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LONG-LIVED RADIONUCLIDES IN THE MARINE ENVIRONMENT OF THAILAND

S. Mainpenyawong

M. Sonsuk P. Polphong

M. Millinawisamat Y. Panyathipsakul

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1. INTRODUCTION

Radionuclides in environment have mainly two sources of origin: one is natural occurrence the other is artificial discharge. The naturally occurring radionuclides are exist in constant level in the time domain. On the other hand, artificial radionuclides discharged from many sources such as nuclear weapon testings, nuclear accidents, satellite burn-up and waste disposal from nuclear facilities may vary in level according to human activity on nuclear affair.

Artificial radionuclides released into marine environment have been changed in level from time to time. To study radioactivity in the marine environment of Thailand, there have been many researches conducted since 1981. Among these Polphong et al.[1] monitored gross beta activity in 256 samples of sea water, sediment, fish and shellfish collected from the Gulf of Thailand during 1981-1983. They[2] also compared levels of natural radioactivity of sediment and water in the gulf and in shrimp rearing farms on the east coast of Thailand. Their reports described mainly the measurement of gross beta activity in the samples. However, reports on analyses of specific radionuclides in marine samples are few.

During the nuclear accident at Chernobyl in Soviet Union in 1986, it is estimated that large amount of radionuclides was released into the world atmosphere and consequently some of these nuclides was injected into marine environment. The inputs of

radionuclides into seas and oceans are leading to a complex distribution and changing overall level to some extent. Without exception, the Gulf of Thailand which is a part of South China Sea is believed to be affected by the accident to some extent. To re-evaluate the distribution pattern of artificial as well as natural radionuclides in this oceanographic region, International Atomic Energy Agency has therefore agreed with Office of Atomic Energy for Peace(OAEP) to conduct a coordinated research program namely "Sources of Radioactivity in the Marine Environment and their Relative Contributions to Overall Dose Assessment from Marine Radioactivity". The research was fulfilled in three year period while each year objective was slightly different.

In the first year program,OAEP is responsible in studying the level and a rough distribution pattern of naturally occurring radionuclides, H^3, C^{14} and K^{40}, and artificial radionuclides ; Sr^{90}, Cs^{137}, $Pu^{239,240}$ and Am^{241} in marine environment of The Gulf of Thailand. The research was conducted mainly on finding the naturally and artificially occurring radionuclides in the marine environment of Thailand emphasizing particularly on The Gulf of Thailand. The research was intended to evaluate a broad view of radioactivity status of the gulf.

Owing to the large fraction of present internal dose expected to be contributed by Cs^{137} and Po^{210} in sea food, the second and the third year research was therefore focussed only on the two mentioned radionuclides. Moreover, types of samples were selected to be sea water from the gulf and some species of marine biota collected from the gulf and the Andaman sea located

on the west coast of the country. The species of biota were selected to be duplicated, if possible, of both seas.

The results obtained from the research are expected to be useful for better understanding of the behavior of the radionuclides in semi-closed and shallow sea represented by the gulf. Furthermore, the dose evaluation by sea food consumption of Thai will also be figured out.

2. GEOGRAPHY

The Gulf of Thailand.[3]

The Gulf of Thailand is a shallow arm of the South China Sea. It locates between latitude 6°-13°, 30'N and longitude 99° -105°. The gulf extends approximately 724 kilometers from the north to the south. Its greatest width is approximately 482 kilometers; its greatest depth, in the center, is slightly more than 80 meters. The central depression has depths greater than 60 meters. Branching out from the central depression are troughs which appear to be narrow, drowned river valleys. Four large rivers; the Chao Phraya, the Bang Pakong, the Tha Chin and the Mae Khlong discharge fresh water into the head of the bight of Bangkok, but many smaller rivers flow into the gulf from both sides. The northeastern coast is generally shallower and flatter than the south-western coast.

The deeper central gulf is separated from the South China Sea by two ridges. One extends southwest at depths of less than 25 meters for more than 96 kilometers from Cape Camau. The second and deeper ridge extends northeast off Kota Bharu for a distance of 145 kilometers at depths of less than 50 meters. In the narrow, deeper channel between the ridges, a sill depth of 67 meters was observed. The bottom topography and depths of gulf are shown in Figures 1. and 2. The gulf bed composes of clay, silt and sand mixed in different ratio in different regions. The composition of sediment at the gulf bed is simply illustrated in Figure 3. Since the gulf bed is shallow comparing to South China Sea, it is regarded as a semi-closed bay situated on a continental shelf of the Pacific Ocean.

The Andaman Sea.(4)

The Andaman Sea is a part of the Bay of Bengal which situates on the upper part of the Indian ocean. It is bounded east by Burma and Thailand,north by the Gulf of Martaban and the Delta of the Irrawaddy river, south by Strait of Malacca and Sumatra and west by the Andaman and Nicobar Islands. The Andaman Sea is linked to the Pacific ocean by the strait of Malacca and the Bay of Bengal by numerous passages through the Andaman and Nicobar Islands. The distance between north to south is about 750 kilometers and approximate area of 218,000 square kilometers.

Sea floor of the Andaman sea occured by the submergence of the west shoreline of Thailand, therefore it is deeper than the Gulf of Thailand. West of Thailand coast its depth is approximately 120 meters, but east of Andaman Islands and Nicobar Islands there are two deep basins. The deepest one is 4426 meters under sea level. Sea floor of the Andaman Sea composes mainly of mud deposited by Irrawaddy river in the north and sedimentary rocks in the south. However, along the west coast of Thailand the shoreline floor composes of mud which is drained by many short rivers on the peninsula of Thailand.

3. OCEANORGRAPHIC CHARACTER

The Gulf of Thailand.

The Gulf of Thailand may be characterized as a classical two-layered, shallow water estuary on the basis of distribution charts. Low salinity water which has been diluted by heavy precipitation and water runoffs flows out of the gulf at the surface. There is inflow of high salinity, relatively cool water

from the South China Sea into the gulf over the 67-meter sill in the entrance channel. This high salinity water fills the deep, central depression below a depth of approximately 50 meters. Superimposed on this two-layered system is a complex circulation composed of the wind-driven currents related to the monsoon wind and tidal currents whose velocities in many locations exceed one knot. Neither the northeast nor the southwest monsoon winds were found to be constant in direction or velocity over the gulf as a whole. The interplay of forces due to the variable winds, tidal currents, fresh water runoffs and excessive precipitation gives rise to localized areas of divergence where low temperature, high salinity water, usually of low oxygen content, is upwelled. These forces also establish areas of convergence where high temperature, low salinity and highly oxygenated water sinks.

Classical oceanography has shown that ocean currents in the northern hemisphere flow at right angles to the slope of the density surfaces so that the lighter lies on the right hand of an observer looking in the direction of the current, and the denser water lies on the left hand. In shallow estuarine waters the assumptions upon which current velocities are calculated from the distribution of density based on temperature and salinity observations in the open ocean may not be valid. However, density distributions do give an indication of the direction of water movements both horizontally and vertically, and density gradients give evidence of relative current strength since horizontal motion must take place if horizontal density differences exist.

Density of a body water may be changed at the surface by river runoff, precipitation and effective incoming radiation.

Because of the earth's rotation, wind blowing over the sea surface will transport water at 45^{o} to the right of the wind. The wind may produce areas of coastal upwelling or sinking, convergence or divergence and currents; the wind-induced motion will alter the distribution of density both horizontally and vertically. Wind also alters density by mechanical mixing. In the shallow Gulf of Thailand where strong winds occur such mixing may extend to the bottom or it may be limited at the halocline by the extreme stability of the high salinity deep water at times of light winds.

Without wind fresh water river runoff would stay on the surface of the ocean water. In addition to the main channel of the Chao Phraya River discharging into the Bight of Bangkok, there are also one river to the east, the Klong Dan, draining the same basin and three other rivers draining different regions. These latter are from east to west; the Bang Pakong, draining southeastern Thailand, the Tha Chin and the Mae Khlong, draining the Bilauktaung Mountains on the western border of Thailand. According to the 1970-1971 surface salinity observations compiled by the Royal Thai Navy[3] indicate that river and very rapidly in the Bight of Bangkok. In the delta region, the penetration of the tides up the rivers is a function of the river flow. In winter at times of low river runoff, saline water penetrates the Chao Phraya River channel as far north as Bangkok at high tide. There is little mingling of the two water masses at the interface except when mixed by outside forces. Most of the mixing appears to take place beyond the mouths of the rivers where the wind provides the force.

The Andaman Sea.

The Andaman sea water is a typical Southeast Asian Water that the light water of low salinity and high temgerature is formed at the surface and the water masses of higher density in and below the discontinuity layer must originate outside the region. The surface circulation in the Andaman Sea is controlled by the monsoon. The winds produce the characteristic seasonal and annual current pattern, which in turn influences the distribution of all the hydrological parameters espectially the upper layer. The winter winds cause south-southwesterly currents to flow in the Andaman Sea. The northwesterly flow from the Strait of Malacca into the southern part of the sea during most of the year. This flow is strongest during the northeast monsoon from January to April.

The surface water mass of the Andaman Sea extends down to 75 to 130 meters. This part of water is affected by the changing of the monsoon seasons. The water shows the high temperatures typical of tropical region 26-30^{0} C. In addition the influence of the low salinity reduces the density. The large excess of rainfall over evaporation and a large continental runoff causes an average salinity of less than 34 o/oo. The properties of the surface water normally extend a certain distance into the depth, before the transition to the colder water take place.

Regional Climate.

