Accumulation of organochlorinated pesticides by triolein-containing semipermeable membrane device (triolein-SPMD) and rainbow trout

Yibing Lu, Zijian Wang*

*State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences,
Chinese Academy of Sciences, Beijing 100085, China*

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Abstract

An important advance in aquatic ecotoxicology is the development and application of biomimetic sampling technology for hydrophobic contaminants. In this paper, accumulation kinetics of five organochlorinated pesticides, i.e. hexachlorobenzene, lindan, aldrin, heptachlor epoxide and 4,4'-DDT, by triolein-containing semipermeable membrane device (triolein-SPMD) and by fish (rainbow trout) were compared in a laboratory continuous flow system. Accumulation kinetics of organochlorinated pesticides by the triolein-SPMD and by rainbow trout were linear during the exposure period of 20 d, except for lindan. Approach of an asymptote could be observed for lindan at the end of the exposure for rainbow trout. When the kinetic data were used to calculate the first-order uptake rate constants, it was found that the uptake rates of the chemicals in triolein-SPMD were 1–2.5 times higher than those in the fish. The estimated bioconcentration factors of the five pesticides in rainbow trout were in the range of 1000–7000 and the device concentration factor in the range of 1500–18 000. Although equilibrium was not reached for most pesticides under 21-d exposure period, correlations between C_L/C_w and K_{ow} ($R^2 = 0.887$) and C_f/C_w and K_{ow} ($R^2 = 0.931$) could be observed which was a clear indication that triolein-SPMD accumulates chlorinated pesticides in a quite similar way as fish do. Our results show that triolein-SPMD could serve as a good surrogate for rainbow trout for simulating accumulations of chlorinated pesticides and may be used as a universal surrogate for fish in natural waters.

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1. Introduction

There is a clear need for rapid, easy to use, effective and low-cost integrative methods that allow not only the direct monitoring of the fate and concentrations of trace level of chemical-contaminant mixture in the environment, but also assessment of their hazards to aquatic organisms and human health. Membrane-based passive samplers seem to be a promising tool for the time-integrated monitoring of hydrophobic pollutants in

aquatic ecosystems. In these devices the uptake of chemicals is based on the process of passive partitioning of a compound between water and a lipophilic solvent enclosed in a semipermeable polymeric membrane. Thus, the passive samplers can be used as indicators of bioavailability of chemical pollutants. Furthermore, in contrast to most living organisms, they can be exposed to harsh environmental conditions for long time periods and still remain operative [1]. Among the several designs of membrane samplers that have been used, triolein-containing semi-permeable membrane devices (triolein-SPMD), proposed by Huckins et al. [2], consisting of a thin film of neutral lipid, enclosed in thin-walled layflat tubing, made of low-density polyethylene, proved to be

*Corresponding author. Tel.: +86-10-628-49-1-40; fax: +86-10-629-23-5-43.

E-mail address: wangzj@mail.rcees.ac.cn (Z. Wang).

most effective in their capacity to accumulate lipophilic substances. The molecular size-exclusion limit of the polyethylene membrane is similar to that of biological membranes while triolein, usually used in triolein-SPMD, constitutes a significant fraction of the lipid pool of most aquatic organisms. Triolein-SPMD has received wide recognition as an effective tool for environmental research and analysis. It has been successfully used for monitoring chemicals in aquatic environments [2–8], and in air [9,10]. Triolein-SPMD has also been used for screening and preliminary identification of emission sources in wastewater systems and groundwater [11–14], and appears to simulate the bioconcentration of dissolved chemicals by aquatic organisms [15–21]. The procedure for dialytic recovery of pesticides can be used separately to remove lipids and other interferences from environmental sample extracts [22,23].

Studies have been carried out on the comparisons of accumulations of a variety of organic contaminants by triolein-SPMD and by several types of aquatic organisms, but different results were obtained. In the present work, Five chlorinated pesticides were exposed to triolein-SPMD and rainbow trout, which is a model fish for standard tests for chemicals [24]. We report the uptake kinetics and bioconcentration factors (BCFs) of five chlorinated pesticides in rainbow trout and in triolein-SPMD and the similarity and differences between the two sampling approaches to examine how well triolein-SPMD mimics rainbow trout in terms of discriminatory uptake of hydrophobic chemicals and further explore the possibility of using triolein-SPMD to concentrate bioavailable environmental contaminants.

