

Transfer of polychlorinated biphenyls from sediment to benthic fish in laboratory tanks

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Abstract

To evaluate the transfer of polychlorinated biphenyls (PCBs) from sediment to aquatic organisms, we examined the temporal trend of PCB concentrations in various parts of benthic fish by laboratory experiment. As experiment tanks, control (only seawater), SS (seawater containing suspended solid 100 mg/l) and sediment tanks (sediment laid on the bottom of the tank) were prepared, and marbled soles *Pleuronectes yokohamae* were placed for 14-28 days. The total PCB concentrations in the water were 47-99 pg/l, 2,100-2,200 pg/l and 44,000-50,000 pg/l for the control, the SS tank and the sediment tank for 0-28 days, respectively. More than 90 % of the total PCB concentration in the SS tank and the sediment tank was the particle phase. The mean PCB concentrations in each part of the fish increased 2-5-fold in the sediment tank during the experiment period whereas those did not increase in the control and the SS tank. It was suggested that PCBs originating from the sediment transferred to the fish. In addition, the composition of the PCB homologue in the fish from the control and the SS tank reflected that in the food, while that in the fish from the sediment tank reflected those in the sediment and water.

Introduction

In the aquatic environment, the majority of persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are present in the sediment. Therefore, sediments may play a role as secondary sources of these compounds to other media such as water and aquatic organisms. In particular, the transfer potential of POPs from the sediment to aquatic organisms needs to be studied because it is known that the major route of exposure to PCBs and PCDD/DFs for the Japanese population is through uptake of fish and shellfish^{1,2}). In this report, to evaluate the transfer of PCBs from the sediment to aquatic organisms, we examined the temporal trend of PCB concentrations in various tissues of benthic fish by laboratory experiment.

Materials and Methods

Experimental scheme

We used as test organism farmed marbled sole *Pleuronectes yokohamae* (hereafter called marbled sole) in this experiment. The marbled soles (one year; wet weight: 5.4-32.7 g, avg. 14.7 g; n=15) were placed for 14-28 days at 20 °C in glass tanks containing 60 l of seawater. As the experiment tanks, control (only seawater), SS tank (seawater containing 100mg/l (nominal) of suspended solid (SS)) and sediment tank (with a bottom sediment layer of approximately 1 cm thickness) were prepared. Two of the total fifteen marbled soles were sampled just before the start of the experiment. Then, four to five marbled soles were placed in each tank. After 14 days and 28 days from the start of the experiment, two or three marbled soles were sampled from each tank. Ten liters of seawater from each tank was replaced every 2 days. Five liters of water samples were collected during each water replacement for the two periods of 0-14 days and 15-28 days. For the SS tank and sediment tank, the SS concentration in the water of each tank was measured at each water replacement, and sediment particles were added to compensate for the loss brought about by the replacement. The sediment that was collected from Tokyo Bay in April 2004 was clay silt (mean particle diameter of 4 µm) and contained 1.7 % organic carbon. The seawater used was deep seawater (salinity of 36, pH 8.0), which was collected from Sagami Bay in November 2005. The food used was a commercial fish food, which was fed to the fish at 1 % of the body weight per day.

PCB analysis

The sampled marbled soles were washed to remove sediment particles on the body surface by running water and Milli-Q water, and then were sectioned into muscle, liver, internal organs (except liver), and the remaining parts (head, skin, bone and so forth). These parts were homogenized with a homogenizer and stored at <-20 °C until analysis. Prior to extraction, the homogenized samples were thawed and dehydrated by powdered anhydrous sodium sulfate or diatomite (hydromatrix, Varian, Inc., USA). The dehydrated samples were spiked

with $^{13}\text{C}_{12}$ -labeled internal standards (MBP-MXP, Wellington Laboratories Inc., Canada), and Soxhlet-extracted with 100 ml of hexane:methylene chloride mixture (1:1 v/v) for 16 hours.

Five liters of the water samples were separated into the particle phase and dissolved phase using a pressurized filtration system (Advantec Toyo Kaisha, Ltd., Japan). The glass fiber filters used were precombusted (2 hours, 400 °C). In this report, the particle phase corresponds to those retained on a 0.3 μm (nominal pore size) glass fiber filter (GF-75, Advantec Toyo Kaisha, Ltd., Japan).

The water and sediment samples were spiked with $^{13}\text{C}_{12}$ -labeled internal standards (MBP-MXP, Wellington Laboratories, Canada) prior to extraction. The particle phases of the water samples and sediment samples were extracted using a Soxhlet/Dean-Stark extractor with 300 ml of toluene for 16 hours. The dissolved phase of the water samples was liquid-liquid extracted twice each with 100 ml of hexane. The extracts of fish, water and sediment samples were cleaned by multilayer silicagel column chromatography. The eluates were then spiked with $^{13}\text{C}_{12}$ -labeled injection internal standard (PCB-IS-A, Wellington Laboratories Inc., Canada). All of the 209 congeners of PCBs were identified and quantitated using a HRGC/HRMS (HP6890, Agilent Technologies, USA/JMS 700, JEOL Ltd., Japan) system equipped with a HT8-PCB column (60m \times 0.25 mm (i.d.), Kanto Chemical Co., Inc., Japan).

