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Abstract

The concentration and distribution of mercury in environmental samples (sea water, rock, sea sand, sediment, soil, fish and etc.) taken from various coastal sea areas and its ambient area in Malaysia were investigated.

Mercury in environmental samples was subjected to the reduction-vaporization using of tin (II) chloride and heating-vaporization method. The mercury vapor was concentrated with a porous gold collector. Afterwards, mercury vapor evolved from the heated collector was determined by cold vapor atomic absorption spectrometry.

In coastal sea water of Malaysia, the geometric mean of inorganic mercury and inorganic mercury plus organic mercury were 3.5_2 and 3.9_8 ng l^{-1} which are 1.9 and 1.7 times higher than those for the East China Sea, respectively.

Arithmetic and geometric mean values of total mercury concentration in sea and river sediments were 30.4 and 20.1 μg kg^{-1}, respectively, and the polluted river sediments in Port Kelang was about one order higher than both of these mean.

In fish meat, the chemical form of mercury was mainly organic mercury and the inorganic mercury was negligible. Mercury concentration was high in carnivorous fish such as sharks and crabs compared to other fish species.

Key words :Mercury, Sea water, Sediment, fish, Distribution.

Introduction

The mercury concentration of sea water is usually very low, as little as several nanogram per liter[1), 2)]. Thus, it is very difficult to get precise and reliable analytical results for mercury by direct measurement without separation. Pretreatment and pre-concentration techniques are often required in order to determine the amount of mercury in these kinds of samples. Due to the low concentration of mercury, its chemical form is not well understood.

Mercury in environmental samples was subjected to reduction-vaporization using of tin (II) chloride and heating-vaporization method. The mercury vapor was concentrated with a porous gold collector. Afterwards, mercury vapor evolved from the heated collector was determined by cold vapor atomic absorption spectrometry.

The analytical method has been reported that total mercury[3)-5)] in liquid sample was determined by cold vapor atomic absorption, and the organic mercury[6), 7)] was determined by gas chromatography. Time was needed for using these two methods, and manipulation was complicated.

Authors improved means of Magos[8)], Umezaki *et al.*[9)] and Kamada *et al.*[10)] and carried out concentration of mercury by a porous gold collector[2), 11)].

In this study, we determined the concentration of mercury in environmental samples collected at various coastal sea areas and its ambient area in Malaysia. We discuss, the mercury concentration, chemical form, their mutual relations and distributions based on analytical results of environmental samples from Malaysia.

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Experimental

Collection and preservation of sample

Sampling stations of Malaysian environmental samples (sea water, rock, sea sand, sediment, soil and fish etc.) are shown in Figure 1.

Collection vessel of sample used washed polystyrene bottle for soil, rock, sea-sand, sediment and fish.

