

# **Applications for passive sampling of hydrophobic organic contaminants in water – A review**

Adam C. Taylor<sup>a</sup>, Gary R. Fones<sup>a\*</sup>, Branislav Vrana<sup>b</sup> and Graham A. Mills<sup>c</sup>

*<sup>a</sup>School of Earth and Environmental Sciences, University of Portsmouth, Burnaby Road, Portsmouth, PO1 3QL, UK*

*<sup>b</sup>Faculty of Science, Research Centre for Toxic Compounds in the Environment (RECETOX), Masaryk University, Kamenice 753/5, Pavilion A29, 625 00 Brno, Czech Republic*

*<sup>c</sup>School of Pharmacy and Biomedical Sciences, University of Portsmouth, White Swan Road, Portsmouth, PO1 2DT, UK*

\*To whom all correspondence should be addressed

Phone number: +442392842252

e-mail: [gary.fones@port.ac.uk](mailto:gary.fones@port.ac.uk)

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## **Abstract**

We comprehensively review the current state-of-the-art of environmental monitoring for hydrophobic organic contaminants in aqueous matrices using passive sampling devices. Principles of the theory of passive sampling are presented. Strategies for passive sampler design and operation, limitations in performance and data quality-assurance and quality-control are reviewed. Advances in applications of available passive sampling devices are extensively critiqued. Future trends and current challenges facing practitioners and barriers to further adoption of the devices are discussed.

## **Keywords**

Environmental monitoring; Non-polar pollutants; Passive sampling; Sampler applications; Water analysis

## **Abbreviations**

DM, Diffusion membrane; EDA, Effect driven analysis; EDCs, Endocrine disrupting compounds; EQS, Environmental quality standard; EVA, Ethylene vinyl acetate; HLB, Hydrophobic-lipophilic balanced; HOCs, Hydrophobic organic compounds; LDPE, Low-density polyethylene; LOQ, Limit of quantification; LSER, Linear solvation energy relationship; MESCO, Membrane-enclosed sorptive coating; MoA, Mode(s) of toxic action; PAHs, Polyaromatic hydrocarbons; PBDEs, Polybrominated diphenylethers; PCB, Polychlorinated biphenyl; PDMS, Polydimethylsiloxane; PES, Polyethersulfone; POP, Persistent organic pollutant; POM, Polyoxymethylene; PMMA, Polymethyl methacrylate; PRCs, Performance reference compounds; PSD, Passive sampling device; QSAR, Quantitative structural activity relationship; SBSE, Stir-bar sorptive extraction; SPM, Suspended particulate matter; SPMD, Semi-permeable membrane device; SPME, Solid-phase microextraction SR, Silicone rubber; TWA, Time-weighted average; WBL, Water boundary layer

## 1. Introduction

Hydrophobic organic compounds (HOCs) are present throughout all environmental compartments and may be present in the aquatic environment at trace concentrations ( $\text{ng L}^{-1}$  to  $\text{pg L}^{-1}$ ). The risk posed by certain HOCs (e.g. polyaromatic hydrocarbons (PAHs), polybrominated diphenylethers (PBDEs), polychlorinated biphenyls (PCBs)) is well established and these compounds are included in the priority pollutant lists of the EU's Water Framework Directive, EPA's Clean Water Act and the persistent organic pollutant (POP) list of the Stockholm Convention [1–3]. Typically, monitoring programmes for priority pollutants consist of discrete grab (bottle or spot) samples chemically analyzed for compliance with threshold concentrations. Achieving limits of quantification (LOQ) for all priority HOCs can be expensive, requiring collection of large volumes of water and several sample separation, concentration and analytical steps. Additionally, grab samples only provide a 'snapshot' of the aquatic analyte concentration at the moment a sample is taken, which may not accurately reflect the risk posed to human or aquatic organisms [4]. HOC concentrations in the environment vary dynamically, undergoing mass fluxes between environmental compartments in response to long-range transport and regular and episodic pollution, favoring accumulation in sediment and biotic phases. The risk posed by aquatic HOCs to biota may be magnified by microplastic vectors that are now known to be present in waters globally and interact with biota via a number of pathways [5].

Several strategies have been suggested to better assess exposure risk from aquatic HOCs. These strategies include, frequent grab samples or automated sampling, monitoring in biota and sediment. Each of these strategies has advantages and weaknesses. Frequent grab sampling or automated sampling is often unworkable due to restrictions in sampling at remote locations. In addition, unrealistically high volumes of water have to be processed in order to reach sufficiently low method limits of quantification for compliance monitoring. HOC concentrations in whole water do not directly reflect their chemical activity and associated risk to aquatic organisms. Sediment monitoring is less useful when HOC concentrations in overlying waters and sediments are not in equilibrium or the composition of sediments varies over the sampled area. This complicates the comparability of spatial or temporal data. Likewise, chemical monitoring using analysis of aquatic biota is complicated by the large inherent variability in HOC concentrations related to many factors including exposure pathways, organism lipid content, age, gender and trophic position. Moreover, sampler preparation can involve complex analyte extraction and concentration steps. Another approach is the use of

passive samplers. These devices can provide additional information on freely dissolved aquatic HOCs and provide time-weighted average (TWA) or equilibrium concentrations. Passive sampling overcomes many of the shortcomings of grab, sediment and biota monitoring, caused by variable and poorly defined monitoring matrix composition issues. Materials used in passive sampler construction have constant composition and well-defined diffusion and partition properties. This allows sampling, quantification and the potential to compare HOC concentrations in time, space and across environmental compartments in a reproducible way.

Passive sampling relies on *in-situ* accumulation of analytes within a receiving phase during an exposure in the sampled medium. Since the last comprehensive review of aquatic passive sampling (all pollutant classes) in 2005 by Vrana et al. [6], knowledge of passive sampling of HOCs has advanced, with >300 additional publications since the time of this publication. This review briefly introduces the principles of passive sampling, then presents the new applications of passive sampling for HOCs between 2005-2019.

## **2. Principles of passive sampling**

Passive sampling refers to any technique through which analytes present in a bulk phase of the sampled medium are transferred and retained in a receiving phase, where flux of analytes between phases is driven only by differences in chemical potential [7]. If the receiving phase remains in the bulk phase the spontaneous flux of analytes will continue between phases until the difference in chemical potential disappears i.e. thermodynamic equilibrium is reached. The receiving phase may be an adsorbent or absorbent solid, a solvent or a chemical reagent, which can be loose or stabilized on or in a supporting matrix. Typically, the receiving phase of PSDs for HOCs is either a hydrophobic solvent or an absorbent non-polar elastomer [8]. Adsorbent receiving phases are used less frequently [9].

Partitioning of HOCs between phases typically follows first order kinetics. This can be described by a one-compartment mathematical model, where the analyte concentration in the receiving phase  $C_s$  at a known exposure time ( $t$ ), is proportional to the analyte concentration in the bulk phase  $C_w$ , and the uptake  $k_1$ , and dissipation  $k_2$  constants. Accumulation of analyte in the receiving phase occurs in kinetic followed by equilibrium regimes. The first order model can be described by equation (1):

$$C_s(t) = C_w \frac{k_1}{k_2} (1 - e^{-k_2 t}) \quad (1)$$

PSDs can be operated in either the kinetic or equilibrium regime. Different devices exposed at the same location for an equal time, may not produce comparable results for all HOCs. Before exposure, it is important the design and operation of the PSDs are considered alongside the characteristics of HOCs present in sampled waters. This will ensure the design of the monitoring programme provides the most appropriate results to answer the experimental question [10].

### 2.1 *Equilibrium passive sampling*

Equilibrium PSDs are exposed for sufficient time for the concentration of analyte in receiving and bulk phases to reach thermodynamic equilibrium. In a theoretical system where the analyte concentration in the bulk phase is constant and is not depleted by accumulation in the receiving phase, once equilibrium is reached the concentration of analyte in the receiving phase will not change and the aquatic concentration of HOCs can be derived using receiving phase-water partition coefficients ( $K_{sw}$ ) [11–15]. The time taken to attain this theoretical equilibrium is referred to here as  $t_{eq}$ . Environmental concentrations of HOCs are dynamic. The suitability of PSDs operated in the equilibrium regime will depend on the magnitude and arbitrariness of analyte concentration variability in the bulk phase and the response time of the PSD. This must be shorter than said variations in concentration. Environmentally significant concentrations of certain HOCs in aqueous media are often trace ( $\text{ng L}^{-1}$  to  $\text{pg L}^{-1}$ ) and relatively stable and in these circumstances, PSDs operating in the equilibrium regime are appropriate [16]. When PSDs are operated in the equilibrium regime equation (1) can be altered to equation (2):

$$C_s = C_w \frac{k_1}{k_2} = C_w K_{sw} \quad (2)$$

### 2.2 *Kinetic passive samplers*

Kinetic passive samplers are designed and operated so that accumulation of target HOCs is time integrative and responsive to concentration changes in the sampled water (kinetic regime). In the kinetic regime, initial accumulation in the receiving phase is linear (if  $C_w$  is constant), as the HOC dissipation rate ( $C_s k_2$ ) from the sampler is negligible compared to the uptake rate ( $C_w$

$k_1$ ) [8]. Increases in analyte concentration in the receiving phase and the dissipation rate are proportional. Accumulation of analyte is integrative until the theoretical time at which the magnitude of the dissipation rate is no longer negligible in relation to the uptake rate, referred to here as  $t_{lin}$ . After exposure time  $t_{lin}$ , accumulation is curvilinear approaching an asymptote at thermodynamic equilibrium ( $C_s k_2 = C_w k_1$ ) at exposure time  $t_{eq}$ . The kinetic regime ends at  $t_{eq}$  (equilibrium). Kinetic PSDs are operated between  $t_0$  and  $t_{lin}$  in the linear uptake stage of the kinetic regime. During this stage the uptake rate is linearly proportional to the concentration in the bulk phase and sampling is time-integrative [8]. Here equation (1) can be reduced to equation (3):

$$C_s(t) = C_w k_1 t \quad (3)$$

Kinetic PSDs are used to measure TWA concentrations of analytes in the aqueous phase. In this case equation (3) can be rearranged in order make the analyte mass in the receiving phase ( $M_s$ ) at the end of the exposure ( $t$ ) the subject:

$$M_s(t) = C_w R_s t \quad (4)$$

Here  $R_s$  represents the sampling rate (unit volume of water sampled per unit time). If the analyte mass in the receiving phase is measured and the sampling rate is known it is possible to calculate the average analyte concentration ( $C_{TWA}$ ) over exposure time ( $t$ ) by rearranging equation (4):

$$C_{TWA} = \frac{M_s}{R_s t} \quad (5)$$

In order to determine TWA analyte concentrations in the bulk phase the  $R_s$  is required. The sampling rate is a product of the overall mass transfer coefficient and sampler surface area  $k_o A$  [8]. In case of water boundary layer controlled HOC uptake, the mass transfer coefficient  $k_o$  is affected by water flow velocity and turbulence. In such cases site specific sampling rates can be derived from the release rate of performance reference compounds (PRCs) covering the hydrophobicity range of analyzed compounds [17, 18]. Models are available that relate water boundary layer controlled sampling rates with molar mass [19]. For compounds slowly diffusing in the membrane or receiving phase material, diffusion in those media may be rate-

limiting. Knowledge of diffusion coefficients of analyzed HOC in those media is therefore important for assessment of the main barrier controlling the uptake [20].

### 2.3 *Passive sampler design*

An ideal passive sampler design is inexpensive with a simple construction, easy to prepare, deploy, retrieve and analyze, and has selectivity and sensitivity for a wide range of analytes [21]. In practice, passive sampler design is optimized according to several objectives and no device is suitable for all applications. Devices are either single or dual phase. Single phase polymer PSDs form the simplest designs. Here polymer formulation and surface area to volume ratios can be selected to alter sampler performance [22]. Polymers such as polyoxymethylene (POM) are favored for sampling in the equilibrium regime because of their high resistance to mass transfer in the polymer (low polymer diffusion coefficient ( $D_p$ )) and low  $K_{sw}$ . This compares to the use of low-density polyethylene (LDPE) or silicone rubber (SR), which results in a faster (apparent) equilibrium in POM (surface layer only). Migration of HOCs within the POM polymer cross-section can bias this apparent equilibrium with increasing storage time of retrieved samplers [23, 24]. Dual phase passive samplers such as the Chemcatcher<sup>®</sup> contain a receiving phase and a diffusion membrane (DM). The DM effectively extends the kinetic regime by slowing diffusion between the aqueous and receiving phases [25]. In the semi-permeable membrane device (SPMD), the DM retains the liquid receiving phase (triolein). Polymer-water partition coefficients increase with the hydrophobicity of HOCs. For very hydrophobic compounds ( $\log K_{ow} < 5.5$ ) equilibrium cannot be attained even with deployment times in excess of several months [26]. Where equilibrium is unlikely within the exposure time, the extent of equilibration must be quantified [8]. Passive sampler design considerations differ when quantitative TWA concentrations (kinetic regime) are desired. Ideally these designs should have a  $D_p$  that does not limit uptake, demonstrate isotropic exchange of PRCs and have a sufficiently high  $K_{sw}$  and  $R_s$  so that  $t_{lin}$  is longer than the exposure time.  $M_s$  (after extraction) needs to be  $>$  LOQ of the analytical method. The design of deployment apparatus may also influence sampler performance. Novel PSDs for HOCs in aquatic matrices continue to be developed [27, 28]. However, as researchers and legislators try to incorporate passive sampling into frameworks for regulatory monitoring, existing designs for which performance, applications and inter-comparability of data are well established are likely to be preferred.

