

## Uptake of Polychlorinated Biphenyls (PCBs) from an Aqueous Medium by Polyethylene, Polyvinyl Chloride, and Polystyrene Films

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The sorption of selected polychlorinated biphenyl (PCB) congeners (from tri to deca chlorinated) by three food-packaging plastic films [polyethylene, polyvinyl chloride (PVC), and polystyrene] from an aqueous solution was investigated. From the data generated, PCB uptake, partition, and diffusion coefficients were calculated for the various films. Polyethylene exhibited the highest PCB uptake, diffusion, and partition coefficients when compared to the other materials. Although PVC indicated larger sorption diffusion and partition coefficients for the lower chlorinated congeners than polystyrene, a reversal of this trend was observed for the higher congeners. For polyethylene and PVC, the PCB uptake decreased as the chlorine numbers in the congeners increased, confirming the correlation between increasing chlorination and increasing cohesive density within the PCB molecules. For polystyrene, the uptake decreased from tri to penta congeners, but showed an increase for the hexa, and then a decreased uptake until the deca chlorination. A comparison of the molecular sizes of the PCB congeners showed that the partition ( $K_e$ ) and sorption diffusion ( $D_s$ ) coefficients generally decreased with their increasing molar volumes. The resulting  $K_e$  values were used to determine the extent of sorption because these values indicate the affinity of PCBs for the plastic films. Results from this study can be of practical importance for cases of product quality related to the transfer of contaminants from the product to the packaging materials.

**KEYWORDS:** PCBs; packaging; polyethylene; PVC; polystyrene

### INTRODUCTION

Polychlorinated biphenyl (PCB) compounds had widespread industrial uses prior to 1975 (1). The chemical properties of PCB made it an excellent additive to lubricating oils and greases, electrical installations, plastics, and carbonless printing. This is so because of its low electric conductivity, high heat resistance, and chemical stability (2). These same properties are also responsible for the persistence of PCBs in the environment long after they were banned (in 1975) in the United States because of their toxicity. Indeed, studies have shown the presence of PCBs in humans and aquatic, terrestrial, and avian creatures (3–7). PCBs are difficult to eliminate, and concentrated efforts are needed to eradicate or reduce their levels in the environment (8).

PCBs comprise a biphenyl ring with various degrees of chlorine substitutions. This ranges from mono to deca substitu-

tion. This substitution takes place on either or both rings and thus gives rise to a total of 209 congeners. As one ascends the homologous series of congeners from mono to deca chlorination, changes are seen in the properties of these molecules. These changes include increases in the molecular size, polarity, and toxicity (9). The objective of this research was to evaluate the sorption characteristics of PCBs by selected polymers as a means of removing PCBs from water. The polymers studied were films of low-density polyethylene, glassy polystyrene, and flexible polyvinyl chloride (PVC). Because both PCBs and the polymers are relatively lipophilic, if made to contact each other in an aqueous medium, successful partitioning of PCBs toward the polymers would result in a reduction in the concentration of the PCB contaminant in the water. However, due to differences in properties among polyethylene, polystyrene, and PVC, the rates and kinetics of PCB uptake by each of these materials would be expected to differ. This study investigated these differences by exposing PCBs to similar weights of these materials under similar conditions of contaminant concentration, time, and temperature. The results obtained provided an opportunity for the evaluation of the solubility, partition coefficients, and sorption diffusion coefficients of each plastic and one PCB congener from each group—tri to deca chlorinated.

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**Table 1.** Systematic Names of the PCB Congeners Used in This Study<sup>a</sup>

IUPAC no.	chlorobiphenyl systematic name
internal standard 30, 2,4,6-trichlorobiphenyl	
isomer no.	biphenyl compound
tri (31)	2,4',5-trichlorobiphenyl
tetra (47)	2,2',4,4'-tetrachlorobiphenyl
penta (103)	2,2',4,5',6-pentachlorobiphenyl
hexa (128)	2,2',3,3',4,4'-hexachlorobiphenyl
hepta (171)	2,2',3,3',4,4',6-heptachlorobiphenyl
octa (200)	2,2',3,3',4,5',6,6'-octachlorobiphenyl
nona (206)	2,3,4,5,6,2',3',4',5'-nonachlorobiphenyl
deca (209)	2,3,4,5,6,2',3',4',5',6'-decachlorobiphenyl

<sup>a</sup> Ballschmiter and Zell (10).