2. Materials and methods

2.1. Test chemicals and reagents

The pesticides-grade lindan (99.0% purity), heptachlor epoxide (99.0% purity), aldrin (99.9% purity) and hexachlorobenzene (99.0% purity) were purchased from the Institute of Organic Industrial Chemistry (Warsaw, Poland). 4,4'-DDT (99% purity) was purchased from Sigma Chemical Company (St. Louis, MO, USA). Organic solvents employed in the experiment, such as cyclohexane (C_6H_{12}), petroleum ether (boiling point range is 60–90°C), acetone (CH_3COCH_3), analytical grade ethanol (Beijing Chem. Co.) were distilled by all-glass systems before use.

2.2. Semipermeable membrane devices

Triolein-SPMD was prepared in a similar way as described by Huckins et al. [2]. Layflat polyethylene tubing (width 3.0 cm, membrane thickness ranged from 50 to 55 μm , from Beijing Plastics company) was cut into

pieces of 23 cm in length, soaked in cyclohexane for 24 h to remove trace contaminants, and dried with ultra-pure nitrogen. The total mass of tubing was 1.45 g, and the ratio of mass of tubing to volume of solvent was 0.01 g/ml. Then 250 μl (227.5 mg) of triolein was coated onto the polyethylene tubing before the ends were sealed with a plastic sealer (Model SF-300, China). The ratio of triolein-SPMD surface area to triolein volume is 400 cm^2/ml . The tube was formed into twisted loops and glass sticks were used as anchors to keep the device in a vertical position when suspended in the test solution.

2.3. Rainbow trout

The rainbow trout were obtained from Shuntong Aquatic Institute in Huairou, Beijing. The average length and weight of the fish were 9.4 ± 0.6 cm and 8.8 ± 1.5 g ($n = 20$), respectively. The average lipid content calculated on wet weight was $10.0 \pm 2.1\%$. The fish were sterilized and reared under laboratory condition for 2 weeks before experiment. The death rate during the domestication was lower than 2%.

2.4. Exposure experiment

The experiment was conducted in a glass exposure chamber (volume 70 l). A set of 21 triolein-SPMDs and 25 fish was placed in the exposure chamber, which was connected to a continuous-flow system. The flow rate through the chamber was maintained at about 300 ml/min (the flow rate through the test chambers was 5.76 volume additions per 24 h). Concentrated solution of the five pesticides in ethanol (concentration ratio 1:1:1:1:1) was supplied into the main flow up the exposure chamber by a peristaltic pump. The rate of the peristaltic pump was adjusted to obtain concentrations of the pesticides of about 2 $\mu g/l$, with a negligible concentration of ethanol (less than 0.1 ml/l). The water temperature was maintained at 20°C. The experiment lasted for 20 d, during which the rainbow trout received no food. On days 0.5, 1, 2, 4, 8, 14 and 20, three water samples of each 50 ml, three triolein-SPMDs and three fish were taken and processed to determine concentrations of the pesticides. The rainbow trout were stored in glass jars at –20°C until analysis. Control triolein-SPMD (three devices) and rainbow trout (three individuals) were taken at the beginning of the experiment, just before the connection of the exposure chamber to the continuous-flow system.

3. Analytical methods

3.1. Water

The pesticides were spiked with known amounts of surrogate standard before extraction with 20 ml aliquots

of petroleum ether. The extract was reduced in volume to 0.5 ml using nitrogen blow-down technique. Aliquots of the extract were spiked with the instrumental internal standard and analyzed using a gas chromatograph (HP-5890 Series II) equipped with a ^{63}Ni electron capture detector (GC-ECD) and a HP-5 fused silica capillary column (30 m $\times$ 0.32 mm i.d., film thickness of 0.17 μm). The temperatures of the injector and of the detector were kept at 250°C and 260°C, respectively. Column temperature was programmed at 85°C increasing at 10°C/min to 180°C, and then increasing at 20°C/min to 280°C, and a held for 30 min at 280°C. Nitrogen gas of extra purity was used as a carrier gas. The flow rate was 1 ml/min and the pressure was kept at 20 psi.