## 2.4 Calibration of passive samplers

Calibration of PSDs is necessary in order to relate  $C_s$  to  $C_w$  by determining  $R_s$  and  $K_{sw}$ , as required for the calculation according to equations 1-5. Calibration of PSDs for HOCs in aqueous matrices may be undertaken *in-situ* by measuring isotropic exchange of PRCs. Less accurate alternate approaches may involve calibration prior to exposure in simulated conditions, or the formation of empirical [19], mechanistic [29], linear solvation energy relationship (LSER) [30] or quantitative structural activity relationship (QSAR) [31] models. The development of models enables the calculation of the relationship between molecular structural features and substance specific mass transfer coefficients and partition coefficients. The influence of exposure specific uptake limiting factors mean that theoretical uptake kinetics derived from first principles or observations disagree with those realized in practice [6]. As such, calibration approaches which seek to capture analyte and exposure specific variation in uptake are generally favored [21]. Calibration of PSDs requires prior knowledge of the environmental conditions during deployment in order to accurately replicate them in the laboratory. Experiments must be carefully designed to account for depletion of HOCs in the aqueous phase due to transfer to the receiving phase and sorption to surfaces in the calibration system [19]. Laboratory exposures usually consist of either a flow-through system containing freshly spiked water or a closed system where the spiked water is replenished at set intervals. Despite differences in experimental design, variation in derived  $R_s$  and  $K_{sw}$  between studies uncovered no method bias. Practitioners have identified and actioned the responses necessary to reduce this variation as part of the wider coalescence towards improving data quality assurance [32] (see Section 5). Perhaps the best approach is through use of PRCs, which has now been extensively demonstrated for various PSDs [18, 33]. PRCs are stable isotope labelled compounds absent in the sampled phase and added to the receiving phase prior to exposure. The mass of these compounds remaining in the PSD can be used to correct for uptake rate-limiting factors and infer the  $R_s$  and the extent of equilibration [18]. Often PRCs are isotopically labelled analogues of target HOCs. However, PRCs can be other suitable labelled analogues or HOCs (not present at the exposure site) [26, 34, 35]. The dissipation rate is dependent on analyte  $K_{sw}$ , less hydrophobic analytes may dissipate entirely, whilst compounds with high  $K_{sw}$  values may not dissipate sufficiently to calculate  $R_s$  values during a deployment [18]. Booij and Smedes [18] developed the now consensus method (unweighted non-linear least-square regression) to extrapolate the data obtained from the use of PRCs to estimate  $R_s$  and the extent of equilibration. This improved on previous methods by reducing the analytical bias found by the use of individual PRCs.



## 2.5 *Uptake rate-limiting environmental factors*

Rate-limiting environmental factors affecting the uptake of HOCs to PSDs can be divided into those which influence mass transfer resistance by the presence of an intermediary phase at the sampler-water boundary (e.g. biofouling or water boundary layer (WBL)) and the influence the physiochemical condition of the aqueous phase (e.g. temperature) may have on mass transfer [19]. It is the exposure and compound specific effects of these factors that necessitate the calibration of PSDs [36]. For example, an increase in temperature causes an increase in  $R_s$  [37]. For very hydrophobic HOCs, rate-limiting factors at the sampler-water boundary have the greatest influence on  $R_s$  and for HOCs with a low  $K_{sw}$ , temperature is likely to be rate-limiting [8].

## 2.6 *Fouling*

Natural waters contain a variety of microbial flora and fauna able to colonize virgin surfaces, including PSDs [38]. Once colonized a biofilm may form, further incorporating higher organisms (e.g. annelids and crustaceans), colloids and other deposited particles from the water column as it develops. Formation and community structure of fouling films is dependent on the composition of sampled waters. Factors include number of microorganisms with potential to form biofilms and the concurrent presence and abundance of substrates for cell metabolism [39]. Other factors include the ease of colonization of the surface of the PSD for cell/particle attachment, competition and predation between members of the biofilm and the presence of any added biocidal compounds within the PSD [39, 40]. Accordingly, the thickness and composition of fouling films are never uniform. The formation of biofilms between aqueous and receiving phases may increase resistance to mass transfer of HOCs. It is therefore necessary to quantify (with PRCs) any rate-limiting effect attributable to fouling during exposures [41–43]. A review of biofilm formation on aquatic microplastics introduces the possibility that certain HOCs may be metabolized within biofilms [39]. The potential for metabolism of HOCs may differ between aqueous media [44]. Theoretically, such metabolism could obfuscate PRC correction by suppressing HOC accumulation in the receiving phase or altering the dissipation rate of biodegradable PRCs such as DDT [45] or certain PAHs [46]. For instance, if the rate of metabolism creates a functionally greater difference in chemical potential at the biofilm-sampler boundary than generated through PRC transport/elimination through biofilm and WBL phases alone, the dissipation rate would increase. Whilst neither effect has been confirmed to date, Allan and Jenssen [47] observed anisotropic exchange favoring dissipation of PRCs in heavily fouled PSDs and decreased  $M_s$  up to a factor of 27 for certain PAHs, when compared

to co-deployed unfouled devices. This was attributed to the high refractory carbon content of suspended particulate matter (SPM), thought to predominate in the fouling film. This favoured mass transfer from the sampler to the fouling layer and was not thought to result from bacterial metabolism. It should be emphasized that the use of PRCs to correct for the influence of fouling has been demonstrated [40], and remains appropriate. However, in the rare cases where extreme fouling causes anisotropic exchange, the PRC dissipation profile may not indicate this bias [47]. Sampling of material contained on heavily fouled PSDs with SPME (coated with same polymer as the fouled PSD) for the presence of PRCs and HOCs could indicate whether fouling has caused a bias. Determining whether metabolism of sampled HOCs has taken place in the biofilm may be difficult. Readmittance of isotope labelled transformation products of PRCs such as DDD (formed by microbial metabolism of DDT in sedimented material) to LDPE PSDs, has been demonstrated by Tcaciuc et al. [45]. Comparison of fouled and unfouled PSD extracts in toxicological analysis may help identify the metabolism of sampled HOCs through the presence/absence of triggered toxicological endpoints, without prior knowledge or need to identify HOCs.

### ***2.7 Coupling passive sampling to toxicological and qualitative chemical analysis***

To determine the risk posed to biota by pollutant mixtures, tools to reduce the complexity of the sampled matrix and to identify adverse effects are required (effect assessment) [48]. Targeted analysis only focuses on regulated HOCs or those with known or suspected presence/toxicity. This approach neglects the majority of the potentially thousands of chemicals present [49]. Effect assessment workflows vary and may consist of separation, enrichment, dilution, biotesting, analyte fractionation and confirmation steps. Workflow design must be carefully formulated to avoid discrimination of compounds and ideally biotesting, analyte confirmation as well as fractionation or dilution will be iterative and tiered, such as in effect driven analysis (EDA) [49]. In EDA the first tier will identify the mode(s) of toxic action (MoA) with each iteration/tier further resolving the contribution of HOCs exerting the same MoA in mixtures. Biotesting identifies MoA through a range of bioassays. These are *in vitro* or *in vivo* bioanalytical tests eliciting an observable biological response when a toxicological endpoint is triggered [50]. Fractionation of HOCs is generally achieved through chromatographic separation [51]. Analyte confirmation is variously target, suspect or unknown, with primary, ancillary or no quantitation.

Passive sampling in effect assessment of HOCs is nascent [52–60], likewise qualitative screening of PSD extracts is developing [53, 61–63]. A review by Brack et al. [49] highlighted the potential for analysis of extracts from PSDs operated in the equilibrium regime as ‘model organisms’ mirroring the bioaccumulation profile of HOCs. Limitations presented, included long equilibrium times in water and the limited extract available for analysis. Restriction of kinetic PSDs to qualitative assessments was recommended due to incongruent HOC profiles among receiving, aqueous and biotic phases. Variability in data obtained from passive sampling has been demonstrated to result from differences in the  $K_{sw}$  values used [18], and inter-laboratory inconsistency in analysis and calculation methods [64]. Considering this variability, incorporation of passive sampling into effect assessment workflows should be deliberate. This is to ensure no bias is introduced and if toxicological and/or qualitative chemical analysis of PSD extracts is undertaken, limitations and uncertainty should be determined and reported. Passive dosing with PSDs has been suggested in place of spiking with an extract (whole or fraction), because it eliminates the effect of extraction solvents on the biological system. However, the throughput of the system may be reduced [49]. Claessens et al. [56] found passive sampling coupled to passive dosing proved a complimentary tool in a toxicological study and suggested the inclusion of several PSD designs with a broad selectivity of HOCs, to make any characterization as representative possible.

### **3. Types of passive sampling device**

PSDs for HOCs in aquatic matrices can be broadly split between single-phase polymeric PSDs (e.g. LDPE, polyethersulphone (PES), POM, SPME, SR) and dual-phase devices such as the SPMD and Chemcatcher<sup>®</sup>. Among PSDs for HOCs the extant literature on the SPMD remains the most extensive. However, the application of single phase polymeric PSDs has expanded since the first comprehensive study into the application of LDPE by Adams et al. [65] and the establishment of criteria for polymer selection by Rusina et al. [66]. Among single-phase polymeric PSDs the most publications have been on LDPE and SR with a variety of studies on sampler performance and numerous field applications. Factors influencing the increased popularity of single-phase polymeric PSDs include their simple construction and low cost when compared to the SPMD and Chemcatcher<sup>®</sup>. Since the last review by Vrana et al. [6] other PSDs such as the Chemcatcher<sup>®</sup> and solid-phase microextraction (SPME) have seen a number of publications on sampler performance and calibration with a comparatively limited number on other applications. Whilst the use of several other PSDs has fallen out of favor, notably the membrane enclosed sorptive coating (MESCO), the ceramic dosimeter and solvent-filled

dialysis membranes. It is likely that in the future single-phase polymeric PSDs will remain popular and SPME will be increasingly adopted as has been seen in other areas of sample separation and environmental monitoring. Fig. 1 shows a range of PSDs used for HOCs. Figures of merit of the available hydrophobic passive samplers are summarized in Table 1.

[Figure 1 near here]

[Table 1 near here]

### 3.1 *Semi-permeable membrane devices*

SPMDs were first described in 1990 by Huckins et al. [90]. SPMDs consist of a sealed lay flat tube of thin walled non-porous LDPE containing a thin film of liquid receiving phase. Triolein is conventionally used as receiving phase, as it is the major storage lipid found in most organisms and it has low permeability through LDPE membranes. Other advantages of triolein include the similarity in magnitude, and good correlation of, triolein-water and *n*-octanol-water partition coefficients, and low triolein-LDPE interfacial tension. Non-porous LDPE was selected as the diffusive barrier because; the structure of its polymer chains permit dissolution and uptake of the bioavailable fraction of dissolved and vapor phase HOCs, it is stable in the presence of organic solvents, it is relatively resistant to physical damage and it is widely available [91]. The SPMD was intended to be biomimetic of HOC accumulation in biota. This was not demonstrated, however, as true equilibrium within a PSD is never attained for all HOCs present [26] and because of variability of accumulation in biota [92]. Subsequently, the LDPE was found to contribute significantly to uptake capacity [93]. The major milestone in the development of the SPMD was the development of PRCs [17, 93]. This demonstrated that the rate-limiting step in the uptake of very hydrophobic HOCs into the SPMD occurred at the water boundary layer. In addition, uptake was isotropic, allowing site specific uptake rates in response to differing environmental factors to be accurately interpreted though differential dissipation of PRCs [94]. This approach has since been applied to other aquatic passive samplers. Since 2005 there have been > 150 publications on the SPMD on a variety of applications, more than any other passive sampler of HOCs. Recently, updated experimental  $K_{spmd/w}$  values for a range of HOCs were published [11], which show a linear  $K_{spmd/w} - K_{ow}$  relationship in contrast to the parabolic relationship suggested in the USGS “SPMD Water Concentrations Calculator” [68]. Smedes [11] recommended that in combination with revised  $K_{spmd/w}$ , a different model approach

should be used for obtaining accurate aqueous concentrations from passive sampling measured with SPMD.