## EXPERIMENTAL PROCEDURES

**Materials.** *Polyethylene.* Pellets of polyethylene resin were obtained from The Dow Chemical Co. (Midland, MI). Cast films of polyethylene were then prepared by using a 1 in. single-screw four-zone Killion model KLB-100 extruder with an attached chill roll. The temperature zones 1–4 of the extruder were set to operate at 110, 121, 132, and 149 °C, respectively. The extrusion die temperature was set at 143 °C. The chill roll was set at a cooling temperature of 120 °C. The percent crystallinity of the polyethylene film, after casting, was 53.6%, and its thickness was set at approximately  $2.286 \times 10^{-3}$  cm.

*Polyvinyl Chloride.* PVC film with a thickness of  $1.78 \times 10^{-3}$  cm was obtained from Reynolds Metals Co. (Grottoes, VA). It was ~30% plasticized.

*Polystyrene.* Polystyrene film with a thickness of  $3.05 \times 10^{-3}$  cm was obtained from The Dow Chemical Co. (Midland, MI).

*Solvents and Chemicals.* All solvents used were of analytical HPLC grade. Hexane (85.04%) and petroleum ether (99.99%) were obtained from EMD Chemicals Inc. (Gibbstown, NJ). Acetone (99.8%), isooctane (99.91%), toluene (99.99%), diethyl ether (99.99%), and acetonitrile (99.8%) were obtained from Mallinckrodt Specialty Chemicals (Paris, KY).

All chemicals were of analytical grade and obtained from Mallinckrodt Specialty Chemicals (Paris, KY): sodium sulfate (activated by storage overnight at 130 °C); Florisil, 60–80 mesh (activated by storage overnight at 130 °C); silica gel 60, 70–230 mesh (activated by storage for 16 h overnight at 130 °C); sodium chloride; polychlorinated biphenyl congeners were obtained from Accustandard Inc. (New Haven, CT).

PCB congeners selected were on the basis of the occurrence of the congeners in Aroclor 1254 and are shown in **Table 1** (10).

**Methods.** *Exposure of Films to PCB Congeners.* This process began by preparing a standard 0.5 ppm solution of eight PCB congeners in acetone. Deionized distilled water was then spiked with the standard solution to produce an aqueous solution containing 10 ppb of each congener. After mixing, the aqueous solution was heated at 40 °C for 1 h with frequent shaking to evaporate the residual acetone. The solution thus obtained was cooled to 25 °C and set aside for contact with the polymeric films.

Approximately 0.160 g of each film was cut into strips of approximately  $4.0 \times 1.5$  cm. Each sample was then inserted into a 76 mL serum vial supplied by Wheaton Inc., Millville, NJ. Each vial was then brim-filled with the aqueous PCB congener solution, sealed, and stored at 25 °C in the absence of light. At given time intervals samples were removed for PCB congener quantification until an equilibrium concentration of the PCB uptake by each film was achieved.

*PCB Congener Analysis.* The PCB analytical method, including the cleanup procedures and the quantification of PCBs in the plastic materials and the aqueous solutions used in this study, was adapted from electron capture gas chromatographic analyses for PCB as outlined by Price et al. (11). The procedure is given in detail in Pascall et al. (12). All measurements in this study were performed in triplicate.

Because the concentrations of PCBs in water were of the order of parts per billion, an estimate of the sorption of PCB molecules by the

polymeric films was fitted by the equation

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \left[ \exp\left(\frac{-D\pi^2 t}{L^2}\right) + \frac{1}{9} \exp\left(\frac{-9D\pi^2 t}{L^2}\right) \right] \quad (1)$$

where  $M_t$  and  $M_\infty$  are the masses sorbed at time  $t$  and at equilibrium, respectively,  $D$  is the diffusion coefficient of the PCB molecules, and  $L$  is the film thickness (13). The diffusion coefficient ( $D$ ) was estimated by setting  $M_t/M_\infty$  equal to 0.5 and solving to obtain  $D$

$$D = \frac{0.049L^2}{t_{0.5}} \quad (2)$$

where  $t_{0.5}$  is the time required to attain the value  $M_t/M_\infty = 0.5$ .