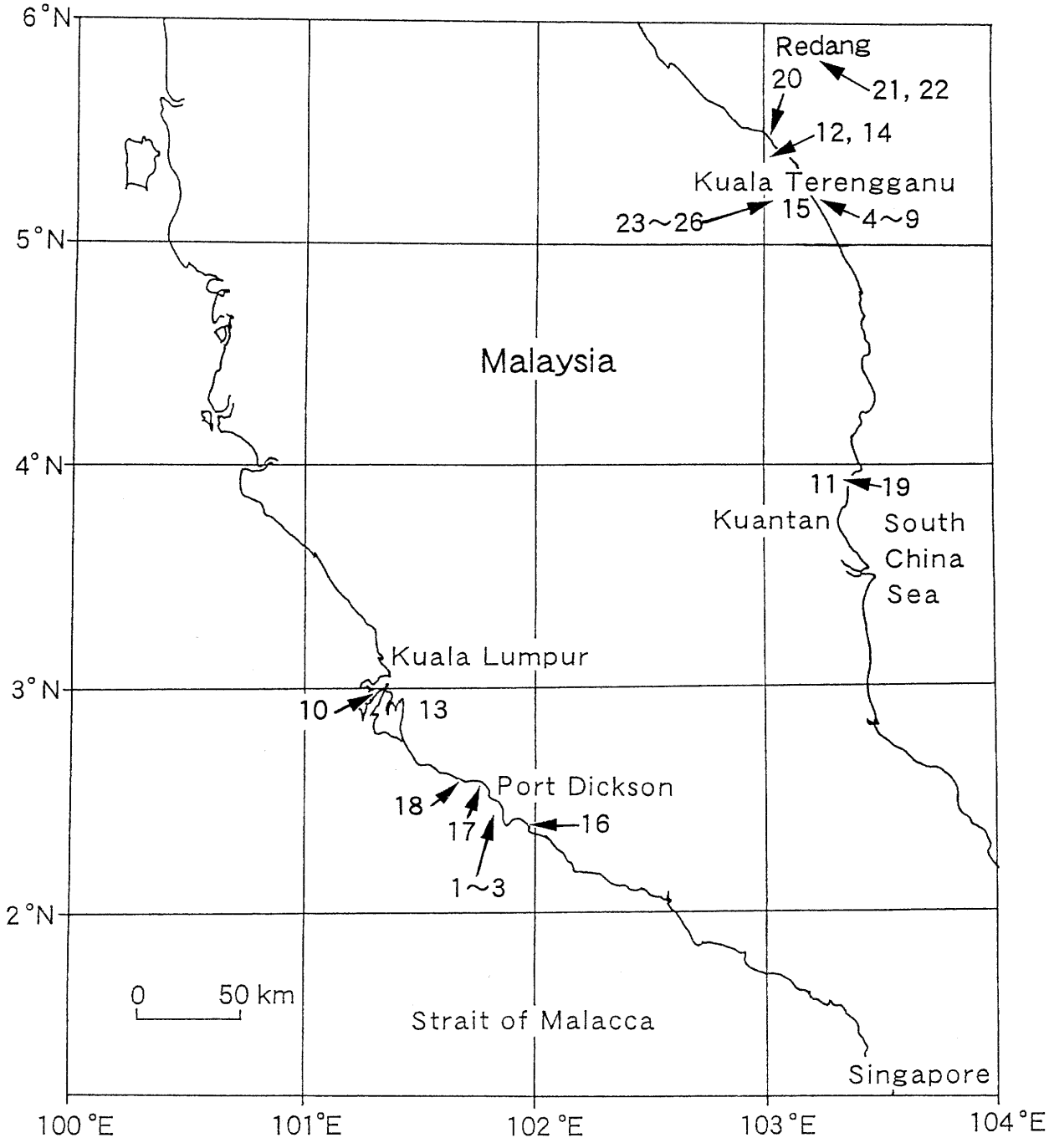

Fig. 1 Map of sampling stations for environmental samples of Malaysia. (1~10: Sea water and river water, 1~8, 10~26: Soil, sea sand and Sediment)

Surface sea water was sampled using a plastic container. Deep sea water samples were obtained by the use of the Bandon Sampler (made from polypropylene) hanging on a rope from shipboard.

As for the preservation of samples, hard glass bottles are desirable, but there is concern of breakage during transport. So we used bottles with a narrow opening made from polypropylene which let exude with nitric acid of about 2 mol l^{-1} beforehand for two weeks or more and are then washed thoroughly with water.

Sea water samples used for mercury determination were kept in hard glass bottles equipped with Teflon packing, which were exuded before hand. 10 ml of (1:1) sulfuric acid was added to 1 liter of sample after returning home. Samples were then brought to the laboratory and analyzed.

Floating particles (suspended matter) in water samples containing heavy metal as mercury separated before sulfuric acid addition. The adsorption and elution of mercury in filter medium was carefully taken cared. In particular mercury, since the mercury concentration is high in the air of laboratory[2], samples were filtered in the clean bench which tried to remove as much mercury as possible through the inside of activated charcoal.