### **3.2 *Silicone rubber devices***

SR devices derive from a number of sorptive extraction techniques (e.g. stir-bar sorptive extraction (SBSE), SPME, rods, tubes and sheets) based on silicone polymers [95]. The popularity of this sampler grew with confirmation of the suitability of SR polymers for sampling, solvent extraction and analysis through thermal desorption coupled to cold injection and GC/MS analysis [66]. HOCs have fast diffusion coefficients in this material [20]. Polymer-water partition coefficients and models relating sampling rates to compound properties [13, 19, 22, 75], field testing [23] and the suitability and quality of PRCs [18] have all been evaluated.

Diffusion coefficients within SR are typically 2-2.5 orders of magnitude greater than in LDPE [20]. This fact elevated the use of SR in passive sampling of HOCs ahead of a range of other polymers. SR PSDs typically consist of strips or sheets of silicone elastomers, such as polydimethylsiloxane (PDMS), however, samplers based on rods or SBSE are also used [96–99]. Cleaning and preparation is required prior to deployment to remove artefacts (e.g. oligomers) within the SR polymers, that, if not carefully removed, may cause instrumental interference during analysis [71]. Since 2005, there have been > 80 publication on SR PSDs of HOCs in aquatic matrices. Their adoption by more practitioners is being facilitated through an annual proficiency testing (QUASIMEME) [100].

### **3.3 *Low-density polyethylene devices***

The development of LDPE PSDs began when Booij et al. [93] suggested using the LDPE membrane from a SPMD as a single phase device, i.e. without internal triolein. This was followed by initial field studies by Müller et al. [78]. However, it was not until more extensive field performance studies were undertaken in 2007 [65] that the use of LDPE gathered momentum. Since 2005 > 80 publications have utilized LDPE PSDs for measuring HOCs in aqueous matrices. A number of these studies combined passive sampling of aqueous phases with other phases such as the atmosphere to determine mass fluxes of HOCs between environmental compartments. LDPE comprises long linear polymer chains with short and long branches at approximate intervals of 25 and 50 monomer units respectively. This results in a crystallinity of 35-55% [101]. LDPE diffusion coefficients are lower than SR and higher than POM [20, 66]. LDPE polymers contain less potential artefacts than SR and are more widely

available at lower cost. Absorption of HOCs within LDPE follows the same process as that in other single phase polymeric devices (e.g. SR), with uptake proportional to LDPE-water partition coefficients [101].

### **3.4 Chemcatcher<sup>®</sup>**

The Chemcatcher<sup>®</sup> PSD was developed by Kingston et al. [102]. Unlike other PSDs the Chemcatcher<sup>®</sup> consists of a reusable three-part PTFE body (base plate, retaining ring and transport lid) which houses a commercially available 47mm SPE disk (Empore<sup>™</sup>/AttractSPE<sup>™</sup>/Atlantic<sup>™</sup>) receiving phase overlain with a DM. The choice of receiving phase and DM is made on the basis of which have the required selectivity and sensitivity for the analyte(s) being monitored. Since the original development of the Chemcatcher<sup>®</sup>, the design of the device has undergone several iterations. The receiving phase in earlier designs sat within a recess. More recent designs have removed this recess to increase uptake rates for HOCs (and other analytes) by reducing the length of the effective diffusional path between bulk and receiving phases [103]. Since 2005 improvements have been made to sampler performance. These included the combination of a C<sub>18</sub> receiving phase and LDPE DM to monitor HOCs, and the addition of *n*-octanol to the interstitial space between receiving phase and DM to reduce internal resistance to mass transfer within the device [80]. PRCs to determine the influence of rate-limiting interactions at the membrane-water boundary [36, 82] have been used as well as comparative field trials alongside other PSDs [69]. Despite this, the application of the non-polar Chemcatcher<sup>®</sup> PSD since 2005 has been modest at > 25 publications. One of the limitations is its small surface area (17 cm<sup>2</sup>). Since the sampler body has a set dimension, upscaling of the device to larger surface area is not possible. In order to reach required limits of quantification (analytes in the range of pg L<sup>-1</sup>) multiple devices can be exposed in parallel and sampler extracts combined into a pool. Such an approach is more costly than the use of SR or LDPE.

### **3.5 Solid-phase microextraction**

SPME was developed by Pawliszyn and Arthur [104] as a sensitive, solvent free, economical and easily automatable sample preparation technique. The distinction between on-site sampling and passive sampling with SPME is not always clear. This review only considers applications of SPME where the aqueous matrix is sampled directly (i.e. without, the addition of internal standards or buffers, filtering or agitation of the sampled matrix). Reviews of on-site sampling [105, 106], analysis of water [107], geometry and coatings [108], and future directions [109,

110] provide an introduction to the diversity of SPME designs and applications. SPME PSDs are formed typically of a narrow glass fiber with a thin polymeric (liquid or solid) coating. Uptake capacity is determined by the polymer-water partition coefficient and fiber thickness (related to polymer mass applied on the fiber). Due to the limited polymer mass only a very small mass of analyte is sampled. Typically, extraction is non-depletive of the analyte in the sampled medium and equilibrium can be achieved in a relatively short time (within hours). If it is desirable to extend integration times (for instance when measuring TWA concentrations of HOCs), polymers with a higher capacity or thicker polymer coating can be selected. Another strategy to extend the integrative sampling period is to have the SPME fiber recessed within in a sheath (such as a needle). This strategy has the added advantage of protecting the fiber during deployments and if the sheath has a narrow enough opening, entry of analytes into the diffusion channel within the sheath will be independent of water flow. Another problem encountered with SPME is the interference of macromolecules when sampling complex matrices (such as contaminated water). This can be overcome with the addition of a selective membrane to exclude molecules based on size. This has the further advantage of extending the integrative period and provides the opportunity to fill the diffusion channel beneath the membrane with a liquid with a higher transfer coefficient than the sampled medium thereby decreasing transfer along the diffusional channel further. When adopting these approaches, it is important efforts to extend the integration time do not sacrifice measurement sensitivity, and that field handling and deployment is practicable [8]. Since 2005 there have been > 25 publications on SPME PSDs of HOCs. Most of these have investigated the performance of the device in a range of field applications including the use of PRCs [111] [112]. In future, the application of SPME PSDs is likely to increase, particularly if devices for field application can be made simpler to use (by non-experts), more reliable and easily coupled to automated analytical methods. These advances have already been seen in the application of SPME in clinical, food and forensic sectors [113].

### **3.6 Other polymeric devices**

Non-polar polymers have been shown to be effective single phase PSDs for HOCs, with most research since 2005 investigating LDPE and SR. Other polymers have been investigated (mainly for niche applications) and include polyurethane (PU), ethylene vinyl acetate (EVA), polymethyl methacrylate (PMMA), nylon, PES and mixed polymers (e.g. PDMS/hydrophobic-lipophilic balanced (HLB) sorbents). However, of the > 30 publications since 2005 most publications have investigated POM. POM PSDs are used when it is desirable to deploy the

same polymer in sediments and overlying waters [114]. POM PSDs are made typically of 0.055 mm thick strips (various lengths), which are deployed as an equilibrium PSD over exposures of at least 4 weeks.  $R_s$  in POM are low and uptake is membrane controlled, as the resistance to mass transfer in the polymer is greater than in the WBL. True equilibrium is not reached in POM PSDs during typical exposures, owing to low  $D_p$ , instead an apparent equilibrium in the surface layer occurs [66]. In future, usage of POM is likely to be restricted due to difficulty in interpreting data due to its functionally biphasic uptake kinetics. Mono-phasic mixed polymers are likely to become more popular, as the multiple affinity for analytes extends the range of hydrophobicity over which devices can operate [28, 115].

#### **4. Applications of passive sampling**

All peer reviewed publications where passive sampling of HOCs in aqueous matrices was investigated between the publication of the critical review by Vrana et al. [6] in 2005 and 2019 are reviewed. Fig. 2 presents number of publications by year for the main types of PSDs. An overview of each application is provided. Delineation of the extant literature identified the following main applications:

- i) Monitoring of process efficacy
- ii) Monitoring discharges and dispersal of pollutants
- iii) Chemical speciation, distribution and degradation
- iv) Monitoring spatial and temporal trends
- v) Toxicity and toxicological monitoring
- vi) Calibration and sampler performance
- vii) Monitoring alongside biota
- viii) Comparison of passive sampling devices

In this review, classification of the application was based on the main subject of a given study. Tables 2-7 review applications differentiated by the PSD together with the HOCs monitored and the matrix sampled. A short description of each application is given, and citation(s) provided. Table 8 presents research and reviews of studies comparing the performance of different passive sampling devices or methods.

**[Figure 2 near here]**



#### **4.1 *Monitoring of process efficacy***

PSDs have been used to monitor various processes designed to reduce HOC concentrations in water (17 applications). This included several unusual processes where conventional tools (e.g. grab or spot sampling) may not have adequately captured said reduction. Namely, inference of sorption of terpenes to carbonaceous geosorbents [116, 117], sediment remediation strategies [118–120], and a bioretention cell for stormwater treatment [121]. Passive sampling coupled to target chemical analysis and toxicological analysis was used to investigate operational multistage treatment process for petroleum wastewater and drinking water [122–126]. Most studies investigated municipal wastewater treatment. Here, passive sampling was used to monitor concentrations of HOCs before and after primary, secondary and advanced treatment processes, including alongside biota, active sampling and numerical simulations [127–133]. These studies were all united by analysis of a predefined list of target analytes and/or MoA. One study investigated a potable wastewater reuse scheme incorporating physical, chemical and biological treatment processes [53]. Here, several versions of the Chemcatcher<sup>®</sup> with selectivity for analytes ranging over a broad hydrophobicity were used to screen against > 1000 organic chemicals and to assign toxicity through several bioassays. An effect-based screening approach is particularly suited to wastewater treatment processes where input water composition and the performance of biological processes (driven by unique microbial communities) are temporally and spatially diverse [134]. Legislation such as the EU Urban Wastewater Treatment Directive recognizes this diversity through site specific discharge permits, stipulating compliance with threshold values for oxygen demand, suspended solids and nutrients [135]. This approach neglects emerging pollutants in wastewater such as the HOC nonylphenol [136]. A review of emerging pollutants in wastewater by Petrie et al. [134] highlights the restrictions posed by grab sampling (snap shot only) and flow proportional composite sampling (logistics and chemical stability) and proposes passive sampling as a possible alternative monitoring method.

#### **4.2 *Monitoring of discharges and dispersal of pollutants***

Waters receiving discharges containing HOCs have been monitored widely with PSDs (29 applications). Many of these studies have monitored the dispersal of produced water from offshore oil and gas installations, including alongside biota [137–143]. Other discharges monitored, resulted from paper mills (dioxins and endocrine disrupting compounds (EDCs)) [144, 145], wastewater treatment (triclocarban, synthetic musk's, PCBs and pesticides) [146–154], smelters (dioxins) [114] and human settlements in Antarctica (PAHs) [155]. Dispersal of

pollutants resulting from activities in the water column included, salvage operations (PAHs) [156], oil spills (PAHs) [157] and pisciculture (PCBs and pesticides) [158, 159]. Dispersal through atmospheric deposition was also monitored at varying distances from oil sands operations (PAHs) [160]. Passive sampling was shown to be a suitable tool for monitoring pollutant discharges and their subsequent dispersal. Data obtained through passive sampling was found to be more representative than grab sampling, particularly where discharges were discontinuous, the composition was variable and/or the dynamics of dispersal (e.g. river hydrology) and distance from source complicated timing of grab sampling.

#### **4.3 *Chemical speciation, distribution and degradation***

Due to the limitations of grab samples coupled to remote analysis, PSDs are often favored for this application (28 applications). PSDs have been used to investigate the distribution of HOCs between particulate, dissolved and colloidal phases in rivers [161, 162]. PSDs have been deployed at depth gradients in the water column to elucidate the vertical distribution of HOCs, including the effects of photodegradation of HOCs within SPMDs [163–170]. Remobilization of HOCs from sediment to aqueous phases has been tested in field and laboratory exposures [171–173]. The influence of matrix complexity (organic matter) on dissolved HOC concentrations was investigated in lab exposures [174]. PSDs were deployed in coastal waters and used to predict the concentrations of HOCs in other environmental compartments through equilibrium models [175, 176]. PSDs have been exposed in sediments, pore waters, waters and the overlying atmosphere (gaseous and aerosol) to determine activity gradients across environmental compartments [177–185]. In future, passive sampling is likely to be applied to other emerging questions concerning the distribution of chemicals between environmental compartments. For example, PSDs deployed in the equilibrium regime can be used to understand the partitioning and half-lives of HOCs absorbed to microplastics [186].