From the data collected in this experiment, the uptake of each congener exposed to each polymer was calculated by substitution into the expression

$$S = M_\infty/w \quad (3)$$

In eq 3,  $S$  is the uptake expressed as mass of PCBs sorbed at equilibrium per mass of polymer.  $M_\infty$  is the total mass of PCBs absorbed by the polymer at equilibrium for a given temperature, and  $w$  is the weight of the polymer sample.

The equilibrium partition coefficient ( $K$ ) is the ratio of the equilibrium concentration of the PCB congeners in the polymeric phase and the equilibrium concentration in an aqueous phase

$$K = C_p/C_{aq} \quad (4)$$

where  $C_p$  is the equilibrium concentration of PCBs in the plastic film and  $C_{aq}$  is the equilibrium concentration of the PCBs in the aqueous solution.

## RESULTS AND DISCUSSION

**Figure 1** shows the diffusion coefficients of each PCB congener in polyethylene, PVC, and polystyrene. Polyethylene appears to have the largest coefficients for all congeners. PVC showed higher diffusion coefficients for the tri- to heptachlorinated congeners when compared with polystyrene. The presence of the nona- and deca-chlorinated congeners was not detected in the PVC material. In general, all plastic materials showed higher sorption coefficients for the lower congeners than the larger ones.

The partitioning of each congener between the aqueous and the polymeric phases is shown in **Table 2**. For polyethylene and PVC, the larger chlorinated congeners demonstrated a lower partitioning potential for the polymers than their lower counterparts, and conversely, the lower congeners had a higher partitioning for the polymers than the larger ones. For polystyrene, the hexa and hepta congeners showed higher partitioning for this polymer when compared with the other congeners.

To better understand the sorption affinity of the different congeners for the polymers, theoretical solubility parameters for PVC, polystyrene, and polyethylene were calculated (**Table 3**). An increase in the solubility parameters indicates a reduction in the solubility potential of the contacting species. Solubility parameters were based on the group contribution method where

$$\delta_d = \sum F_{di}/V \quad (5)$$

$\delta_d$  is the solubility parameter,  $F_{di}$  is the sum of the group molar attraction contribution for the PCB congeners, and  $V$  is their respective molar volumes (14). Calculations based on eq 5 show that the solubilities of PCB congeners exposed to each of the polymeric materials were expected to decrease from tri to deca chlorination. This trend may be the result of the increasing chlorination of these congeners from tri to deca, corresponding