Apparatus and reagent

Mercury RA-1 reduction aeration mercury analyzer and Mercury SP-3 heating vaporization mercury analyzer produced by Nippon Instruments Co. were used. All measurements were made in the peak-height mode.

Pure water was prepared by purifying distilled water with a Millipore Mili-Q SP system.

All reagents used were of analytical reagent grade for the measurement of toxic metals.

Mercury (Ⅱ) chloride standard (Hg 1,000 mg l^{-1} in 0.1 mol l^{-1} hydrochloric acid) was obtained from the Nakarai Co., Japan. A working solution (Hg 0.1 mg l^{-1}) was prepared by diluting this solution with 0.05 mol l^{-1} of sulfuric acid that contained 3% sodium chloride solution.

Determination of mercury in liquid samples (sea water, river water etc.)

Mercury concentration was determined by the established analytical method[11), 12)] as shown below.

Procedure (a): A 200 ml water sample was acidified with 2 ml of sulfuric acid (1:1) and was reduced with 5 ml of 10% tin (Ⅱ) chloride, then nitrogen gas was blown into the sample solution to evolve mercury vapor. The evolved mercury vapor was trapped on a porous gold collector and determined by cold vapor atomic absorption spectrometry. Procedure (b): 12 ml of 10 mol l^{-1} sodium hydroxide, 2 ml of copper (Ⅱ) sulfate solution (which contained Cu of 1,000 mg l^{-1}) and 5 ml of 10% tin (Ⅱ) chloride were added to a 200 ml water sample, and the sample was treated the same as Procedure (a). Inorganic mercury is determined by Procedure (a), and total of inorganic and organic mercury is determined by Procedure (b).

Determination of mercury in solid samples (rock, soil, sediment and fish etc.)

Wet samples of sediment were air dried and the samples were grind with mortar made of stainless steel or agate mortar and perform uniformly.

A solid sample (0.02 to 0.20 g) was weighed out into a porcelain boat previously ignited about 800℃ to exclude any trace of mercury and covered with an addition agent (sodium carbonate ignited about 700℃ and calcium hydroxide ignited about 800℃ and afterward mixed in the volume ratio of 1:1). Thus analytical sample prepared was heated in the furnace of a mercury analyzer at about 300℃ for three minutes, and then at about 700℃ for four minutes to complete the vaporization. At this time, the evolved mercury pass through the air (mercury removed by activated charcoal) at flow rate 0.5 l min^{-1}. Evolved gas passed through the oxidation decomposition furnace at about 900℃ along with copper (Ⅱ) oxide wire. Furthermore, it was washed with phosphate buffer solution of pH 6.86. Mercury vapor collects on porous gold collector as gold amalgam. This collector heats it at about 700℃ with a heater. Evolved mercury vapor was determined by cold vapor atomic absorption spectrometry[13)-15)]. Determination of mercury was performed automatically and total mercury was determined.

Results and discussion

Mercury concentration in sea water of Malaysia coastal area

We collected the samples from Malaysian Port Dickson and Kuala Terengganu coastal sea water and mixed water (river and sea water) in 1995 (from Sept. 17 to Sept. 30) and 1996 (from Jul.21 to Aug. 4).

Mercury in sea water and river water samples was subjected to reduction-vaporization by use of tin (Ⅱ) chloride. The mercury vapor was concentrated with a porous gold collector using Procedure (a) and (b).

The results are shown in Table 1. Samples showing considerably high mercury concentration exist in a part of the samples from 1995, but it is not found in samples from 1996 at all. As for these abnormal values, the possibility that sediment mixed in collection of sample is considered. There was a extremely little mercury concentration of other samples.

