperse the ^{226}Ra into all waters. However, if compared between each station, the activities showed a significant difference, where the activities in station E were higher than those in station D and C, especially for samples dated August 1999 and June 2000 when the thick layer of thermocline was more than 70 m, or the activities increased northward (Table 2 and Figure 5). Probably the ^{226}Ra was supplied from the Japan Sea Proper Water in conjunction with the spreading of oxygen, because the concentration of oxygen was also higher in station E especially in the bottom layer for samples dated June 2000. However, when the layer of thermocline was thin or the thickness of the thermocline layer was only around 70 m, the activities of ^{226}Ra in station were not higher than those in station C and D both vertically and horizontally (Table 2). It may result from the weaker upwelling event in June 1999 compared to that in August 1999 and June 2000.

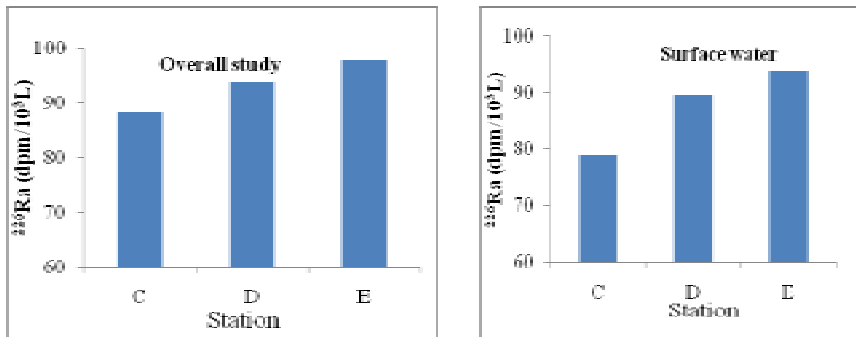


Figure 5. The activities of ^{226}Ra at each station both in the overall study and surface water.

Table. 1. The range and mean activities of ^{226}Ra in the overall study and surface water.

Isotope	Samples	Range activities (dpm/10 ³ L)			Mean activities (dpm/10 ³ L)			Mean dpm/10 ³ L
		1999.6	1999.8	2000.6	1999.6	1999.8	2000.6	
^{226}Ra	overall	58.0 - 110.5	56.3 - 107.7	69.9 - 131.6	84.2 (30)	85.5 (29)	100.1 (30)	89.91
	surface	58.0 - 110.5	56.3 - 98.9	79.0 - 131.6	78.1 (14)	81.4 (15)	103.4 (15)	87.60

The vertical distribution of ^{226}Ra in samples dated June and August 1999 showed that the activities were relatively constant from the surface to the bottom, especially in station E2 and C2. In station D2 the fluctuation of ^{226}Ra was slightly different, where at below 30m in depth the ^{226}Ra slightly increased. For samples dated June 2000, the activities in the surface layer especially in station D2 and E2 were higher than those in the bottom layer, also higher than those in June and August 1999. This indicates that during that time there was mixing process from out place that supplied ^{226}Ra presumably from the Yellow Sea as previous studied by Nozaki *et al* [8] who found that the ^{226}Ra in northern region of the North Pacific increased by two times when the upwelling occurred. Thus, the principle source of ^{226}Ra in the surface water is probably from the upwelled waters and to a lesser extent the runoff from land. According to Shannon and Charry [27], the primary removal mechanism of radium is associated with phytoplankton, there being a subsequent conveyance to deeper water and sediment.

The bottom layer in this study did not show any increase in ^{226}Ra , because the upwelling event was able to spread ^{226}Ra activities from the bottom to the surface. Thus, the ^{226}Ra activities in the study area were different with those in several oceanic areas [24, 28], where the ^{226}Ra increased with depth. This is because in the ocean region the large portion of ^{226}Ra is derived from the bottom sediment, where ^{226}Ra is produced by the decay of ^{230}Th and diffuses out to the bottom water through the sediment at pore-water [29], which is in contrast with the present study where the ^{226}Ra did not show any increase with depth.

