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Determining equilibrium partition coefficients between lipid/protein and polydimethylsiloxane for highly hydrophobic organic contaminants using preloaded disks

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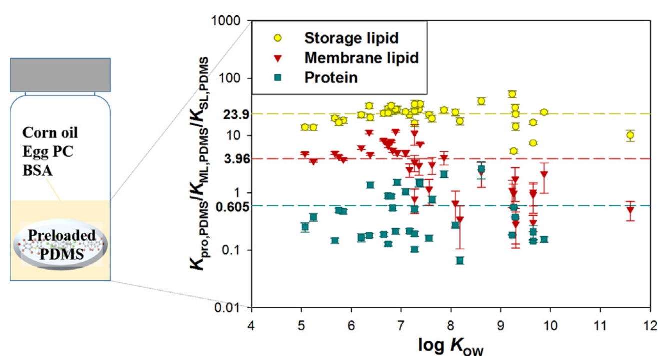
HIGHLIGHTS

- Preloaded PDMS disks showed viable loading efficiency for hydrophobic compounds.
- Partition coefficients ($K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$) were determined.
- Partitioning between biological phases and PDMS was independent of hydrophobicity.
- Pseudo-equilibrium explained different $K_{pro,PDMS}$ for compounds with Log $K_{ow} > 9$.
- The SL had the highest sorption capacity, followed by ML and protein.

GRAPHICAL ABSTRACT



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ABSTRACT

Bioaccumulation of hydrophobic organic contaminants is of great concern and understanding their partitioning to biological phases is crucial for estimating their bioaccumulation potential. The estimation, however, was of large uncertainty for highly hydrophobic organic contaminants (HHOCs) with log $K_{OW} > 9$ due to the challenge of quantifying their water concentrations. In the present study, partition coefficients between polydimethylsiloxane (PDMS) and storage lipid ($K_{SL,PDMS}$), membrane lipid ($K_{ML,PDMS}$) and protein ($K_{pro,PDMS}$) were measured for 21 polychlorinated biphenyls (PCBs), 14 polybrominated diphenyl ethers (PBDEs), dechlorane plus (DP) and decabromodiphenyl ethane (DBDPE), covering log K_{OW} from 5.07 to 11.6, using a preloaded PDMS depletion method. The values of $K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$ were in the ranges of 5.36–52.5, 0.286–11.8 and 0.067–2.62 g/g, respectively, being relatively constant although their K_{OW} values extend more than six orders of magnitude. The relative sorption capacity of the biological phases showed storage lipid was the dominant sorption phase in biota, followed by membrane lipid and protein was the lowest. The $K_{PDMS,pro}$ values of the compounds with log $K_{OW} < 9$ were similar (0.382–14.9 g/g) regardless of the thickness of preloaded PDMS disks (58–209 μm). For HHOCs, however, $K_{PDMS,pro}$ values dropped when thinner PDMS disks were used, as a result of slow diffusion of HHOCs in PDMS. The $K_{PDMS,pro}$ values of HHOCs measured by 58-μm PDMS disks ranged from 1.78 to 6.85 g/g, which was consistent with compounds with log $K_{OW} < 9$. This validated that partition coefficients between PDMS

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and biological phases were independent of chemical hydrophobicity, showing the advantage of using PDMS-based methods to directly estimate bioaccumulation potential of HHOCs.

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1. Introduction

Halogenated hydrocarbons, like polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) were historically used and are still of great concern nowadays owing to their persistence, bioaccumulation and toxicity (Hites, 2004; Diamond et al., 2010). The restriction on penta- and octa-BDEs promoted the use of highly hydrophobic organic contaminants (HHOCs) as flame retardants, including decabrominated diphenyl ether (BDE-209), dechlorane plus (DP) and decabromodiphenyl ethane (DBDPE) (Wang et al., 2016; Yu et al., 2016). Although these HHOCs are less bioavailable compared with PCBs and low-brominated BDEs (Klosterhaus et al., 2011; Zhang et al., 2013), they have been detected in aquatic organisms (Zhang et al., 2013; Sun et al., 2016), calling for monitoring their occurrence in the environment.

Passive sampling provides a cost-effective way to monitor hydrophobic organic contaminants (HOCs) in the environment, showing a potential for developing global aquatic monitoring networks (Lohmann et al., 2017). Polydimethylsiloxane (PDMS) has been successfully used as passive samplers to measure the freely dissolved concentration (C_{free}) of HOCs with moderate hydrophobicity, e.g. polycyclic aromatic hydrocarbons (PAHs) and PCBs (Rusina et al., 2007; Mayer et al., 2014). Nevertheless, its application for HHOCs remains challenging for the high uncertainty of PDMS-water partition coefficients ($K_{PDMS,W}$) (Booij et al., 2015). It becomes increasingly challenging to accurately quantify C_{free} with increasing HOCs' hydrophobicity which results in extremely low C_{free}. Recently, Jahnke et al. (2012, 2014a) directly estimated thermodynamic bioaccumulation potential of sediment-bound HOCs through PDMS concentrations at equilibrium and lipid-PDMS partition coefficients ($K_{lip,PDMS}$), offering a means to estimate bioaccumulation using passive samplers without the necessity of measuring C_{free}. Compared with $K_{PDMS,W}$, measuring $K_{lip,PDMS}$ is more approachable and accurate (Jahnke et al., 2012), because lipid and PDMS have similar sorption capacities for HOCs.

