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## Passive Sampling-based versus Conventional-based Metrics for Evaluating Remediation Efficacy at Contaminated Sediment Sites: A Review

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### Abstract

Passive sampling devices (PSDs) are increasingly used at contaminated sites to improve the characterization of contaminant transport and assessment of ecological and human health risk at sediment sites, and to evaluate the effectiveness of remedial actions. The use of PSDs after full-scale remediation remains limited; however, in favor of evaluation based on conventional metrics, such as bulk sediment concentrations or bioaccumulation. This review has three overall aims: (1) identify sites where PSDs have been used to support clean-up efforts, (2) assess how PSD-derived remedial endpoints compare to conventional metrics, and (3) perform broad semi-quantitative and selective quantitative concurrence analyses evaluating the magnitude of agreement between metrics. Contaminated sediment remedies evaluated included capping, *in-situ* amendment, dredging and monitored natural recovery (MNR). We identify and discuss 102 sites globally where PSDs were used to determine remedial efficacy resulting in over 130 peer-reviewed scientific publications and numerous technical reports and conference proceedings. The most common conventional metrics assessed alongside PSDs in the peer-reviewed literature were bioaccumulation (39%), bulk sediments (40%), toxicity (14%), pore water grab samples (16%), and water column grab samples (16%), while about 25% of studies used PSDs as the sole metric. In a semi-quantitative concurrence analysis, the PSD-based metrics agreed with conventional metrics in about 69% of remedy assessments. A more quantitative analysis of reductions in bioaccumulation after remediation (i.e., remediation was successful) showed decreases in uptake into PSDs agreed with decreases in bioaccumulation (within a factor of 2) 61% of the time. Given

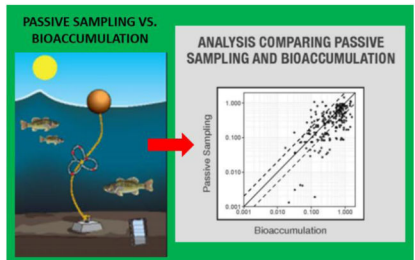
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the relatively good agreement between conventional and PSD-based metrics, we propose several practices and areas for further study to enhance utilization of PSDs through-out the remediation of contaminated sediment sites.

### Graphical Abstract



### Keywords

Remediation; Passive sampling; Contaminated sediments; Superfund; Remedy efficacy

### Introduction

In recent years, passive sampling has gained scientific credibility as a powerful tool for measuring the freely dissolved concentration ( $C_{free}$ ) of anthropogenic contaminants<sup>1, 2</sup> as well as a surrogate for the bioaccumulation of these pollutants.<sup>3-6</sup> In addition, this credibility is buttressed by the recent availability of methodological guidance for using passive samplers and applying passive sampling data<sup>7-9</sup> resulting in the expanding use of this powerful tool for remedial and regulatory purposes at contaminated sediment sites around the world (e.g., North America, Europe, Asia). A clear indication of this progress is the recognition of the need for this type of specific passive sampling guidance<sup>10, 11</sup> and increasing availability of commercial laboratories performing reliable passive sampler analyses.<sup>12</sup>

Environmental passive sampling is commonly used at contaminated sediment sites where the technique can be applied for a range of purposes, from assessing contaminant bioavailability to measuring fluxes from the sediments into the water column. Many of these applications of passive sampling can be used to inform remediation of the contaminated sites.<sup>8, 13</sup> The U.S. EPA<sup>14</sup> document *Contaminated Sediment Remediation Guidance for Hazardous Waste Sites* discusses multiple sediment remediation procedures including dredging, capping, and monitored natural recovery (MNR) for cleaning up contaminated sediment sites. In general, the goal of remediation is to reduce the ecological and human health risks associated with contaminated sediment sites.<sup>14</sup> While the ecological risk driver usually revolves around toxicity to benthic and water column organisms at the site, the human health endpoint is frequently associated with the concern of consumption of fish and shellfish that have bioaccumulated contaminants at the site.<sup>15, 16</sup> Regardless of the specific remediation method or risk driver, post-remediation monitoring is often performed to evaluate remediation effectiveness. Common conventional metrics used for monitoring include (i) measuring contaminant concentrations in the water column, sediments and