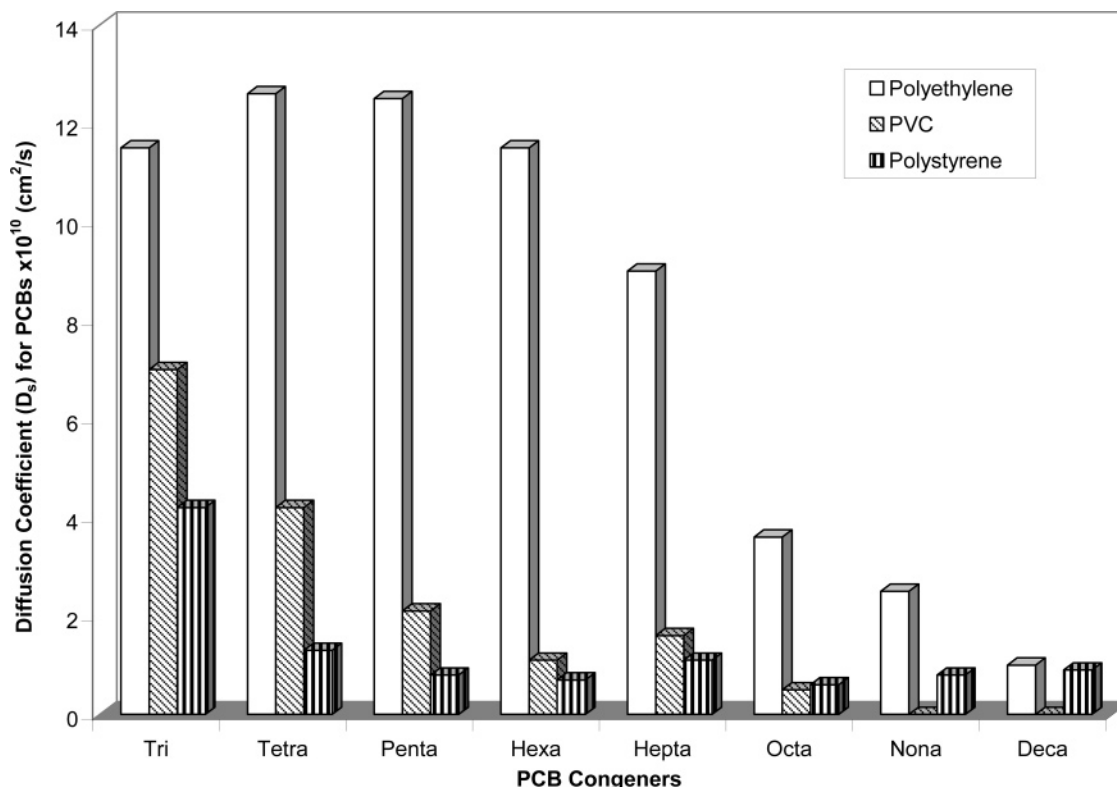


Figure 1. Diffusion coefficients of all congeners in polyethylene, PVC, and polystyrene at 25 °C.

Table 2. Partition Coefficient ( $K_p$ ) for All PCB Congeners Exposed to the Polymeric Materials Stored at 25 °C

congener	polyethylene	PVC	polystyrene
tri	200118	6853	310
tetra	121173	3223	409
penta	104500	4527	411
hexa	114570	720	1341
hepta	2571	545	756
octa	1054	298	430
nona	475	ND <sup>a</sup>	129
deca	307	ND	103

<sup>a</sup> Not detected.

Table 3. Theoretical Solubility Parameters for PCB Congeners Exposed to PVC, Polystyrene, and Polyethylene Films

PCB congener	PVC 0% <sup>a</sup>	PVC 20% <sup>a</sup>	PVC 30% <sup>a</sup>	PVC 40% <sup>a</sup>	polystyrene	polyethylene
tri	7.60	5.71	4.86	4.11	4.43	6.04
tetra	8.21	6.98	6.55	4.61	4.61	7.44
penta	7.82	6.31	5.71	5.28	5.96	8.69
hexa	8.07	6.67	6.13	5.75	6.47	9.32
hepta	8.37	7.05	6.56	6.22	6.91	9.85
octa	8.69	7.45	6.99	6.68	7.30	10.32
nona	9.01	7.82	7.39	7.10	7.66	10.74
deca	9.32	8.18	7.78	7.50	7.98	11.11

<sup>a</sup> Indicates percent plasticizer.

with their increasing molar attraction ( $F$ ) and, thus, an increasing resistance to the solvating action of the polymeric compounds. The information contained in **Table 3** could be used to estimate the solubility potential of a polymer for a diffusing chemical species. The calculated solubilities in this table showed how the addition of plasticizers to PVC could influence its solubility to PCB congeners.

Hernandez et al. (1986) indicated that knowledge of the diffusion, solubility and permeability coefficients of organic

penetrants through polymer structures is of both theoretical and practical importance (15). In terms of theoretical importance, such studies can aid in developing a better understanding of the mechanism of diffusion of organic penetrants through polymer membranes and particularly for the case of permeant molecules that have strong interaction with the polymer. The diffusion, partitioning, and uptake of the PCBs within the polymers studied can be of practical importance for the removal of PCBs from any aquatic medium.