Results and Discussion

PCB concentrations in water, sediment and food

The PCB concentrations in the water from each tank are shown in Table 1. The PCB concentrations in each tank were considered relatively stable during 0-28 days because the PCB concentrations in the water samples of 0-14 days were relatively the same as those of 15-28 days. The total PCB concentration in the water samples in the control ranged from 47 to 99 pg/l, those in the SS tank ranged from 2,000 to 2,200 pg/l, and those in the sediment tank ranged from 45,000 to 50,000 pg/l.

The total PCB concentration in the SS tank was more than 20-fold higher than those in the control, although the PCB concentrations in the dissolved phase in the SS tank were roughly equal to those in the control. Moreover, the PCB concentrations in the dissolved phase in the sediment tank were 8-24-fold higher than those in the control and SS tank.

Approximately 90 % of the total PCB concentration was in the dissolved phase in the control although more than 90 % of the total PCB concentration was in the particle phase in the SS tank and the sediment tank. The mean SS concentration in the SS tank was 54 mg/l and 76 mg/l for 0-14 days and 15-28 days, respectively. The mean SS concentration in the sediment tank was 610 mg/l and 420 mg/l for 0-14 days and 15-28 days, respectively.

The PCB concentration in the sediment was 89 ng/g dry matter, which roughly corresponded to the PCB concentration in the sediment collected from Tokyo Bay, as mentioned in another paper ³⁾. The PCB concentration in the food was 6.8 ng/g wet weight.

Table 1. PCB concentrations in water samples from each tank (pg/l).

	0-14 days			15-28 days		
	dissolved phase	particle phase	total	dissolved phase	particle phase	total
Control	95	4	99	41	6	47
SS tank	100	2,000	2,100	62	2,200	2,200
Sediment tank	780	44,000	45,000	1,000	49,000	50,000

Temporal trend of PCB concentrations in marbled soles

The PCB concentrations in each part increased in fish from the sediment tank during the experiment period whereas those did not increase in fish from the control and the SS tank. The temporal trends of PCB concentrations in each part of marbled soles are shown in Figure 1. Under the condition that the PCB concentration in the dissolved phase is approx. 40-100 pg/l and/or the SS concentration is 54-76 mg/l (the control and/or the SS tank), it was suggested that PCBs did not apparently transfer from the dissolved phase and the suspended particle to the fish. On the other hand, under the conditions that the sediment is present, the PCB

concentration in the dissolved phase is approx. 800-1,000 pg/l and the SS concentration is 420-610 mg/l (the sediment tank), it was suggested that PCBs transfer from the dissolved phase, the suspended particle and the sediment to the fish. However, both the dissolved phase and the suspended particle essentially originated from the sediment.

The variation in the mean PCB concentration in each part was of the following order: liver > internal organs > remaining parts > muscle. The mean PCB concentration in each part of fish from the sediment tank increased by 2-5-fold from day 0 to day 14. Then the mean PCB concentration in the muscle, liver and internal organs decreased or roughly leveled off from day 14 to day 28, although those in the remaining parts increased to approx. 3-fold from day 14 to day 28. However, the temporal trend of the PCB concentrations from day 14 to day 28 was not clear because the difference between the minimum and maximum concentrations in the samples collected after 28 days is large. In the case of the whole body (all tissues combined), the mean concentration increased to 4-fold from day 0 to day 28. However, the temporal trend of from after day 14 to day 28 was unclear because the difference between the minimum and maximum concentrations in the samples collected after 28 days is large.

In terms of PCB homologues, the T4CB and P5CB concentrations tended to increase considerably in all the parts of the fish from the sediment tank during the experiment period. The temporal trends of PCB homologue concentrations in each part of the fish from the sediment tank are shown in Figure 2. H6CB concentrations were the highest among the PCB homologues in all the parts of the body before the start of the experiment. However, day 14 and day 28, the T4CB and P5CB concentrations in all the parts of the body were higher than the H6CB concentration. The composition of PCB homologues to total PCB concentrations was relatively constant regardless of the part. Before the start of the experiment, the compositions of T4CB, P5CB and H6CB in each part were $14\pm 0.7\%$, $20\pm 3.5\%$ and $45\pm 5.0\%$, respectively. On the other hand, after day 14 and day 28, the compositions of T4CB, P5CB and H6CB in each part were $43\pm 1.5\%$, $26\pm 1.0\%$ and $18\pm 1.7\%$, respectively.