Table 1 Analytical results of mercury in sea water and river water samples of Malaysia

Station No.	Date	Depth (m)	Temp. ℃	pH	Cl^- mg l^{-1}	Inorg-Hg ng l^{-1}	Total-Hg ng l^{-1}
1	Sept. 22.'95	0	29.8	7.93	15,240	1.4	1.6
1	Jul. 25.'96	0	29.9	7.83	16,600	3.2	3.2
1	Sept. 22.'95	10	29.8	7.97	15,200	27.0	28.0
1	Sept. 22.'95	20	29.9	7.92	15,310	20.8	22.5
1	Jul. 25.'96	25	30.0	7.97	16,700	3.3	3.4
1	Sept. 22.'95	0	29.8	7.89	15,340	16.7	24.2
2	Sept. 22.'95	0	29.8	7.94	15,850	6.3	6.3
2	Jul. 25.'96	0	29.9	8.03	16,700	2.9	3.0
2	Sept. 22.'95	2	29.8	7.97	15,450	1.6	1.9
2	Sept. 22.'95	4	29.5	7.86	15,160	3.7	4.2
2	Jul. 25.'96	6	30.0	8.01	16,600	3.2	3.2
3	Sept. 22.'95	0	29.4	7.95	15,490	11.3	13.7
3	Jul. 25.'96	0	30.0	7.98	16,500	2.8	2.8
3	Sept. 22.'95	2	29.8	7.90	15,380	9.1	8.9
3	Jul. 25.'96	4	30.0	7.99	16,200	2.9	2.9
4	Sept. 26.'95	0	28.3	7.07	84.0	0.5	3.1
4	Jul. 31.'96	0	30.2	8.06	17,700	3.2	3.2
4	Sept. 26.'95	4.1	28.3	6.65	90.0	0.8	2.0
4	Jul. 31.'96	5.3	30.2	8.07	16,500	3.1	3.1
5	Sept. 26.'95	0	29.1	7.41	10,720	2.0	2.0
5	Jul. 31.'96	0	30.2	8.18	17,600	2.6	2.7
5	Sept. 26.'95	5.1	29.1	7.89	17,110	5.2	5.0
5	Jul. 31.'96	12.8	29.4	8.05	18,200	2.8	2.8
6	Sept. 26.'95	0	28.8	7.43	7,528	2.0	2.0
6	Jul. 31.'96	0	30.2	8.12	17,800	3.0	3.0
6	Sept. 26.'95	4.5	28.8	7.56	12,070	2.8	2.9
6	Jul. 31.'96	10	29.6	8.04	17,900	3.0	3.1
7	Sept. 26.'95	0	28.9	7.68	17,220	41.0	52.0
7	Jul. 31.'96	0	29.9	8.20	16,800	2.6	2.7
7	Sept. 26.'95	6.3	29.2	8.06	17,400	1.9	2.0
7	Jul. 31.'96	27.9	25.7	7.96	18,200	2.8	2.8
8	Sept. 26.'95	0	28.9	8.05	16,280	1.6	1.7
8	Jul. 31.'96	0	30.2	7.96	12,700	3.4	3.4
8	Sept. 26.'95	5.5	28.9	8.06	16,710	2.0	2.1
8	Jul. 31.'96	4.8	30.2	8.10	16,700	2.8	2.9
9	Jul. 31.'96	0	29.9	7.12	10,400	3.3	3.3
10	Jul. 23.'96	0	29.4	8.76	15,690	4.7	5.4

Inorganic mercury is found to be the major chemical form of mercury in this coastal area, and rate of organic mercury was extremly less than inorganic mercury and inorganic plus organic mercury.

Table 2 shows comparative data of mercury concentration of Malaysian Port Dickson and Kuala Terengganu coastal sea water and mixing water (river and sea water) and the open sea (1980～1983).