#### **4.4 *Monitoring spatial and temporal trends***

Spatial and temporal trends in HOC concentrations have been investigated in many different aquatic matrices (59 applications). Temporal investigations have occurred over time periods (several years) as prolonged as the impoundment of the Three Gorges Dam (China) [187–189]. Other studies have investigated the ability of methods such as grab sampling and passive sampling to integrate sporadic fluxes in HOC concentrations within river catchments [70, 190–207]. Monitoring of spatial trends have occurred at distinct locations and different levels of resolution, ranging from a raft expedition across an ocean [208] to the distribution of HOCs in

surface waters at different altitudes in a mountain range [209]. PSDs deployed in the kinetic regime are typically favored for applications investigating temporal variation, due to their ability to integrate variations in concentration. Equilibrium and kinetic samplers have both been applied in a number of spatial investigations. Current regulatory monitoring programs (surveillance mode), which routinely monitor spatial and temporal trends in concentrations of priority pollutants currently exclude passive sampling. However, the Environmental Quality Standards Directive 2013/39/EU [210] recognizes the potential for future application of passive sampling and promotes method development. Efforts to demonstrate the utility of passive sampling are underway, such as the creation of a strategic global network of passive sampling stations (AQUA-GAPs project) [72, 211]. As methods become more robust and are adopted by more laboratories, the justification for the establishment of environmental quality standard (EQS) for freely dissolved concentrations (as measured by PSDs) in compliance monitoring will increase (see section 5) [10].

#### ***4.5 Toxicity and toxicological monitoring***

PSDs have been shown to be an adaptable tool for assessing the toxicity of HOCs in aquatic matrices (50 applications). A great diversity in study design is observed with applications ranging from the use of SPME as a biomimetic extraction procedure applied to simulated oil spills [212, 213] to using Chemcatcher<sup>®</sup> to monitor the exometabolome of fish under different husbandry conditions [214]. Studies have also used sampler extracts in effect-based assessment, such as EDA with iterative fractionation coupled to bioassays (e.g. microtox, AhR agonist, Vtox) to determine HOC toxicity. Recognition of the need to quantify the toxicity of HOC mixtures is increasing. The working group on the Water Framework Directive (Chemicals) have proposed the adoption of holistic monitoring incorporating analytical and effect assessment and the establishment of EQS for groups of substances exerting the same MoA [215]. The EU SOLUTIONS project investigated the suitability of passive sampling and spot sampling to toxicologically profile a European river and found each method to be complimentary [54]. Standardization and expanded use of passive sampling in toxicological monitoring is likely in the future [216].

#### ***4.6 Calibration and sampler performance***

Before a PSD can be used in environmental monitoring it is necessary to qualify sampler performance with numerous laboratory and field studies have been undertaken to this end (98 applications). The reliability of data obtained from passive sampling is contingent on accurate

values for analyte specific coefficients describing kinetics and partitioning in bulk and receiving phases. Equilibrium polymer-water partition coefficients have been determined through PSD exposures in reference media and co-solvent solutions [14, 75, 217]. Polymer diffusion coefficients have been investigated through film stacking experiments and exposures that overcome the rate-limiting effect of the WBL [12, 20, 218–220]. In addition, several theoretical models have been developed which attempt to predict these values (both  $K_{sw}$  and  $D_p$ ) from first principles [30, 221–223]. Building on these fundamentals, other studies investigated the influence of polymer selection [66], formulation [95], thickness [89] and PSD geometry [103] on performance. Rate-limiting environmental factors such as temperature [224, 225], fouling [40, 42, 43, 47, 202] and flow velocity [224, 226] and the suitability and data quality of PRC correction [18] have been extensively reported. Several inter-laboratory studies have demonstrated between laboratory variability greater than within laboratory variation, with still greater degrees of variation at lower concentrations. This has been attributed to analytical and calculation errors [64, 227]. The uncertainty of results obtained from passive sampling are approximately a factor of two [69], and efforts to improve data quality assurance and control are ongoing.

#### **4.7 Monitoring alongside biota**

PSDs have been deployed alongside trophically diverse biota species to assess bioaccumulation and biomagnification of HOCs (29 applications), including the development of models [29, 228–230]. Studied biota included, bivalves, fish, crustaceans, insects, annelids and seagrass. Captive and resident biota have been used with approaches ranging from analysis of caged mussel tissues and co-deployed PSDs [137–140, 142], to PSDs deployed on the surface of fish [231]. Extrapolation of concentrations in biotic phases is best described for biota at lower trophic levels with increasing uncertainty at higher positions. Analysis of PSD extracts is considerably simpler than biota tissues or lipids, likewise passive sampling eliminates the need to collect or introduce representative biota. Biota monitoring is permissible in the Water Framework Directive if spot sampling cannot achieve the required sensitivity and  $EQS_{biota}$  has replaced  $EQS_{water}$  for several priority substances [210]. Given the inherent limitations of biota monitoring, passive sampling may represent an improvement in method reliability [232]. Updated guidance acknowledges this and permits the development of methods using passive sampling as part of a tiered approach, to prioritize biota monitoring at subsequent tiers [233] e.g. the tiered sampling approach proposed by Miège et al. [32].

[Table 2 near here]

[Table 3 near here]

[Table 4 near here]

[Table 5 near here]

[Table 6 near here]

[Table 7 near here]

[Table 8 near here]

## 5. Incorporation of passive sampling into regulatory monitoring of HOCs

Improvement and standardization of passive sampling methodologies to enable their inclusion in regulatory monitoring has been a goal of practitioners for some time. This goal was advanced in 2011 with the publication of ISO 5667-23:2011 [380]. A 2015 workshop involving representatives from academia, industry and regulatory agencies, discussed this theme and agreed the next steps towards inclusion in regulatory monitoring [32]. These seek to ensure quality assurance and control (i-vi) and demonstrate method applicability (vii-ix):

- i) Uptake should be absorption based with sufficiently high  $K_{pw}$  and  $D_p$ , to allow for good analyte accumulation and WBL controlled uptake.
- ii) Development of harmonized guidelines for, measurement of  $K_{pw}$  and  $D_p$ , PRC correction and calculation of  $C_w$  with validated models.
- iii) Provision of certified reference materials (standard spiked polymers).
- iv) Commercial availability of passive sampling products.
- v) Determination of lipid-polymer partition coefficients to enable conversion of  $EQS_{\text{biota}}$  from  $C_w$ .
- vi) SR and LDPE are best candidate PSDs, however, there are currently no commercial suppliers.
- vii) Multi-phase inter-laboratory studies to test participant proficiency, then compare and validate methodologies for field deployments, analysis and  $C_w$  calculation (for hydrophobic EU Water Framework Directive priority substances).

- viii) Example field deployments comparing the application of passive sampling alongside grab sampling and biota monitoring.
- ix) Development of assessment criteria in relation to EQSs.

The obstacles these steps seek to overcome have been discussed by others [4, 10, 232, 360], particularly data quality requirements. A comprehensive description of which is provided by Booij et al. [10]. Progress towards inclusion in compliance monitoring continues and an initial inter-laboratory study has taken place [64]. As the 2019 Water Framework Directive review approaches, a realignment to holistic monitoring of a water body's chemical status is underway. Discussing this, Brack et al. [215] recommend the adoption of passive sampling in revisions of the Directive - particularly as a proxy or to compliment biota monitoring and encourage the development of quality assessment criteria and procedures to convert passive sampling measurements into EQS.

## **6. Conclusions and future trends**

Over the last 15 years, theoretical and technical advances within the field of passive sampling have incrementally increased the utility and performance of the method with respect to monitoring HOCs in aqueous environments. These advances have been derived from the efforts of a global, but relatively small, research community. Several devices have sustained interest and remain in use, however, as novel devices have been proposed and adopted, others have fallen into obsolescence. Trends in the appropriateness of the remaining devices for various applications have emerged. It was expected that the commercial availability of SPME would reduce the barriers to its adoption in the passive sampling of aqueous phase HOCs. However, only a modest number of applications have been described, focusing on determining the analytical performance of the device. It is unclear why this is the case, particularly considering the established advantages that SPME provides. SPMDs still remain popular, despite their relative complexity of construction and extraction; this seems counterintuitive. The existence of a critical mass of data arising from previous studies may be promoting their adoption in ongoing and in future research activities. Single-phase polymeric PSDs, such as LDPE and SR are becoming increasingly used due their simplicity and high performance and compatibility with simplified analytical methods for their extraction.

The value passive sampling can add to existing monitoring programs is through its representativeness of aqueous concentrations compared to most other methods. However, if passive sampling is to displace incumbent tools, the ease, cost and risk of adoption cannot be prohibitive. Currently the availability of diverse passive sampling products and lack of certified reference materials, prohibits the use of commonly agreed values for water-polymer partition and polymer diffusion coefficients. Until such uniform materials and values are available, capacity building will be restricted and inter-laboratory variation will persist. The use of *in-silico* methods to rapidly determine these values for novel compounds, risks being undermined by lack of consistency in polymer formulation or by ambiguity in polymer properties between batches and over time. This is particularly important, as the growing interest in emerging contaminants will require tools to identify and attribute the toxicity of HOCs alone and in complex mixtures. This could lead to the expanded use of passive sampling coupled to qualitative chemical and toxicological analysis in effect-based screening approaches.

The accessibility of passive sampling as a monitoring tool is reduced by the current lack of consolidation and clarity in device design/application. This may also reduce the commercial incentive for suppliers to introduce certified reference materials. If pursued in isolation, future improvements to sampler performance through optimization of device design are unlikely to increase the wider appeal of passive sampling. Sacrificing a component of performance for straightforwardness (i.e. devices standardized by formulations, dimensions, durations, calculations and reporting for different applications) could increase the adoption in the short-term, augmenting the capacity for improvement in future. It is difficult to see how this could be achieved other than through parallel deployment of standardized devices in ongoing research activities. Routine deployment of LDPE devices to develop a global network of knowledge and proficiency has been proposed in the past and may still hold merit. The timeline for future inclusion of passive sampling in regulatory monitoring programs is uncertain but will rely, however, on legislator acceptance of unconventional tools and validation and demonstration of these methods by practitioners.

