

Bioconcentration and redeposition of polychlorinated biphenyls by zebra mussels (*Dreissena polymorpha*) in the Hudson River

Young-Cheol Cho^a, Robert. C. Frohnhoefer^a, G-Yull Rhee^{a,b,*}

^aWadsworth Center, New York State Department of Health, Empire State Plaza, Albany, NY 12201, USA

^bSchool of Public Health, State University of New York at Albany, Albany, NY 12201, USA

Received 14 February 2003; received in revised form 6 August 2003; accepted 27 October 2003

Abstract

The potential impact of zebra mussel infestation on the dynamics of polychlorinated biphenyls (PCBs) in the Hudson River was determined by investigating the biodeposition and bioconcentration of PCBs, using algal food contaminated with 2,5,2'- and 2,4,2',4'-chlorobiphenyls (CBPs) in the laboratory. Approximately 46–90% of the total food was ingested depending on the supply rate. The highest proportion of ingested congeners was found in biodeposits ($64 \pm 11\%$ for 2,5,2'-CBP, and $52 \pm 6\%$ for 2,4,2',4'-CBP), followed by tissues ($17 \pm 3\%$ for 2,5,2'-CBP, and $23 \pm 5\%$ for 2,4,2',4'-CBP), and the lowest in shells. The clearance rate decreased with increasing food concentration, but increased with dilution rate. On the other hand, ingestion rate (IR) increased with food concentration and dilution rate. IR also increased with food supply rate (food concentration \times dilution rate) following the same linear function whether the supply rate was varied through food concentration or dilution rate. Therefore, the dilution rate- or food concentration-dependent variation in IR was due to the change in the food supply rate. IR was independent of the kind of PCB congeners. The trend of bioaccumulation in mussel tissues from food ingestion was similar to that of IR; bioaccumulation increased linearly with food supply rate, whether the supply rate was varied through the dilution rate or the food concentration. The bioaccumulation of 2,4,2',4'-CBP was significantly higher than that of 2,5,2'-CBP ($p < 0.05$). The bioaccumulation was linearly related to the IR or to the total amount of food ingested. Assimilation efficiency, PCB incorporated in the tissue per total ingested PCB, was higher for 2,4,2',4'-CBP than for 2,5,2'-CBP ($p < 0.05$). The congener concentration in biodeposits increased with food supply rate. However, the concentration of 2,5,2'-CBP was significantly greater than that of 2,4,2',4'-CBP in a mirror image of bioaccumulation. These results indicate that zebra mussels may significantly alter PCB dynamics in the Hudson River through redeposition from the water column and through bioconcentration.

© 2003 Elsevier Ltd. All rights reserved.

Keywords: Zebra mussel; Bioconcentration; Biodeposition; Assimilation efficiency; PCB

1. Introduction

The Hudson River has been suffering from two major stresses resulting from human activities: contamination

by polychlorinated biphenyls (PCBs) and colonization by zebra mussels (*Dreissena polymorpha*). PCB contamination has impaired the use of the river for decades because of potential public health risks [1]. Zebra mussels have become an important and probably permanent part of the Hudson River ecosystem. Their ecological impacts on the river have been extensively investigated in recent years [2]. The invasion of the river by these hardy filter feeders has also caused significant

*Corresponding author. Wadsworth Center, New York State Department of Health, Empire State Plaza, Albany, NY 12201, USA. Tel.: +1-518-473-8035; fax: +1-518-486-2697.

E-mail address: rhee@wadsworth.org (G.-Y. Rhee).

changes in the river ecosystem, including alterations of the food chain [3,4].

Zebra mussels filter a wide range of particle types and sizes with approximately equal efficiencies [5–7]. The filtration rate of zebra mussels in the Hudson River is estimated to be equivalent to the entire volume of the tidal freshwater portion being turned over every 1.2–3.6 days [2,7]. The total PCB concentration in water (in both particulate and aqueous phases) in the upper Hudson River ranged from 26 to 101 ng/L [8]. A theoretical calculation based on these upper-river values shows that, at a filtration volume of 48 m³/m² mussel bed/d [9], the total amount of PCBs passing through zebra mussels would range from 1.3 to 4.9 µg/m² mussel bed/d. These estimates indicate that zebra mussels should have a significant impact on particle fluxes, which in turn control PCB dynamics.

Zebra mussels may influence the dynamics through two major routes: i.e., bioconcentration in mussel tissue through direct partitioning of PCBs and ingestion of contaminated particulates, and the transfer of PCB-contaminated particulates from the water column to the sediments through the biodeposition of feces and pseudofeces. Filter-feeding bivalves can remove a significant amount of PCBs [10–12] and other hydrophobic organic contaminants [13,14] from the water column. In a Baltic coastal system, blue mussels increased the redeposition of PCBs by 50% [15]. Therefore, zebra mussels can have potentially significant impact on the PCB dynamics in the Hudson River through their bioconcentration of PCBs and transfer of PCB-laden particulates from the water column to sediments, which otherwise would have escaped from the system with river flow.

In the present study, we have investigated, under laboratory conditions, the extent of biodeposition and bioconcentration of PCBs by zebra mussels collected from the Hudson River.

2. Materials and methods

2.1. Zebra mussels and algal food

Zebra mussels were collected in the Hudson River (NY, USA) near North Germantown. They were sorted by size (average tissue dry weight 38 ± 2 mg). They were maintained until use in an aquarium in a constant temperature room (18°C). They were continuously fed with heat-killed unicellular alga, *Nanochloris* sp., in a modified medium of Wright et al. [16] using a peristaltic pump. The medium contains 109 mg CaCl₂•2H₂O, 91 mg MgSO₄•7H₂O, 166 mg NaHCO₃, and 8 mg KCl/L (pH 7.0). *Nanochloris* sp. was mass-cultured in Bold basal medium [17], harvested by centrifugation, and heated in a water bath (80°C) for 20 min.