3.2. Semipermeable membrane devices

After sampling, triolein-SPMD was first rinsed under running tap water. After soft papers drying, they were transferred into glass jars with ground edge caps. Cyclohexane (50 ml) was added, and the membranes were dialyzed for 48 h at 20°C. Five milliliter of the dialysates was taken up and was reduced to about 2 ml using rotary evaporation and then reduced to 1 ml by applying a gentle stream of high-pure nitrogen gas. GC-ECD was used for analysis at the same conditions as analysis of water extracts.

3.3. Rainbow trout

The rainbow trout were thawed at room temperature for several hours. After length and wet weight determination, each fish was sawed into small pieces, ground with a meat grinder, and then blended with anhydrous Na_2SO_4 (baked at 475°C prior to use). The mixture was packed into Soxhlet extractor and then was extracted with mixture of cyclohexane, acetone and petroleum ether (1:1:1 = v:v:v) for 24 h. The extract was divided into two parts. One part (50% of the total) was concentrated by rotation evaporator to about 1 ml then was passed through a silica gel column (60–80 mesh, activated at $180 \pm 2^\circ\text{C}$ for 12 h before use) and then a glass-column (10 mm I. D) filled with 12 cm of silica gel. The column was eluted with 50 ml cyclohexane. After

concentrating to a final volume of 1 ml using a gentle stream of extra purity nitrogen and adding in the internal standards, 1 μl of the purified extract was injected into GC/ECD in splitless mode to analyze the concentrations of pesticides in the same way as water samples analysis procedure. The other part was used to measure the lipid content, using the method described previously [2]. Analytical procedure was the same as that for water extract.

4. Quality control

For quality control of gas chromatographic conditions, a checkout procedure was performed before sample analysis. In the procedure, a standard mixture with DDT content of 15% was used and the calibration was carried out when the percentage of DDT in standard mixture deviated significantly from 15%.

One triolein-SPMD blank (a freshly prepared triolein-SPMD) and two reagent blanks (100 ml of cyclohexane and 100 ml of dichloromethane, blow down to 1 ml, respectively) were used for the entire clean up and enrichment procedure. No chemicals of concern were found in these blanks. 2,4,5,6-tetrachloro-*m*-xylene (TMX) and decachlorobiphenyl (PCB209) were used as surrogate and 4,4'-dichlorobiphenyl (PCB15) was used as an internal standard.

To 1 g of fish or 1 g of triolein-SPMD, 1 μg of each organochlorinated pesticide was spiked before extraction. Three replicates were used. The recoveries of all the pesticides were in the range of 76–102% with variation coefficients not exceeding 15%. There were no significant differences between spiking levels and recoveries, and the concentration estimates were not corrected for these recoveries.

5. Results and discussion

5.1. Uptake kinetics

Properties of the five pesticides are shown in Table 1. The time-dependent bioconcentration processes of

Table 1
Selected physicochemical properties of the test organochlorinated pesticides and their concentrations in water

Compound	Molecular weight	Water solubility $\mu\text{g/l}$	Log K_{ow} ^a	Concentration in water ($\mu\text{g/l}$, mean + SD) ^b
Lindan	291	7800	3.9	2.36 ± 1.70
Aldrin	365	180	5.3	1.31 ± 0.69
Heptachlor epoxide	389.2	350	5.4	1.52 ± 0.53
4,4'-DDT	354.5	5.5	5.7	1.14 ± 1.20
Hexachlorobenzene	284.8	—	6.2	1.07 ± 0.99

^a Cited from [22].

^b Mean value of all samples taken over the entire exposure period, 21 samples in total.

Fig. 1. The uptake kinetics of selected pesticides by triolein-SPMD and rainbow trout, where (■) and (◆) indicate triolein-SPMD and rainbow trout, respectively.

pesticides by the triolein-SPMD and by fish are shown in Fig. 1. A quick sequestration, especially at the beginning of the exposure, and a linear increase during the entire exposure period of 20 d was observed for all tested chemicals, excepting for the bioaccumulation of lindan by the fish (Fig. 1). For lindan in the fish, after 20 d a steady state of exposure could be observed.