porewaters, (ii) quantifying sediment and water column toxicity or benthic habitat impacts, and (iii) determining contaminant fluxes between environmental media (e.g., sediment to water, water to air).<sup>17–19</sup> In addition, monitoring can include the use of biomonitoring organisms to provide a bioaccumulation endpoint to assess remedial effectiveness.<sup>20, 21</sup> Biomonitoring can be challenging if organisms are not available for deployment or site conditions are stressful to the organisms. More recently, pressure has been exerted to minimize the use of experimental organisms for ethical reasons.<sup>22</sup> For these reasons, alternative approaches, including passive sampling, have been considered for monitoring and assessing the effectiveness of remediation.

Passive sampling measures the  $C_{\text{free}}$  of contaminants and this value is a useful surrogate for the bioavailable concentration of organic contaminants; for example, it can be used in risk assessment as a measure of exposure.<sup>8, 9, 13</sup> As a result, in recent years, as discussed above, passive sampling has been applied as a component of remedial monitoring. To date, a comprehensive review of passive sampling for evaluating remedial effectiveness has not been performed. The objective of this investigation was to perform that evaluation to provide regulatory scientists and managers, including remedial project managers (RPMs), with the information needed to determine the usefulness of passive sampling at their contaminated sediment sites. Specifically, this review had three components: (1) a search of the scientific literature was performed to identify peer-reviewed remedial investigations in which passive sampling was used, (2) selected passive sampling-based metrics (e.g.,  $C_{\text{free}}$ ) were compared with co-occurring conventional remedial metrics (e.g., water column and porewater concentrations), and (3) a concurrence analysis was performed to display the degree of agreement and disagreement between passive sampling metrics and bioaccumulation when assessing remedial efficacy. As noted above, minimizing risks to human health is one of the goals of remediation, however, for this investigation, the focus was on evaluating remedial effectiveness based on ecological endpoints in aquatic environments.

## Materials and Methods

We searched the peer-reviewed, scientific literature using the *Web of Science* and *ProQuest* databases (Supporting Information (SI) Table S1). The last search was performed in January 2022. An article was included in this review if the following criteria were met by the investigation:

1. Historically- and anthropogenically-contaminated sediment from a polluted site including tidal mud flats (i.e., studies related solely to soil remediation or laboratory studies utilizing spiked sediment were not included).
2. Contaminant(s) included were considered legacy contaminants of concern (CoC) such as hydrocarbons like polycyclic aromatic hydrocarbons (PAHs), halogenated hydrocarbons (e.g., polychlorinated biphenyls (PCBs)), and metals/metalloids (e.g., cadmium, copper, nickel, lead, zinc, mercury, arsenic, chromium). Nutrients and bulk organics (e.g., oil and grease) were not included.<sup>23</sup>

3. Passive sampling was applied before, during, or after sediment remediation. Studies using passive sampling devices (PSDs) only during remedial investigations (e.g., for source identification) were not included unless similar data were available during and/or after remedial actions were taken.

References cited within identified articles and articles citing those included (as identified by *Web of Science*) were also screened for inclusion in this review. Additionally, we searched various references not subject to peer review to build a comprehensive contaminated site and data source list where PSDs were used to monitor remedy effectiveness. Data sources screened included the following:

1. Draft and final reports from studies funded by the U.S. Department of Defense, U.S. Department of Energy and U.S. EPA Strategic Environmental Research and Development Program (SERDP) and the Environmental Science and Technology Certification Program (ESTCP), specifically grants in the *Environmental Restoration* field;
2. Abstracts and presentations (when available) from the Battelle International Conference on Remediation and Management of Contaminated Sediments (2003 to 2019);
3. Abstracts and presentations (when available) from the Society of Environmental Toxicology and Chemistry (SETAC) North American annual meeting (2009 to 2021 except 2015);
4. Abstracts and presentations from other conferences, including the North American Environmental Monitoring Conference (NEMC) (2011 to 2021) and DIOXIN conference series (1990 to 2021);
5. U.S. EPA internal database of Superfund site documents with search terms “*passive sampling*” and “*SPMD*” (semi-permeable membrane device).