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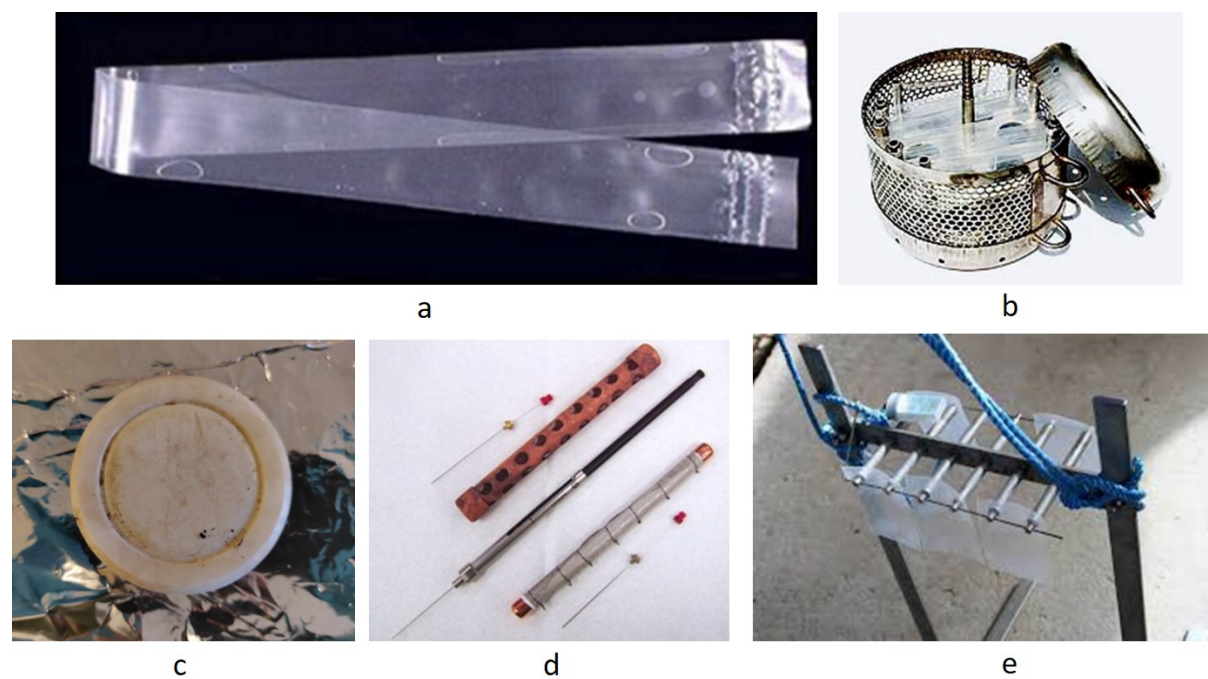
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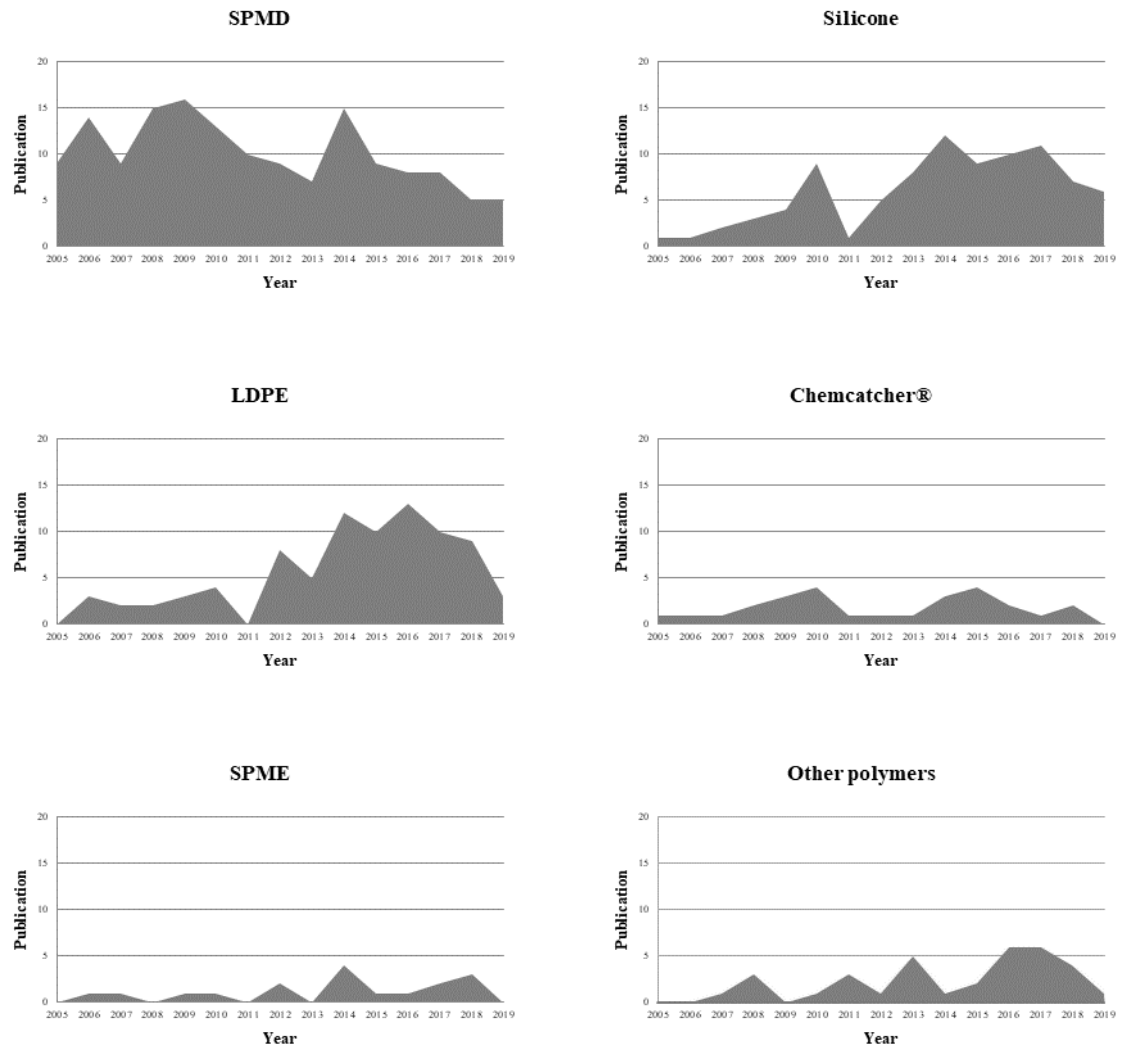
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## Figures and Tables



**Figure 1.** Passive sampling devices together with deployment apparatus used for monitoring hydrophobic organic compounds in water (a) semi-permeable membrane device, (b) semi-permeable membrane device mounted in deployment canister, (c) Chemcatcher<sup>®</sup>, (d) a selection of solid-phase microextraction devices, (e) silicone rubber membranes mounted in a deployment rig.



**Figure 2.** Number of publications per year (2005-2019 in part) that described the use of passive sampling for measuring HOCs in water.

**Table 1**

Comparison of design, performance and availability of PSDs used for monitoring HOCs

\*typical range over which device is used.

**Abbreviations:** DCM, dichloromethane;  $D_p$ , polymer diffusion coefficient;  $\log D_{sw}$ , log distribution coefficient sampler-water;  $\log K_{sw}$ , log partition coefficient sampler-water; LOQ, limit of quantification; 4-NP, 4-nonylphenol; OCPs, organochlorine pesticides; OPFRs, organophosphorus flame retardants; PAHs, polycyclic aromatic hydrocarbons; PBDEs, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; PDMS, polydimethylsiloxane; PSD, passive sampling device;  $t_{eq}$ , time to equilibration; WBL, water boundary layer.

Passive sampling device	Practical aspects			Performance						Availability		R e f
	Specification	Versatility	Preparation and extraction	Uptake and selectivity* (log $K_{ow}$ )	Analyte	$R_s$ (L d <sup>-1</sup> cm <sup>-2</sup> )	Log $K_{sw}$	$t_{eq}$ (days)	LOQ (ng L <sup>-1</sup> )	Commercially available	Cost	
SPMD	Standard - 106 × 2.54 cm LDPE lay flat tube (thickness 70-95 μm) containing 1 mL triolein (sampling area/triolein ratio = 460 cm <sup>2</sup> mL <sup>-1</sup> ). Miniaturized devices or devices with alternate membrane and/or internal receiving phases are sometimes used.	Not reusable. Deployment in protective cage to avoid damage during deployment. Degradation of sequestered analytes may occur if PSD is exposed to sunlight.	Complicated sample clean-up; dialysis, accelerated solvent extraction, microwave assisted extraction ultrasonic extraction. (solvents: <i>n</i> -hexane, <i>n</i> -hexane-DCM, <i>n</i> -hexane-acetone, toluene, cyclohexane or cyclopentane).	WBL controlled (3-10)	PAHs	~0.04-0.40	2.28-5.70	7-< 28	0.03	Yes - constructed devices and component parts can be purchased from several suppliers.	\$\$\$	[11, 37, 67-70]
					PCBs	~0.09-0.40	3.89-7.85		0.01			
					OCPs	~0.17-0.30	2.94-5.70		0.02			
					PBDEs	-	4.48-5.69		0.001			
Silicone rubber	Single phase PSD comprised of multiple sheets (approx. 0.5 mm thick) with a combined sampling area of 300-600 cm <sup>2</sup> .	Can be reused if cleaned. Multiple sheets can be combined to form one device.	Oligomers must be removed before use this takes time and solvent. Soxhlet extraction (solvents: methanol-acetonitrile, 1:2 v/v or methanol).	WBL controlled (3-7)	PAHs	0.007-0.032	3.03-6.24	Weeks-months	0.001	No - Polymer formulation may differ between suppliers.	\$\$	[12, 14, 19, 26, 71-74]
					PCBs	0.006-0.015	3.63-7.12		0.002			
					OCPs	0.150	2.28-6.27		-			
					PBDEs	-	4.29-5.29		-			
					Musks	-	4.29-5.37		40-1500			
					4-NP	-	4.62		570			
					Triclosan	-	3.89		4			
OPFRs	-	3.05-6.36	10									
LDPE	Single phase PSD comprised of multiple sheets (approx. 0.1 mm thick) with a combined sampling area of 324 cm <sup>2</sup> .	Non-reusable sheets can be damaged, use in aquatic, sediment and atmospheric phases well described.	Preparation and extraction easier than SPMD with similar sensitivity and selectivity.	WBL controlled (3-7)	PAHs	0.17-10	2.74-7.84	7-< 56	0.2	No - Polymer formulation may differ between suppliers.	\$	[37, 69, 72, 75-79]
					PCBs	~0.1-0.4	4.19-7.77		3			
					OCPs	~0.15-0.22	2.8-5.59		-			
					PBDEs	-	4.2-7.6		-			
Chemcatcher®	PTFE housing 47 mm C <sub>18</sub> receiving disk	Reusable (housing)	Ultrasonic bath (5 min) using acetone (5	WBL controlled,	PAHs	0.009-0.325	3.66-5.4 (log $D_{sw}$ )	< 28 - (14	0.1-83	Yes - Housing,	\$\$	[36, 69, 80-83]

Passive sampling device	Practical aspects			Performance						Availability		R e f
	Specification	Versatility	Preparation and extraction	Uptake and selectivity* (log <i>K<sub>ow</sub></i> )	Analyte	<i>R<sub>s</sub></i> (L d <sup>-1</sup> cm <sup>-2</sup> )	Log <i>K<sub>sw</sub></i>	<i>t<sub>eq</sub></i> (days)	LOQ (ng L <sup>-1</sup> )	Commercially available	Cost	
	(600 µL) and 450 µL <i>n</i> -octanol, overlain with LDPE DM, sampling area = 17 cm <sup>2</sup> .	only). Multiple devices may be needed to achieve low LOQ.	mL) followed by 5 min in 50:50 (v/v) ethyl acetate: 2,2,4-trimethylpentane (5 mL).	<i>n</i> -octanol added to increase internal diffusion (3.5-7)	PCBs	0.19-0.89	-	days linear for most compounds)	-	DM and receiving phases can be purchased.		
PBDEs					0.15-0.58	-	0.04-1					
OCPs					0.44-0.81	-	0.2-40					
SPME	1 cm long, 7, 30 or 100 µm thick sorbent coated fibre (typically PDMS) which may be recessed in a sheath. Other SPME may be rods of larger dimension.	Reusable (damage may occur over time, due to thermal desorption).	Thermal desorption (head space), this can be automated and requires no solvent. Matrix effects common and LOQ is higher than other PSDs. No extract retained so reanalysis not possible.	SPME sheath can be altered to control uptake (2-7, PDMS)	PAHs	0.005-6.6 (mL d <sup>-1</sup> )	3.32-5.13	>1-63	-	Yes - Can be purchased, used widely in sample separation.	\$\$	[22, 23, 79, 84, 85]
					PCBs	5.68-6.48 (mL d <sup>-1</sup> )	4.09-7.05		300-8000			
POM	Long strips (approx. 0.055-0.5 mm thick), length tailored to deployment needs.	Deployed directly in the water column, no cages or housing needed.	Cleaned by sequential extraction in <i>n</i> -hexane followed by methanol for 2 h with extraction in 1:1 acetone: <i>n</i> -hexane, or other solvent such as DCM. Extraction can be aided by sonication and may need to be repeated to enhance recovery. Analyte migration in stored PSDs can bias results. Very clean extracts due to low <i>D<sub>p</sub></i> .	Diffusion (polymer) controlled (3-7)	PAHs	-	3.23-5.15	> 1-119 (> 1 month for many compounds)	> 0.01-1	Yes - Conditioned polymer strips can be purchased.	\$\$-\$\$\$	[23, 72, 79, 86-89]
					PCBs	-	4.44-6.2		> 0.001-0.08			
					OCPs	-	3.67-5.66		-			

**Abbreviations (Table 2-8):** BTEX, benzene, toluene, ethyl benzene and xylene; CFIS, constant flow integrative sampler; cVMS, cyclic volatile methylsiloxanes; DGT, diffusive gradient in thin films; DOM, dissolved organic matter; FRs, flame retardants; GC, gas chromatography; HCBd, hexachlorobutadiene; HDPE, high-density polyethylene; HPLC, high-performance liquid chromatography; LC, liquid chromatography; MS, mass spectrometry; OPEs, organophosphate esters; PA, polyacrylate; PAN, polyacrylonitrile; PC, polycarbonate; PFASs, polyfluoroalkyl substances; POCIS, polar organic chemical integrative sampler; PS, polystyrene; PPCPs, pharmaceuticals and personal care products; PU, polyurethane; PVC, polyvinyl chloride; ToF, time-of-flight; TPHs, total petroleum hydrocarbons.