For the present studies, two PCB congeners, 2,5,2'- and 2,4,2',4'-chlorobiphenyl (CBP; IUPAC No. 18 and 47, respectively; AccuStandard, New Haven, CT, USA) were used. 2,5,2'-CBP is a major component of Aroclor 1242 (8.8 mol%), the primary PCB mixture discharged into the Hudson River. 2,4,2',4'-CBP was detected at high concentration (5.0 mol%) in contemporary river sediments [18], probably because they are resistant to microbial dechlorination [18,19]. The log-octanol–water partition coefficients (K_{ow} 's) of 2,5,2'- and 2,4,2',4'-CBP are 5.55 and 6.29, respectively [20]. A concentrated solution of algal cells contaminated with each congener (3.3 µg PCB/mg algae) was prepared without a carrier solvent according to Lederman and Rhee [21]. This suspension was allowed to equilibrate for at least 4 days before use. For experiments, the food supply was made up daily using this equilibrated stock solution at desired concentrations. Most PCBs were found in the food, with the aqueous concentration accounting for less than 1% of the total.

2.2. Experimental apparatus

Experiments were carried out using a flow-through glass chamber (beaker) modified from Madon et al. [22] in a constant temperature room (18°C) (Fig. 1). Each chamber had an inner chamber consisting of a smaller beaker with an open bottom that was fitted with a detachable 120-µm stainless-steel mesh (TWP Inc., Berkeley, CA, USA). This mesh size was large enough for algal food to pass through freely, yet small enough to retain biodeposits. Each experimental chamber contained four mussels. They were glued to the horizontal part of an L-shaped glass rod, at the outer edge of one shell, with cyanoacrylate adhesive. The vertical end of the glass rod was attached to the inside bottom surface of an inverted petri-dish so that, when the chamber was covered by this dish, the mussels would be kept suspended at the center of the chamber. The food suspension, in an 8-L reservoir, was constantly mixed to maintain homogeneity, and was fed into the mussel chamber by a peristaltic pump at a predetermined rate. Fresh food supply was made daily, because most of the 8-L was consumed each day to supply duplicate experimental chambers with mussels and two identical chambers without mussels as the control. To minimize surface sorption, the feed line was made of glass tubing, except for a short segment of Tygon tubing at the pump to allow peristaltic pumping. The control was fed from the same reservoir through a second channel of the pump at the same rate.

2.3. Experiments

Before introduction of PCB-laden food, mussels in experimental chambers were acclimated to the algal food

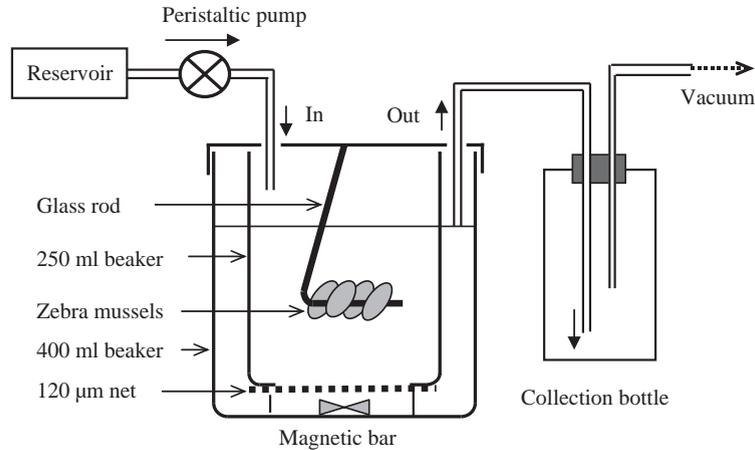


Fig. 1. Diagram of the apparatus used in the experiment.

by feeding them with PCB-free algae for 3 days. The effect of food concentration were investigated at five different levels using PCB-contaminated algae, ranging from 1 to 5 mg algae/L, at a constant flow rate (F , L/d) to displace the media volume in the chamber 3 times a day, or at a dilution rate ($F/\text{media volume}$, /d) of 3/d. The effects of feeding rate were studied at five dilution rates ranging from 1 to 5/d, while food concentration was maintained constant (3 mg algae/L).

The concentration of algae was determined daily by monitoring chlorophyll *a* fluorescence using a fluorometer (TD-700, Turner Designs, Sunnyvale, CA, USA) in both experimental and control chambers. A preliminary study showed that this pigment in heat-killed cells did not degrade at least for one month. The chlorophyll *a* reading was calibrated against algal dry weight to convert its value into dry weight. A periodic check also showed no change in the curve. The biodeposit on the stainless-steel mesh was collected daily, and its dry weight was measured.

The algal clearance rate (CR; mL of water cleared/mussel/d) was determined as

$$\text{CR} = \left(\frac{C_c - C_e}{C_c} \right) F / M, \quad (1)$$

where C_c and C_e are the concentrations of algae (mg algae/L) in the control and experimental chambers, respectively. F is the flow rate (L/d), and M is the number of mussels in the chamber (the value of C_c is the same as C_f , the food concentration in the reservoir). All these parameters were measured daily during a 4-day experimental period.

Ingestion rate (IR, μg of algae consumed/mussel/d) was calculated as

$$\text{IR} = (C_c - C_e)F / M. \quad (2)$$

Assimilation efficiency (AE) of PCBs, the amount of PCB incorporated into the tissue (P_t) from total ingested PCB ($P_a \text{ IR } t$), was calculated at the end of a 4-day experimental period by the equation for each dilution rates and food concentration

$$\text{AE} = \frac{P_t}{P_a \text{ IR } t}, \quad (3)$$

where P_a is the amount of PCB associated with algae (μg PCB/mg algae) and t is time (d). An average AE was calculated as the slope of a plot of P_t against the total ingested PCB ($P_a \text{ IR } t$) for all dilution rates and food concentrations.