Several factors are known to influence the uptake of a compound by a sorbent material such as plastic. These include (1) the physical and chemical nature of the sorbent, (2) the physical and chemical nature of the sorbate, (3) the sorbate concentration in contact with the sorbent phase, (4) the characteristics of the phase in contact with the absorbent, (5) the environmental temperature, and (6) the contact time of the system. This study focused on the physical–chemical nature of the sorbent phase (polymer) and the sorbate (PCBs).

A close examination of the physical and chemical structure of the PCB congeners used in this study reveals other factors responsible for the results obtained. These factors include dimensional issues such as molecular size, molar volume, and cross-sectional area of the PCB molecules. Comparing the tri- to the deca-chlorinated congeners, an increase in molecular weight and an increase in molecular size are observed. However, Perry et al. reported that molecular weight is not an effective description of molecular size but that parameters such as the parachlor, permachlor, and the space factor (or molar volume) can be used to correlate molecular properties with the passage of a migrating species through the membrane of a given polymer (16). Lyman reported that parachlor ( $P_c$ ) is an empirical relationship between molar volume and surface tension and gives rise to eq 6, where the surface tension is  $\sigma$  and  $V$  is the molar

$$V = \frac{P_c}{\sigma^{1/4}} \quad (6)$$

**Table 4.** Calculated Molar Volumes  $V_B$  and  $V_B'$  for PCB Congeners<sup>a</sup>

biphenyl compound	calcd $V_B$ LeBas molar vol (cm <sup>3</sup> /mol)
2,4',5-trichlorobiphenyl	229.96
2,2',4,4'-tetrachlorobiphenyl	247.48
2,2',4,5',6-pentachlorobiphenyl	265.00
2,2',3,3',4,4'-hexachlorobiphenyl	282.52
2,2',3,3',4,4',6-heptachlorobiphenyl	300.04
2,2',3,3',4,5',6,6'-octachlorobiphenyl	317.56
2,3,4,5,6,2',3',4',5'-nonachlorobiphenyl	335.08
2,3,4,5,6,2',3',4',5',6'-decachlorobiphenyl	352.60

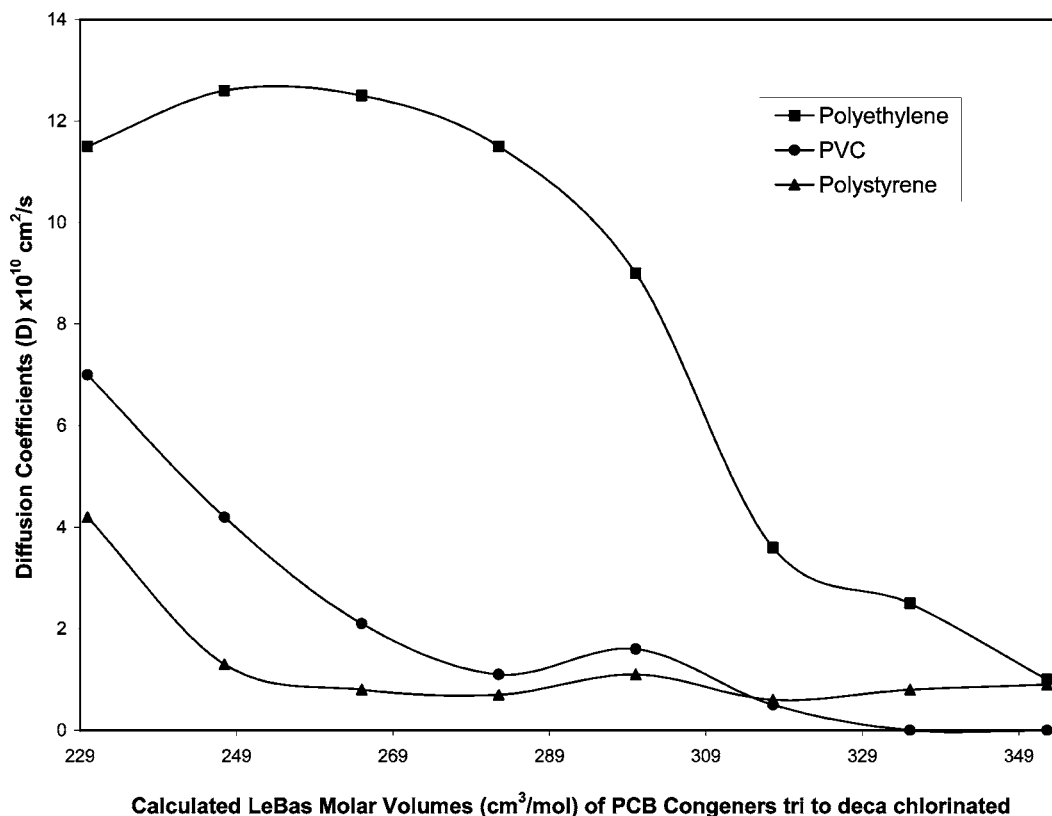
<sup>a</sup> Adapted from Tucker and Nelken (18).