Monsoon winds are the dominant factor in the climate of South-east Asia. The onset of the monsoons varies to some extent, but the southwest monsoon is usually well established in May and ends of September. These winds, having blown across the Indian Ocean and Bay of Bengal, come laden with moisture and bring

abundant rainfall to Thailand, Cambodia and Viet Nam between July and October. Annual rainfall peaks occur at different times at different locations due to the irregular distribution of sea, land and mountain chains, and the prevailing southwest direction of the winds of the Indian Ocean become more variable in speed and direction over Southeast Asia land and water. The annual peaks of the Chao Phraya and Mekong River occur August to November. The area drained by the Chao Phraya River system is fairly small and simple ; increased rainfall over Thailand is reflected in the river flood peak about on month later. Rainfall data from locations around the Gulf coast,where rivers are short and drainage areas small, are good indicators of expected fresh water dilution of near shore Gulf areas.

The dry season is that of the northeast monsoon which normally begins in November and ends in February,but occasionally surges of the northeast monsoon may still be experienced in March or even in early April. The period is one of variable moderate winds over the Gulf and mild pleasant temperatures on land.

There are two periods of transition between the opposing monsoons, one of two month duration (March-April) and one of one month(October). The northeast monsoon winds blow across China and contain little moisture by the time they reach the coastal waters of Viet Nam and the Gulf of Thailand. The northeast direction is steady over the South China Sea but is variable in the Gulf of Thailand.

4. SEA FOOD PRODUCTION POTENTIAL

Thailand has two seas located on both sides of its

southern tip : one is the gulf of Thailand and the other is Andaman Sea which is part of the Indian Ocean. Thailand is therefore one of countries that have high capability of catching marine biota and producing sea food. Comparing between major marine product producing countries, its capability of producing approximately 2 millions tons per year makes it stand in the ninth position as shown in Fig.4. However, the total catches have markedly increased since 1985 as shown in Fig.5 and reach 2.6 millions tons in 1987. Apart from this quantity, Thailand has also produced sea food by coastal aqua-culture farming especially of shrimp and prawn. The production capacity from capture marine fisheries by group of species has increased from 2,064 x 10^3 tons in 1977 to 2,370 x 10^3 tons in 1989(5). Due to the deterioration of marine environment which consequently decrease natural food web for fish and biota in recent year, the method for producing sea food by farming is getting important role and expected to increase sea food production potential of Thailand in the near future.

5. SAMPLE COLLECTION

In the cruise during 9-16 March 1989, samples were collected on board of the Pramong-1 research vessel of Fishery Department, Ministry of Agriculture and Cooperative. Five sampling locations were selected according to spatial distribution and the depth of sea floor. At each location, some physical parameters of the sea such as the depth of sea floor, surface water temperature and salinity of surface water were measured then 200 liters of sea water was suctioned and kept in

20 liter polyethylene carboys. Sea sediment was randomly bored by a Gravity Corer of 2 inch diameter,kept in polyvinyl chloride tubes and frozen before bringing back to a laboratory for separating profile and drying. Ten to fifteen cores were obtained from eachlocation depending on the hardness of the sea bed. The sampling locations and related physical parameters of the sea at collection time are shown in Fig.6 and Table 1. respectively.

For marine biota, 5 species were selected according to their feeding behavior. Indo-Pacific mackerel was selected to represent pelagic fishes, Snappers was selected to represent demersal fishes, Bloody cockle was selected to represent shellfish, shrimp was selected to represent crustacea and squid was selected to represent cephalopoda. Twenty to twenty five kilograms wet weight of each species was taken from Samut Prakan sea food market which is a big fresh sea food distributor close to Bangkok.

Table 1. Physical Parameters of Sampling Locations of the gulf.

Location	Sampling Date	Latitude (N)	Longitude (E)	Depth m	Temperature °C	Salinity o/oo
ST-5	16 Mar.89	13° 15'	100° 35'	13	27.7	32.4
ST-1	9 Mar.89	13°	100° 30'	22	24.9	32.6
ST-2	11 Mar.89	12°	100° 25'	32	26.6	33.4
ST-3	13 Mar.89	10° 25'	100° 30'	57	26.2	32.5
ST-4	14 Mar.89	9° 28'	100° 30'	43	26.0	32.7

6. SAMPLE PREPARATION AND ANALYTICAL PROCEDURES

Before conducting radiochemical analyses of H^3, C^{14}, Sr^{90}, Cs^{137}, $Pu^{239,240}$, and Am^{241}, various types of samples were prepared as follows:

6.1 Sample Preparation

Sea water

Sea water was filtered through a capsule filter 0.2 um of Gelman Sciences Inc.

Sediment

Sea sediment was defrozen and then dissected to obtain top 10 centimeter section and dried at 105 ^{o}C in an electric oven until constant weight was obtained. The sample to be analyzed for Sr^{90} and transuranium elements was ashed at 450^{o}C in an electric muffle furnace for 24 hours.

Biota

Each spicies of marine biota was dissected to separate the edible part from bone or shell. The edible part was then dried at 105 ^{o}C in the oven until constant weight was obtained. The sample to be analyzed for Sr^{90} and transuranium elements was ashed as sediment sample.

After preparation, the samples were separated to be analyzed for the interested radionuclides by different procedures. A brief description of radiochemical procedures for analyzing each radionuclide is shown in the appendix I.

7. RESULTS AND DISCUSSION

The natural and artificial radionuclide activity in sea water collected from the gulf at different locations between 1989-1991 are shown in Tables 2. and 3. respectively. Since the activity of the nuclides between locations and collecting time is not significantly different, the averaged activity of K^{40} is estimated to be 12.55 Bq/l and C^{14}, Po^{210}, Sr^{90}, Cs^{137} are 5.33, 0.71, 1.40, 3.66 mBq/l and $Pu^{239,240}$ is 3.23 uBq/l respectively. These figures are then used for calculating the bioaccumulation factors, the ratio of radionuclide activity in biota to its living media, of different types of marine biota. The estimated factors are shown in Table 11.

The factors were calculated by using radionuclides activity concentrated in muscle and activity in filtered sea water. If taking into account total activity of the nuclides in unfiltered samples, the factors are expected to be lower. When comparing between species and types of biota Po^{210} and $Pu^{239,240}$ tend to be discriminately concentrated in muscle of bloody cockle which is a bottom feeder. Other types of biota do not clearly have different factors among species.

Natural and artificial radionuclides in sediment collected from 5 locations in the gulf are shown in Tables 4 and 5. The ranges of C^{14}, K^{40}, Th^{232}, U^{238}, Sr^{90}, Cs^{137} and $Pu^{239,240}$ are 5.21-7.52, 313.86-504.23 , 31.2-48.9 , < 35.5-41.9, 0.001-0.004, 0.82-2.14 and 0.017-0.114 Bq/kg respectively. Speaking for artificial radionuclides,the degree of strong absorption on sediment are arranged in the order Cs^{137} > $Pu^{239,240}$ > Sr^{90}. Since the composition of sediments collected from the sampling locations are different as indicated in Fig.3, this is expected to affect

the activity of each nuclide at the location. But unfortunately, the analyses of sediment composition have not been performed. On the other hand, the data of c^{14} at each location are obtained from the analyses of carbonate fraction of sediment which is composed mainly of shell and calcium carbonate precipitated from sea water. Therefore, the percent carbonate content tends to be correlated with the C^{14} activity at each location.

Radionuclide activity in biota collected from the gulf and the Andaman sea during 1989-1991 are shown in Tables 6, 7, 8, 9 and 10. Generally speaking, the level of these nuclides in the samples from both seas are not significantly different. Many nuclides are not highly concentrated in specific type of organisms except Po^{210} and $Pu^{239,240}$ which show tendency to concentrate in shellfish such as squid, bloody cockle and mussel. The detail investigation of these trends will be performed in the separated work.

It should be noted that the data of Po^{210} in biota in the first and second year reports were adjusted to some extent in the final report because of the differences between Po^{209} standards used as a tracer in the analytical procedure of Po^{210} of each year.

The results of Po^{210} in surface sea water and marine biota of 1991 are considered to be more reliable because of the confidence of the exact activity of Po^{209} standard tracer given by Harwell Laboratory, England. In the Polonium method, Po^{210} contents in samples are not measured that is the reason why Po^{210} decay factors are not corrected because of the unknown contributions from Pb^{210}. The sampling, measuring dates and

counting periods of Po^{210} in surface sea water and marine biota are shown in Appendix II and III for supporting interpretation of the Po^{210} results.

Table 2. Natural Radionuclides in Surface Sea Water Collected from the Gulf of Thailand during 1989-1991. The sampling, measuring dates and counting periods of Po^{210} are shown in Appendix II.

Location	Carbon Content	C^{14} Activity		K^{40}	Po^{210}	
	g/l	PMC	mBq/l±2σ	Bq/l±2σ	mBq/l±2σ	
Collection Period			1989	1989	1990	1991
ST-5	0.0198	107	4.87±0.18	13.27±0.34	1.10±0.40	0.12±0.04
ST-1	0.0214	121	5.96±0.20	12.68±0.06	1.31±0.38	0.34±0.08
ST-2	0.0198	115	5.24±0.18	12.29±0.48	1.04±0.38	0.29±0.06
ST-3	0.0206	112	5.31±0.18	11.78±0.34	1.26±0.40	0.25±0.06
ST-4	0.0201	114	5.27±0.18	12.75±0.36	1.15±0.34	0.16±0.04

Table 3. Artificial Radionuclides in Surface Sea Water Collected from the Gulf of Thailand during 1989-1991.