To obtain values per mussel, shell-free tissues from four mussels were pooled, freeze-dried and weighed before analysis. From the analytical data, the value per mussel was calculated based on the average tissue weight of $38 (\pm 2)$ mg.

2.4. PCB extraction and analysis

The amounts of PCBs in the liquid phase, algal cells and biodeposits were determined by gas chromatography. Algae and biodeposit samples were filtered through pre-washed GF/F filters. The filtrate was used to measure the amount of PCB in the aqueous phase. The filter was sliced and then mixed with 2 g of Celite 521 (Aldrich Chemical, Milwaukee, WI, USA). For extraction of PCBs from mussel tissue, the mussels were frozen, and the shells were removed. The shell-free tissue from four mussels was pooled into one sample, freeze-dried, and weighed before analysis. The tissue or shells were mixed with Celite 521, and put into an Accelerated Solvent Extractor (ASE; Dionex, Sunnyvale, CA, USA) cell. 2,5,2',5'-CBP (IUPAC No. 52) was added into all

samples before sample extraction to assure extraction efficiency and to serve as an internal standard.

PCBs were extracted using an ASE system [19,23]. Extraction was done with hexane for 15 min operating at 100°C and 2000 psi. The hexane extracts were treated with an equal volume of sulfuric acid for fat removal, and cleaned up on a 4% deactivated Florisil (60–100 mesh; Sigma Chemical, St. Louis, MO, USA) column. The eluent was concentrated in Rapid Vap N₂ system (Labconco, Kansas City, MO, USA).

To measure PCBs in the aqueous phase, 10 mL hexane was added into 1-L of filtrate in a separatory funnel. 2,5,2',5'-CBP was added to serve as an internal standard. The mixture was then shaken for 18 h. After phase separation, the solvent phase was removed, and the water phase was extracted again in the same manner. The hexane was then combined with the previous extract. The combined fraction was treated with anhydrous sodium sulfate and purified using a Florisil column. The extraction efficiency of the internal standard was greater than 91%.

PCB analysis was performed on a Hewlett Packard 5890 gas chromatograph equipped with a ⁶³Ni electron-capture detector, Apiezon-L column (Restek, Bellefonte, PA, USA), and a computerized data acquisition system (ChromPerfect, Justice Innovations, Mountain View, CA, USA) as previously described [19]. PCBs were quantitated with a calibration standard containing single congener standards (AccuStandard, 99% purity).

2.5. Statistical analysis

Differences in algal clearance rate or IR were determined by comparing regression coefficients by the *F*-test [24]. Any difference between 2,5,2'- and 2,4,2',4'-CBPs was tested by the paired *t*-test using the SigmaStat program (SPSS, Chicago, IL, USA).

3. Results

3.1. Clearance rate

Clearance rate was determined by measuring algal cells contaminated with 2,5,2'- or 2,4,2',4'-CBPs according to Eq. (1). At a constant dilution rate (*D*), the CR decreased with the concentration of algae (Fig. 2a). No difference in CR was seen between the two congeners (*F*-test, *p* > 0.05). When regression was made between CR and food concentration (*C_f*) using combined data points for both congeners, the slope was significantly different from 0 (linear regression, *p* < 0.01), with the rate decreasing as *C_f* increased. On the other hand, when *D* was increased while *C_f* was maintained constant (3 mg algae/L), the CR showed a linear increase (Fig. 2b). The different trends with these two variables remained even when the CR was plotted against the rate of daily food supply which was calculated as the product of *C_f* and *D* (mg algae/L/d) (Fig. 2c). These results indicate that the variation in CR is a function of both *C_f* and *D*, and is not related to the food supply rate.

3.2. Ingestion rate

Unlike the CR, the IR increased with *C_f* as with *D*. As expected, no difference was found between the two congeners (*F*-test, *p* > 0.05; Fig. 3). At a constant *D*, the IR was a linear function of *C_f*. At a constant *C_f*, the IR also increased linearly with *D*. When IR was plotted against food supply rate, an identical linear relationship was found (linear regression, *R*² > 0.96) regardless of whether the variable was *C_f* or *D* unlike CR (Fig. 3c). Therefore, the underlying mechanism of the *D*- or *C_f*-dependent variation of the IR appeared to be variation in the food supply rate.

The amount of ingested food (and thus, congeners) increased linearly with total amount of food (or

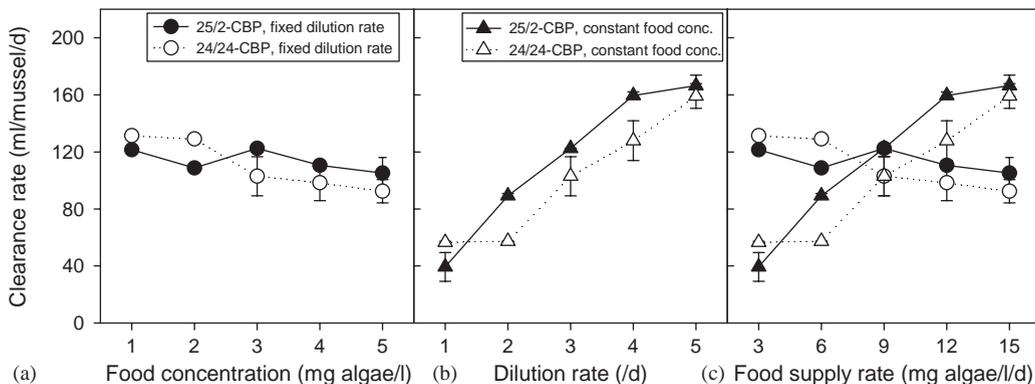


Fig. 2. Clearance rate (CR) as affected by algal food concentration, dilution rate, and food supply rate. CR vs. food concentrations at a constant dilution rate (3/d) (a); CR vs. dilution rates at a constant food concentration (3 mg algae/L) (b); and CR vs. the food supply rate (food concentration × dilution rate) (c). The mussel value was calculated based on a tissue weight of 38 mg.