To predict chemical concentration in water by using its concentration in triolein-SPMD, the sampling rate (R_s) is required, which could be obtained from the linearity of uptake kinetics. To obtain the R_s , a mathematical model was used to fit the experimental data. In the model, the concentration of a chemical in the triolein-SPMD, or C_L , should be proportional to R_s

when it is linearly proportional to the concentration of the chemical in water, or C_w , as shown by the following equation:

$$C_L = C_w R_s t / V_L, \quad (1)$$

where V_L is the volume of the triolein-SPMD and t is the exposure period (in d). By least-squares linear regression for the linear part of the experimental data, a good fit was obtained for all pesticides with r^2 values from 0.80 to 0.93 for rainbow trout and from 0.79 to 0.95 for triolein-SPMD. Variation coefficients for R_s were less than 50% for rainbow trout and less than 30% for triolein-SPMD, respectively (Table 2). The uptake rate

Table 2
Calculated sampling rates of pesticides by rainbow trout and by triolein-SPMD

Compounds	R_s (whole body based)				R_s (lipid based)			
	Rainbow trout		SPMDs		Rainbow trout		SPMDs	
	L/d	CV (%)	L/d	CV (%)	L/d	CV (%)	L/d	CV (%)
Lindan	—	—	0.09	11	—	—	0.3	12
Aldrin	0.12	28	0.34	21	1.26	5	1.07	20
Heptachlor epoxide	0.18	37	0.45	14	1.69	21	1.43	12
4,4'-DDT	0.24	26	0.56	1	2.61	19	1.14	6
Hexachlorobenzene	0.28	12	0.61	7	3.69	18	1.94	5

of pesticides by rainbow trout was defined as R_f , which could be obtained by using Eq. (1) in a similar way.

By definition, R_s is indicative of the potentiality of a chemical entering the triolein-SPMD and R_f is for fish, respectively. Generally, R_s and R_f could be expressed as wet weight base (in case of triolein-SPMD, it includes weights of membrane and triolein). In Table 2, R_s 's were 1–2.5 times higher than R_f 's. The rates followed the increasing order of lindan < aldrin < heptachlor epoxide < 4,4'-DDT < hexachlorobenzene. This order is also the order of their lipophilicity or octanol/water partitioning coefficients (K_{ow}). Moreover, a lower variability could be noted in the case of triolein-SPMD, because triolein-SPMD was a reproducible standardized design, while individual discrimination of fish is unpredictable. In the work of Meadows et al. [25], the uptake rate constants of polychlorinated biphenyls (PCBs) by triolein-SPMD were 1–2 times of those by brown trout. Their and our results showed that triolein-SPMD concentrates hydrophobic organochlorinated compounds more quickly than fish do.

As regards triolein-SPMD, R_s depends not only on the absorption of targets in triolein, but also on the ability of a chemical to be absorbed by the polyethylene membrane. R_s could be lipid normalized as well. As shown in Table 2, when lipid-normalized, the rate constants for fish were larger than those for triolein-SPMD. The accumulation of a chemical in triolein-SPMD could be an integration of its accumulation in membrane (polyethylene in this case) and in triolein. Both processes are related to the lipophilicity of the compound. In the work of Gale [26], three-compartment model was used for evaluating contaminant accumulation by triolein-SPMD. The model predicts aqueous film control for compounds with moderate to high K_{ow} , rather than polymer film control. Although accumulation in the triolein may be subjected to polymer film control, the overall accumulation in triolein-SPMD should remain under aqueous film control, the lipid-normalized rate constant is therefore much smaller. The diffusivity of hydrophobic chemicals through biomembrane-like polymeric membrane is also controlled by

physicochemical properties of the diffusant molecules. The hydrophobic pesticides used in the present study have similar functional groups, size and steric effects and they are most likely the major factors controlling diffusion rates in the membrane. Since uptake behaviors for different pesticides, when compared on the basis of wet weight, were similar for triolein-SPMD and for rainbow trout, it appears that polyethylene membrane may serve as a good model of biological membrane with respect to the uptake of truly dissolved hydrophobic chemicals.