Horizontally, the ^{226}Ra decreased with increasing distance from the coastal. This case was very clear for samples dated June 1999 (Figure 4) which indicates that for long distance from coastal (offshore) the ^{226}Ra was not supplied from estuarine area. It was similar too in Tampa Bay, Florida, where submarine groundwater contributed high ^{226}Ra to the bay water [4]. For samples dated August 1999 the horizontal distribution of surface ^{226}Ra was relatively constant, which means that the mixing processes only promoted homogenous ^{226}Ra , because the activities were not different with

the mean activities in samples dated June 1999. For samples dated June 2000 the activities of surface ^{226}Ra horizontally fluctuated, but higher than those in the previous samples in June 1999 and August 1999 (Table 2), especially in station D and E. It may be caused by supply from out place possibly from the Yellow Sea or by the upwelling event which at that time was stronger than that for the previous samples. Nozaki *et al* [30] had used $^{228}\text{Ra}/^{226}\text{Ra}$ ratio for measuring residence time of the shelf water in the East China and the Yellow Sea because the activities of this radionuclide are relative high in those places. Besides that, Yang *et al* [31] found that in Japan Sea (East Sea) there are various water masses distributed in this region such as Japan Sea Proper Water (JSPW), Tsushima Surface Water (TSW), Yellow Sea Cold Water (YSCW) in which all them will form a strongly seasonal thermocline that supports the increase of ^{226}Ra .

From the above explanation, it can be said that the upwelling event affected the distribution of ^{226}Ra activities, however, the temperature and salinity did not show any direct effect on the distribution of ^{226}Ra both in the overall study and surface seawater (Table 3) even though the anomaly of temperature and salinity in the open sea can be used to detect upwelling event [12, 13].

Table 2. Mean activities (dpm/1000L) of ^{226}Ra vertically and horizontally.

Sample date	Station	Vertically	Horizontally
		^{226}Ra	^{226}Ra
June 1999	C	-	78.74
	D	89.61	79.48
	E	88.93	75.34
August 1999	C	88.88	71.42
	D	88.70	83.81
	E	-	88.94
June 2000	C	87.72	86.80
	D	102.55	105.52
	E	106.78	117.54

Table 3. Correlation coefficients between ^{226}Ra and the temperature and salinity both in the overall study and surface seawater.

Sample date	Isotope	Overall study		Surface seawater	
		Temp	Salinity	Temp	Salinity
1999.6	^{226}Ra	0.19	0.005	0.74	0.16
1999.8	^{226}Ra	0.46	0.24	0.002	0.05
2000.6	^{226}Ra	0.59	0.66	0.57	0.65

CONCLUSION

The water temperature, salinity and DO did not indirectly influence the ^{226}Ra distribution, but the thermocline which caused upwelling affected it. Vertically, ^{226}Ra activities were relatively homogeneous from the bottom to the surface layer. Horizontally, however, the activities decreased with increasing distance from the coastal.

The exponential decay of ^{226}Ra in this study was not effective to reduce the ^{226}Ra in the water column, because after elapsed time of one year the activity of ^{226}Ra did not show any decrease, in fact, it increased. With regard to these phenomena, it can be concluded that the source of the ^{226}Ra came from the coastal and bottom areas. Moreover, the ^{226}Ra activity also increased northward, which indicates that the ^{226}Ra activity is dependent upon location.

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Appendix 1. Temperature, salinity, DO and activities of ^{226}Ra for samples dated June 1999.

Station	Depth (m)	Temp (°C)	Sal. (psu)	DO	^{226}Ra (mL/L)
	(dpm/1000L)				
C1	0	19.48	33.86	5.36	100.88±3.12
C2	0	19.96	33.81	4.73	73.24±2.92
C3	0	21.25	33.89	4.63	74.17±3.04
C4	0	21.33	33.99	4.61	66.66±2.78
C5	0	21.75	34.15	4.63	ND
D1	0	17.80	33.90	6.41	110.46±3.18
D2	0	20.32	33.89	4.77	81.68±3.10
	10	18.27	33.81	5.02	89.94±2.34
	20	15.34	34.33	4.42	86.47±2.05
	30	14.96	34.39	4.14	81.64±2.21
	50	11.13	34.34	4.29	92.15±1.98
	75	3.26	34.09	5.33	105.08±2.63
	100	1.86	34.06	5.51	92.27±2.94
	118	1.81	34.06	5.67	87.64±1.66
D3	0	20.60	33.82	4.46	79.44±3.03
D4	0	21.15	34.00	4.42	67.08±1.73
D5	0	21.10	34.12	4.40	58.75±2.13
E1	0	19.14	33.86	5.88	88.73±2.05
E2	0	19.01	33.86	5.86	89.16±2.16
	10	17.76	33.90	6.17	95.09±3.23
	20	12.47	34.22	5.20	93.01±2.91
	30	10.56	34.33	4.51	83.34±2.10
	50	6.06	34.15	5.25	89.02±3.01
	75	3.16	34.06	5.93	85.85±3.30
	100	1.96	34.05	6.18	80.89±3.08
	125	1.46	34.05	6.19	91.30±1.85
	150	1.22	34.05	6.14	91.98±3.07
	200	0.99	34.06	5.85	89.61±2.31
E3	0	20.66	33.90	4.57	73.39±1.97
E4	0	20.56	33.83	4.55	67.42±2.85
E5	0	20.55	33.88	4.49	58.00±2.62
Mean					84.15

ND: not determined

Appendix 2. Temperature, salinity, DO and activities of ^{226}Ra for samples dated August 1999.