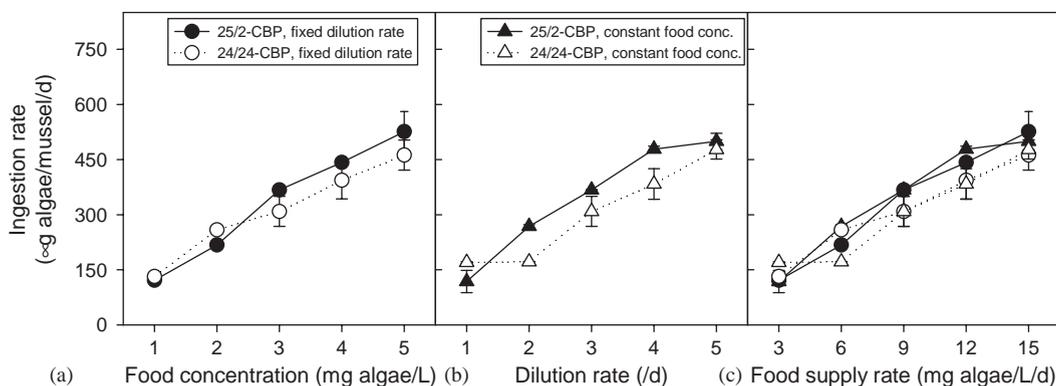


Fig. 3. Ingestion rate (IR) as affected by algal food concentration, dilution rate, and food supply rate. IR vs. algal food concentrations at a constant dilution rate (3/d) (a); dilution rates at a constant food concentration (3 mg algae/L) (b), and IR vs. food supply rates (food concentration \times dilution rate) (c).

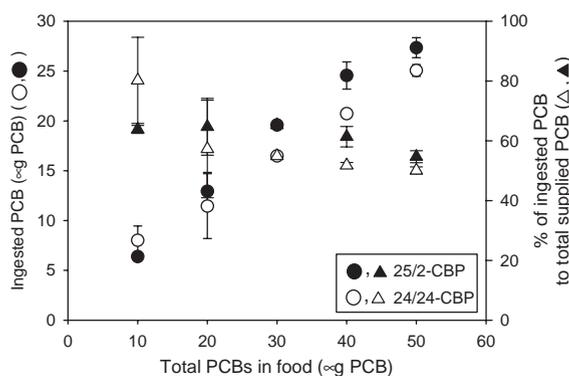


Fig. 4. Relationship between the amount of PCB ingested and total amount of PCB supplied. Ingested PCBs were determined as the difference between the total in the food reservoir and the residual amount in the overflow.

congeners) supplied, although PCB ingestion as a percent of the total PCB provided decreased (Fig. 4). There was no difference in this linear trend between 2,5,2'- and 2,4,2',4'-CBP (F -test, $p > 0.05$).

3.3. Bioaccumulation and assimilation efficiency

PCBs in the food supply were mostly associated with algae, since their concentration in the aqueous phase was less than 1% of the total. Others also showed that bioaccumulated PCBs mostly came from food particles [25]. Therefore, we used the total concentration, without differentiating between soluble and particulate PCBs, when we calculated bioaccumulation in mussel tissues.

The bioaccumulation of congeners in zebra mussel tissues increased linearly with both C_f and D . It also increased with food supply rate whether the supply rate was varied through D or C_f (Fig. 5). Significantly, the level of bioaccumulation was higher for 2,4,2',4'-CBP

than for 2,5,2'-CBP (paired t -test, $p < 0.05$) (Fig. 5c), probably reflecting the higher K_{ow} of the tetrachlorobiphenyl (see below). The bioaccumulation of congeners in the mussel tissue increased with total amount of food ingested; here again, the level of bioaccumulation was significantly higher for the tetrachlorobiphenyl (Table 1).

When PCB concentrations in mussel tissues were plotted against the concentrations of the total ingested PCBs, the relationship was linear for both congeners (Fig. 6a). The regression slope of this plot represents AE, and its value for the tetrachlorobiphenyl (0.20 ± 0.03) was significantly higher (F -test, $p < 0.05$) than that for the trichlorobiphenyl (0.13 ± 0.01).

3.4. Biodeposits

Biodeposits (and therefore, biodeposit-associated PCBs) increased as C_f and D increased (Figs. 7a and b). They also increased with food supply rate (Fig. 7c). They were also higher at higher values of IR. When plotted against the total food (or PCB) ingested, biodeposits showed a linear increase (Fig. 6b). It is interesting that the congener concentration in biodeposits showed the opposite of that found in the mussel tissue and that the biodeposit level of less hydrophobic 2,5,2'-CBP was significantly greater than that of more hydrophobic 2,4,2',4'-CBP.