Previous work has shown that triolein-SPMD could accumulate organic pollutants to the levels similar to or even higher than those in aquatic organisms. For example, after 28 d of exposure of triolein-SPMD adjacent to caged channel fish in the water at the mouths of five Lake Michigan tributaries, they accumulated a greater number of chemicals and at higher concentrations than the fish [17]. However in the work of Prest et al. [16], levels of sequestered pesticides and PCBs were approximately 1.6 times higher in freshwater clams (*Corbicula fluminea*) than those in triolein-SPMD after 2 months of exposure in the river water (10°C). Moreover, another result showed that blue mussels were more efficient at sequestering polycyclic aromatic hydrocarbons than were SPMDs [4]. The lipid content in mussels (*Anodonta piscinalis*) is about 10 times that in clam [22]. It has been demonstrated that for organochlorinated pesticides whose molecules are often characterized by relatively high bond rotational and conformational freedom, each 5°C increase in water temperature results in 1.3–2-fold increase in uptake rates. Therefore, both types of organism and temperature could affect the uptake kinetics of pesticides by triolein-SPMD or by aquatic organisms.

5.2. Comparison of pesticides uptake by triolein-SPMDs and rainbow trout

Generally in steady state, the accumulation of hydrophobic organic compounds, such as pesticides, by aquatic organisms could be predicted through the

Fig. 2. Relationship between $\log K_{ow}$ values and the concentration factors of rainbow trout and SPMD.

linear relationship between BCF (or BAF) and K_{ow} [27,28]. For compound whose $\log K_{ow}$ varies from 5 to 7, the prediction according to linear relationship between $\log BCF$ and $\log K_{ow}$ could result in a better fit. However for compounds whose $\log K_{ow} < 5$, the prediction would be affected by its strong tendency to remain in the water phase and biodegradation in biota. For a compound whose $\log K_{ow} > 7$, its association with organic matter would make it less bioavailable to biota [29]. Although equilibrium was not reached for most pesticides under 21-d exposure period, correlations between C_L/C_w and K_{ow} ($R^2 = 0.887$) and C_f/C_w and K_{ow} ($R^2 = 0.931$) could be observed in Fig 2, where $\log C_L/C_w$ or $\log C_f/C_w$ was plotted versus $\log K_{ow}$. This figure was a clear indication that triolein-SPMD accumulates chlorinated pesticides in a quite similar way as fish do and accumulations of individual pesticide either by triolein-SPMD or by fish could be predicted by its $\log K_{ow}$.

Biomonitoring of aquatic organisms has been used as supplementary to monitoring aqueous concentration, because it provides not only information on environmental pollution, but also the potential ecological risks of an anthropogenic chemicals. However, the use of aquatic organisms in biomonitoring work involves certain problems, such as individual and species variability by sex, age, and physiological state of the organism. All aquatic species are confined to their geographic distribution areas, which makes it difficult to compare data from locations not covered by the distribution area of a single species. In addition, very few, if any, organisms may survive harsh environmental conditions such as deposits of highly contaminated and toxic sediments or industrial effluents. The significant similarity between bioconcentration by rainbow trout and biomimetic concentration by triolein-SPMD for chlorinated pesticides at concentration level close to that observed in natural environment confirms the applicability of the device as surrogate for biomonitoring

work. In addition, monitoring with triolein-SPMD is less complex and cost-effective than those using aquatic organisms.

6. Conclusion

In the laboratory continuous-flow system and at a concentration level close to that in natural environment, the uptake kinetics of five representative chlorinated pesticides by triolein-SPMD and by rainbow trout were quite similar. The time-dependent accumulation of the pesticides in triolein-SPMD or in fish was linear and only lindan could reach a steady state in fish when exposed for 21 d. However after 21-d exposure, there were good correlations between $\log C_f/C_w$ or $\log C_L/C_w$ and $\log K_{ow}$, indicating the accumulation process was controlled by the lipophilicity of the chemical. Our results provide additional information on the applicability of triolein-SPMD as a standard testing device for biomonitoring work.

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