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Characteristics of the bioaccumulation process of mercury in the benthic ecosystem in the enclosed coastal seas

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The authors review the previous studies on the bioaccumulation process of mercury in the marine ecosystem, focusing on the benthic one in enclosed coastal seas. According to the conventional ideas, the mercury content of animals tends to increase discontinuously as the elevation of the trophic level in the biological community. This typical pattern of the bioaccumulation of mercury has been described in the pelagic ecosystem. The mercury contaminated in the water is absorbed by the phytoplankton, and is further biologically magnified in the aquatic animals located at higher trophic levels such as planktivorous fish, piscivorous fish, and sea birds and marine mammals. In the benthic ecosystem in enclosed coastal seas, the mercury transference pathway tends to be more complex due to a variety of diets available for the benthic invertebrates, and the presence of a kind of bioconcentration process of mercury into the sediment. A large amount of organic particles, which are referred to as POM such as dead bodies of phytoplankton and zooplankton and feces excreted from the zooplankton, are always settling on the sea floor as detritus before they have been degraded fully by microorganisms in the water, due to shallow water. The detritus tend to be degraded by microorganisms, but its non-degradable parts, including mercury contaminants, tend to accumulate in the sediment. Some benthic invertebrates burrow into the sediment, and take in the mercury condensed in the sediment through their deposit-feeding activities or suspension-feeding activities to the re-suspended sediment just above the sea floor. The mercury accumulated in the benthic invertebrates is apt to be transferred to benthic and benthopelagic fishes that feed on the infaunal benthic invertebrates preferentially. Here, an accelerated bioaccumulation pathway of mercury exists in the benthic ecosystem, and brings further biologically magnified mercury to the pelagic ecosystem in the enclosed coastal seas.

Keywords: benthic ecosystem, bioaccumulation, bioconcentration, enclosed coastal seas, epifauna, infauna, mercury

1. Introduction

In Japan, people have established residential areas along the coasts of the enclosed coastal seas such as Tokyo Bay, Osaka Bay, Seto Inland Sea, Ariake Bay, etc. since antient times, partly because they have depended on sea foods collected from the coasts of the bays as one of the most important protein-based food resources. The sea foods still keep the important position in Japanese meals in the present days. In 2011, 2,397,000 tons of various marine organisms were caught in the natural fisheries grounds or harvested in aquaculture farms in the coastal seas. It accounts for about 45 % of the total year national fishery catch and production[1)], and indicates that about 19 kg of the sea foods collected from the coastal seas are utilized for diets per person per year in Japan.

If the enclosed coastal seas were once polluted heavily by the discharge of harmful heavy metals and/or chemicals, it would cause a serious negative impact on the water environment and aquatic ecosystem of the seas due to limited exchange of the water with the open sea, and environmental disturbance would last for a long time[2)-7)]. Furthermore, among the countries as Japan, which highly depends on protein-based food resources in seafood, the residential people along the coastal seas are subject to food poisoning through the intake of polluted ones, since the concentrations of the harmful substances tend to increase markedly in the body tissues of higher animals such as omnivorous or carnivorous fishes due to the effects of bioaccumulation in the food web of the aquatic ecosystem[8)-10)].

In Japanese history, we experienced serious food poisoning incidents caused by mercury twice around Minamata Bay facing Yatsushiro Sea, located in the middle part of Kyushu, in the 1950s to 1960s[11) 12)], and in the estuary and lower reach of Agano River facing the Sea of Japan, located in the middle of Honshu, in the 1960s[13)]. The effluent containing methylmercury was commonly discharged from the chemical plants to the rivers connected to the seas. The organic mercury accumulated in the body tissues of the fish occurring in the rivers and coastal seas through the bioaccumulation process in the aquatic food web system[14)-16)]. Not only animals such as sea birds and street cats that fed on the polluted fish, but also people living along the rivers and/or the coastal areas suffered seriously from the food poisoning caused by the intake of methylmercury[17)],

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which are referred to as “Minamata disease” and “Niigata Minamata disease”, respectively[14].

Methylmercury tends to accumulate in various organs of human including the brain, liver, kidney etc., and caused sensory disturbances, ataxia, dysarthria, constriction of the visual field, auditory disturbances, and tremors[18]. It was also transferred from the mothers to their infants through the placenta or mother’s milk, and brought them similar symptoms commonly such as intelligence disturbance, primitive reflex, cerebellar symptoms, disturbance of body growth and nutrition, dysarthria, deformity of limbs, etc.[19) 20] The wildlife such as fish, bird, and mammals, which are exposed to elevated levels of methylmercury through diets, often suffered from behavioral, neurochemical, hormonal, and reproductive disturbances[21)-24].

Thus, mercury is one of the most hazardous heavy metals for higher animals including human as mentioned above. However, 2,200 to 4,000 tons of mercury is still being emitted into the atmosphere throughout the world per a year, due to the anthropogenic causes (mainly mining, extraction and burning of fossil fuels, and intentional use of mercury in industrial processes, products, and gold mining operations, etc.)[24)-27]. It is also being added to the atmosphere naturally due to volcanic eruptions and geothermal activities, which are estimated to be 114 to 700 tons year^{-1} [27)-29], except the natural cycling of mercury in the global environment. In the Japanese Archipelago, most of which has been shaped by the volcanic activities, the background levels of mercury in the environment tend to be fairly elevated to those of the stable continental areas, in particular in the nearer places to the volcanos[30)-32].