3.5. PCB distribution

The ingested PCBs ranged from $\sim 46\%$ to 90% of the total supplied PCBs. The total average recovery of 2,5,2'-CBP from mussel tissue, biodeposit, and shell was, on average $86 (\pm 10)\%$ of the calculated total ingestion and the average recovery of 2,4,2',4'-CBP was $80 (\pm 7)\%$ (Table 1). The discrepancy between the ingested total and the sum of recovered PCBs was probably due

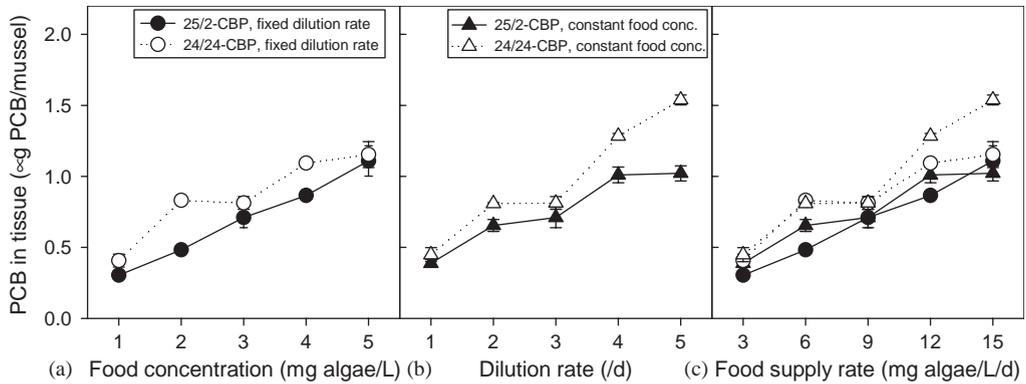


Fig. 5. PCBs in mussel tissues as affected by algal food concentration, dilution rate and food supply rate. Tissue PCB vs. algal food concentrations at a constant dilution rate (3/d) (a); tissue PCB vs. dilution rates at a constant food concentration (3 mg algae/L) (b); and tissue PCB vs. food supply rate (food concentration \times dilution rate) (c).

Table 1
Distribution of 2,5,2'- and 2,4,2',4'-chlorobiphenyl (CBP) in zebra mussels

Congener	Dilution rate (/d)	Food conc. (mg algae/L)	Tissue ^a (%)	Biodeposit ^a (%)	Shell ^a (%)	Total recovery ^a (%)	Total recovered PCB ^b ($\mu\text{g PCB/mussel}$)
2,5,2'-CBP	3	1	18.8 (1.2) ^c	56.1 (1.0)	6.6 (0.3)	81.5 (2.0)	1.32 (0.04)
		2	16.6 (1.3)	59.3 (0.7)	5.3 (0.2)	81.2 (1.5)	2.36 (0.06)
		3	14.5 (1.5)	59.1 (5.0)	4.9 (0.4)	78.5 (6.7)	3.85 (0.33)
		4	14.7 (0.1)	72.3 (0.5)	6.5 (0.1)	93.5 (0.8)	5.51 (0.05)
		5	15.8 (1.5)	76.1 (7.5)	5.1 (0.6)	97.0 (6.6)	6.80 (0.67)
	3	1	24.6 (1.7)	55.5 (3.9)	1.5 (0.2)	81.6 (4.7)	1.28 (0.09)
		2	18.3 (1.2)	46.7 (3.0)	3.5 (0.4)	68.6 (5.3)	2.45 (0.16)
		3	14.5 (1.5)	59.1 (5.0)	4.9 (0.1)	78.5 (5.6)	3.85 (0.33)
		4	15.8 (0.9)	77.1 (0.5)	5.5 (0.5)	98.5 (2.0)	6.28 (0.12)
	3	1	23.2 (2.6)	54.4 (1.1)	2.4 (0.2)	80.0 (4.7)	1.40 (0.07)
		2	24.2 (0.0)	45.4 (0.9)	6.5 (0.4)	76.0 (3.3)	2.62 (0.05)
		3	19.7 (1.1)	45.0 (1.9)	7.1 (0.2)	71.8 (4.6)	2.96 (0.13)
4		20.9 (0.3)	56.4 (0.8)	6.5 (0.3)	83.8 (1.4)	4.40 (0.07)	
2,4,2',4'-CBP	3	5	18.7 (1.5)	56.4 (7.5)	6.7 (0.3)	81.9 (8.1)	5.04 (0.57)
		1	19.8 (2.2)	47.2 (1.6)	3.7 (0.1)	70.7 (5.7)	1.60 (0.09)
		2	35.4 (0.3)	48.7 (2.3)	4.0 (0.2)	88.0 (3.6)	2.01 (0.06)
		3	19.7 (1.1)	45.0 (1.9)	7.1 (0.2)	71.8 (5.1)	2.96 (0.13)
		4	25.1 (0.4)	59.1 (3.5)	4.7 (1.6)	88.9 (3.7)	4.55 (0.28)
5	24.2 (0.5)	58.0 (9.1)	4.3 (0.3)	86.4 (6.8)	5.50 (0.63)		

^a% of the total ingested PCB.

^bSum of PCBs in tissues, biodeposits, and shells after a 4-day experimental period.

^cNumbers in parentheses are SD.

mostly to problems associated with a complete recovery of biodeposits. Biodeposits are generally loose pellets and are difficult to recover quantitatively. Therefore, values in the biodeposits were most likely underestimated. The total recovery appeared to depend largely on the recovery of these loose pellets, since when the total recovery was plotted against the recovery in biodeposits,

the relationship was linear (linear regression, $p < 0.001$) (not shown).