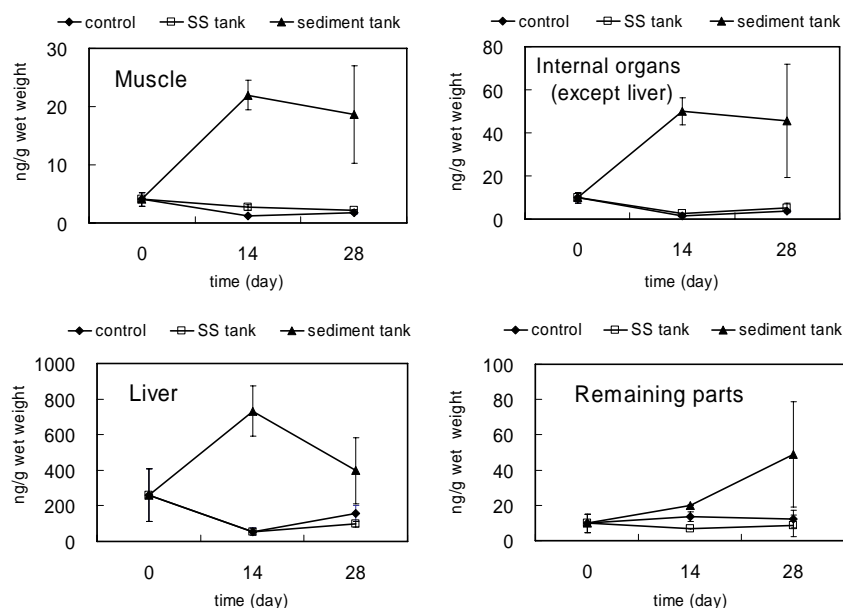


Fig. 1. Temporal trend of PCB concentrations in the various parts of marbled soles (error bars denote range of concentrations).

Comparison of composition of PCB homologues between fish and other media

To estimate the transfer route of PCBs from the various media to the marbled soles, the composition of PCB homologues in the marbled soles was compared with those of the water, sediment and food. Figure 3 shows comparison of the composition of PCB homologues among the various media. The composition in the marbled soles was average in the muscle because the composition was relatively constant regardless of part.

In the food, the compositions of T4CB, P5CB and H6CB were 17 %, 26% and 36%, respectively. In the water and the sediment, the compositions of T4CB, P5CB and H6CB were 38-43 %, 21-24 % and 13 %, respectively. On the other hand, the compositions of T4CB, P5CB and H6CB in the marbled soles at day 0 from the control and the SS tank were 15-24%, 22-26 % and 38-40 %, respectively, and corresponded well to the composition in the food. The compositions of T4CB, P5CB and H6CB in the marbled soles from the sediment tank were 44 %, 28 % and 16 %, respectively, which reflected the composition in the sediment and the water. Thus, the transfer of PCBs from the sediment and the water to the marbled soles was also supported by the comparison of homologues among the various media. It is considered that the transfer routes of PCBs are by uptake through the gill of dissolved phase and particle and by uptake through mouth of sediment and particle. However, in the present study, their contributions could not be explained.

Acknowledgements

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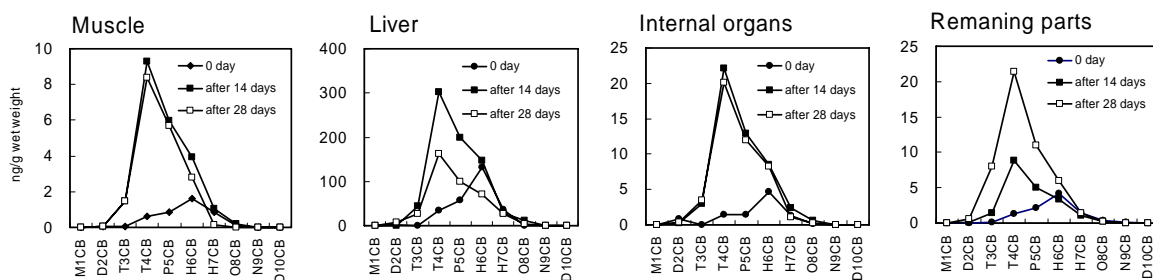


Fig. 2. Temporal trend of mean PCB homologue concentration in each part of fish from the sediment tank.

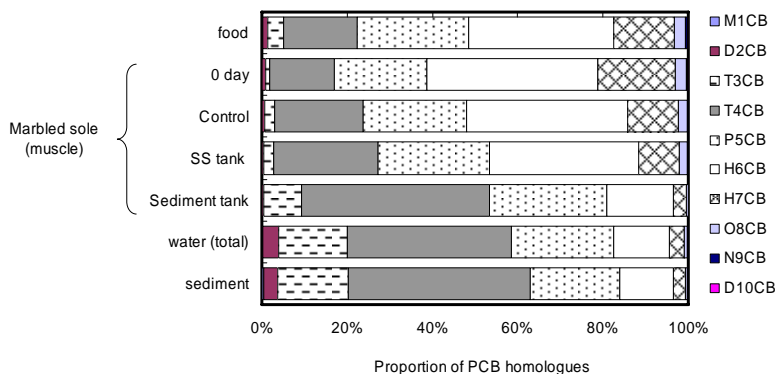


Fig. 3. Comparison of composition of PCB homologues in fish, food, sediment and water samples.