The authors have studied the bioaccumulation process of total mercury (THg), which includes elemental mercury (Hg^0), divalent mercury ions (Hg^{2+}) and methylmercury, in the aquatic ecosystem of the bay (a part of enclosed coastal seas), where the background levels of THg tend to be elevated due to the emission of mercury from an active volcano beside the bay[33) 34]. Most of the previous studies on the bioaccumulation process of mercury in the aquatic ecosystem in the enclosed coastal seas have described the simple elevation of mercury contents in the species located at the higher trophic levels in the biological communities, mainly focusing on the pelagic organisms. However, our studies revealed the presence of much more complex process of bioaccumulation of mercury on the benthic ecosystem[33) 34]. Here, we review the recent achievements on the research of bioaccumulation process of THg of the aquatic organisms occurring in the coastal seas, and try to clarify further the characteristics of its bioaccumulation process in the benthic ecosystem and its influence to the pelagic ecosystem in the enclosed coastal seas.

2. Estimation of the trophic positions of aquatic organisms with stable isotope ratios of carbon and nitrogen and bioaccumulation of THg in the food web in the aquatic ecosystem

In the previous studies on the bioaccumulation of harmful substances in the aquatic ecosystem, two different processes that increase the concentrations of the substances of the organisms have been recognized. One is referred to as “bioconcentration “ that the primary producers such as phytoplankton increase the concentrations of the substances in their cells through absorbing them from the water. Another one is referred to as “biomagnification” meaning that the consumers increase the contents of the substances in their body tissues though the predation of the organisms located at the lower trophic positions in the food web of the biological communities[8) 35].

To follow up the process of the bioaccumulation of the harmful substances, at first the trophic position of each species in the biological community must be classified properly. In the conventional methods, we have depended on the observations of the morphological structure of the feeding apparatus, feeding behaviors, and stomach contents to determine its trophic position deterministically. In the recent studies, it is estimated more probabilistically, using the technique of stable isotope analysis of carbon and nitrogen of the body tissues[36]. The stable isotope ratios of the body tissues of the organism are represented as following equations.

$$\delta^{13}\mathrm{C} = \left(\frac{{}^{13}\mathrm{C}/{}^{12}\mathrm{C}_{\mathrm{sample}}}{{}^{13}\mathrm{C}/{}^{12}\mathrm{C}_{\mathrm{standard}}} - 1\right) \times 1000\ (‰),$$

$$\delta^{15}\mathrm{N} = \left(\frac{{}^{15}\mathrm{N}/{}^{14}\mathrm{N}_{\mathrm{sample}}}{{}^{15}\mathrm{N}/{}^{14}\mathrm{N}_{\mathrm{standard}}} - 1\right) \times 1000\ (‰)$$

Pee Dee Belemnite (PDB) and atmospheric nitrogen are generally used as references for ^{13}C and ^{15}N, respectively

In general, the primary producers that are exploited as food resources for the animals as the consumers can be classified roughly into algae, benthic microphytes, phytoplankton, and terrestrial plants by the values of δ^{13}C of their tissues (Table 1), due to the different capacity for discrimination of $^{13}CO_2$ from $^{12}CO_2$ in the process of fixation of CO_2 from the environment[38]. Since the molecules of heavier stable isotopes of carbon and nitrogen are apt to be retained longer in the body tissues of the animals, they are more enriched in the animals located at higher trophic levels in the community. According to the empirical data of the previous studies on the stable isotope ratios of carbon and nitrogen between the animals linked with the prey-predator relationship, the values of δ^{13}C and δ^{15}N increase +1.0 ‰ and +3.4 ‰, respectively, in the body tissues of the predator side located at one step higher trophic position in the food web system[36] (Fig. 1).

If the values of stable isotope ratios of carbon and nitrogen of the primary producers are fixed, we are able to identify the primary consumers that exploit the primary producers as diets and the consumers located at the further higher trophic positions on the same food chain, being based on the increase trend of the values of δ^{13}C and δ^{15}N between the prey and predator. We are able to determine the trophic position (TP) of each of their species , using the following equation.

Table 1 The mean values and ranges of the stable isotope ratios of carbon and nitrogen of the primary producers expressed by $\delta^{13}C$ and $\delta^{15}N$. The ratios of isotopic shift of $\delta^{13}C$ and $\delta^{15}N$ are based on Yokoyama (2008)[37].

Organisms	$\delta^{13}C$ (‰)		$\delta^{15}N$ (‰)	
	Mean	Range	Mean	Range
Vascular plant --- C_3 terrestrial	-28.2	-30.0 to -26.7	0.8	-0.6 to -3.1
Vascular plant --- C_3 salt marsh	-26.4	-28.0 to -24.9		
Vascular plant --- C_4 salt marsh	-13.8	-16.7 to -12.0		
Vascular plant --- C_4 sea grass	-11.4	-14.9 to -7.2		
Phytoplankton	-21.0	-24.4 to -17.8	6.6	1.5 to 10.8
Benthic microphytes	-16.4	-22.0 to -12.8	6.8	-0.7 to 17.7
Algae	-18.5	-29.7 to -9.4	7.8	1.8 to 12.0

Table 2 The relationship between the nitrogen stable isotope ratio and content of total mercury (THg) in the aquatic ecosystem. *BMF (Biomagnification factor) values: It was calculated as the increase ratios of the mean THg content of the secondary consumers to that of the primary consumers.