**Table 2**  
Applications of the semipermeable membrane device

Environment	Analytes	Description	Ref
<b>Monitoring of process efficacy</b>			
Drinking water treatment	PAHs	Long- and short-term monitoring at various stages of drinking water treatment through a range of chemical analysis and bioassays.	[122–124]
Wastewater treatment	EDCs; PAHs; PBDEs; PCBs; pesticides; synthetic musk's	SPMDs deployed alongside biota and PSDs (artificial mussels, active sampling, numerical simulations, fish, DGTs and POCIS,) at various stages in primary, secondary and advanced wastewater treatment processes to measure process efficacy through the concentration (freely dissolved and particulate phase) of a range of HOCs including the effects of factors such as of temporal fluctuation in influent quality and climatic conditions.	[127–133]
Petroleum wastewater treatment	PAHs; TPHs	SPMDs used to monitor the efficacy of a range of processes (activated carbon, activated sludge, anthracite and zeolite) in petroleum wastewaters through chemical analysis and ecotoxicological assessment.	[125, 126]
<b>Monitoring of discharges and dispersal of pollutants</b>			
Wastewater treatment	PCBs; pesticides; synthetic musk's; triclocarban	SPMDs deployed, including alongside POCIS, at various locations in raw and receiving waters to identify sources, characterize concentrations and profiles and to describe the fate of wastewater associated HOCs at downstream locations.	[149–152]
Wetlands	PAHs	SPMDs deployed alongside resident wood frog tadpoles in boreal wetlands near Alberta's oil sands. Sampler extracts and collected tadpoles were analyzed to monitor the atmospheric deposition of PAHs in water bodies at varying distances from oil sand mining operations.	[160]
Oil and gas produced water	PAHs	Deployments of SPMDs in impacted marine waters, including alongside POCIS and biota (fish, oysters, scallops and mussels) to monitor the freely dissolved concentration, dispersal and bioaccumulation of HOCs associated with produced water and research and methodological gaps for passive sampling within Norwegian produced water monitoring guidance.	[137–142]
Paper mill wastewater	Dioxins; EDCs	SPMDs deployed at sites upstream and downstream of the effluent outfalls of paper mills on the Androscoggin River (USA) and the Biobio River (Chile), to monitor HOCs produced during the bleaching of paper and pulp, through chemical analysis and bioassays of SPMD extracts respectively.	[144, 145]
Oil spill (marine waters)	PAHs	SPMDs deployed at three locations underneath ice flows over a six-day period following a 7000 L oil slick (fresh crude oil) in the Barents Sea, to study oil transport and oil, water, ice interactions.	[157]
Coastal waters (salvage)	PAHs	SPMDs and DGTs deployed at sites in near the Costa Concordia shipwreck (Italy) to monitor the release and dispersal of PAHs and trace metals over each stage of the 2.5-year salvage operation.	[156]
Pisciculture	PCBs; pesticides	SPMD were deployed at sites upstream and downstream of a salmon spawning site on the Credit River (Canada) to determine whether the death and decay of salmon after spawning causes the release to the aqueous phase of bioaccumulated non-polar compounds at detectable concentrations.	[158]
<b>Chemical speciation, distribution and degradation</b>			
River waters	PAHs; PBDEs; PCBs	SMPDs used alongside methods including grab sampling and statistical analysis of land use to attribute phase specific (freely dissolved, apparently dissolved and particulate phase) sources, occurrence, concentrations and fate of HOCs within river catchments.	[161, 162]
River, lake, coastal and marine waters	EDCs; PAHs; PBDEs; PCBs; pesticides; synthetic musk's	SPMDs, including alongside other PSDs such as POCIS and XAD-2 resin were deployed at depth intervals in the water column at various sites to establish the vertical distribution in the bioavailability, concentration and photo-degradation (within SPMDs) of a range of HOCs.	[163–166]

Environment	Analytes	Description	Ref
Coastal waters	Dioxins; PAHs; PCBs	SPMDs deployed near activities causing disturbance of sediments (trawling and sediment relocation) to determine whether said activities facilitate transfer of HOCs from the sediment to aqueous phase.	[171, 172]
Calibration tank	PAHs	The bioavailability of HOCs was tested in a closed system by deploying SPMDs and <i>Daphnia magna</i> at a range of different organic matter concentrations.	[174]
Calibration tank	PAHs	The effect of <i>Nereis diversicolor</i> bioturbation on the remobilization of PAHs from the sediment to aqueous phase was determined through measuring release fluxes and mass transfer coefficients with SPMDs in laboratory exposures.	[173]
<b>Monitoring spatial and temporal trends</b>			
River waters	Dioxins; PAHs; PBDEs; PCBs; pesticides; synthetic musk's	SPMDs deployed, including alongside other methods and PSDs (grab sampling, POCIS and DGT), at various locations on a number of rivers to measure the spatial and temporal variation in occurrence and concentration of a range of HOCs through chemical analysis and bioassays.	[70, 190, 200–206]
Coastal waters	Dioxins; halogenated organic chemicals; PAHs; PCBs; pesticides	SPMDs deployed, including alongside other PSDs (Chemcatcher <sup>®</sup> , POCIS and SR) and grab sampling, in coastal waters to measure the spatial and temporal variation in occurrence and concentration of various HOCs.	[234–240]
Impounded reservoir	PAHs; PCBs; pesticides	PRC spiked SPMDs deployed, in the Three Gorges Dam (China) over the impoundment of the reservoir and after impoundment to measure the spatial and temporal variation in HOC concentration.	[187–189]
Marine waters	PAHs; PBDEs; PCBs; pesticides	SPMDs, grab samples and fish bile collection used to monitor a range of HOCs during the Norwegian Tangaroa balsa raft expedition in the Pacific in 2006.	[208]
Lake waters	PCBs; synthetic musk's; triclosan	SPMDs alone and alongside POCIS were deployed at lake sites to monitor the bioavailable concentration of a range of HOCs, this was compared to results obtained from grab samples or assessed in relation to limnology and pollution sources of sampled waters.	[241, 242]
Ground and surface waters	Dioxins; PAHs; PCBs; pesticides	Monitoring of temporal and spatial variation in HOC concentration with SPMDs, alone and alongside POCIS, in karstic systems.	[243, 244]
<b>Toxicity and toxicological monitoring</b>			
Drinking water treatment	Dioxins; PAHs; PCBs; pesticides	Toxicity assessment of water with extracts from SPMDs deployed at different stages of drinking water treatment using microtox, AhR agonist, cytotoxic and genotoxic bioassays.	[59, 245, 246]
River waters	Dioxins; PAHs; PCBs; pesticides; PPCPs	Ecotoxicological risk assessment using a variety of methods such as active sampling, passive sampling (SPMD and POCIS), Vtox, biota monitoring (mussel and fish tissue), to monitor for a variety of HOCs and biomarkers of exposure in various rivers.	[247–251]
River Waters	Dioxins; EDCs; PAHs; PCBs; pesticides; triclosan	Toxicological assessment of various rivers through chemical analysis (quantitative and screening) and various bioassays of/with PSD extracts (SPMD and POCIS), biota and composite sampling, including bioassay driven chemical analysis and observation of mortality and fertility.	[55, 57, 58, 60, 252–254]
Lake water	Dioxins; EDCs; PAHs; pesticides	SPMDs including alongside biota (caged carp, resident fish and Common toad larvae), deployed in several natural and impounded lakes to evaluate concentrations and toxicity potential of HOCs through combination of chemical analysis, bioassays, and observation of mortality and fertility.	[255–257]
Groundwater	PAHs; PCBs; pesticides	SPMDs used to monitor HOCs in groundwater to determine natural background concentrations and any secondary contamination and the associated toxicity of each.	[258]
Coastal waters	PAHs	SPMDs deployed alone and alongside mussels and analyzed chemically and with several bioassays to monitor the concentrations of a range of environmental toxicants	[259, 260]
<b>Calibration and passive sampler performance</b>			



<b>Environment</b>	<b>Analytes</b>	<b>Description</b>	<b>Ref</b>
Calibration tank	Alkylphenols; PAHs; pesticides; UV filters	Uptake rates and kinetics for PRC spiked and unspiked SPMDs and other PSDs (SR, POCIS and altered SPMDs) are determined in various exposures with known analyte concentration for a range on HOCs, including the influence of a range of factors such as water velocity, DOM, pH, alkalinity, water hardness, fouling and membrane and receiving phase composition.	[42, 43, 73, 261–266]
Marine, river and lake waters	PAHs; PCBs; pesticides	In situ calibration of PRC spiked SPMDs in a range of waters and for various exposures to determine the uptake rates of a range of HOCs, including through deployment alongside biota (crayfish), with addition of antifouling agents, under differing water velocities and the development of predictive models.	[40, 224, 267–269]
Calibration tank, river and lake waters	PAHs; pesticides	Investigations of the performance of variations of the SPMD with altered membranes and/or receiving phase.	[270–272]
Modelled data	PAHs; pesticides	A mechanistic model developed and applied to two previous studies. The effect of volume of sampled water on the equilibration rate constant and laboratory field extrapolation errors are discussed.	[273]
River waters	PAHs; PBDEs; PCBs; pesticides	A method for the fast isolation of HOCs from exposed SPMDs using hexane as an extraction solvent was tested for samplers deployed in various aquatic ecosystems.	[274]
Calibration tank	PAHs; PCBs; pesticides	Calculation of new, and recalculation of reported, polymer water partition coefficients for SPMD and SR, through cosolvent, lipid and aqueous exposures in batch experiments, to facilitate conversion of equivalent analyte concentrations between phases with only partition coefficients.	[11]
<b>Monitoring alongside biota</b>			
Coastal waters	Dioxins; EDCs; PAHs; PCBs pesticides	Monitoring with SPMD and biota (mussels, oysters, Atlantic cod) for a range of HOCs in coastal waters with a range of climates and varying uses (e.g. shipyards, marinas, harbors, estuaries, estuarine lagoons), including concurrent sediment monitoring.	[275–280]
Calibration tank	PAHs	SPMDs exposed alongside biota (Atlantic cod and <i>Daphnia magna</i> ) to known analytes in an exposure system to determine correlation of analyte accumulation in biotic and sampler receiving phases.	[281, 282]
Lake and river waters	PAHs; PBDEs; pesticides; triclosan	Monitoring with SPMD and trophically diverse biota (various fish) for a range of HOCs in lake waters to determine concentrations and in sampled waters and at different trophic levels (biomagnification)	[228, 283–286]
Coastal waters	PAHs; PCBs; pesticides	Nine studies published in the decade prior to 2006 where mussels and SPMDs used to monitor HOCs were evaluated, to investigate differences and similarities between the sampling methods including development of a model to compare concentration ratios and comments on method reliability.	[92]
Marine waters	PAHs; pesticides	PSDs including SPMDs, POCIS, and DGT deployed alongside biota (mussels and fish) to monitor a range of HOCs that may bioaccumulate in seafood flesh.	[287, 288]

**Table 3**

Applications of the silicone rubber passive sampling device

Environment	Analytes	Description	Ref
<b>Monitoring of discharges and dispersal of pollutants</b>			
Wastewater	PCBs	SR deployed at outfalls and at downstream locations in receiving waters, including alongside other PSDs (LDPE) to monitor the occurrence, concentration and fate of a range of HOCs found in treated wastewater through targeted and qualitative chemical analysis.	[148, 153]
Calibration tank	PAHs; PCBs	SR and naked Empore™ C <sub>18</sub> disks were deployed to monitor simulated discharges from the offshore oil and gas industry during three exposures of fourteen days, each corresponding to one of three discharge scenarios (continuous, discontinuous and short abrupt).	[143]
Coastal waters	Dioxins	Active sampling and SR and POM PSDs were deployed in pore water and the overlying water column at sites in Frierfjord (Norway) to determine the freely dissolved equilibrium concentration of dioxins originating from a historic Mg smelter.	[114]
<b>Chemical speciation, distribution and degradation</b>			
Coastal waters	PAHs; PCBs	SR deployed in the water column at a number of points along the Belgium coast to monitor the freely dissolved concentration of a range of HOCs. Equilibrium models used to predict concentrations in sediment, suspended particulate matter and biotic phases.	[175]
Coastal waters	PAHs; PCBs; pesticides; synthetic musk's; triclosan	SR exposed alongside resident clams and caged mussels in mangroves (Singapore) and were measured alongside sediments to determine the distribution of HOCs between environmental compartments.	[176]
<b>Monitoring spatial and temporal trends</b>			
River waters	PAHs; PCBs; pesticides; phthalates	SR alongside Speedisk PSDs were used to monitor the spatial and temporal variation in sources, occurrence, concentrations and fate of a range of HOCs within various river catchments.	[191–195, 207]
Coastal waters	PAHs; PCBs; pesticides	SR and other PSDs (LDPE, Chemcatcher® and SPMD) were deployed in various exposures at coastal sites to investigate temporal and spatial trends in the concentration of a range of target HOCs and to screen for the occurrence of HOCs and use of detection frequency to inform a fugacity model.	[61, 237, 289]
Various (global)	Various	Proposals for the global aquatic passive sampling network (AQUA-GAPS) employing SR and LDPE PSDs to monitor HOCs at strategically important locations.	[211]
<b>Toxicity and toxicological monitoring</b>			
River waters	EDCs; PAHs; PCBs; pesticides	SR and other PSDs (LDPE, POCIS-pharms, POCIS-pest) and biota (mussels), deployed in various rivers and sampler extracts used in chemical and toxicological analyses and bioassays, including in EDA using HPLC fractionalization coupled to bioassays as a way of profiling anti-androgenic activity.	[216, 290–295]
Coastal waters	PAHs; PCBs; pesticides	SR including alongside POCIS deployed in field exposures and sampler extracts and grab samples underwent chemical and toxicological analysis to assess water quality, including in EDA using HPLC fractionalization coupled to LC-ToF-MS and bioassays (photosystem II activity, microalgae).	[74, 290, 296–298]
Marine waters	Toxicological only	SR used to sample marine waters and then passively dose an ecotoxicological test medium as part of the development of an approach to aquatic toxicity monitoring.	[56]
Produced water (oil and gas)	PAHs	LDPE and SR exposed to a sample of produced water to determine the concentration of HOCs, with subsequent use of sampler extracts and diluted produced water samples in zebrafish bioassays.	[299]
<b>Calibration and sampler performance</b>			
River waters	HCBD; PAHs; PBDEs; PCBs; pesticides	SR alone and alongside other PSDs (SPMDs, LDPE and POM) underwent various exposures in rivers to monitor a range of HOCs to evaluate the effect of PSD selection, design and deployment strategy on performance. To determine the influence of SPM on the surface of SR PSDs on the reliability of <i>in-situ</i>	[12, 26, 47, 89, 227, 300]