Table 2 Average values of mercury concentration in sea water and river water samples of Malaysia and the open sea (East China Sea)

Locality		Mercury concentration (ng l^{-1})			
		Inorg-Hg		Total-Hg	
	Range	1.4～41.0 (0.8～41.0)		1.6～52.0 (1.6～52.0)	
Strait of Malacca	XA	6.0_3	(5.7_6)	7.3_3	(6.5_7)
South China Sea	XG	3.4_8	(3.5_2)	4.0_9	(3.9_8)
(River water)	n	19	(37)	19	(37)
	Range	1.0～2.6 (1.0～2.9)		1.4～2.9 (1.4～2.9)	
Open Sea	XA	2.0_4	(1.9_4)	2.4_3	(2.3_4)
(East China Sea)	XG	1.9_8	(1.8_9)	2.3_8	(2.2_8)
	n	15	(27)	15	(27)

XA: Arithmetic mean, XG: Geometric mean, n: No. of samples.
( ): Inclide each class of depth, Without ( ): Surface water.

Levels of mercury determined by Procedure (a) and (b) were found to be in the range of 0.8～41.0 ng l^{-1} (geometric mean 3.5_2 ng l^{-1}), 1.6～52.0 ng l^{-1} (geometric mean 3.9_8 ng l^{-1}) for 37 samples taken from Malaysia and 1.0～2.9 ng l^{-1} (geometric mean 1.8_9 ng l^{-1}), 1.4～2.9 ng l^{-1} (geometric mean 2.2_8 ng l^{-1}) for 27 samples taken from the East China Sea, respectively. Mean levels of mercury [Procedure (a), (b)] for Malaysia were 1.9, 1.7 times higher than those for the East China Sea. This might be contributed by human activities (industry, vehicle to the natural environment).

Total mercury concentration in sediment etc. of Malaysia

We collected soil, rock, sea sand, river and sea sediment in the same location where samples of sea water of Malaysia were collected and the peripheral area as well.

Total mercury was determined by heating-vaporization and gold amalgam-cold vapor atomic absorption spectrometry.

The results are shown in Table 3. Table 4, total mercury concentration divided into soil, sea sand, sediment (river and sea sediment) samples in Malaysia.

As for Table 4, total mercury concentration in soil (containing fine particle) sample is high.

Arithmetic mean value and geometric mean value of total mercury concentration of sea sand (containing comparatively bigger particle) were 10.1, 5.4 μg l^{-1}, respectively. Arithmetic mean value and geometric mean value of total mercury concentration in river and sea sediment were 30.4, 20.1 μg l^{-1}, respectively. These sea sediment values were not so high compared with total mercury concentration in general sediments, but mercury concentration in river sediment of Port Kelang compared with other areas as well as its mercury concentration was high. The reason is because an industrial area exists in the upper stream.

Mercury concentration in Malaysian fish

We examined mercury concentration in possible appetite part of fish (fish meat) obtained in Port Dickson and Kuala Terengganu. Mercury in fish meat was divided into inorganic mercury and organic mercury and determined by cold vapor atomic absorption spectrometry[16].

The results are shown in Table 5. It seems to be clear that organic mercury holds the most as a chemical form in fish meat from Table 5. There was an extremely small amount of inorganic mercury compared with the total mercury.