Location	Sr^{90}	$Pu^{239,240}$	Cs^{137}			Am^{241}
	mBq/l±2σ	μBq/l±2σ	mBq/l±2σ			μBq/l±2σ
Collection Period	1989	1989	1989	1990	1991	1989
ST-5	6.38±0.56	0.84±0.008	3.43±0.62	3.40±0.49	3.14±0.44	*
ST-1	< 16.3	2.31±0.09	4.25±0.50	3.00±0.43	3.89±0.50	79.6±41.9
ST-2	< 4.0	6.49±2.23	4.13±0.72	4.01±0.47	3.72±0.34	*
ST-3	5.12±4.24	3.74±1.40	3.76±0.58	3.56±0.46	4.33±0.52	171±9.47
ST-4	< 19.0	2.81±1.07	3.72±0.56	2.94±0.44	3.60±0.56	3.1±0.25

* data are not available.

Table 4. Natural Radionuclides in Sediment Collected from the Gulf of Thailand in 1989.

Location	Carbonate Content	C^{14}		K^{40}	Th^{232}	U^{238}
	% dry wt.	PMC	Bq/kg±2σ	Bq/kg±2σ	Bq/kg±2σ	Bq/kg±2σ
ST-5	18.8	87	-7.52±0.18	346.43±4.72	31.3±1.0	<50.5
ST-1	17.5	65	5.23±0.16	350.56±4.26	31.2±1.0	41.9
ST-2	15.5	73	5.21±0.14	313.86±3.32	46.5±0.78	<35.5
St-3	16.8	69	5.33±0.16	379.31±5.66	48.9±1.22	<62.3
ST-4	17.1	77	6.06±0.16	504.23±5.74	42.3±1.26	<35.0

PMC is percent modern carbon.

* data are not available

Table 5. Artificial Radionuclides in Sediment Collected from the Gulf of Thailand in 1989.

Location	Sr^{90} Bq/kg dry wt. ±2σ	Cs^{137} Bq/kg dry wt. ±2σ	$Pu^{239,240}$ Bq/kg dry wt. ±2σ	Am^{241} mBq/kg dry wt. ±2σ
ST-5	<0.30	2.14 ± 0.24	0.114 ± 0.008	*
ST-1	<1.54	0.82 ± 0.22	0.017 ± 0.003	294 ± 71.5
ST-2	<0.67	1.04 ± 0.20	0.033 ± 0.004	95.5 ± 3.37
ST-3	<0.25	1.59 ± 0.32	0.074 ± 0.006	*
ST-4	<10.86	1.66 ± 0.26	*	*

* data are not available.

Table 6. Natural Radionuclides in Marine Biota Collected in 1989.

Species	the Gulf of Thailand				
	C^{14}			K^{40}	
	Carbon Content % dry wt.	PMC	Bq/Kg dry wt. ± 2σ	% dry wt	Bq/kg dry wt ± 2σ
Indo-pacific					
Mackerel	22.9	108	57.0 ± 2.10	20.7	459.94 ± 5.09
Snapper	21.7	104	52.0 ± 2.00	16.0	489.90 ± 2.52
Squid	26.7	103	64.0 ± 2.46	8.7	185.52 ± 2.86
Shrimp	25.3	108	63.0 ± 2.32	17.1	375.79 ± 4.34
Bloody cockle	22.7	109	57.0 ± 2.08	19.8	296.07 ± 2.20

Table 7. Natural Radionuclides in Marine Biota Collected in 1990.

The sampling, measuring dates and counting periods of Po^{210} are shown in Appendix III.

Species	the Gulf of Thailand			the Andaman Sea		
	% dry wt	K^{40}	Po^{210}	% dry wt.	K^{40}	Po^{210}
		Bq/kg dry wt. ±2σ			Bq/kg dry wt. ±2σ	
Indo-pacific Mackerel	38.53	294.94±4.9	8.43±1.74	23.27	396.69±4.9	14.63±3.80
Snapper	22.31	496.32±5.25	2.44±0.62	21.31	443.74±4.71	2.29±0.64
Rakegilled Mackerel	24.09	305.06±4.34	12.77±2.40	22.82	403.11±10.08	11.98±4.62
Threadfin bream	23.30	370.73±4.98	2.36±0.54	*	*	*
Bluemackerel scad	23.12	32.92±5.43	12.45±2.74	*	*	*
Tuna	*	*	*	25.61	382.67±34.23	36.22±9.96
Squid	16.13	293.43±4.03	21.00±4.74	16.10	226.17±3.62	103.04±19.26
Shrimp	17.99	242.33±3.58	5.57±1.18	20.27	363.39±5.21	14.56±2.72
Bloody cockle	19.79	284.13±3.78	21.95±16.02	*	*	*
Sea Mussel	12.78	248.65±3.53	92.81±18.44	13.83	362.76±6.13	102.18±21.76

* data are not available.

Table 8. Natural Radionuclides in Marine Biota Collected in 1991.

The sampling, measuring dates and counting periods of Po^{210} are shown in Appendix III.

Species	the Gulf of Thailand			the Andaman Sea		
	% dry Wt.	K^{40}	Po^{210}	% dry Wt.	K^{40}	Po^{210}
		Bq/kg dry wt.±2			Bq/kg dry wt.±2	
Indo-pacific Mackerel	25.06	318.04±3.64	4.62±0.56	*	*	*
Snapper	22.59	545.57±8.88	1.04±0.20	23.24	487.57±4.34	0.89±0.18
Rakegilled Mackerel	25.28	366.83±5.25	5.22±0.46	24.93	474.97±5.29	9.12±1.18
Threadfin bream	24.55	420.94±8.92	1.27±0.16	*	*	*
Bluemackerel scad	28.08	310.56±4.01	7.36±0.54	28.50	377.45±4.77	16.37±2.10
Tuna	*	*	*	26.70	436.62±4.15	27.26±2.28
Squid	16.99	235.79±3.05	19.26±1.40	16.45	361.54±3.69	9.05±0.84
Shrimp	20.77	423.53±5.71	3.21±0.32	19.67	365.57±4.29	15.17±1.22
Bloody cockle	19.78	336.52±3.64	15.69±1.20	*	*	*
Sea Mussel	16.12	314.01±4.49	32.01±3.52	14.36	*	47.73±7.38

* data are not available.

Table 9. Artificial Radionuclides in Marine Biota Collected from the Gulf of Thailand during 1989-1991.

Species	Sr^{90}	$Pu^{239,240}$		Cs^{137}		Am^{241}
	Bq/kg dry wt. ±2σ	mBq/kg dry wt. ±2σ		Bq/kg dry wt. ±2σ		mBq/kg dry wt. ±2σ
	1989	1989	1989	1990	1991	1989
Indo-pacific						
Mackerel	<0.39	0.87±0.03	0.61±0.19	0.34±0.13	0.19±0.10	2.46 ± 0.56
Snapper	<1.26	1.38±0.45	0.74±0.09	0.49±0.14	0.45±0.08	33.8 ± 4.15
Rakegilled						
Mackerel	*	*	*	0.56±0.01	0.09±0.10	*
Threadfin						
bream	*	*	*	0.96±0.13	0.94±0.18	*
Bluemackerel						
scad	*	*	*	0.88±0.18	0.32±0.10	*
Squid	0.36±0.09	0.62±0.30	0.18±0.08	0.21±0.12	0.18±0.06	7.47 ± 0.12
Shrimp	*	1.39±0.50	0.10±0.15	0.27±0.10	<0.75	1.80 ± 0.76
Bloody						
cockle	<0.15	5.92±1.21	0.12±0.09	0.23±0.11	0.10±0.08	*
Sea Mussel	*	*	*	0.29±0.12	0.16±0.08	*

* data are not available.

Table 10. Radionuclides Cs^{137} in Marine Biota Collected from the Andaman Sea during 1990-1991.

Species	Cs^{137} Bq/kg dry wt. ± 2	
	1990	1991
Indo-pacific Mackerel	0.56±0.04	*
Snapper	1.74±0.06	0.47±0.06
Rakegilled Mackerel	0.29±0.10	0.45±0.12
Threadfin bream	*	*
Bluemackerel scad	*	0.46±0.10
Tuna	0.66±0.12	0.72±0.09
Squid	0.22±0.11	0.15±0.09
Shrimp	0.61±0.14	<0.75
Bloody cockle	*	*
Sea Mussel	0.32±0.01	*

* data are not available.

Table 11. *The Estimated Bioaccumulation Factors(l/Kg.) of Various Radionuclides in Different Species of Marine Biota Collected from the Gulf of Thailand.*

Radionuclide / Species	C^{14}	Po^{210}	Sr^{90}	Cs^{137}	$Pu^{239,240}$
Indo-pacific Mackerel	10,694	12,009	156	317	269
Snapper	9,756	3,475	248	362	426
Rakegilled Mackerel	*	18,191	*	86	*
Threadfin bream	*	3,362	*	264	*
Bluemackerel scad	*	17,735	*	168	*
Squid	12,007	29,915	248	222	191
Shrimp	11,820	7,934	*	116	429
Bloody cockle	10,694	31,267	57	47	1,827
Sea Mussel	*	132,208	*	64	*

* data are not available.