Environment	Analytes	Description	Ref
		sampling rates calculated with PRCs, additionally polymer water partition coefficients derived from laboratory exposures and film stacking experiments were validated.	
Calibration tank	Biotoxins; BTEX; EDCs; HCBd; PAHs; PDBEs; PCBs; pesticides; triclosan; 2-nonylphenol	SR alone and alongside other PSDs (POM, LDPE and SPMD) exposed to known HOC concentrations in various laboratory exposures (in tanks and flow through systems). To evaluate sampler performance, including factors such as polymer formulation, polymer selection, polymer surface area, salinity, temperature, water velocity, PRC accuracy, accuracy of passive flow monitors, composition of sampled phase and the difference between theoretical and experimentally derived partition coefficients.	[13, 19, 301–305, 22, 73, 75, 89, 95, 225, 226, 264]
Film stacking	Various	Film stacking experiments were undertaken to measure diffusion coefficients within SR and LDPE polymers to interpret and understand mass transfer resistance within each polymer during passive sampling of waters including development of a QSAR model.	[20, 218, 219, 221]
Methods	PAHs; removal of oligomers	Novel methods for sampler preparation, cleaning, extraction evaluated for SR sheets and rods.	[306, 307]
Calibration tank	Various	SR-water partition coefficients were determined for various HOCs through cosolvent methods (water/methanol) and compared to $\log K_{ow}$ .	[14, 217]
Calibration tank	PAHs; PCBs; pesticides	Calculation of new, and recalculation of reported, polymer water partition coefficients for SPMD and SR, through cosolvent, lipid and aqueous exposures in batch experiments, to facilitate conversion of equivalent analyte concentrations between phases with only partition coefficients.	[11]
Coastal and river waters	PAHs; PCBs	Performance of deuterated and $^{13}\text{C}$ labelled PRCs during equilibrium monitoring of fresh and marine waters was investigated through field exposures of LDPE and SR.	[308]
<b>Monitoring alongside biota</b>			
Sediments and overlying waters	PCBs	SR and LDPE deployed in various lake and river sediments and overlying waters alongside biota to monitor the concentration of a range of HOCs and assess the bioaccumulation prediction capacity of each device for biota at a range of trophic levels.	[229, 309]
River waters	PAHs; PCBs; pesticides	SR deployed in rivers alongside resident macroinvertebrates and attached to captured fish (flathead catfish) to measure water concentrations and exposure of biota through chemical analysis of PSD extracts and biota tissues for a range of HOCs.	[231, 310]
Coastal waters	PAHs; PCBs	SR alone and alongside other methods (DGT, sediment and grab samples) deployed with biota (mussels and seagrass) in coastal waters to monitor concentrations and bioaccumulation of HOCs.	[220, 311]
Calibration tank	PAHs	Partition equilibrium and extraction rates for freely dissolved PAHs and SR and blackworms were determined to better understand the principles of bioconcentration of HOCs in aquatic organisms.	[312]

**Table 4**  
Applications of the low-density polyethylene passive sampling device

Environment	Analytes	Description	Ref
<b>Monitoring of process efficacy</b>			
Calibration tank	Limonene; pesticides; pinene	LDPE calibrated for HOCs and exposed in three phase systems (LDPE, water and sorbent), to infer analyte uptake to several carbonaceous geosorbents and virgin and regenerated activated carbon.	[116, 117]
<b>Monitoring of discharges and dispersal of pollutants</b>			
Wastewater	PCBs; PFASs	LDPE deployed at outfalls and at upstream and downstream locations in receiving waters (rivers), including alongside other PSDs (SR) and in the overlying atmosphere to monitor the occurrence, concentration and fate of HOCs found in treated wastewater.	[146–148]
Lake waters	PAHs	LDPE deployed in water and the overlying atmosphere at locations around the lower Great Lakes (USA) to monitor the freely dissolved and gaseous concentration of PAH and determine the influence of proximity to sources (population centers and wastewater treatment works effluent) and impact of vectors (river discharges and precipitation) on measured concentrations.	[313]
Antarctic lakes	PAHs	LDPE deployed in seven lakes in Antarctica to track human footprints through PAH concentrations.	[155]
<b>Chemical speciation, distribution and degradation</b>			
River waters	PBDEs; PCBs	LDPE deployed in various environmental compartments including air, water sediment, and pore water, to study the partitioning and mass fluxes of various HOCs between compartments.	[181, 182]
Marine waters	PAHs; PBDEs; PCBs; pesticides	LDPE, POM and SPME deployed along depth gradients and in sediments and overlying waters to investigate the vertical distribution and mass flux between environment compartments of HOCs.	[167, 168, 185]
Lake waters	FRs; PBDEs; PCBs; pesticides	LDPE deployed in water and the overlying atmosphere at sites in the Great Lakes (USA and Canada), to determine the spatial variation in concentration, partitioning and mass fluxes of a range of HOCs between the air and water compartments.	[178–180]
Coastal waters	Dioxins	LDPE deployed at five sites in the water column and overlying atmosphere in Newark Bay (USA) to monitor concentrations of dioxins and mass fluxes between air and aqueous phases.	[183]
Coastal waters	FRs; PAHs; PBDEs; pesticides; PPCPs	A variety of methods were used to investigate the vertical distribution of a range of HOCs in coastal waters, including grab samples and LDPE deployed along depth gradients and in sediments.	[169, 170]
Marine waters	PBDEs	LDPE passive and active sampling devices were deployed on an east west transect of the tropical Atlantic Ocean in the water column and overlying atmosphere to monitor the spatial distribution in concentration and mass fluxes between air and aqueous phases.	[184]
<b>Monitoring spatial and temporal trends</b>			
River waters	PAHs; PBDEs; PCBs pesticides	LDPE deployed, including alongside other methods (SPMD, POCIS and High volume grab sampling), to measure spatial and temporal trends in HOC sources, occurrence and concentration.	[196–199]
Wetland waters	Pesticides	LDPE deployed in surface waters and the overlying atmosphere to determine the altitudinal variation in the concentration of organochlorine pesticides in mountain ranges in southern Brazil.	[209]
Marine, costal and lake waters	OPEs	LDPE deployed on deep water moorings at a number of locations in the Fram Strait (Canada) and in several surface water sites in lakes and coastal waters of the Canadian Arctic to monitor concentrations of a range of organophosphate esters (OPEs).	[314]
Various (global)	Various	Proposals for the global aquatic passive sampling network (AQUA-GAPS) employing SR and LDPE PSDs to monitor HOCs at strategically important locations.	[72, 211]
<b>Toxicity and toxicological monitoring</b>			

Environment	Analytes	Description	Ref
Coastal waters	PAHs; pesticides; phthalates; synthetic musk's	LDPE exposed in several coastal waters and sampler extracts (without and after iterative fractionalization) were used in several bioassays and analyzed chemically, to determine and attribute associated toxicity for a range of HOCs.	[52, 315]
River waters	EDCs; PAHs	LDPE, SR and POCIS exposed in a number of rivers and sampler extracts were chemically analyzed and used in a range of bioassays including in effect driven analysis (HPLC fractionalization coupled to bioassays) to profile toxicity in sampled waters.	[291, 316]
Produced water (oil and gas)	PAHs	LDPE and SR exposed to produced water to determine the HOC concentrations and subsequent use of sampler extracts and diluted produced water samples in several zebrafish bioassays.	[299]
<b>Calibration and sampler performance</b>			
Coastal, lake and river waters	PAHs; PCBs; pesticides	The performance of LDPE alone and alongside other PSDs (SR, SPMD and SPME), and influence of factors such as use of deuterated and <sup>13</sup> C labelled PRCs, PSD design and deployment methodology, and PRC correction, was tested in a range of field exposures.	[300, 317–321]
Modelled data	Various	Two mechanistic models to predict partition coefficients for HOCs between LDPE and aqueous phases developed using data available in the extant literature.	[30]
Calibration tank	PAHs; PBDEs; PCBs; pesticides	Model to account for non-equilibrium exposure conditions developed based on HOC, uptake rate constants, elimination rate constants and water polymer partition coefficients in batch experiments.	[322]
Modelled data	PAHs; PCBs	A QSAR model was developed to predict the diffusion coefficient for various HOCs in LDPE PSDs.	[323]
Calibration tank and field exposures	Dioxins; PAHs; PBDEs; PCBs; pesticides	The performance of a range of PSDs (LDPE, SR, PU, PMMA, POM and SPMDs) to monitor HOCs based on polymer selection and thickness. Tested in laboratory experiments and selected PSDs were then tested in field exposures, including the development of predictive models.	[65, 76, 89, 324, 325]
Film stacking	PAHs; PBDEs; PCBs	Film stacking experiments measuring resistance to mass transfer ( $D_p$ ) in SR and LDPE.	[20, 219]
Wastewaters	cVMS	<i>In-situ</i> calibration of LDPE spiked with PRCs, for cVMS, during exposure in wastewater effluent.	[326]
Calibration tank	Biotoxins; PAHs; PCBs; pesticides;	LDPE alone and alongside other PSDs (SR and HDPE) were exposed to known HOC concentrations in laboratory exposures (in tanks and flow through systems) to evaluate sampler performance, and the influence of factors such as flow velocity, polymer selection and thickness, PRC accuracy, temperature and salinity, including the development of a predictive model.	[75, 77, 226, 303, 305, 327–329]
<b>Monitoring alongside biota</b>			
Coastal waters	Dioxins; FRs; PAHs; PBDEs; PCBs; pesticides	LDPE including alongside PSDs (POCIS, SPME and DGT) and biota (gulf killifish, mussels and shellfish) deployed in coastal waters to evaluate the application of each method to monitor HOCs and predict concentrations in biota.	[330–334]
River and lake waters	PAHs; PCBs	LDPE alone and alongside SR co-deployed with biota in a range of surface waters and sediments to monitor concentrations of a range of HOCs and assess the bioaccumulation prediction capacity of each device for sampled biota at various trophic levels.	[229, 309, 335]

**Table 5**  
Applications of the Chemcatcher® passive sampling device

Environment	Analytes	Description	Ref
<b>Monitoring of process efficacy</b>			
Wastewaters	Database of 1250 organic chemicals	Bioassays and qualitative chemical analysis (GC-MS screening) coupled to spot and Chemcatcher® (various receiving phases), used to test the efficacy of the processes in a direct potable reuse wastewater recycling system in Antarctica.	[53]
<b>Monitoring spatial and temporal trends</b>			
River waters	PAHs; pesticides	Chemcatcher® (C <sub>18</sub> and SDB-XC receiving phases) deployed on several rivers and analyzed with quantitative and qualitative methods to measure spatial and temporal trends in the presence and concentrations of a range of HOCs, including comparison to grab samples.	[9, 62]
<b>Toxicity and toxicological monitoring</b>			
River waters	EDCs	Chemcatcher® (C <sub>18</sub> receiving phase) deployed in rivers and sampler extracts analyzed for toxicity with luminescence and recombinant receptor reporter gene bioassays.	[336]
Calibration tank	Endogenous fish metabolites	Chemcatcher® (C <sub>18</sub> and SDB-RPS receiving phases) deployed to monitor metabolites excreted by two fish species to characterize the influence of fish husbandry conditions on the fish exometabolome.	[214]
<b>Calibration and sampler performance</b>			
Calibration tank	PAHs; PBDEs; PCBs; pesticides	Chemcatcher® alone and alongside other PSDs (MESCO, SPMD, SR rods and strips) exposed to known analyte concentrations in a range of laboratory experiments to determine sampler performance and the influence of factors such as, composition of the sampled matrix, addition of <i>n</i> -octanol to receiving phase, flow velocity, exposure time, DM selection and sampler design.	[33, 80–83, 103]
Calibration tank and field exposures	Nonylphenol-ethoxylates; nonylphenol	The performance of three versions of the Chemcatcher® (C <sub>18</sub> , SDB-XC and SDB-RPS receiving phases) to monitor nonylphenol ethoxylate and nonylphenol mixtures evaluated in two laboratory trials and subsequent field exposures.	[337]
River waters	PAHs; pesticides	An empirical relationship that allows calculation of <i>in-situ</i> sampling rates in Chemcatcher® ( <i>n</i> -octanol saturated C <sub>18</sub> overlain with LDPE) spiked with PRCs was established through modelling of sampler analyte exchange kinetics determined in previous calibration experiments and through comparison with spot samples during field exposures.	[36]
<b>Monitoring alongside biota</b>			
Coastal waters	PAHs; PCBs; pesticides	Chemcatcher® (C <sub>18</sub> receiving phase) and Ecoscope PSDs were deployed alongside resident biota (mussels) and grab samples at several harbor sites to monitor for a range of HOCs.	[338]
Calibration tank	PAHs; PCBs; pesticides	Chemcatcher® (C <sub>18</sub> receiving phase overlain with either LDPE or PES DM) deployed alongside mussels in a flow through system to evaluate the ability of each method to sequester HOCs.	[339]