Table 3 Analytical results of total mercury in solid samples of Malaysia

Station No.	Station	Sampling Date	Hg μg kg^{-1}*	Remarks
1	Port Dickson Stn.A	Sept. 22.'95	59.1	Marine sediment (0-5cm)
1	Port Dickson Stn.A	Jul. 25.'96	56.7	Marine sediment (0-5cm)
2	Port Dickson Stn.B	Sept. 22.'95	26.7	Marine sediment (0-5cm)
2	Port Dickson Stn.B	Jul. 25.'96	41.4	Marine sediment (0-5cm)
3	Port Dickson Stn.C	Sept. 22.'95	22.3	Marine sediment (0-5cm)
3	Port Dickson Stn.C	Jul. 25.'96	35.2	Marine sediment (0-5cm)
4	Terengganu No.1	Sept. 26.'95	17.3	Marine sediment (0-5cm)
4	Terengganu No.1	Jul. 25.'96	6.4	Marine sediment (0-5cm)
5	Terengganu No.2	Sept. 26.'95	24.8	Marine sediment (0-5cm)
5	Terengganu No.2	Jul. 31.'96	23.0	Marine sediment (0-5cm)
6	Terengganu No.3	Sept. 26.'95	6.0	Marine sediment (0-5cm)
6	Terengganu No.3	Jul. 31.'96	27.0	Marine sediment (0-5cm)
7	Terengganu No.4	Sept. 26.'95	15.6	Marine sediment (0-5cm)
7	Terengganu No.4	Jul. 31.'96	4.2	Marine sediment (0-5cm)
8	Terengganu No.5	Sept. 26.'95	27.2	Marine sediment (0-5cm)
8	Terengganu No.5	Jul. 31.'96	6.1	Marine sediment (0-5cm)
10	Port Kelang	Jul. 23.'96	163	River sediment (0-5cm)
11	Dungun Paka	Jul. 30.'96	30.7	Soil (0-5cm)
12	Terengganu Airport	Aug. 02.'96	137	Soil (0-5cm)
13	Serdang UPM	Aug. 06.'96	113	Soil (0-5cm)
14	Terengganu Airport	Aug. 02.'96	2.2	Sand(0-5cm)
14	Terengganu Airport	Aug. 02.'96	5.0	Sand (0-5cm)
15	Chenering	Jul. 30.'96	3.7	Rock (0-5cm)
16	Blue Lagon	Sept. 21.'95	8.8	Sea sand (0-5cm)
16	Blue Lagon	Jul. 25.'96	8.2	Sea sand (0-5cm)
16	Blue Lagon	Sept. 21.'95	9.8	Sea sand (0-5cm)
16	Blue Lagon	Jul. 25.'96	5.3	Sea sand (0-5cm)
17	Port Dickson Batu 4	Sept. 21.'95	8.9	Sea sand (0-5cm)
17	Port Dickson Batu 4	Jul. 25.'96	1.2	Sea sand (0-5cm)
17	Port Dickson Matu 4	Sept. 21.'95	10.2	Sea sand (0-5cm)
17	Port Dickson Batu 4	Jul. 25.'96	5.1	Sea sand (0-5cm)
18	Port Dickson Esso Jetty	Sept. 21.'95	16.9	Sea sand (0-5cm)
18	Port Dickson Esso Jetty	Jul. 25.'96	12.0	Sea sand (0-5cm)
18	Port Dickson Esso Jetty	Sept. 21.'95	22.9	Sea sand (0-5cm)
18	Port Dickson Esso Jetty	Jul. 25.'96	66.1	Sea sand (0-5cm)
19	Kampong Rantau Abang	Jul. 30.'96	1.1	Sea sand (0-5cm)
20	Trengganu	Aug. 01.'96	1.4	Sea sand (0-5cm)
21	Redang TSL	Aug. 02.'96	1.3	Sea sand (0-5cm)
22	Redang IS	Aug. 02.'96	1.7	Sea sand (0-5cm)
23	Trengganu No.1	Sept. 25.'96	9.0	River sediment (0-5cm)
24	Trengganu No.2	Sept. 25.'96	8.4	River sediment (0-5cm)
25	Trengganu No.3	Sept. 25.'96	46.8	River sediment (0-5cm)
26	Trengganu No.4	Jul. 31.'96	12.0	River sediment (0-5cm)

*: 110℃, 6 hrs dry basis

Table 4 Average values of mercury concentration in solid samples of Malaysia

Sample	Mercury concentration (μg kg^{-1})	
Soil	Range	30.7～137
	X_A	93.6
	X_G	78.0
	n	3
Sand (Rock)	Range	1.1～66.1
	X_A	10.1
	X_G	5.4_2
	n	19
Sediment	Range	4.2～163
	X_A	30.4
	X_G	20.1
	n	21

XA: Arithmetic mean, XG: Geometric mean, n: No. of samples.