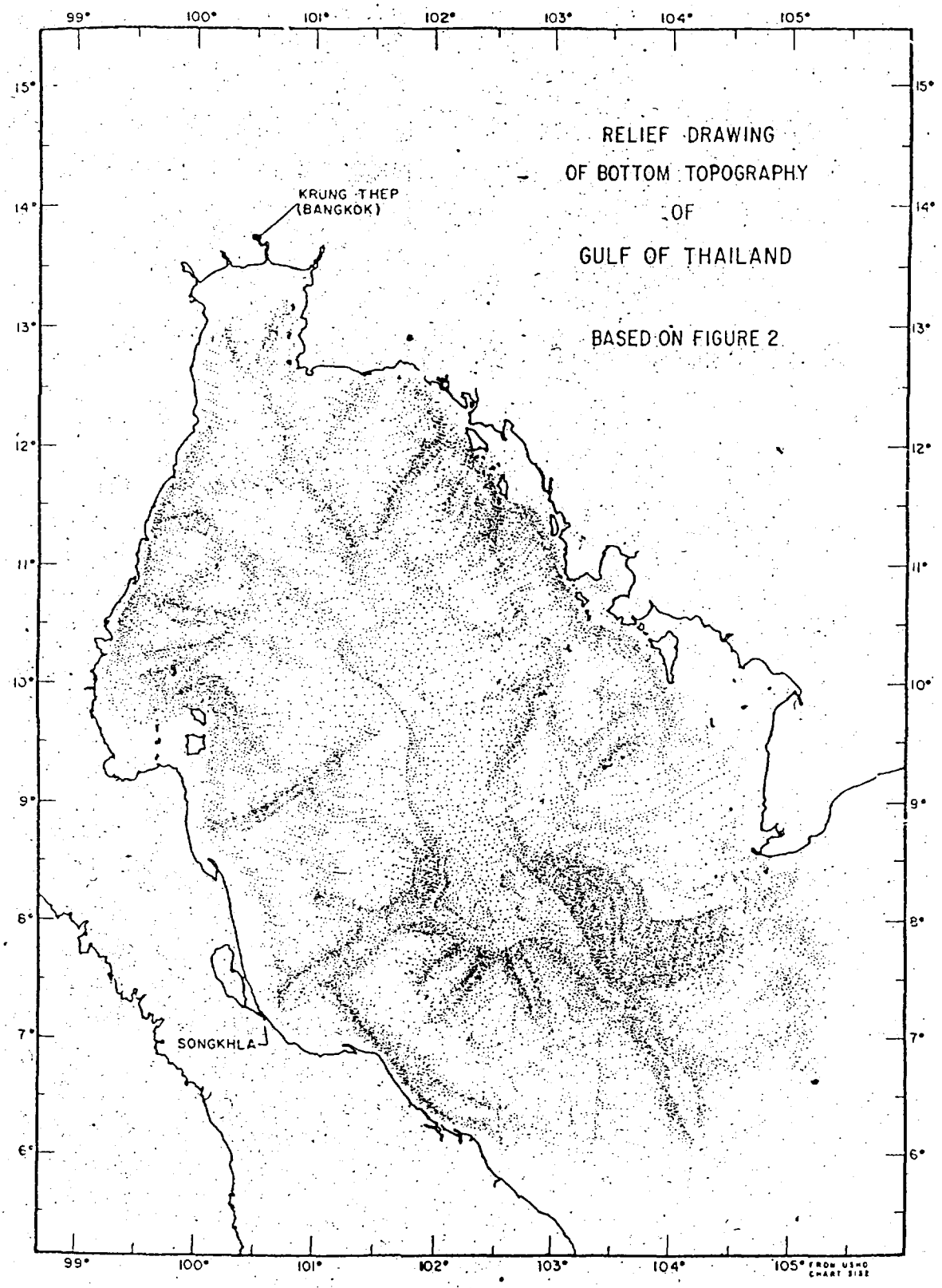

Fig.1 Bottom Topography of the Gulf of Thailand.

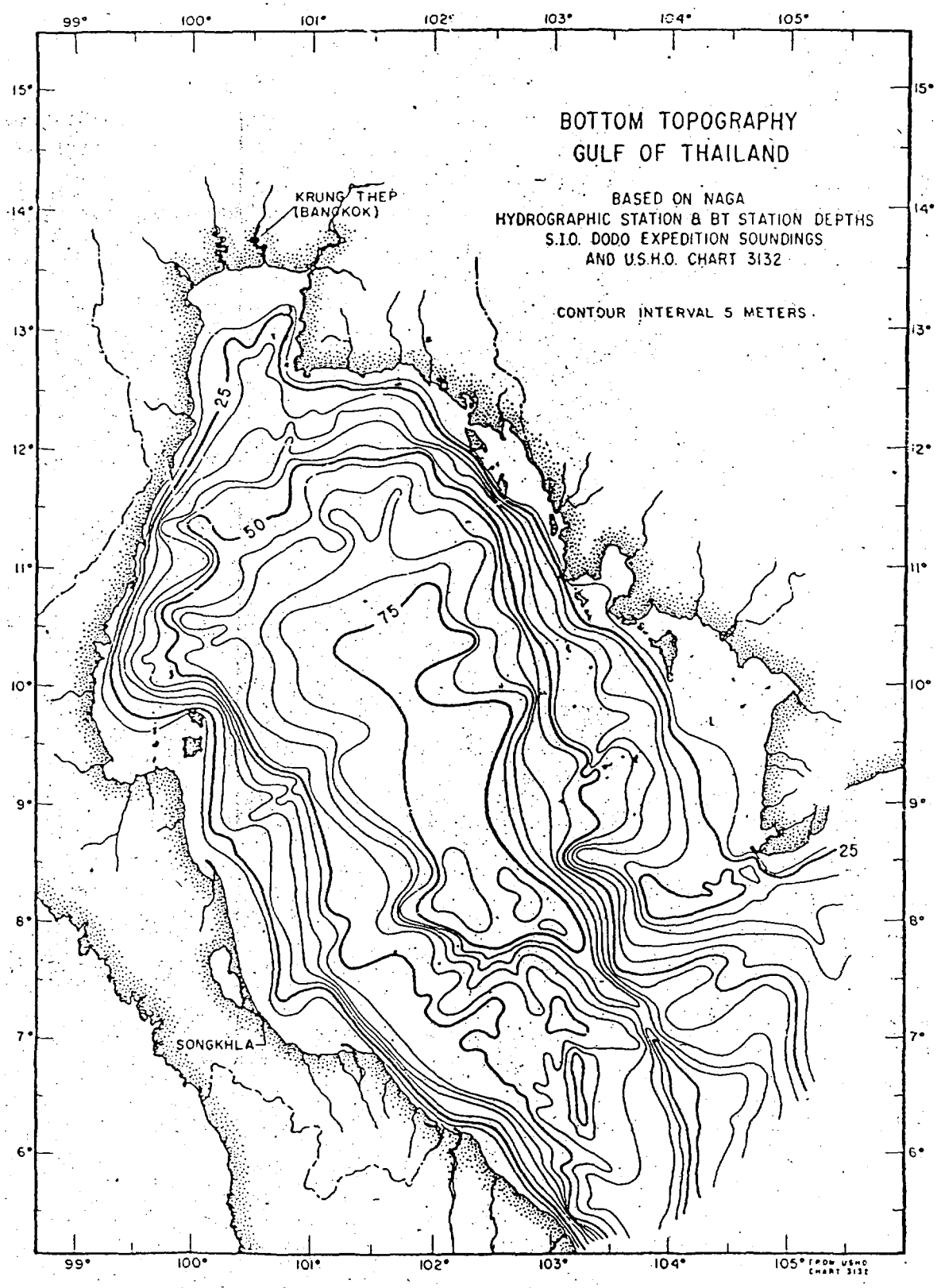

Fig.2 Depths of Floor of the Gulf of Thailand.