volume (17). Parachlor values can be obtained from eq 6 if data on the chemical properties of the functional groups, length of the carbon chains, types of unsaturation, and the degree of branching of the permeating molecules are known (17).

In addition to obtaining values for  $V$  from eq 6, they can also be obtained from atomic and structural diffusion volume increments (18). An example of this calculation adapted from Tucker and Nelkon was used to determine the  $V$  values for the PCB congeners as seen in Table 4 (18). A correlation between these values and their diffusion coefficients was investigated and can be seen in Figure 2, which shows good correlation with PVC and polystyrene. This figure shows that the diffusion coefficients for the tri, tetra, penta, and hexa PCBs were fairly constant but that the relationship decreased sharply from hepta to deca chlorination when exposed to polyethylene. This result may have been influenced by the pore sizes of the polyethylene when compared to the molar volumes of the tri to hexa PCB congeners used in this study. If the molar volumes of these congeners are smaller than the pore spaces of polyethylene, then this may have been the reason for the results we obtained. If our selection of the PCB congeners resulted in their having larger differences between the molar volumes of the tri to hexa

chlorination, the trend of our result would have been more consistent with those obtained for PVC and polystyrene as seen in Figure 1. However, the general trend of Figure 2 was that an increasing chlorination of the biphenyl ring from tri- to deca-chlorinated corresponded with a reduction in diffusion from tri to deca PCBs. This finding was corroborated by Chern et al., who reported that large nonspherical compounds, cross-sectional area, and volume of the penetrant molecule may be the most important factors in determining the contaminant's ability to pass through an adequately sized gap (19). Heindoff also supported this by showing that an inverse correlation exists between the size of a penetrant and its diffusion rate through a given polymer (20). The result for the uptake of PCB by polystyrene in Figure 3 seems to defy the explanation given above. For polystyrene, the hexa and hepta congeners showed higher uptake values than the other congeners. We believe that the molecular volumes of the PCB congeners were not the only factor that influenced the results for polystyrene. The difference between the physiochemical properties of polyethylene and polystyrene may have contributed to the difference in the PCB uptake results. The polymeric backbone of the styrene polymer has a benzene molecule in place of hydrogen, as is the case for polyethylene. As a result, segmental mobility within the polystyrene chains is restricted at temperatures that do not restrict such movements in polyethylene. Also, the presence of benzene increases the distance between adjacent polymeric chains. The greater the segmental mobility and the greater the distance between the polymeric chains, the easier it is for a diffusing chemical to transverse the matrix of a given polymer (all other factors being constant). However, because segmental mobility within polystyrene is reduced by the presence of benzene, the material will be expected to behave differently from polyethylene.

Other factors about the nature of the polymeric matrix also affect the diffusion of a chemical species in contact with the



**Figure 2.** Comparison of the LeBas molar volumes of each PCB congener with their diffusion coefficients in polyethylene, PVC, and polystyrene.