For both congeners, the percent distribution in each compartment did not exhibit any discernable trend with respect to D or C_f (Table 1). The highest proportion of the ingested congeners was found in biodeposits (~ 45 – 78%), followed by tissues (~ 15 – 35%) (Table 1). When

the level of 2,5,2'-CBP in biodeposits was compared to that of 2,4,2',4'-CBP using an average value of pooled data at all values of C_f and D , the concentration of this less hydrophobic congener was significantly higher (paired t -test, $p < 0.05$) than that of more hydrophobic

tetrachlorobiphenyl ($64 \pm 11\%$ vs. $52 \pm 6\%$). However, the level in mussel tissues showed the opposite (paired t -test, $p < 0.05$), with the level of 2,4,2',4'-CBP ($23 \pm 5\%$) being significantly higher than that of 2,5,2'-CBP ($17 \pm 3\%$). The value recovered from shells was not significantly different between the two congeners (5.0 ± 1.5 and 5.3 ± 1.7).

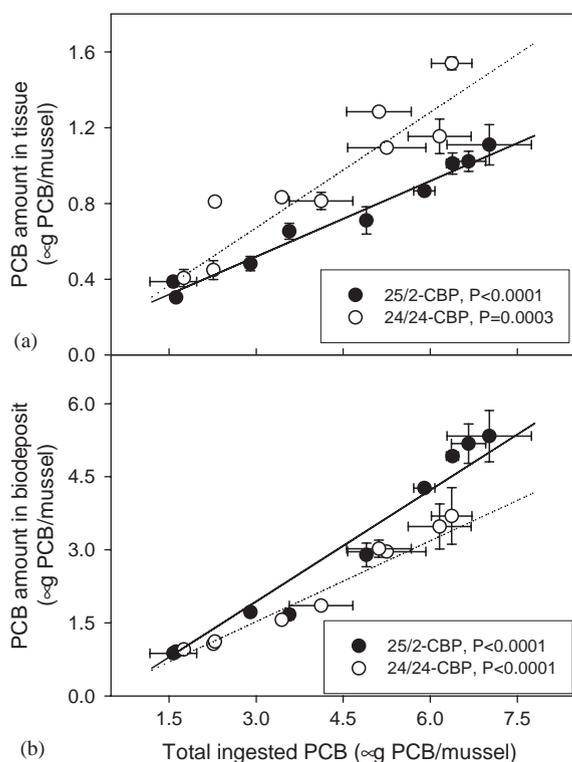


Fig. 6. Correlation between total ingested PCB and PCBs in mussel tissues (a) and PCBs in biodeposits (b). The total ingested amount of PCBs was integrated values for a 4-day experimental period. The regression slope in Fig. 6(a) represents assimilation efficiency.

4. Discussion

The bioaccumulation of hydrophobic compounds, either through direct partitioning or food ingestion, depends on the K_{ow} ; higher the value, greater is the bioaccumulation. The $\log-K_{ow}$ values of PCB congeners ranges from an average of 4.5 for mono-chlorinated biphenyls to 8.1 for hepta-chlorobiphenyls [20]. A model prediction suggests that in PCB bioaccumulation in fishes in the Great Lakes, the contribution of direct partitioning from the aqueous phase increases with decreasing K_{ow} while the contribution of food ingestion increases with increasing K_{ow} [26]. The relative contribution of each exposure route to the total bioaccumulation in mussel tissue is influenced largely by the concentration in the aqueous phase and food particles [11].

Earlier studies of zebra mussels [27–29] and blue mussels [11] demonstrated a negative correlation between clearance rate and the concentration of suspended particles as found in the present investigations. However, the present study was unable to determine whether the correlation is exponential as reported earlier because of the relatively narrow concentration range that we tested.

Previously reported clearance rates ranged from 2.3 to 3.2 L/mussel/d [7]. These values are higher than 0.04 to 0.17 L/mussel/d we found in the present study. However, it is difficult to compare the two studies, because Roditi

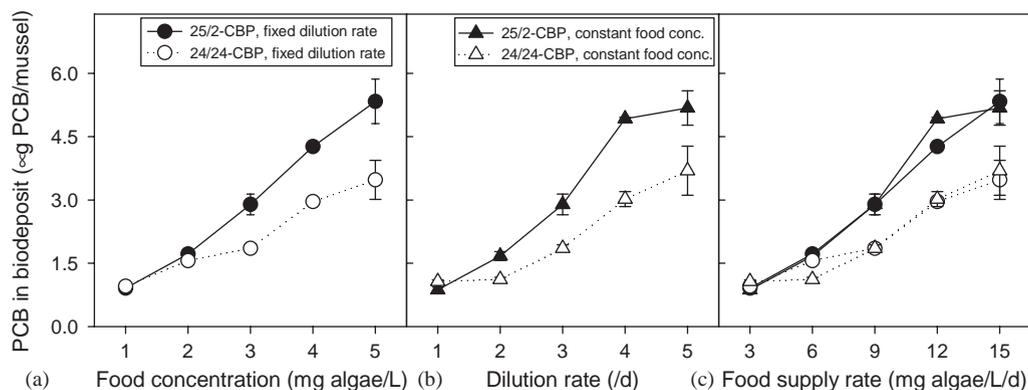


Fig. 7. PCBs in biodeposits as a function of algal food concentration, dilution rate, and food supply rate. PCBs in biodeposits vs. algal food concentrations at a constant dilution rate (3/d) (a); PCBs in biodeposits vs. dilution rates at a constant food concentration (3 mg algae/L) (b); and PCBs in biodeposits vs. food supply rates (food concentration \times dilution rate) (c).

et al. [7] used a recirculating system whereas we used a flow-through system. As the present study shows, CR is a positive function of flow rate, but at the same time negatively correlated to food concentrations. In a recirculating system, the food concentration would continuously decrease over time. It is also known that CR varied with the size of mussels, its value generally higher in smaller ones [30]. Mussels used in our study were bigger than those employed by Roditi et al. [7]. Considering the differences in the experimental system and the mussel size, the difference in CR values does not appear inordinate. In a natural lake, CR ranged from 0.3 to 1.9 L/mussel/d [29].