The focus of this report is the peer-reviewed literature, but we do pull from non-peer-reviewed sources sparingly when appropriate. All data sources and sites are included as an \*.xlsx file in the Supporting Information along with a plot showing cumulative publications over time used in this review (SI Figure S1).

Using these search parameters, 113 peer-reviewed journal articles were identified utilizing PSDs to evaluate or monitor the effectiveness of remediation of historically and anthropogenically-contaminated sediment sites. These articles discussed the results of 89 distinct studies (i.e., some studies resulted in multiple articles). The earliest study used dialysis samplers (also known as peepers) to monitor the migration of toxic metals (TMs) through a 34 cm pilot-scale sand cap placed in Hamilton Harbour, Canada in 1995.<sup>24, 25</sup> More recent studies have generally focused on using polymeric samplers,<sup>8</sup> such as low-density polyethylene (LDPE) film, to determine the  $C_{\text{free}}$  of hydrophobic organic contaminants. However, some researchers have used other types of PSDs, namely diffusive gradient in thin film (DGT) samplers, to evaluate labile toxic metal behavior.

Table 1 compiles the contaminants, conventional metrics, and passive sampling-based metrics including the specific PSDs and endpoints. Contaminants included PCBs,

PAHs, the pesticide dichloro-diphenyl-trichloroethane (DDT) and its primary degradation products (DDXs), pyrethroid pesticides, polychlorinated dibenzo-dioxins/furans (PCDD/F), hexachlorobenzene (HCB), several toxic metals (e.g., cadmium, chromium, copper, nickel, lead, selenium, zinc, mercury), and metalloids antimony and arsenic. Conventional metrics for assessing remedial efficacy included measuring concentrations of contaminants in bulk sediments, the water column, air samples, and sediment porewaters, as well as measuring toxicity to aquatic organisms, elevated bioaccumulation, and degradation of the benthic community condition. Along with the LDPE and DGT passive samplers mentioned above, other equilibrium passive samplers included polydimethylsiloxane (PDMS) fibers, polyoxymethylene (POM) and silicone rubber films, and the older passive sampler semi-permeable membrane devices (SPMDs).<sup>8</sup> In some instances, dialysis samplers and peepers were mentioned for sampling toxic metals when discussing an experimental design and used alongside DGTs. Specific passive sampling metrics included biomimetic (i.e., mass of contaminants taken-up by the polymer),  $C_{\text{free}}$  (including air values), flux of contaminants between sediments and the water column, and derivation of site-specific partition coefficients (SS- $K_p$ ) such as the bulk sediment-water partition coefficient ( $K_d$ ) or organic carbon normalized  $K_p$  (i.e.,  $K_{OC}$ ). Several studies used the resin Tenax and the endpoint of desorption kinetics as a proxy for bioaccessibility to assess remedy effectiveness. Additionally, several researchers employed the resins XAD or large polymeric samplers as infinite sinks for contaminants, usually to estimate sediment-to-water fluxes in static systems. For this review, applications of these materials (i.e., Tenax, XAD, infinite sinks) and the desorption kinetics endpoint were not included to focus on passive samplers that are equilibrium sampling-based. In a few cases, the use of infinite sink sampling is noted as part of an experimental design being described. In addition, the conventional metric gut fluid extraction was not included as it is not a routinely-used measurement. The *Background* section offers an introduction to remedial techniques, contaminant transport mechanisms, and equilibrium passive sampling methods at contaminated sediment sites (see the SI section).

### Data Extraction and Analysis

Data from peer-reviewed studies were extracted and analyzed to compare passive sampling-based remedial endpoints to conventional remedial endpoints. Experimental metadata (e.g., sediment characteristics, remedy information) were manually transcribed from peer-reviewed articles. Remedial endpoint information (e.g., concentrations of CoCs in various media) were taken from the text or tabulated data, if available, or manually extracted from figures using WebPlotDigitizer.<sup>26</sup> Files containing all the raw extracted data are available in the SI. Authors were not contacted for missing information.