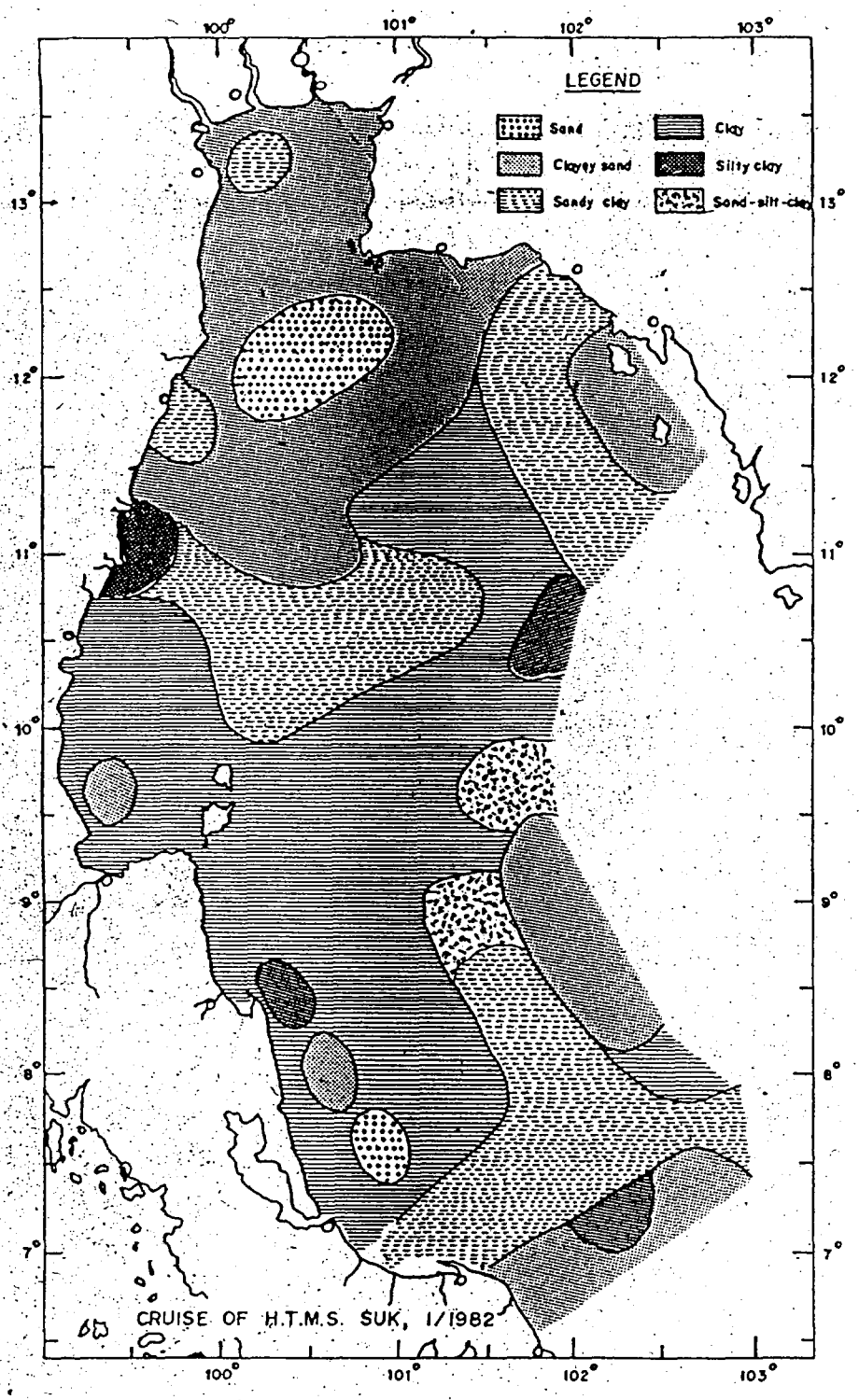

Fig.3 The sea bed composition of the Gulf of Thailand.

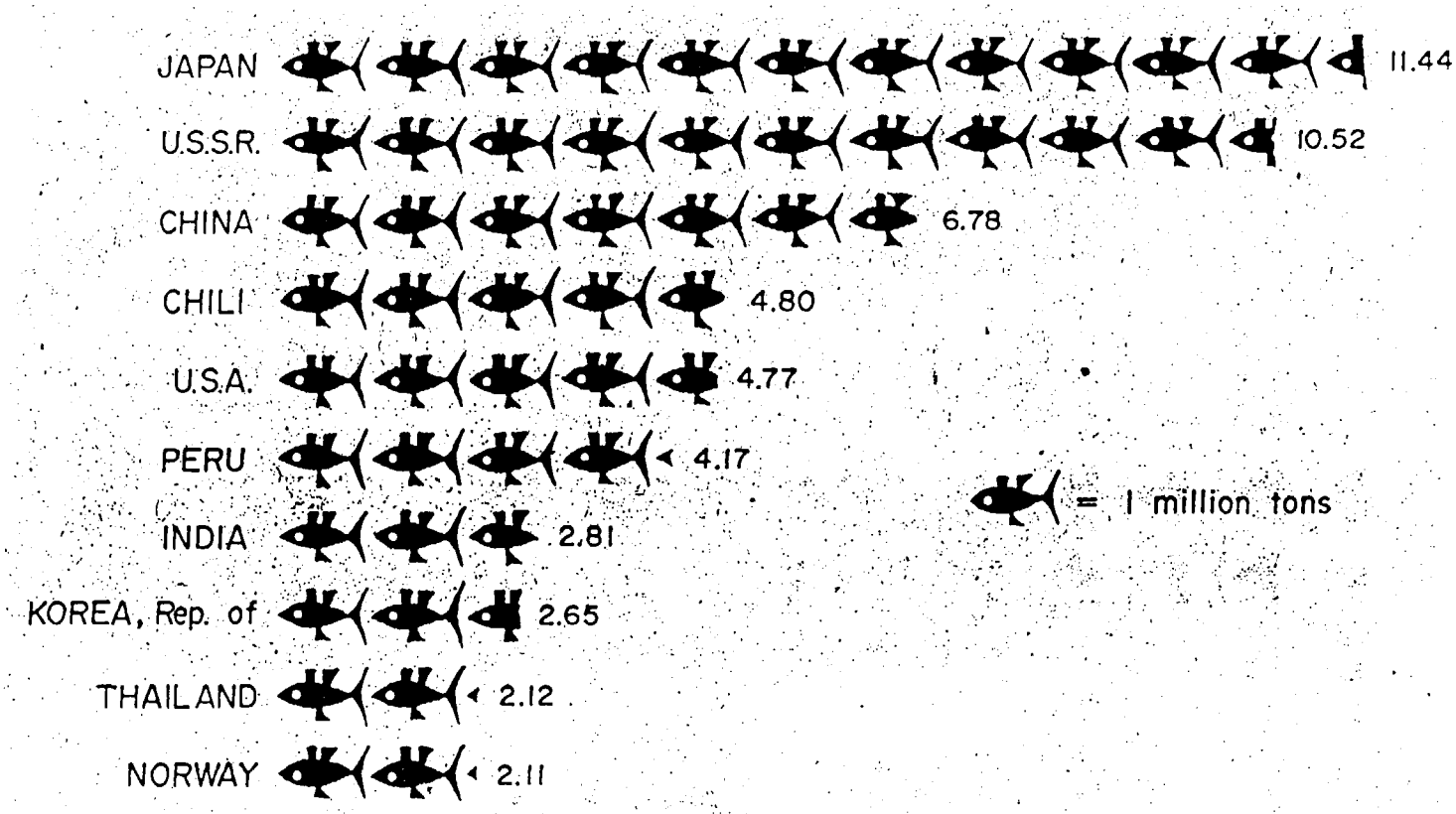

Fig. 4 Catches by Major Fishing Country (1985)