Location	Sample type	Feeding habitat	Linear	r^2		TMF	References
Masan Bay, Korea	Invertebrates, Fish	Benthic	Log [Hg] = 0.119 ($\delta^{15}N$) + 0.041	0.37	dw	2.54	41
Gulf of St. Lawrence, Canada	Invertebrates, Fish, Birds	Pelagic	Log [Hg] = 0.218 ($\delta^{15}N$) - 1.20	0.65	dw	5.51	40
		Benthopelagic	Log [Hg] = 0.194 ($\delta^{15}N$) - 0.40	0.59	dw	4.57	40
		Benthic	Log [Hg] = 0.111 ($\delta^{15}N$) + 0.28	0.35	dw	2.38	40
Lancaster Sound, Canada	Invertebrates, Fish, Birds, mammals	mixed	Log [Hg] = 0.20 ($\delta^{15}N$) - 3.33		dw	4.79	43
Coast of northern Rio de Janeiro State, Brazil	Plankoton, Invertebrates, fish	mixed	Log [Hg] = 0.246 ($\delta^{15}N$) - 0.708	0.801	dw	6.86	45
Isahaya Bay, Kyushu, Japan	Inveretebrates, High THg content group				dw	2.13*	33
Isahaya Bay, Kyushu, Japan	Inveretebrates, Low THg content group				dw	2.50*	33

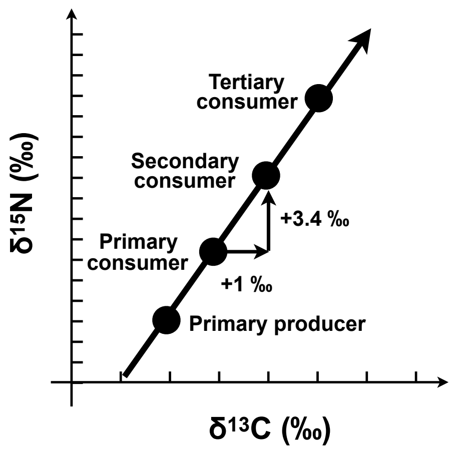

Fig. 1 Empirical trend on the increase of the values of the stable isotope ratios of carbon and nitrogen expressed by $\delta^{13}C$ and $\delta^{15}N$ of the body tissues among the organisms linked with a food chain in the community. This figure was illustrated being based on the statement of Wada et al. (1991)[36].

$$TP_i = 1 + (\delta^{15}N_i - \delta^{15}N_{p.p.}) / 3.4\ (‰)$$

TP_i : trophic position of species i, $\delta^{15}N_i$: $\delta^{15}N$ value of species i, $\delta^{15}N_{p.p.}$: $\delta^{15}N$ value of the primary producer, 1: the value of trophic position of the primary producer, 3.4 (‰): isotopic shift of $\delta^{15}N$ value between the prey and predator.

However, in many studies, the values of $\delta^{15}N$ of the animals which seem to belong to the same trophic position in the community being judged from their feeding behaviors are not necessarily concentrated on a narrow range[9) 34) 39)-42)], although the values of their stable isotope ratios of carbon and nitrogen indicate that they are distributed near the line of stable isotopic shift of a single food chain. These facts suggest that the consumers tend to prey on a variety of diets on the same food chain.

A combined analysis with the nitrogen stable isotope ratios and the contents of the harmful substance of the body tissues of the organisms has been recently adopted to describe the bioaccumulation process of the harmful substance in the biological communities[9) 39)-41) 43)-47)]. The relationship between the nitrogen stable isotope ratio and the content of THg of the organism is expressed as the following equation:

$$Log_{10}(THg) = a * (\delta^{15}N) + b,\ \ TMF = 10^{a*3.4}$$

TMF: Trophic Magnification Factor, 3.4 (‰): isotopic shift of $\delta^{15}N$ value between the prey and predator.

According to the previous studies on the bioaccumulation process of THg in the marine ecosystem, TMF values of THg range between $10^{0.38}$～$10^{0.84}$ (2.38～6.86) (Table 2).

3. Bioaccumulation of THg along the prey-predator relationship in the food web in the pelagic and benthic ecosystem

Figure 2 shows the outline of bioaccumulation process

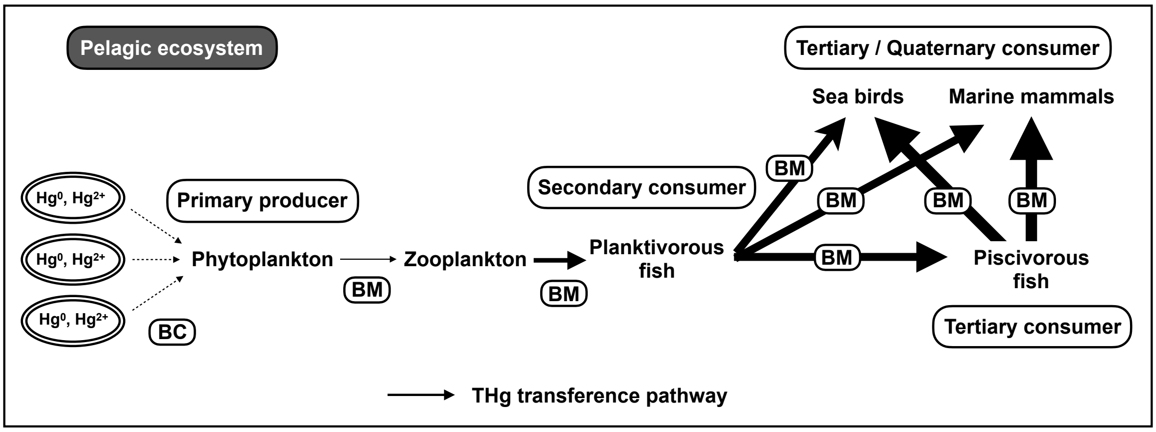

Fig. 2 The bioaccumulation pathways of total mercury (THg) along the food web in the pelagic ecosystem in the enclosed coastal seas. BC: bioconcentration, BM: biomagnification