Both the IR and congener concentrations in mussel tissues were the same linear function of food supply rate, regardless of whether the supply rate was varied by changing C_f or D . These results clearly indicate that it is not higher values of C_f or D per se, but rather higher food supply rates that are responsible for higher values of IR and the higher bioaccumulation. Maden et al. [22] also reported that, at lower flow rates and food concentrations, the IR was lower and resulted in lower bioaccumulation of PCBs.

The bioconcentration of PCB appears to be non-linear over a long period of time [31,32]. When blue mussels were transplanted to a heavily contaminated area, the tissue PCB concentration increased linearly during the initial 40 days and leveled off thereafter [31]. These results suggest that AE may decrease after an extended period of exposure.

The bioaccumulation of congener 2,4,2',4'-CBP was greater than that of the less hydrophobic 2,5,2'-CBP. A regression of bioconcentration data against total ingested food showed a significantly higher elevation for 2,4,2',4'-CBP (Fig. 6a). This probably reflects the difference in the $\log-K_{ow}$ with the value higher for 2,4,2',4'-CBP (6.29) than for 2,5,2'-CBP (5.55) [20]. AE was also higher for the tetrachlorobiphenyl, probably for the same reason, although Bruner et al. [12] were unable to correlate it to the K_{ow} values of hydrophobic compounds.

Zebra mussels filter a wide range of particle types and sizes [5–7]. In the present study, ~46–90% of the total food was ingested, as estimated by the amount of recovered congeners, depending on the food supply rate. The highest proportion of congeners was found in biodeposits, followed by tissues, and lastly shells. The quantity of biodeposits (and the amount of PCB congeners in them) increased with filtration rate. It also increased with total amount of food ingested, increasing from 1.9 to 5.1 times the bioconcentrated amount in tissue, as the total amount of ingested food increased. The congener concentration in biodeposits indicates that 45–78% of PCBs in the ingested food is being redeposited in fecal pellets and pseudofeces. These results indicate that zebra mussels can have potentially

significant impact on the PCB dynamics in the Hudson River through their redeposition of PCBs in suspended particulates, which otherwise would have escaped the system with river flow. This redeposition is also likely to significantly increase the food-chain transfer of PCBs, because fecal pellets and pseudofeces constituted a major food resource for benthic organisms [2]. Bruner et al. [12] reported that the AE of PCBs by the benthic invertebrate *Gammarus fasciatus* was very high, at between 80–90% of tetra- and hexa-chlorobiphenyls in biodeposits.

5. Conclusions

Zebra mussels ingested approximately 46–90% of PCB-contaminated algal cells, depending on the supply rate. Of the total ingested food (therefore, PCBs), ~45–78% was found in biodeposits and 15–35% was recovered in mussel tissues.

When an average value of biodeposits at all values of C_f and D was compared between the 2,5,2'-CBP and 2,4,2',4'-CBP, the concentration of less hydrophobic trichlorobiphenyl ($64 \pm 11\%$) was significantly higher than that of more hydrophobic tetrachlorobiphenyl ($52 \pm 6\%$), whereas it was the opposite in mussel tissues, with the level of 2,4,2',4'-CBP ($23 \pm 5\%$) being significantly higher than that of 2,5,2'-CBP ($17 \pm 3\%$).

Since zebra mussels filter a wide range of particles they can clearly increase not only the residence time of PCBs in the Hudson River through redeposition to sediments, but also the transmission of PCBs to food chain organisms, through exposure to benthic organisms as well as significant bioconcentration in mussel tissues.

Acknowledgements

This work was supported by a grant from the Hudson River Foundation (017/00A). We thank Jennifer L. Pikor for technical assistance.

References

- [1] Sanders JE. The PCB-pollution problem in the upper Hudson River: from environmental disaster to "environmental gridlock". *Northeastern Environ Sci* 1989;8(1): 1–86.
- [2] Strayer DL, Caraco NF, Cole JJ, Findlay SEG, Pace ML. Transformation of freshwater ecosystems by bivalves—A case study of zebra mussels in the Hudson River. *Bioscience* 1999;49(1):19–27.
- [3] Caraco NF, Cole JJ, Raymond PA, Strayer DL, Pace ML, Findlay SEG, Fischer DT. Zebra mussel invasion in a large, turbid river—phytoplankton response to increased grazing. *Ecology* 1997;78(2):588–602.