Station	Depth (m)	Temp (°C)	Sal. (psu)	DO (mL/L)	^{226}Ra (dpm/1000L)
C1	0	22.50	31.69	4.68	56.30±1.92
C2	0	25.31	33.02	3.90	81.56±2.11
	10	23.79	33.07	4.06	80.00±2.07
	20	22.89	33.20	4.10	82.52±2.09
	30	20.26	33.19	3.96	79.83±2.06
	50	10.55	34.20	4.00	91.59±2.11
	75	5.85	34.14	5.17	93.72±2.10
	100	3.71	34.07	5.72	101.14±2.30
	115	2.90	34.06	5.72	100.69±2.15
C3	0	24.90	32.88	3.81	73.67±2.03
C4	0	25.54	32.78	3.73	74.68±2.05
C5	0	25.95	32.75	3.96	70.91±2.02
D1	0	23.24	31.69	5.37	81.25±2.09
D2	0	24.32	31.46	5.13	80.34±2.07
	10	22.74	31.86	4.72	75.40±2.03
	20	19.02	33.10	4.31	71.24±1.96
	30	15.74	33.64	4.52	74.74±2.05
	50	8.54	34.16	4.99	99.09±2.10
	75	5.16	34.10	5.03	99.04±2.11
	100	3.64	34.07	5.23	107.69±2.72
	115	3.38	34.07	5.25	102.02±2.46
D3	0	23.03	31.60	5.25	98.85±2.17
D4	0	25.05	32.95	3.64	80.10±2.36
D5	0	24.10	32.67	3.77	78.50±2.91
E1	0	23.16	32.20	4.89	91.86±2.46
E2	0	25.09	32.17	4.14	94.18±2.46
E3	0	26.04	32.21	3.76	86.12±2.38
E4	0	25.12	32.03	4.14	92.88±2.45
E5	0	25.45	32.61	3.82	79.66±2.38
Mean					85.5

ND: not determined

Appendix 3. Temperature, salinity, DO and activities of ^{226}Ra for samples dated June 2000.

Station	Depth (m)	Temp (°C)	Sal. (psu)	DO (mL/L)	^{226}Ra (dpm/1000L)	
C1	0	16.07	34.06	6.53	79.00±3.14	
C2	0	15.81	34.11	6.32	87.36±3.14	
	10	15.71	34.11	6.29	93.90±2.83	
	20	15.21	34.10	6.06	92.52±2.56	
	50	12.39	34.18	5.46	69.90±2.74	
	80	8.07	34.22	5.54	88.79±3.14	
	100	4.44	34.10	5.73	93.83±3.20	
C3	0	15.88	34.10	6.43	83.02±3.12	
C4	0	15.28	33.89	6.49	86.00±2.23	
C5	0	15.61	33.88	6.56	98.64±2.39	
D1	0	17.34	33.49	6.32	117.46±3.35	
D2	0	17.32	33.60	6.31	116.41±3.40	
	10	15.49	33.91	7.15	96.64±2.79	
	20	13.76	34.03	5.26	105.62±3.29	
	40	11.22	34.33	4.90	105.64±3.66	
	80	4.27	34.08	6.18	100.53±2.79	
	100	3.44	34.08	5.75	90.47±3.19	
D3	0	16.79	33.97	6.80	99.22±3.15	
D4	0	16.87	34.00	6.72	98.23±2.28	
D5	0	16.68	33.88	6.67	96.27±2.26	
E1	0	16.97	33.32	6.12	131.61±3.34	
	0	17.47	33.21	5.91	125.84±3.67	
	20	13.66	33.78	5.63	128.44±3.42	
	40	15.13	34.54	4.36	101.68±3.29	
	50	14.44	34.47	4.47	105.68±2.29	
	75	6.60	34.13	6.24	96.26±2.47	
E2	175	1.23	34.04	6.87	82.75±3.13	
	0	17.55	33.10	5.84	106.10±3.23	
	0	17.78	33.17	5.74	108.55±3.25	
	0	17.45	33.21	5.84	115.59±3.37	
	Mean					100.07