of THg in the pelagic ecosystem. The mercury discharged to the seas is absorbed by the phytoplankton, being condensed in its cells, and transferred to the animals located at three or four different higher trophic levels in the food web in the surface layer of the water, which include zooplankton, planktivorous fish, piscivorous fish, sea birds, and marine mammals[40) 42) 43) 46)]. Although the TMF values of THg have been noted in the range between 2.38 and 6.86 in the previous studies in the aquatic ecosystem (Table 2), the THg contents of the bodies of the aquatic organisms enable to be magnified to max. about 2,210 times higher levels at the top predators (the quaternary consumer) of the food web in the communities to those of the primary producers.

In the benthic ecosystem in the enclosed coastal seas, much more complex biomagnification processes exist due to a variety of food resources available for the benthic invertebrates as the primary consumers (Fig. 3). Not only phytoplankton but also microphytobenthos such as benthic diatoms are some of the main members of the primary producers exploitable for them[47)-49)], since the solar radiation with enough light intensity for photosynthesis can reach the sea floor in the shallow coastal waters, in particular on the intertidal zones of the coasts. In the water, a large amount of organic particles tend to be suspended, and are always settling on the sea floor before they have been fully degraded by the micro-organisms in the water column[50) 51)], due to the shallow water as well. The organic particles suspended in the water and ones deposited on the sediment of the sea floor are referred to as “POM (Particulate of Organic Matter)” and “detritus”, respectively. They are derived from dead bodies of phytoplankton and zooplankton, feces excreted from the zooplankton, fragments of macro-algae and terrestrial plants, etc., and various micro-organisms attached on their surfaces for degradation[52)]. Among the benthic invertebrates as the primary consumers, suspension-feeding ones exploit the phytoplankton, microphytobenthos, and POM suspended in the overlying water on the sea floor, while deposit-feeding ones feed on the sediment with the detritus.

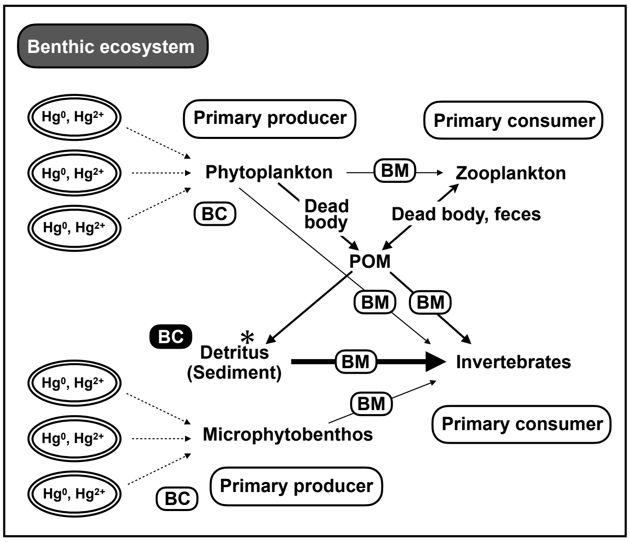

Fig. 3 The bioaccumulation pathways of total mercury (THg) in the primary producers and primary consumers along the food web in the benthic ecosystem in the enclosed coastal seas. BC: bioconcentration, BM: biomagnification

* Undefined pathway of the biococentration of total mercury (THg) on the sediment.

Here, we pay attention to the concentration process of THg on the sediment of the sea floor, since mercury is readily absorbed to the particulate matters in the water and tends to accumulate in the sediment of the sea floor[53)]. The detritus contained in the sediment, which is derived from the POM settled from the water column, is degraded by the respiratory activities of micro-organisms on the sediment. Its bio-degradable components are mineralized to inorganic substances, and recycled into the overlying water just above the sediment, while the non-bio-degradable contaminants such as mercury tend to retain on the sediment. Jaingam et al. (2018)[33)] reported that the sediment on the sea floor contained 4.9 time denser THg (133 ± 23 ng g^{-1} d.w. , n = 19) than that of the POM suspended in the overlying water on the sediment (27.2 ± 9.7 ng g^{-1} d.w., n = 9) at the areas with the depths of less than 10 m in Isahaya Bay, Japan, under the influence of mercury emission from a neighboring active volcano (Mt. Unzen). The concentration rate of THg between the POM and sediment is almost equivalent to the middle of the range of TMF values (2.38～6.86) in the prey-predator relationship of the aquatic organisms described in the previous studies (Table 2).

Therefore, it is deduced that a kind of "bioconcentration process", strictly speaking a biologically accelerated concentration process, of mercury works on the sediment, and promotes the accumulation of mercury into the sediment (not in the bodies of the aquatic organisms). Although Kennish (1998)[53] pointed out that ppm levels of mercury accumulated in the sediment in some bays and harbors in the U.S. where particular discharging sources of mercury such as chemical plants, pulp and paper mills, etc. did not exist in the neighboring areas, the accumulation process of mercury into the sediment has not been defined yet as a kind of bioconcentration process in the benthic ecosystem in the enclosed coastal seas in the previous studies. For the benthic invertebrates occurring in the sediment, which are referred to as infaunal species, they are always exposed the enhanced contaminant levels of mercury in their habitats, and the sediment is one of the main diets for the deposit-feeding species among them. These animals are classified as primary consumers, but it is very likely that mercury tends to accumulate in their bodies as much as those of one-step higher trophic level.