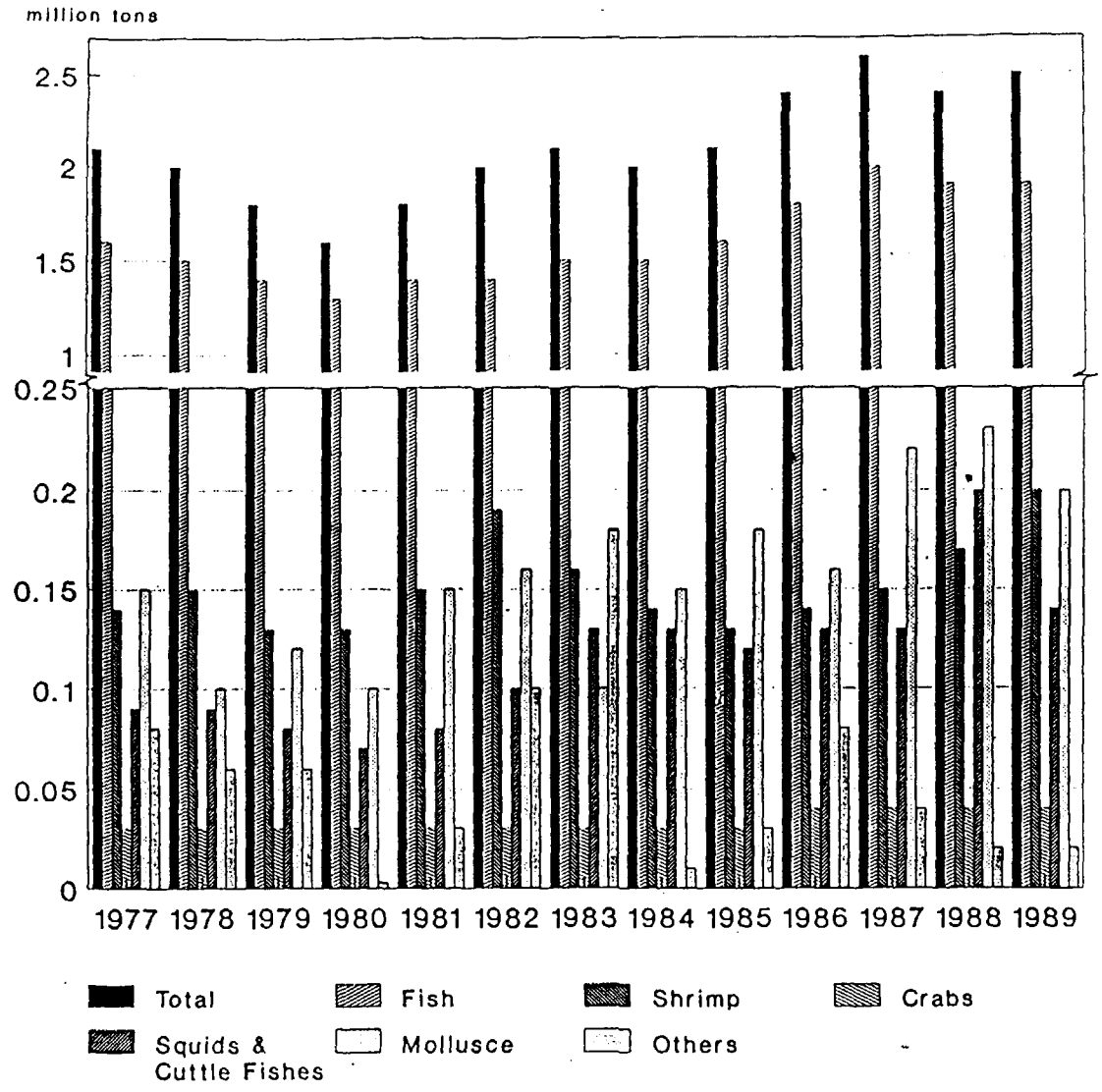

Fig.5 Estimated Anual Catch of Principal Marine Biota during 1977-1989

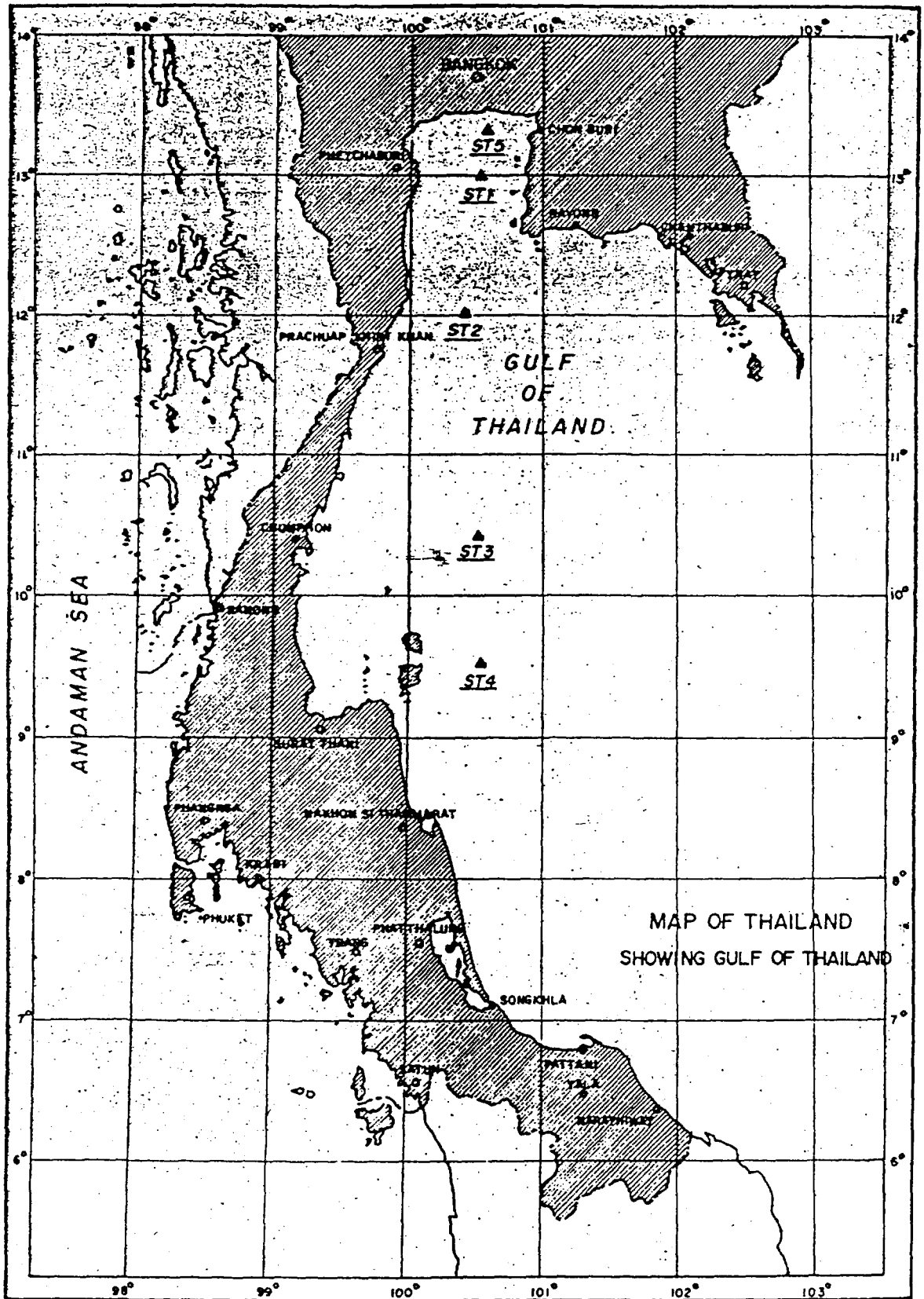

Fig. 6. Sampling Stations in the Gulf of Thailand.

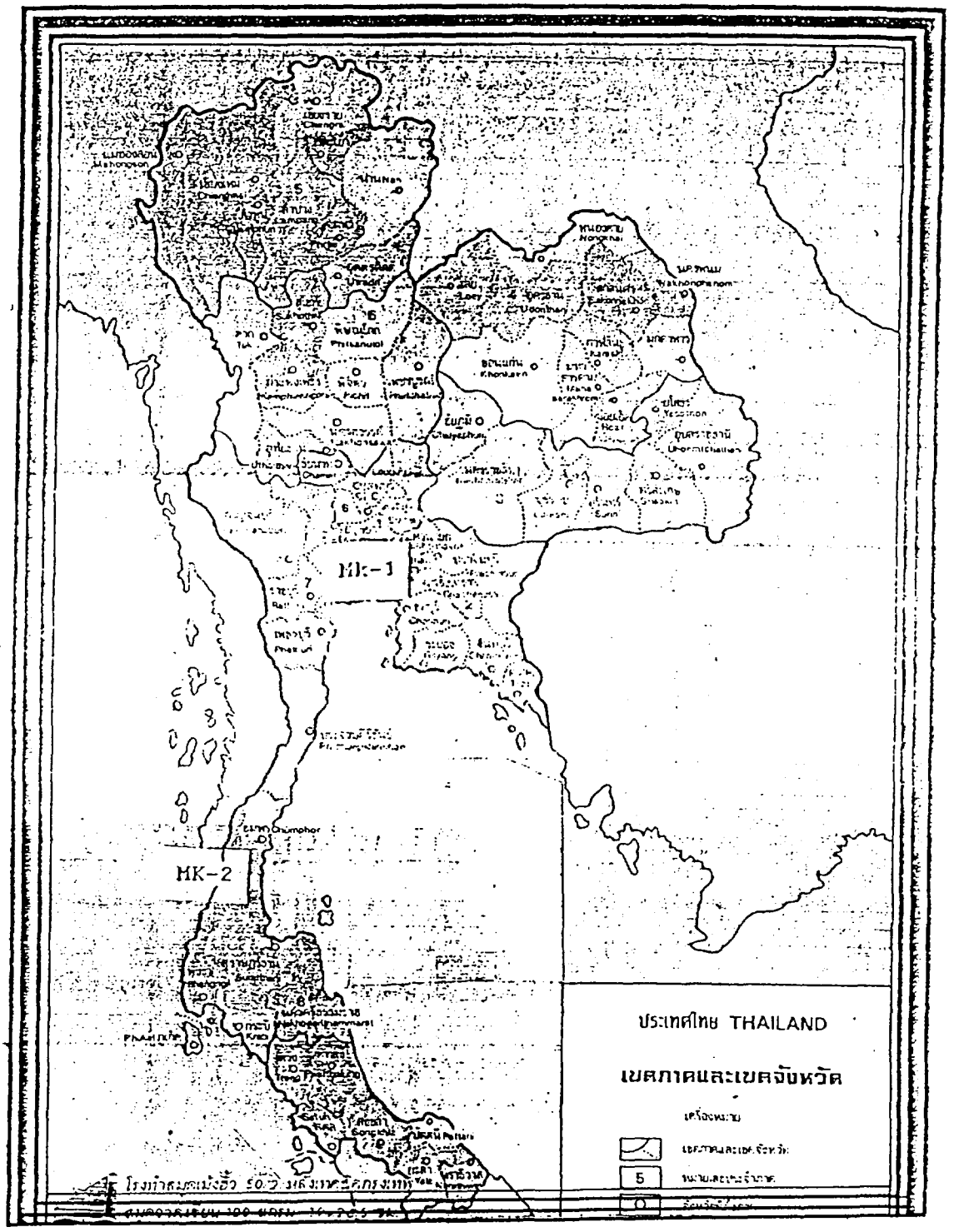

Figure 7. Sampling Markets of Marine Biota. MK-1 is Samutprakarn Sea Food Market. MK-2 is Ranong Sea Food Market.