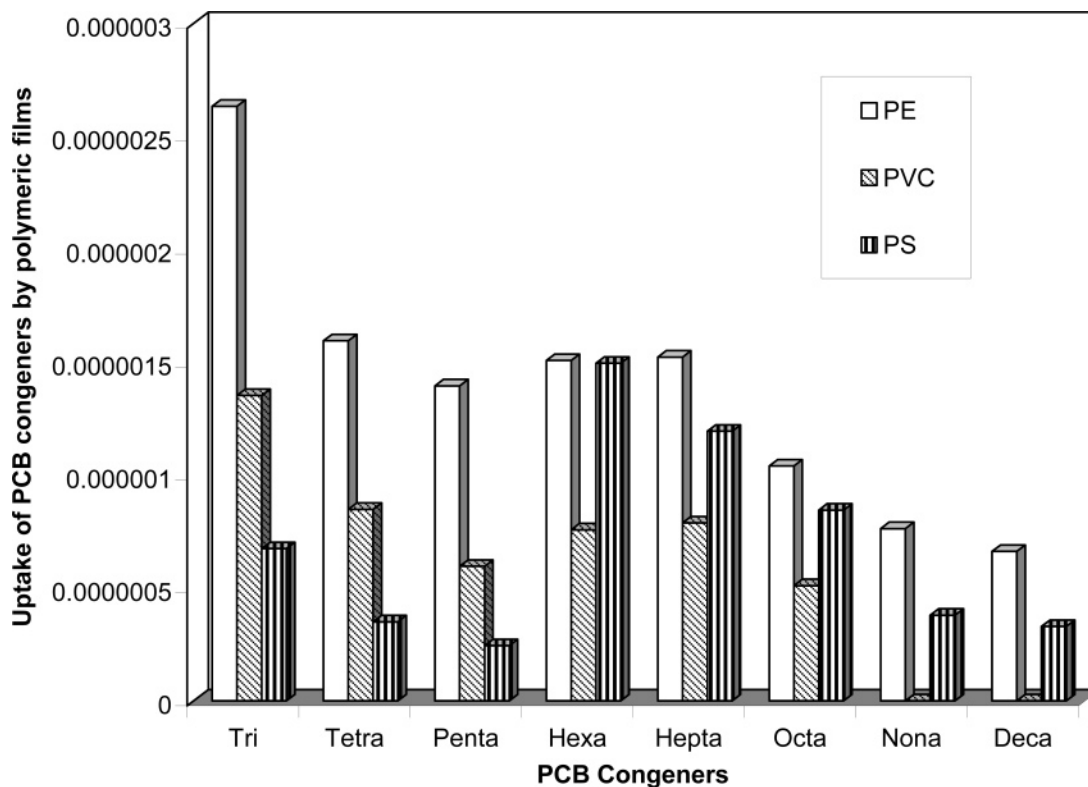


Figure 3. Uptake of PCBs by polyethylene, PVC, and polystyrene.

polymer. This can be seen if the molecular structure of the PVC is examined and compared with polyethylene, for example. Because the presence of the chlorine atoms within the PVC polymeric structure introduces a polar influence, the resultant increase in cohesive density creates attractive forces between the individual PVC chains. This higher density may influence a reduction in its free volume when compared with polyethylene, which does not have chlorine atoms. If the higher density of PVC reduces its free volume, then it would be more difficult for a potential migrating species to diffuse through it. We believe this may be the case in this study if the synergistic effect of the lower free volume of PVC and the increased molecular size of the larger PCB congeners are correlated. Our results reflect this relationship because the results show that the presence of the nona- and deca-chlorinated congeners is not detected when compared with the other lower congeners. These results were confirmed by partition coefficients obtained for PCBs exposed to the PVC. **Table 2** shows that the lower chlorinated congeners exhibited a higher partitioning for the plastic than the higher analogues. Leggett and Parker obtained similar results for the partitioning of *cis*-dichloroethane, *trans*-dichloroethane, 1,1,2-trichloroethane, tetrachloroethane, chlorobenzene, *o*-dichlorobenzene, *m*-dichlorobenzene, and *p*-dichlorobenzene between groundwater and PVC in one instance and for polytetrafluoroethylene (PTFE) in another instance (21). These results showed that an increase in the size of the molecular volumes of the penetrants may have caused a decrease in the partition coefficients for the PVC and PTFE.