4. The effects of bioconcentration of THg into the sediment on benthic invertebrates

According to the conventional ideas on the bioaccumulation of THg in the aquatic ecosystem, the content of the animal tends to increase discontinuously as the elevation of the trophic level in the biological community (Fig. 1). However, Kim et al. (2012)[41] reported that clamworm (polychaete, *Perinereis aibuhitensis*) recorded the highest THg content (139 ± 37.2 ng g^{-1} d.w.) among the benthic invertebrates that occurred in the coasts of Masan Bay, Korea. This species is one of the deposit-feeding primary consumers, inhabiting the sediment, and feed on it[54]. Its THg content exceeded that of a carnivorous secondary consumer of the benthic invertebrate, octopus (*Octopus* sp., 109 ± 34.8 ng g^{-1} d.w.). Sizmur et al. (2013)[55] also found an inverse relationship between $\delta^{15}N$ values and bioaccumulation factors for THg among the polychaetes occurring on the mud flats in the coast of Bay of Fundy, Canada. Deposit-feeding species (primary consumers) such as Maldanidae spp., and Capitellidae spp. had higher values of bioaccumulation factor for THg to carnivorous species (the secondary consumers), Glyceridae spp. These deposit-feeding polychaetes burrow into the sediment, and swallow it to digest the detritus in the sediment[56]. These facts coincide with our prediction based on the presence of the undefined bioconcentration process of THg into the sediment (Fig. 3) after the deposition of POM with THg contaminants on the sediment of the sea floor.

Jaingam et al. (2018)[33] classified each of the primary and secondary consumers of the benthic invertebrates occurring in Isahaya Bay, Japan, into two different groups with their THg contents, which are referred to as "High THg content group" and "Low THg content group" (Fig. 4, Table 3). In the primary consumers, the benthic invertebrates of "High THg content group" contained 101 ± 23 ng g^{-1} d.w. (mean ± S.D.) of the mean THg, and were mainly made up of deposit-feeding small bivalves (*Theora lata, Ratellops pulchella*)[57) 58] and polychaete (*Sternaspis costata*)[56], and suspension-feeding bivalves (*Ruditapes phillinarum, Arculatula senhousia, Tegillarca granosa, Crassostrea gigas*)[59)-62]. All of these species except *C. gigas* live in the sediment which is referred to as "infauna", and *C. gigas* occurs on the mud flats, being partly buried in the mud, in Isahaya Bay[63]. They rely on the items of their main diets to the detritus of the sediment and microphytobenthos increased on the surface sediment or their re-suspensions in the overlying water just above the sediment[47)-49]. Contrastively, the benthic invertebrates of "Low content group" contained 25.2 ± 8.5 ng g^{-1} d.w. (mean ± S.D.) of the mean THg. They were occupied by the epifaunal species such as amphipods, *Cerapus* sp. and *Photis longicaudata*, which live on the surface of the sediment, and move around on it actively, and mussels (*Mytilus galloprovincialis*) attached to the hard substrates. These species exploit the diets mainly from the suspensions of the water and much less from the deposits of the sediment[64) 65].

In the secondary consumers, "High THg content group" contained 215 ± 47 ng g^{-1} d.w. (mean ± S.D., n=4) of the mean THg. They consisted of carnivorous crabs (*Hemigrapsus* sp., *Charybdis japonica, Portunus trituberculatus*) and sea star (*Asterias* sp.). Their high THg contents were transferred to their bodies through their preferential predation on the clams[66)-68] belonged to "High THg content group" of the primary consumers. "Low THg content group" contained 63.1 ± 39.3 ng g^{-1} d.w. (mean ± S.D., n=3) of the mean THg. They were made up of shrimps (*Metapenaeus* cf. *joyneri, Metapenaeus* cf. *affinis*) and carnivorous polychaete, *Glycera*

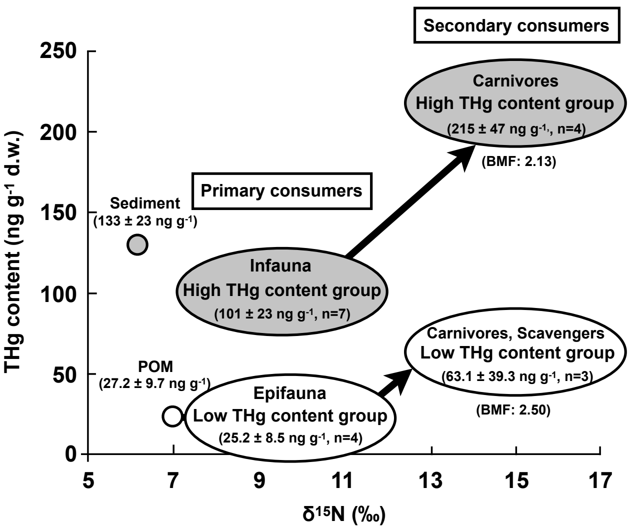

Fig. 4 Two different biomagnification pathways of total mercury (THg) between the primary and secondary consumers in the benthic ecosystem described in the field surveys conducted in the inner parts of Isahaya Bay, Japan. (modified from Jaingam et al. 2018)[33].