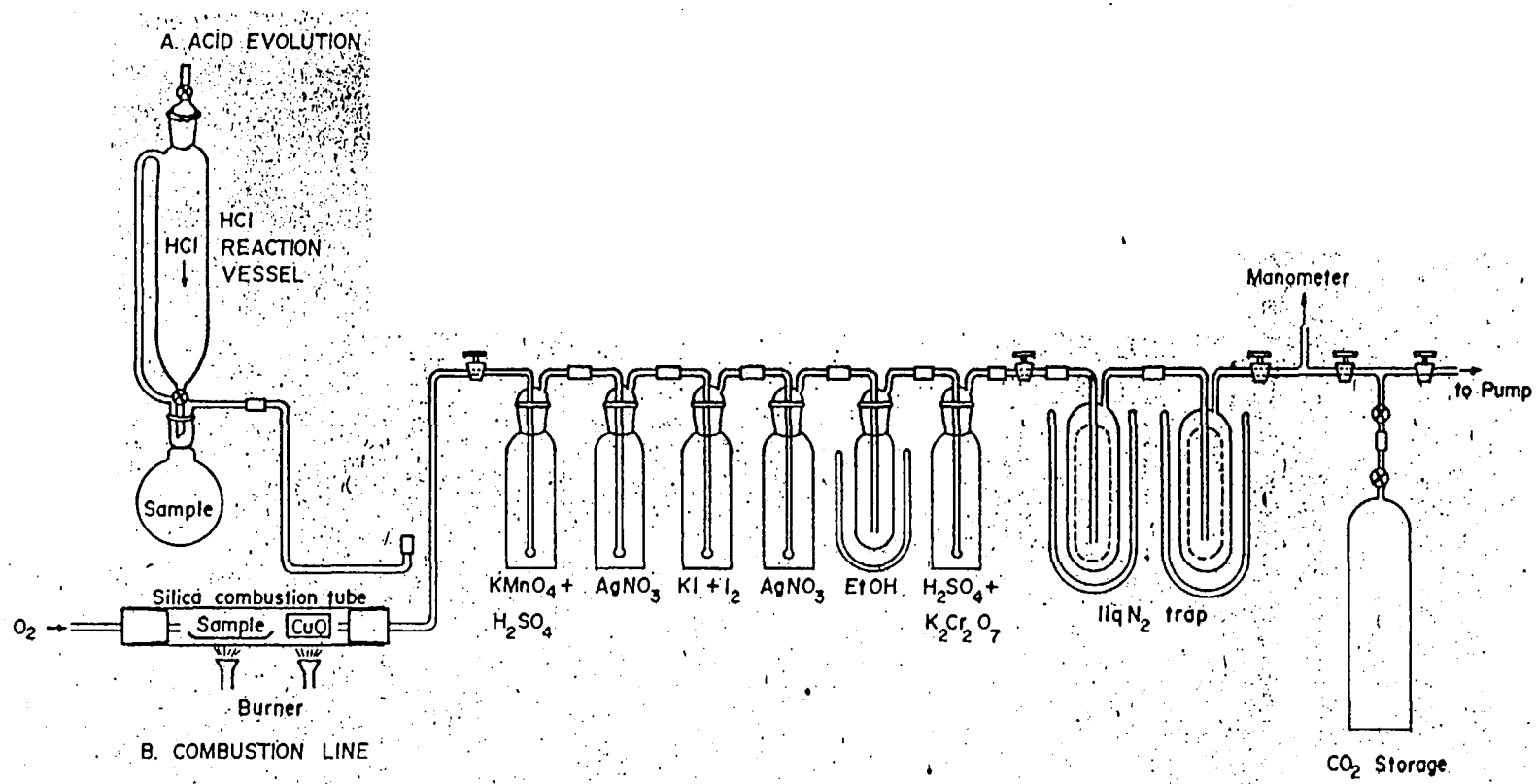

Fig. 8 Carbondioxide Generation Lines

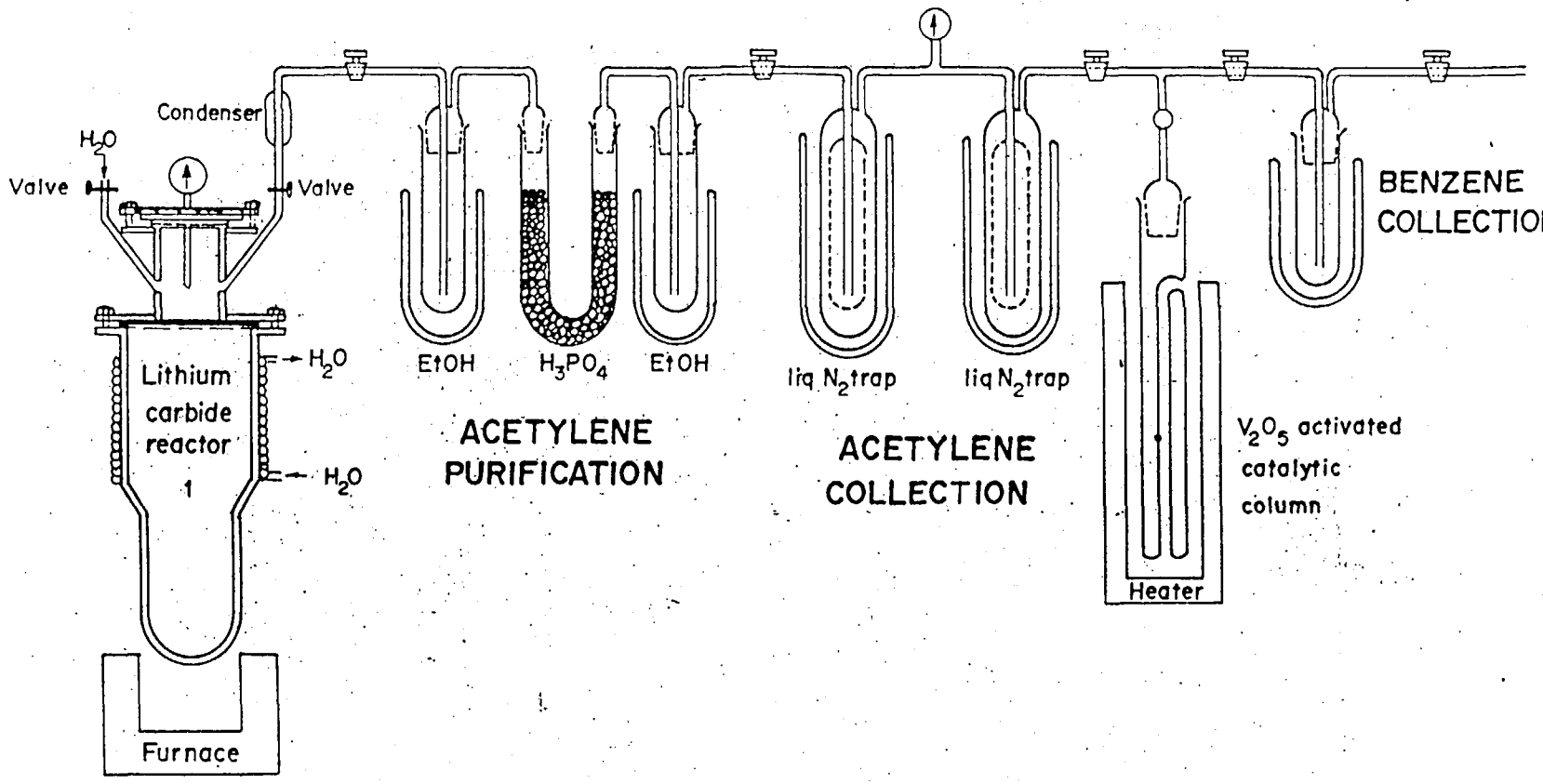

Fig. 9 Lithium Carbide Reactor, Acetylene Generation and Benzene Reaction Line.

Appendix I

Analytical Procedures

H^3

Tritium activity of surface sea water samples was determined by converting the samples to benzene and counting in a liquid scintillation counter. The conversion of water to benzene has been described in detail by Tamer, M. et al.(6)

The samples were distilled in an almost closed distillation system to prevent atmospheric contamination. The distillated water sample was added to commercial calcium carbide and then generated acetylene gas was collected. The acetylene was converted to benzene by using vanadium-activated silica--alumina catalyst. The tritium activities in the samples, standard and background have been determined in a liquid scintillation counter Packard Tricarb Model 2425.

C^{14}

Liquid scintillation counting of benzene was selected for radiocarbon measurement. The method involves benzene preparation and scintillation counting of benzene. Quantitative synthesis of benzene from sample material follows three well established chemical stages namely:

(I) oxidation or hydrolysis of samples to carbondioxide.(Fig.8) About 25 gm. of marine biota sample was combusted in fused silica combustion tube with oxygen stream. Impure carbondioxide was purified as it passes through a purification train and finally collected in two liquid nitrogen cold traps.

About 500 gm. of sediment was hydrolysed by addition of 2 N hydrochloric acid and carbon dioxide collected after

purification in two liquid nitrogen cold traps. About 200 liters of surface sea water was collected in 10 of twenty-liter plastic containers. Two hundred ml. of concentrated hydrochloric acid was added to each of containers. The evolved carbondioxide was carried by carbon dioxide free nitrogen gas in close circuit into saturated solution of sodium hydroxide to absorb the gas. The saturated solution of barium chloride was added to the alkali solution and filtered the barium carbonate precipitate. The barium carbonate was hydrolysed by adding 2 N hydrochloric acid and the carbondioxide was collected in the same way as of sediment samples.

(II) conversion of carbon dioxide to acetylene.(Fig.9)

The conversion of carbondioxide to acetylene has been described in detail by Polach and Stipp.[7] Carbondioxide was initially converted to lithium carbide in a carbide reactor. The lithium carbide was then hydrolysed to acetylene with the gases passed through a phosphoric acid trap to remove any impurities, in particular, ammonia through two ethanol/liquid nitrogen traps to remove water vapor and collected in two liquid nitrogen traps.

(III) catalytic trimerization of acetylene to benzene. (Fig. 9)

The frozen acetylene from the liquid nitrogen collection traps was allowed to sublime and passed onto a vanadium--activated silica-lumina catalyst column on which the acetylene was converted to benzene. Following completion of the trimerization reactor, the catalyst was heated to 110°C and the benzene collected under vacuum in two liquid nitrogen traps.

Synthesized benzene samples were transferred into 5 ml. , low-potassium silica vials into approximately 75 gm. scintillator (t-butyl.PBD). The vials were then sealed with stopper and the benzene weight (approximately 4.5 gm.) was accurately determined.

The carbon-14 activities in the samples 0.95 NBS oxalic acid standard and background have been determined in a Packard Tricarb Model 2425, modified for low-level counting.

Carbon-14 activity is expressed in term of PMC(percent of modern carbon)

$$PMC = \frac{A(s)}{A(std)} \times 100$$

where A(s) is specific activity of the sample

A(std) is specific activity of standard 0.95 NBS oxalic acid

note The standard specific activity of 0.95 NBS oxalic acid in AD1950 was found to be 13.56 $\pm$ 0.07 dpm/gm of carbon. Therefore 1 PMC = 0.0023 Bq/gm of carbon

Sr^{90}

One hundred liters of sea water was added with 10 gm. of strontium carrier and ammonium hydroxide to pH 10. The solution was mixed by bubbling with nitrogen gas for one hour, the ammonium carbonate was added into solution to precipitate alkali earth elements. The precipitate was separated from sea water

solution by suction and is ready for further steps of strontium separation.