Table 5 Mercury concentration in fish meat from Port Dickson and Kuala Terengganu of Malaysia

No.	Name	Inorg-Hg* μg kg^{-1}	Org-Hg* μg kg^{-1}	Residue* μg kg^{-1}	Sum* μg kg^{-1}	Total-Hg* μg kg^{-1}	Total-Hg** μg kg^{-1}	Remarks
1	Scolopsis monograma	<0.1	147	6.7_4	$153._7$	168	719	Port Dickson
2	Lutjanus johnii	<0.1	42.5	0.4_3	42.9	53.6	239	Port Dickson
3	Mempinang	<0.1	34.1	0.1_8	34.3	43.9	183	Port Dickson
4	Siganus canaliculatus	<0.1	4.0_9	<0.1	4.0_9	4.0_4	18.5	Port Dickson
5	Scatohagus argus	<0.1	34.6	0.6_0	35.2	41.4	193	Port Dickson
6	Pfafax	1.8_7	15.4	<0.1	17.3	17.1	134	Port Dickson
7	Siganus jayus	<0.1	10.0	<0.1	10.0	9.4_5	51.4	Port Dickson
8	Crab	0.7_8	146	4.5_0	$151._3$	163	909	Port Dickson
9	Gnathonodom specious	0.6	46.9	10.0	57.5	56.9	216	Terengganu
10	Chiloscyllium sp.	1.7	154	21.5	$177._2$	229	810	Terengganu
11	Sardinella fimbriata	2.0	34.8	3.2	40.0	42.2	157	Terengganu
12	Rastrelliger kanagurta	<0.1	14.1	1.1	15.2	18.7	76.7	Terengganu
13	Sciaenidae	0.8	26.5	3.8	31.1	43.1	189	Terengganu
14	Lutjanus malabaricus	0.5	16.9	2.7	20.1	21.6	94.9	Terengganu
15	Nemipterus furcosus	2.7	157	19.2	$178._9$	191	778	Terengganu
16	Laligo sp.	<0.1	21.4	0.9	22.3	19.6	94.6	Terengganu
17	Euthynnus affinis	2.0	17.2	2.9	22.1	24.4	73.3	Terengganu
18	Penaeidae	0.4	9.5	1.5	11.4	10.6	49.4	Terengganu
19	Portunus pelagicus	<0.1	8.7	1.0	9.7	13.2	69.1	Terengganu

*: Wet basis, **: 110℃, 6 hrs dry basis.

Mercury concentration was high in carnivorous fish such as sharks and crabs compared with other fish. It is thought that mercury concentration in fish meat reflects the food-chain of the fish, and we examined the relation to the weight and length of fish[17] of Japan. The difference of growth rate and fish diet influences the mercury concentration of fish.

Conclusions

The authors collected environmental samples such as Malaysian coastal sea water, sediment, soil, and fish

during the period 1995～1996. Mercury was determined in the samples and concentration, chemical form and its distributions were investigated. The results are summarized as follows;

1) Major chemical form of mercury in coastal sea water was inorganic mercury, and there was an extremely small amount of organic mercury.
2) Geometric mean of inorganic and inorganic plus organic mercury for coastal Malaysian sea water were 3.5_2, 3.9_8 n g l^{-1} and were 1.9, 1.7 times higher than those for the East China Sea, respectively.
3) Arithmetic mean value and geometric mean value of total mercury concentration of sea sand (comparatively largely particle) were 10.1, 5.4 μg kg^{-1}, respectively.
4) Arithmetic mean value and geometric mean value of total mercury concentration in river and sea sediment were 30.4, 20.1 μg kg^{-1}, respectively, but mercury concentration in Port Kelang which refrained industrial area was about one order higher in comparison with the ordinary river sediment.
5) Mercury chemical form in fish meat was mainly organic mercury. There was an extremely small amount of inorganic mercury.
6) Mercury concentration was high in carnivorous fish such as sharks and crabs compared with other fish species.
7) Levels of mercury in river water, sea water and sea sand (rock) were relatively low, but mercury concentration of soil, sediment and biological materials (fish) were higher in comparison with the other environmental samples.

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