Table 3 Carbon and nitrogen stable isotope ratios and THg contents of (a) the benthic invertebrates collected in the field surveys conducted in the inner parts of Isahaya Bay, Japan. The data are based on Jaingam et al. (2018)[33].
B: Bivalvia, M: Malacostraca, P: Polycheata, A: Asteroidea
Living mode; INF: infauna, EPI: epifauna, Feeding type; D: deposit feeder, F: filter feeder,
SF: suspension feeder, SD: surface detritivore, S: scavenger, C: carnivore
N_1: No. of data of the stable isotope analysis, N_2: No. of data of THg content analysis

Species	Living mode	Feeding type	$\delta^{13}C$ (‰) mean ± S.D.	$\delta^{15}N$ (‰) mean ± S.D.	N_1	THg content (ng g^{-1} d.w.) mean ± S.D.	N_2	References
Primary consumers: High THg content group								
B: *Theora lata*	INF	D	-18.0 ± 0.5	7.7 ± 0.6	13	141	1	57
B: *Raetellops pulchella*	INF	D	-18.2 ± 0.7	7.9 ± 1.2	12	122	1	58
P: *Sternaspis costata*	INF	D	-17.6 ± 0.2	8.6 ± 0.2	3	97.2	2	56
B: *Ruditapes philippinarum*	INF	F	-17.5 ± 0.3	9.7 ± 0.4	6	80.8	2	59
B: *Arcuatula senhousia*	INF	F	-17.0 ± 0.4	10.4 ± 1.2	8	77.9	2	60
B: *Tegillarca granosa*	INF	F	-15.9 ± 1.0	11.1 ± 0.5	6	91.7 ± 19.2	6	61
B: *Crassostrea gigas*	EPI/INF	F	-16.6 ± 0.4	11.8 ± 0.5	8	94.1 ± 22.3	8	62, 63
Mean			-17.3 ± 0.8	9.6 ± 1.6		101 ± 23		
Primary consumers: Low THg content group								
M: *Ampelisca bocki*	EPI	SF/SD	-17.4 ± 0.5	8.1 ± 0.4	3	13.8	1	64
M: *Photis longicaudata*	INF/EPI	SF/SD	-17.2 ± 0.2	9.3 ± 1.0	3	28.0	1	64
M: *Cerapus* sp.	EPI	SD	-18.7 ± 0.3	9.8 ± 0.8	3	25.1	1	64
B: *Mytilus galloprovincialis*	EPI	F	-18.0 ± 0.9	9.8 ± 0.4	6	34.1 ± 4.3	10	65
Mean			-17.8 ± 0.7	9.3 ± 0.8		25.2 ± 8.5		
Secondary consumers: High THg content group								
M: *Hemigrapsus* sp.	EPI	C/S	-15.8 ± 0.3	12.6 ± 0.4	3	271 ± 22	3	66
M: *Charybdis japonica*	EPI	C/S	-15.9 ± 1.3	14.1 ± 0.6	5	183 ± 43	5	67
M: *Portunus trituberculatus*	EPI	C/S	-15.7 ± 1.7	14.0 ± 0.8	4	170 ± 48	4	67
A: *Asterias* sp.	EPI	C	-15.1 ± 0.6	12.8 ± 0.3	4	234	2	68
Mean			-15.6 ± 0.4	13.4 ± 0.8		215 ± 47		
Secondary consumers: Low THg content group								
M: *Metapenaeus* cf. *affinis*	EPI	S/C	-16.1 ± 0.8	13.5 ± 0.7	9	102 ± 49	9	69
M: *Metapenaeus* cf. *joyneri*	EPI	S/C	-16.7 ± 1.1	14.4 ± 0.4	5	63.2 ± 20.0	5	70
P: *Glycera* sp.	INF	C	-15.8	12.6	2	23.7	1	56
Mean			-16.2 ± 0.5	13.5 ± 0.9		63.1 ± 39.3		

sp. These shrimps get foods mostly by scavenging[69) 70)], and *Glycera* feeds on small benthic animals[56)]. Most of their foods seemed to be derived from the epifaunal benthic invertebrates belonged to "Low THg content group" of the primary consumers.

Thus, these facts indicate that the condensed mercury into the sediment is transferred to infaunal benthic invertebrates and their predators through their feeding activities.

5. Accelerated accumulation pathway of THg to fish in the benthic ecosystem

The presence of "High THg content group" in both of the primary and secondary consumers of the benthic invertebrates as shown in Fig. 4 suggest the presence of further accelerated THg accumulation pathway to the fish community. Fishes occurring in the coastal seas are classified into three types on their living conditions, "pelagic", "bentho-pelagic", and "benthic"[40)]. Pelagic fish feeds on plankton and/or small fish in the water. Benthopelagic and benthic fishes rely on diets to the benthic invertebrates partly or fully. Although most of them swim in the water, but are easily accessible for diets of those such as invertebrates occurring on the sea floor in the coastal shallow waters. If some of these fishes preyed on particular species belonged to "High THg content group" of the primary and secondary consumers of the benthic invertebrates preferentially, the THg condensed into the sediment would be transferred to the fishes efficiently via predation of the benthic invertebrates of "High THg content group"[33)].