- [4] Smith TE, Stevenson RJ, Caraco NF, Cole JJ. Changes in phytoplankton community structure during the zebra mussel (*Dreissena polymorpha*) invasion of the Hudson River (New York). *J Plankton Res* 1998;20(8):1567–79.
- [5] Bastviken DTE, Caraco NF, Cole JJ. Experimental measurements of zebra mussel (*Dreissena polymorpha*) impacts on phytoplankton community composition. *Freshwater Biol* 1998;39(2):375–86.
- [6] Lei J, Payne BS, Wang SY. Filtration dynamics of the zebra mussel, *Dreissena polymorpha*. *Can J Fish Aquat Sci* 1996;53(1):29–37.
- [7] Roditi HA, Caraco NF, Cole JJ, Strayer DL. Filtration of Hudson River water by the zebra mussel (*Dreissena polymorpha*). *Estuaries* 1996;19(4):824–32.
- [8] US Environmental Protection Agency. Phase 2 report—further site characterization and analysis volume 2C—data evaluation and interpretation report Hudson River PCBs reassessment RI/FS. New York, NY: US EPA; 1997.
- [9] Roditi HA, Strayer DL, Findlay SEG. Characteristics of zebra mussel (*Dreissena polymorpha*) biodeposits in a tidal freshwater estuary. *Arch Hydrobiol* 1997;140(2):207–19.
- [10] Björk M, Gilek M. Bioaccumulation kinetics of PCB 31, 49, and 153 in the blue mussel, *Mytilus edulis* L. as a function of algal food concentration. *Aquat Toxicol* 1997;38(1–3):101–23.
- [11] Björk M, Gilek M. Efficiencies of polychlorinated biphenyl assimilation from water and algal food by the blue mussel (*Mytilus edulis*). *Environ Toxicol Chem* 1999;18(4):765–71.
- [12] Bruner KA, Fisher SW, Landrum PF. The role of the zebra mussel, *Dreissena polymorpha*, in contaminant cycling: II. zebra mussel contaminant accumulation from algae and suspended particles and transfer to the benthic invertebrate, *Gammarus fasciatus*. *J Great Lakes Res* 1994;20(4):735–50.
- [13] Hendriks AJ, Pieters H, Deboer J. Accumulation of metals, polycyclic (halogenated) aromatic hydrocarbons, and biocides in zebra mussel and eel from the Rhine and Meuse Rivers. *Environ Toxicol Chem* 1998;17(10):1885–98.
- [14] Roper JM, Cherry DS, Simmers JW, Tatem HE. Bioaccumulation of toxicants in the zebra mussel, *Dreissena polymorpha*, at the Times Beach Confined Disposal Facility, Buffalo, New York. *Environ Pollut* 1996;94(2):117–29.
- [15] Björk M, Gilek M, Kautsky N, Näf C. In situ determination of PCB biodeposition by *Mytilus edulis* in a Baltic coastal ecosystem. *Mar Ecol Prog Ser* 2000;194:193–201.
- [16] Wright DA, Setzlerhamilton EM, Magee JA, Harvey HR. Laboratory culture of zebra (*Dreissena polymorpha*) and quagga (*D. bugensis*) mussel larvae using estuarine algae. *J Great Lakes Res* 1996;22(1):46–54.
- [17] Nichols HW. Growth media—fresh water. In: Stein JR, editor. *Handbook of phycological methods*. New York, NY: Cambridge University Press; 1973. p. 7–24.
- [18] Cho YC, Kwon OS, Sokol RC, Bethoney CM, Rhee GY. Microbial PCB dechlorination in dredged sediments and the effect of moisture. *Chemosphere* 2001;43:1119–26.
- [19] Rhee G-Y, Sokol RC, Bethoney CM, Cho Y-C, Frohnhoefer RC, Erkkila T. Kinetics of polychlorinated biphenyl dechlorination and growth of dechlorinating microorganisms. *Environ Toxicol Chem* 2001;20(4):721–6.
- [20] Rapaport RA, Eisenreich SJ. Chromatographic determination of octanol–water partition coefficients (K_{ow} 's) for 58 polychlorinated biphenyl congeners. *Environ Sci Technol* 1984;18:163–70.
- [21] Lederman TC, Rhee G-Y. Bioconcentration of a hexachlorobiphenyl in great lakes planktonic algae. *Can J Fish Aquat Sci* 1982;39(3):380–7.
- [22] Madon SP, Schneider DW, Stoeckel JA, Sparks RE. Effects of inorganic sediment and food concentrations on energetic processes of the zebra mussel, *Dreissena polymorpha*—implications for growth in turbid rivers. *Can J Fish Aquat Sci* 1998;55(2):401–13.
- [23] Dionex. Extraction of lipids, polychlorinated biphenyls from fish tissue in a single run using accelerated solvent extraction (ASE). Application Note 337. Sunnyvale, CA: Dionex Corporation; 1999.
- [24] Sokal RR, Rohlf FJ. *Biometry: the principles and practice of statistics in biological research*. 3rd ed., San Francisco, CA: W.H. Freeman and Company; 1995. 887pp.
- [25] Gilek M, Björk M, Broman D, Kautsky N, Kautsky U, Näf C. The role of the blue mussels, *Mytilus edulis*, in the cycling of hydrophobic organic contaminants in the Baltic proper. *Ambio* 1997;26:202–9.
- [26] Thomann RV, Connolly JP. Model of PCB in the Lake Michigan lake trout food chain. *Environ Sci Technol* 1984;18(2):65–71.
- [27] Berg DJ, Fisher SW, Landrum PF. Clearance and processing of algal particles by zebra mussels (*Dreissena polymorpha*). *J Great Lakes Res* 1996;22(3):779–88.
- [28] Fanslow DL, Nalepa TF, Lang GA. Filtration rates of the zebra mussels (*Dreissena polymorpha*) on natural seston from Saginaw Bay, Lake Huron. *J Great Lakes Res* 1995;21(4):489–500.
- [29] Reeders HH, de Vaate AB, Slim FJ. The filtration rate of *Dreissena polymorpha* (Bivalvia) in three Dutch lakes with reference to biological water quality management. *Freshwater Biol* 1989;22:133–41.
- [30] Bruner KA, Fisher SW, Landrum PF. The role of the zebra mussel, *Dreissena polymorpha*, in contaminant cycling: I. The effect of body size and lipid content on the bioconcentration of PCBs and PAHs. *J Great Lakes Res* 1994;20(4):725–34.
- [31] Voie ØA, Johnsen A, Rossland HK. Why biota still accumulate high levels of PCBs after removal of PCB contaminated sediments in a Norwegian fjord. *Chemosphere* 2002;46:1367–72.
- [32] Yu KN, Lam PKS, Cheung CCC, Yip CWY. Mathematical modeling of PCB bioaccumulation in *Perna viridis*. *Mar Pollut Bull* 2002;45:332–8.