**Table 6**

Applications of the solid-phase microextraction passive sampling device

<b>Environment</b>	<b>Analytes</b>	<b>Description</b>	<b>Ref</b>
<b>Monitoring of process efficacy</b>			
Contaminated sediments	PAHs; PCBs	Two sediment remediation strategies (capping and addition of activated carbon) assessed through measurement of HOCs in sediments and overlying waters (with PDMS-SPME) and resident biota.	[118]
<b>Chemical speciation, distribution and degradation</b>			
Marine waters	PCBs	<i>Ex-situ</i> analysis of PCB concentrations in sediments and bottom waters with PDMS-SPME to assess spatial variation in diffusion gradients at the sediment water interface, site specific mixture compositions and baseline toxicity potentials.	[340]
Marine waters	PCBs; pesticides	LDPE and SPME deployed along depth gradients to investigate vertical distribution of HOCs.	[168]
Marine waters	PCBs; pesticides	SPME, LDPE and POM deployed in sediment and overlying water to measure HOC mass flux.	[185]
<b>Toxicity and toxicological monitoring</b>			
Calibration tank	Hydrocarbons (various)	The use of PDMS-SPME fibers as a biomimetic extraction procedure to access bioavailability and predict toxicity of petroleum substances, was tested in a simulated spill and laboratory exposures.	[212, 213]
<b>Calibration and sampler performance</b>			
Calibration tank	PAHs; PCBs; pesticides	PDMS-SPME fibers exposed to known HOCs in laboratory exposures to access sampler performance, including the influence of factors such as, DOM, water velocity, temperature, and to create predictive models, develop new methods for the use of PRCs, new sampler designs and to determine the $K_{OW}$ for several HOCs.	[31, 84, 341–343]
Calibration tank and river waters	PPCPs	C <sub>18</sub> (thin film) SPME PSDs used in lab and field exposures to evaluate the ability to measure TWA concentrations wastewater associated HOCs, alongside HLB SPME PSDs for polar compounds.	[344]
Marine, coastal and lake waters	EDCs; PAHs; PCBs; pesticides	The performance of a range of SPME fibers coatings (acrylate, nylon and SR) and SPME PSD designs to monitor a range of HOCs assessed through field exposures, including comparison to other methods such as LDPE, SPMD and POM, grab samples and biota monitoring.	[85, 112, 308, 320, 345]
<b>Monitoring alongside biota</b>			
Coastal waters	FRs; PAHs; PBDEs; PCBs; pesticides	SPME fibers, POCIS and LDPE were deployed at coastal sites to monitor the concentration and relative abundance of a range of HOCs and compared to concentrations in mussel tissues.	[331]

**Table 7**

Applications of other polymeric passive sampling devices

<b>Environment</b>	<b>Analytes</b>	<b>Description</b>	<b>Ref</b>
<b>Monitoring of process efficacy</b>			
Calibration tank	PAHs; pesticides	Performance of POM and PU determined in batch experiments followed by laboratory scale exposures to test efficacy of a biochar treatment method (contaminated sediments) and bioretention cell (storm water treatment system).	[119, 121]
River waters	PCBs	POM deployed in sediment and the overlying water column at river sites to monitor the bioavailable fraction and mass fluxes of PCBS during sediment remediation with activated carbon.	[120]
<b>Monitoring of discharges and dispersal of pollutants</b>			
Calibration tank and coastal waters	Dioxins; PAHs; PCBs; pesticides	PSDs made from EVA, SR and POM polymers used to monitor the dispersal of HOCs associated with pisciculture (after laboratory calibration), a historic Mg smelter (compared with active sampling) and sediment dredging and relocation.	[114, 159, 346]
Wastewaters	Various	A novel mixed polymer PSD (PDMS and HLB) and POCIS deployed in effluent and receiving waters to monitor the discharge and dispersal of 44 organic compounds with a broad hydrophobicity.	[154]
<b>Chemical speciation, distribution and degradation</b>			
Marine waters	Dioxins; PCBs	POM deployed in the water column and high-volume air samplers in the overlying atmosphere at marine sites to determine the aerosol water distribution of PCBs and dioxins.	[177]
Marine waters	PCBs; pesticides	POM, LDPE and SPME deployed in sediment and overlying water to measure HOC mass flux.	[185]
<b>Monitoring spatial and temporal trends</b>			
Marine waters	Dioxins; PCBs	POM were deployed at two depths (5m above the seafloor and 25m below the surface) at five sites in the Baltic Sea to investigate the spatial distribution in the concentration of dioxins and PCBs.	[347]
Coastal waters	Dioxins; PCBs; pesticides	POM and EVA deployed in a range of coastal waters, including alongside PSDs deployed in the overlying atmosphere to monitor seasonal and spatial trends in various HOCs.	[348–350]
<b>Calibration and sampler performance</b>			
Calibration tank and field exposures	Various	The performance of a range of PSDs (a novel form of POCIS containing a sorbent mixture and nylon membrane, a PES hollow fiber, LDPE, PU, PMMA, POM and SR), in laboratory experiments followed by field exposures to a broad range of HOCs in coastal, river and wastewaters.	[89, 324, 351, 352]
Calibration tank	Various	The performance of a range of PSDs (silicone matrix containing HLB beads, POCIS, PDMS, POM, PDMS stir bars, MESCO and PES tubes) were evaluated in laboratory exposures to organic chemicals over a broad hydrophobicity.	[15, 28, 87, 302, 353]



**Table 8**

Comparison of different passive sampling devices

Environment	Analytes	Description	Ref
<b>Reviews of passive sampling for HOCs in aquatic matrices</b>			
-	-	Reviews on the development and state of the art of a range of PSDs.	[67, 94, 354, 355]
-	-	Review of active and passive sampling (POCIS, SMPDs, LDPE, POM and SR) and analytical consideration and coupled methods such as bioassays.	[356]
-	-	Reviews on advances in analysis of pharmaceuticals in aquatic environments including SPMD, Chemcatcher <sup>®</sup> and SPME PSDs for HOCs.	[357]
-	-	Reviews of methods to access the bioavailability of HOCs in a range of environmental matrices including LDPE, SR, POM, SPMD and SPME.	[88, 230]
-	-	Review of dynamic accumulation processes for HOCs in PSDs (Chemcatcher <sup>®</sup> , SPMD, LDPE, SR-SPME) and trophically diverse biota.	[358]
-	-	Reviews of the use, shortcomings and strengths of passive sampling methods and presentation of the potential for passive sampling in compliance and regulatory monitoring requirements (of HOCs) of the United States, EU and Oslo-Paris Convention for the protection of the marine environment of the North East Atlantic, including scrutiny on the applicability, and uncertainties of the information produced from passive samplers, within this context.	[4, 10, 359–361]
-	-	Review of the use of SR in analytical chemistry including uses in SR PSDs, and PDMS SPME and stir bar PSDs.	[98, 360, 362]
-	-	A review of polyethylene water equilibrium partitioning constants for HOCs in the extant literature and the implications for the use of polyethylene receiving phases in passive sampling.	[101]
-	-	Reviews of the principles, calibration, preparation, field applications and analysis of various chemicals with the Chemcatcher <sup>®</sup> .	[363–365]
-	-	Position papers on the current state of the art for environmental monitoring with PSDs in aquatic matrices and future challenges.	[32, 366]
-	-	Review of current calibration methods in passive sampling, including HOCs in aquatic matrices.	[21]
-	-	Reviews of SPME for on-site sampling, in analysis of water samples and geometries and coatings.	[105–110]
<b>Comparison of different passive samplers</b>			
Stormwater	PAHs	SPMDs and LDPE PSDs (of varying surface area) and grab sampling used to monitor the occurrence and concentration of HOCs in stormwater.	[367]
Modelled data and GCxGC	Various	A model to predict partitioning behavior and diffusion coefficients of HOCs in biota and PSDs (POM, LDPE, PA, PU, PDMS and SPMD) developed and validate based on GCxGC retention times and data from the extant literature.	[29]
Modelled data	Various	Novel statistical interpretation of the error structure of PRC data to enable an improved method for estimating <i>in-situ</i> sampling rates of HOCs during passive sampling with various devices (including SPMDs, LDPE, SR and Chemcatcher <sup>®</sup> ).	[18]
Ground water (managed aquafer recharge)	Pesticides; synthetic musk's	Monitoring of HOCs in an urban storm water recycling system, using SPMD, XAD resin, SR and Chemcatcher <sup>®</sup> PSDs alongside passive flow monitors deployed at sample wells located at varying distances from the recharge well.	[368]
River waters (field exposures and flow through system)	Alkylphenols; PAHs; PBDEs; PCBs; pesticides; synthetic musk's	Various exposures in river waters of multiple PSDs (SR sheets and rods, LDPE, SPMD, POM, PVC, PU, PC, MESCO, Chemcatcher <sup>®</sup> , POCIS and naked SDB-PRS Empore <sup>™</sup> disks), including alongside composite sampling and autosamplers, and inside an exposure cell in a novel dynamic sampling device, to compare the application of each method to monitor a range of HOCs.	[33, 54, 63, 69, 300, 369–373]

Environment	Analytes	Description	Ref
Calibration tank	PAHs; PCBs; pesticides	Exposure of a range of PSDs (SR, LDPE, SPMD, POM, PVC, PU, PC, POCIS-pharms, POCIS-pest and Chemcatcher®) and active samplers (CFIS), to known analyte concentrations in laboratory exposures, to investigate performance and the application of each device to monitor a range of HOCs.	[33, 371, 374, 375]
Coastal waters	PAHs; PBDEs; PCBs; triclosan	Comparison of the application of a range of PSDs (SPMD, SR, SPME, LDPE, POM, POM-55 and POM-500) to monitor HOCs in coastal waters.	[23, 79, 308, 319, 376]
Treated wastewaters	Various	LDPE, SR, Chemcatcher® (fitted with both SDB/RPS and SDB/XC Empore™ disks, overlain with a DM and naked), SPMD and POCIS PSDs tested alongside composite sampling as part of an interlaboratory study (NORMAN Network). Investigated a range of PSDs exposed in a parallel deployment at a single site to compare and verify the analytical standards in participating laboratories and identify the current weak points of adsorption based PSDs and suggest procedures for future method validation.	[64]
Calibration tank	PAHs	The application of thirteen polymers as single-phase passive samplers of HOCs investigated during batch experiments measuring four critical properties namely, release of oligomers, swelling in solvents, diffusion coefficients and partition coefficients.	[66]
Modelled data	PAHs	Contaminant uptake models for single phase PSDs (LDPE, POM and PDMS) informed by polymer and chemical structure were developed and validated with data from the extant literature.	[222, 223]
Calibration tank and river waters	Pesticides	The performance of five PSDs: pharms-POCIS, pest-POCIS, two versions of the Chemcatcher® (C <sub>18</sub> and SDB-RPS receiving phases) and SR, to monitor mass fluxes of 124 legacy and current use pesticides, was evaluated through laboratory calibration experiments (POCIS and Chemcatcher® devices) and field exposures (all devices), including protocols for PSD preparation, calibration, extraction methods and instrumental analysis.	[377]
River and coastal waters	PAHs; pesticides	Interlaboratory study (24 laboratories) comparing various PSDs (DGT, POCIS, non polar, polar and metals versions of the Chemcatcher®, SR, LDPE, SPMD and MESCO) performance in surface waters.	[378]
Calibration tank	Dioxins; PCBs; pesticides	The application a range novel PSDs (either polymeric electrospun nanofiber mats formed of a number of polymers (PAN, PMMA and PS) or composite SR polymers with embedded SPE sorbents) were evaluated in laboratory sorption experiments in water spiked with chemicals of a broad hydrophobicity.	[115, 379]

\*Calibration tank encompasses all exposure systems ranging from artificial rivers to test tubes. \*\*Coastal waters encompass near shore marine waters, harbors and transitional waters.