Jaingam et al. (2018)[34)] found that the THg contents of the muscles of six species of benthic or benthopelagic fishes as the secondary consumers or intermediate ones between the secondary and tertiary consumers (*Hemitrygon akajei* (red stingray), *Acanthopagrus schlegelii* (blackhead seabream), *Takifugu niphobles* (grass puffer), *Nuchequula nuchalis* (spotnape ponyfish), *Takifugu rubripes* (yellowfin pufferfish), *Planiliza haematocheila* (so-iuy mullet) exceeded those of two species of the pelagic ones as the tertiary consumers (*Paralichthys olivaceus* (bastard halibut): 249 ng g^{-1} d.w., n=1, *Lateolabrax japonicus* (Japanese seabass): 266 ± 99 ng g^{-1} d.w., n=5) in Isahaya Bay, Japan (Fig. 5).

In particular, *H. akajei* showed symbolic changes of THg content of the muscles following its growth in the relationship between the trophic position and THg content. Its small individuals (30.6 ± 2.4 cm, 98.2 ± 29.1 g, mean ± S.D., n=6) were located at 3.7 in the trophic position (TP), which is classified as the intermediate consumer but close to the tertiary one, and contained 142 ± 76 ng g^{-1} d.w. of THg. However, its THg content increased drastically as it grew. Its large ones (63.6 ± 4.9 cm, 1,847 ± 145 g, n=4) and

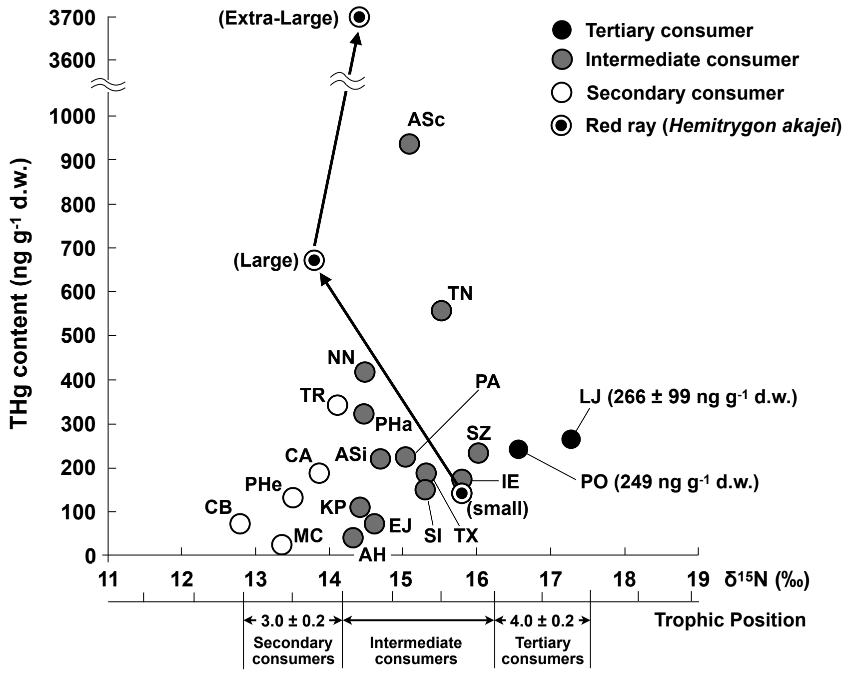

Fig. 5 The relationship between the $\delta^{15}N$ values and THg contents of the body tissues of the fishes collected in the inner part of Isahaya Bay. LJ: *Lateolabrax japonicus* (Japanese seabass), PO: *Paralichthys olivaceus* (Bastard halibut), SZ: *Sardinella zunasi* (Japanese sardinella), IE: *Llisha elongata* (elongate ilisha), TN: *Takifugu niphobles* (Grass puffer), SI: *Sebastes inermis* (Dark-banded rockfish), TX: *Takifugu xanthopterus* (Yellowfin pufferfish), ASc: *Acanthopagrus schlegelii* (Blackhead seabream), PA: *Pennahia argentata* (Silver croaker), ASi: *Acanthopagrus sivicolus* (Okinawa seabream), EJ: *Engraulis japonicus* (Japanese anchovy), PHa: *Planiliza haematocheila* (So-iuy mullet), NN: *Nuchequula nuchalis* (Spotnape ponyfish), KP: *Konosirus punctatus* (Dotted gizzard shad), AH: *Amblychaeturichthys hexanema* (Pinkgray goby), TR: *Takifugu rubripes* (Japanese pufferfish), CA: *Cynoglossus abbreviatus* (Three-lined tongue sole), PHe: *Pseudopleuronectes herzensteini* (Yellow striped flounder), MC: *Mugil cephalus* (Flathead grey mullet), CB: *Callionymus beniteguri* (Whitespotted dragonet). (modified from Jaingam et al. (2018)[34])

extra-large one (72.0 cm, 5,150 g, n=1) contained 671 ± 340 ng g^{-1} d.w. and 3.700 ng g^{-1} d.w. of THg, respectively, although their trophic positions descended to 3.1 (the secondary consumer) and 3.3 (the intermediate consumer, close to the secondary one). The small individuals tend to prey on crustaceans including small shrimps and mysids and fishes due to undeveloped teeth[71]. In this stage, their main diets are "Low THg content group" of the secondary consumers of the benthic invertebrates and/or small pelagic fish. Its larger individuals prefers a bivalve, *Ruditapes philippinarum* (short-neck clam), exclusively[72) 73)], which is one of the members of "High THg content group" of the primary consumers of the benthic invertebrates in Isahaya Bay[33]. Here, the THg condensed into the sediment was transferred to the clam, and it caused drastic acceleration of bioaccumulation of THg to the red stingray by its preferential predation on the clam.