The glass transition temperature ( $T_g$ ) of a polymer is the temperature above which segmental mobility of the polymeric chain begins. Compared with polyethylene ( $T_g = -120$  °C), the  $T_g$  of PVC is  $\sim 85$  °C. The  $T_g$  of PVC can be affected by the degree of plasticization that is done to the initial PVC that is synthesized. Because polyethylene at room temperature is above its  $T_g$ , it is expected to have a higher free volume than

PVC. This fact is also responsible for the increased uptake of the PCB congeners when compared with the uptake by the PVC polymer.

In addition to the effect of molar volume on the properties of a given polymer, Carroll et al. showed that diffusion coefficient can also be influenced by factors such as the Permachor

$$P = A \exp(-s\pi) \quad (7)$$

value and whether the polymer is in a rubbery or a glassy state (22). The Permachor value is the force within a polymer that is holding its chains together. Giacini and Hernandez reported that the Permachor equation could be used to predict the gas permeability of a polymeric structure, where  $P$  is the permeability,  $A$  and  $S$  are constants for any given gas at temperature  $T$ , and  $\pi$  is the Permachor constant (23). Giacini and Hernandez further stated that the Permachor constant is based on the cohesive energy density and free volume of the polymer (23). A rubbery polymer is one that could become swollen (rubberlike) by the moving front of a diffusing penetrant. These rubbery polymers are usually above their  $T_g$  values when unplasticized. A glassy polymer, on the other hand, is one that is more difficult to plasticize by a diffusing penetrant. These glassy polymers are usually below their  $T_g$  and are also referred to as being in a condensed (glasslike) state. Because both unplasticized PVC and polystyrene ( $T_g = 100$  °C) are glassy polymers, the response of these polymers to the plasticizing effect of a penetrant can be described as a time-dependent diffusion process that is reliant on the relaxation rate of the polymeric chains. In such cases, an immediate but partial response, followed by a slow asymptotic approach to equilibrium, occurs. A rubbery polymer, on the other hand, is expected to respond rapidly if similarly treated by an appropriate plasticizer and will facilitate an easier migration pathway for the potential penetrant. Crank reported that rubbery polymers such as polyethylene adjust quickly to the presence of a penetrant and do not cause diffusion anomalies

(13). In such cases, the rate of the diffusing species is less than the rate of relaxation of the polymeric chains. This is a case of a concentration-dependent relationship and tends to be Fickian in nature.

Because plasticizers are usually used in PVC to facilitate its film-making process, this may have influenced the results obtained in this study. Thus, the uptake, diffusion, and partition coefficients for the PCB congeners would be higher with our PVC film when compared to unplasticized PVC. The effects of plasticization of the PVC would result in a lowering of its  $T_g$ . This would influence a more rubbery behavior in the PVC polymer used in this research. The PVC used in this study was ~30% plasticized, and it appears that the plasticizer did not cause the PVC to become 100% amorphous in nature. Had this been the case, the results might have been closer to what was obtained for polyethylene.

In this study, the results show higher coefficients for all congeners exposed to polyethylene when compared to PVC and polystyrene. This is consistent with the rubbery/glass state theory explained earlier. This is so because polyethylene is known to be a rubbery polymer. Yasuda and Stannett also confirmed this principle when they investigated the diffusion of cyclohexane and benzene through polyethylene (24).

In conclusion, this study showed that polyethylene removed all of the PCB congeners from the water much better than either PVC or polystyrene. However, polystyrene removed the higher chlorinated congeners better than it did the lower analogues. When compared with PVC, polystyrene was more efficient in removing the high PCB congeners. For polyethylene and PVC, the lower chlorinated congeners generally showed a higher partitioning for the polymeric materials when compared with the higher analogues. For polystyrene, the hexa congener had the highest partition coefficient.

Industrial applications of this study can be for removal of PCBs from aqueous products. Because PCBs possess a higher partition coefficient for plastic than for water as a solvent, engineering applications of selected plastics for use as adsorbent and absorbent materials in water treatment plants can be investigated for possible use as a purification tool. This technique could also be used to concentrate PCBs in a polymeric matrix if the level of the PCBs in the extrapolymeric environment is too low for quantification. This technology can be used in water-processing facilities and by environmental protection operations as a tool in monitoring low levels of PCBs in groundwater.

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