When assessing the bioaccumulation potential of HOCs, lipids are regarded as the main accumulation phase in organism and HOC concentrations are normalized to a unified lipid, which is defined operationally based on experimental approaches and commonly contains both storage lipid (SL) and membrane lipid (ML). In fact, SL and ML are significantly different in structure and affinity for HOCs (Endo et al., 2011; Quinn et al., 2014) and the binding of chemicals to ML may cause adverse effects to organisms (Escher and Schwarzenbach, 2002). Besides lipid, other biological phases, e.g. protein, also involve in chemical sorption and form an important sorptive phase, particularly in lean organisms which contain high protein contents, such as benthic invertebrates (de Bruyn and Gobas, 2007, Elissen et al., 2010). Therefore, it is good to include SL, ML and protein as individual biocomponents and measure their respective partition coefficients to PDMS ($K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$) when estimating HOC bioaccumulation using PDMS-based methods.

The main aim of the present study was to measure the values of $K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$ for a variety of HOCs, including PCBs, PBDEs and HHOCs. To determine partition coefficients, both uptake methods (chemicals uptake from spiked biological phases to PDMS until equilibrium, e.g. Jahnke et al., 2008, Li et al., 2014 and Mäenpää et al., 2015a) and depletion methods (chemicals being released from spiked PDMS to biological phases until equilibrium, e.g. Escher et al., 2011 and Endo et al., 2013) have been applied. For some PCBs and low-brominated BDEs which have moderate hydrophobicity, both methods worked equally in measuring their partition coefficients (Jahnke et al., 2008; Endo et al., 2013; Li et al., 2014; Mäenpää et al., 2015a), but no data are available for HHOCs between lipid and PDMS. Endo et al. (2013) tried to quantify $K_{ML,PDMS}$ and $K_{pro,PDMS}$ for PBDEs using the depletion method, but failed for chemicals more hydrophobic than BDE-183. Before depletion, HOCs were required to be loaded onto PDMS, which was usually achieved by soaking PDMS in methanol/water solution containing HOCs. This loading technique, however, was not applicable for HHOCs as suggested by the high disk-to-disk variability of BDE-209 in PDMS (>40%) (Endo et al., 2013). Alternatively, Li et al. (2014) used an uptake method for estimating PBDEs' $K_{pro,PDMS}$. Although both studies (Endo et al., 2013; Li et al., 2014) obtained similar $K_{pro,PDMS}$ values for low-brominated BDEs, the values for BDE-183 were quite disparate. Therefore, it is necessary to develop additional methods to determine $K_{lip,PDMS}$ ($K_{SL,PDMS}$ and $K_{ML,PDMS}$) and $K_{pro,PDMS}$ values for HHOCs and interpret the deviation of $K_{pro,PDMS}$ values for BDE-183 between the studies (Endo et al., 2013; Li et al., 2014).

To achieve this, a depletion method with preloaded PDMS disks was developed to determine $K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$ for HOCs with a broad range of hydrophobicity (logarithm of octanol-water partition coefficients (log K_{OW}) from 5.07 to 11.6, HHOCs included). Furthermore, the impacts of HOC hydrophobicity and PDMS thickness on the partitioning process were assessed. The divergent data among studies (Endo et al., 2013, Li et al., 2014 and the present study) were explained. Lastly, the application of $K_{SL,PDMS}$, $K_{ML,PDMS}$ and $K_{pro,PDMS}$ in estimating bioaccumulation potential of HOCs was discussed.

2. Materials and methods

2.1. Surrogates for lipids and proteins

Commercial corn oil, egg phosphatidylcholine (PC) and bovine serum albumin (BSA) were selected as the surrogates for SL, ML and protein, respectively. Geisler et al. (2012) found different SLs showed similar affinity for HOCs, which was further validated by the fact that there was no difference among the $K_{SL,PDMS}$ values for PCBs when vegetable oil, fish oil or seal oil was used as the surrogate for SL (Jahnke et al., 2008). The PC is the main component of biological membranes and serum albumin is the most abundant protein in the serum, thus they were previously selected as the surrogates for ML and protein, respectively (Endo and Goss, 2011; Escher et al., 2011; Endo et al., 2013; Mäenpää et al., 2015a).

Corn oil (Arawana Brand, Shanghai, China) was bought in a local supermarket and egg PC (purity > 98%) and BSA were purchased from Shanghai Advanced Vehicle Technology Pharmaceutical Ltd. (Shanghai, China) and Boao biotech (Shanghai, China), respectively. Egg PC suspension (100 mg/mL) and BSA solutions (20, 60 and 100 mg/mL) were prepared in solution containing 0.9% NaCl and 8 mmol/L NaN_3. Before use, the egg PC suspension was vigorously stirred for 2 h to create liposomes (Mäenpää et al., 2015a).

2.2. Preparation of preloaded PDMS disks

The PDMS disks preloaded with HOCs were homemade from MDX4–4210 BioMedical Grade Elastomer kit which was purchased from Dow Corning (Auburn, MI, USA). According to the manufacturer's manual, elastomer component and curing agent (10:1, w/w) were thoroughly mixed. In the meantime, 2 mL of HOC solution in acetone was added to 23 g elastomer mix. To avoid oversaturation, HOC concentrations in PDMS were kept much lower than their solubility in PDMS (Grant et al., 2016). Then, the mixtures were coated on clean smooth

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