Ten to twenty gm. of ash sample was added with 50 gm. of strontium carrier and digested with 8 M nitric acid/hydrogen peroxide and hydrofluoric acid several times until clear solution was obtained. The clear solution was then increased pH to 10 and ammonium carbonate powder was added to precipitate alkali earth elements. The precipitate was separated from clear solution by decanting and proceeded to further strontium separation steps.

Sediment sample was added with 50 gm. strontium carrier and leached with 8 M nitric acid/hydrogen peroxide 2 to 3 times. The leachate was raised pH to 10 and added with ammonium carbonate powder to precipitate alkali earth elements. The precipitate was separated from clear solution by decanting and proceeded to further strontium separation steps.

The carbonate precipitate of sea water, marine biota and sediment was dissolved in dilute nitric acid and fuming nitric acid was added to precipitate strontium as strontium nitrate. The strontium nitrate was then washed with acetone to remove calcium nitrate. The purification of strontium by fuming nitric precipitation was done for 2-3 times. Barium contaminated in precipitate was removed as barium chromate, the yttrium was separated from strontium by coprecipitation with ferric hydroxide. The solution was kept for 15 days for yttrium 90 milking then yttrium-90 was precipitated as yttrium oxalate for beta activity counting with a gas-flowed proportional counter BERTHOLD LB 2711. Efficiency and background of the counter is 25 % and 0.9 cpm respectively.

Cs^{137} (8,9)

Fifty liters of sea water sample was mixed with 50 gm. of ammonium phosphomolybdate (AMP) and mixed for 3 hours. The AMP precipitate was then left overnight and separated from sea water by suction. The precipitate was dried at 105°C and then pressed into a disc shape of 5.7 cm. diameter with pressure of 4000 lb/m^2.

Fifty kilograms of marine biota sample was dissected to separated edible part from bone or shell. The edible part was dried at 105°C until constant weight was obtained. The dry sample was filled in a Marinelli beaker.

Sediment samples of the weight ranging from 740 to 1670 gms. were dried, ground and filled in Marinelli beakers.

The AMP precipitate in a disc shape, the dry biota and sediment in Marinelli beakers were measured Cs^{137} peak with a 15% efficiency HPGe detector attached to a multichannel analyzer CANBERRA SERIES 35 PLUS. The detector was calibrated with standard reference material IAEA-154 (whey powder) and IAEA-152 (milk powder) in various geometry.

K^{40}, U^{238} and Th^{232}

Twenty liters of sea water was evaporated to 4 liters and filled in a Marinelli beaker. The sample was measured gamma radiation at peaks of 1461, 1001 and 911 kev. for K^{40}, U^{238} and Th^{232} respectively with the HPGe detector.

Samples of sediment and biota ash were prepared as the sample for Cs^{137} measurement. The standard NBS, Peruvian soil 4355 was used for calibrating the detector.

$Pu^{239,240}$(13,14)

One hundred liters of sea water was spiked with 1.5 dpm of plutonium-242 and 2.6 dpm of americium-243 tracers and mixed by bubbling with nitrogen gas. After reducing with sodium bisulfite at pH 9, the transuranic elements were coprecipitated ferrous hydroxide. The precipitate was then separated from clear solution by suction and proceeded to the further steps.

Ash sample was spiked with plutonium and americium tracer and digested with 8 M nitric acid and hydrogen peroxide or hydrofluoric acid. The clear solution obtained was proceeded for further plutonium purification steps.

Sediment sample was spiked with plutonium and americium tracers and leached with 8 M nitric acid and hydrogen peroxide. The transuranium elements were co-precipitated with ferric hydroxide at pH 9. The precipitate was dissolved in 8 M nitric acid and proceeded for further steps of plutonium purification.

Plutonium in 8 M nitric acid was purified by passing through a Dowex 1-X8 (100-200 mesh) column. The column was washed with 8 M nitric acid. The effluent and the wash was kept for americium purification procedures. The plutonium adsorbed on the column was eluted with 9 M hydrochloric acid and 0.1 M ammonium iodide. The eluted plutonium was co-precipitate with cerium trifluoride and filtered on a MILLIPORE membrane filter paper. The sample was then measured for alpha spectrum by a silicon surface barrier detector of ORTEC.

Am^{241}(13,14)

The americium in effluent of plutonium purification procedures was purified by co-precipitating with calcium oxalate.

The oxalate was ignited a 550°C overnight. The ignited precipitate was dissolved in concentrated hydrochloric acid and added with ferric carrier. The solution was raised pH to 9 to co-precipitate americium with ferric hydroxide. The precipitate was dissolved hydrochloric acid then passed through a double ion exchange column of Dowex 1-X8 and Dowex 50W-X8(100-200 mesh) to remove trace of Th, Fe, Po, U and Pu. The effluent from the column was then dried and dissolved in 8 M nitric acid. The solution was passed through a Dowex 1-X8 column to remove thorium from americium. The americium in the effluent was passed through a Dowex 1-X4 column and americium was eluted from the column with 1.5 M hydrochloric acid and 86 % methanol. The americium was prepared for measuring alpha spectrum as the same procedure of plutonium.

Po^{210}

Sea Water[15]

Collect 2 liters of sea water sample in a polyethylene container containing 40 ml of 11.7 M hydrochloric acid. The sample was transfered into a glass beaker and known amounts of polonium-209 was added. Carefully adjust the pH to 2.0 by adding ammonia solution and 20 ml of freshly prepared 4 % (w/v) APDC solution. Stir vigorously for 1 minute, then allow to stand for 5 minutes and stir again for 1 minute. Transfer 500 ml of sea water sample into a 1 l separating funnel and 100 ml of solvent(MIBK) was added and shaked the contents for 5 minutes, after which leave to separate out for 30 minutes. Discard the aqueous portion and other 500 ml of sea water into a separating funnel and repeat above procedure until 2 liters of sea water

was used. Add 6 ml of 11.7 M hydrochloric acid to the organic extract. After shaking, transfer the acid and organic layer to a glass beaker and evaporate nearly to dryness on a hot plate. Then follow the procedure as describes for marine biota sample.

Marine Biota[16]

Weigh 2 g of a sample and add known amount of polonium-209 into a 250 ml beaker. Add 5 ml of hydrochloric acid, 15 ml of nitric acid and 10 ml of perchloric acid into the beaker. Evaporate the solution to dryness at 150°C on a hot plate while adding about 5 ml of hydrogen peroxide occasionally until the clear solution was obtained. The residual was then dissolved in 50-100 ml of 0.5 M hydrochloric acid with warming. After 0.5 g of ascorbic acid was added to the solution, a silver disc coated on one side with heat resistance tape was immerged into the solution. The temperature of solution was maintained at 70-80°C for more than six hours on a hot plate while being stirred occasionally. After plating the silver disc was washed with distilled water and acetone respectively. The sample was counted for alpha spectrum by an ORTEC alpha spectrometer 576 A.

Appendix II

The sampling and measuring date of Po^{210} in Surface Sea Water

Location	Sampling Date		Measuring Date	
	1990	1991	1990	1991
ST-5	26/09/1991	09/08/1991	30/11/1990	13/11/1991
ST-1	26/09/1991	09/08/1991	23/11/1990	03/11/1991
ST-2	26/09/1991	09/08/1991	04/12/1990	04/12/1991
ST-3	26/09/1991	09/08/1991	04/12/1990	04/12/1991
ST-4	26/09/1991	09/08/1991	30/11/1990	16/12/1991

The range of counting period is $2x10^5$ - $5x10^5$ seconds

Appendix III

The sampling and measuring dates of Po^{210} in Marine Biota collected in 1990 and 1991.

Species	The Gulf of Thailand				The Andaman Sea			
	Sampling date		Measuring date		Sampling date		Measuring date	
	1990	1991	1990	1991	1990	1991	1990	1991
Indo-pacific Mackerel	15/06/90	15/08/91	20/11/90	19/11/91	22/06/90	*	04/02/91	*
Snapper	15/06/90	15/08/91	02/01/91	19/11/91	22/06/90	05/08/91	17/01/91	29/10/91
Rakegilled Mackerel	15/06/90	15/08/91	12/12/90	26/11/91	22/06/90	05/08/91	17/01/91	08/10/91
Threadfin bream	15/06/90	15/08/91	20/11/90	26/11/91	*	*	*	*
Bluemackerel scad	15/06/90	15/08/91	15/11/90	13/11/91	*	05/08/91	*	08/10/91
Tuna	*	*	*	*	22/06/90	05/08/91	15/04/91	18/10/91

Species	The Gulf of Thailand				The Andaman Sea			
	Sampling date		Measuring date		Sampling date		Measuring date	
	1990	1991	1990	1991	1990	1991	1990	1991
Squid	15/06/90	15/08/91	15/11/90	08/11/91	22/06/90	05/08/91	08/04/91	30/09/91
Shrimp	15/06/90	15/08/91	20/12/90	08/11/91	22/06/90	05/08/91	08/04/91	18/10/91
Bloody cockle	15/06/90	15/08/91	15/04/91	29/10/91	*	*	*	*
Sea Mussel	15/06/90	15/08/91	12/12/90	29/10/91	22/06/90	05/08/91	03/04/91	30/09/91

The range of counting period is 2×10^5 - 5×10^5 seconds.

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