Figure 6 illustrates the outline of the routes of THg transference between the primary and tertiary consumers in the aquatic ecosystem in the enclosed coastal areas, being based on the statements in this review. At a glance, much more complex THg transference pathways exist between the primary and tertiary consumers in the benthic ecosystem than those in the pelagic ecosystem (Fig. 2). The mercury accumulated in the sediment on the sea floor is not retained there intact, but is apt to return to the fishes swimming in the water of the bay by the feeding activities of the benthic invertebrates, in particular infaunal species occurring in the sediment, and preferential predation on them by the benthic and benthopelagic fishes. The value of BMF (Biomagnification Factor) of THg between the clam (*R. philippinarum*, 80.8 ng g^{-1} d.w.)[33] and its predator, red stingray, (*H. akajei*, 3,700 ng g^{-1} d.w.)[34] reached 45.8 (3,700/80.8). It is about 6.7 times larger than the maximum value of TMF (6.86) noted in the marine ecosystem in the previous studies (Table 2), and the THg content of the red stingray far exceeded those of sea birds (*Sterna paradisaea* (Arctic tern), 2,109 ± 531 ng g^{-1} d.w. (mean ± S.D.)); *Rissa tridactyla* (black-legged kittiwake), 1,930 ± 1,140 ng g^{-1} d.w.) and marine mammals (*Monodon monoceros* (narwhals), 2,320 ± 970 ng g^{-1} d.w.; *Delphinapterus leucas* (beluga whale), 2,250 ng g^{-1} d.w.) occurring in the Arctic region in Canada, although they inhabited uncontaminated areas[42) 43)].

Thus, through this review, we propose to recognize the presence of a bioconcentration process of mercury into the sediment on the sea floor, the transference pathway of the condensed mercury of the sediment to some particular infaunal species of the benthic invertebrates, and highly accelerated mercury transference pathway to the fishes due to their preferential predation on them in the enclosed

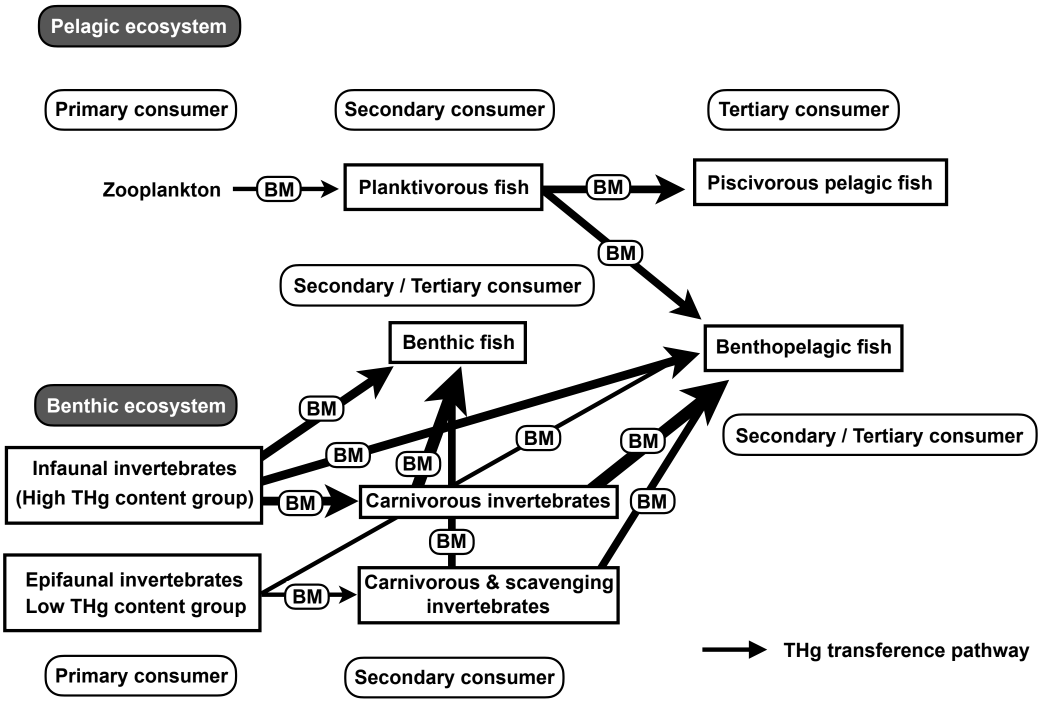

Fig. 6 Outline of the THg transference routes between the primary and tertiary consumers in the aquatic ecosystem in the enclosed coastal areas. BM: biomagnification

coastal seas.

6. Conclusions

An accelerated bioaccumulation pathway of mercury exists in the benthic ecosystem in the enclosed coastal seas. POM with mercury contaminants, which is made up of the dead bodies of phytoplankton and zooplankton and feces excreted from the zooplankton, is always settling on the sediment of the sea floor as detritus before it has been degraded fully in the water, due to shallow water. On the sea floor, the degradable parts of the detritus are degraded by microorganisms in the sediment, while some of the non-degradable parts including mercury tend to be retained in the sediment. The mercury contaminated in the POM tends to highly accumulate into the sediment, and is transferred to the infaunal benthic invertebrates, which burrow into the sediment, and feed on the sediment or take in its re-suspension into the overlying water just above the sea floor. The mercury accumulated in the infaunal benthic invertebrates is transferred to the benthic and benthopelagic fishes that prey on them preferentially. Thus, the mercury that once deposited in the sediment on the sea floor is moved finally to the fishes occurring in the pelagic ecosystem, further highly accelerating its bioaccumulation.

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