Two types of concurrence analyses were performed on the extracted data: (1) semi-quantitative and (2) quantitative. In the semi-quantitative analysis, all relevant conventional metrics were compared to passive sampling metrics for the same treatment. In this analysis, for example,  $C_{\text{free}}$  values from passive sampling might be compared to bioaccumulation, biota survival, or bulk sediment concentrations. The change in studied metrics, for both passive sampling and conventional methods, resulting from the applied remedy was categorized as “increase”, “decrease”, or “no change” based on statistical significance tests

performed in the compared studies. Nominal changes were used if statistical significance tests were not conducted by study authors. Because of the number of comparisons, the semi-quantitative analysis was subdivided into HOCs and TMs.

For the second, quantitative concurrence analysis, all bioaccumulation data were compared to passive sampler uptake and associated  $C_{\text{free}}$  estimates from the sites on a fully quantitative basis. Bioaccumulation and passive sampler uptake have similar units (e.g.,  $\text{ng g}^{-1}$  lipid,  $\text{ng g}^{-1}$  polymer) and can be readily compared. Further, these metrics have been compared in the literature<sup>5, 6</sup> providing precedence for this type of analysis at the contaminated sediment sites identified in this investigation. In addition, we retained PSD-based  $C_{\text{free}}$  estimates in this analysis as a passive sampling metric because it is a common endpoint used in remedial practice. To be able to compare changes in  $C_{\text{free}}$  (e.g.,  $\text{ng L}^{-1}$ ) to bioaccumulation ( $\text{ng g}^{-1}$ ), post-remedy data were divided by the corresponding un-remediated (i.e., baseline or control) data to result in a fraction of initial condition ( $f$ ):

$$f = \frac{\text{metric}_{\text{post}}}{\text{metric}_{\text{pre}}} = 1 - \frac{\% \text{reduction}}{100} \quad (1)$$

where  $0 \leq \% \text{reduction} \leq 100$  was reported in many studies instead of the values of the metric before and after remediation. For example, if the concentrations of total PCBs in biota tissue before and after remediation were  $100 \text{ ng g}^{-1}$  and  $20 \text{ ng g}^{-1}$ , respectively, then  $f = (20 \text{ ng g}^{-1}) / (100 \text{ ng g}^{-1}) = 0.2$  or the percent reduction is 80%. Note, we use  $f$  instead of  $\% \text{reduction}$  in our analysis because  $f \geq 0$  (and thus amenable to log transformation) even when the metric increased as a result of remediation, whereas the  $\% \text{reduction}$  would be negative in this case and not log-transformable. For example, for  $f = 0.2$ ,  $\% \text{reduction}$  would be 80%, but for  $f = 1.5$ ,  $\% \text{reduction}$  would be  $-50 \%$ . Ultimately, because the goal of remediation is to reduce the magnitude of the conventional or passive sampling metrics, we found most  $f$  to fall in the range of  $0 \leq f \leq 1$ .

The agreement of  $f$  values for passive sampling-derived endpoints with  $f$  values for measured bioaccumulation was explored using multiple metrics. First, a standard Pearson correlation coefficient [ $r$ ] was calculated to assess the correlative strength ( $\alpha = 0.05$ ) of the relationship between passive sampler uptake and bioaccumulation. Second, the Lin's concordance correlation coefficient (Lin's CCC) was calculated. Lin's CCC ( $-1 \leq \rho_c \leq 1$ ) is a measure of how closely the compared methods fall on the 1:1 line with  $\rho_c = 1$  indicating perfect agreement.<sup>27, 28</sup> Next, the arithmetic mean of ratios between paired observations was calculated (i.e., mean ( $y/x$ )); this gives an estimate of the relative bias. Lastly, the percentage of paired observations falling within a factor of 2 of each other was calculated to approximate the spread of the agreement. Confidence intervals (95 % CI) were estimated for each of these metrics using a percentile bootstrap procedure.<sup>29</sup> Briefly, each investigated dataset was resampled with replacement and the metrics were computed for the resampled bootstrap dataset. The process was repeated 2000 times and the results for each metric was sorted in ascending order. The lower and upper 95% CI were taken as the value of the metric closest to the lowest and highest 2.5 % and of the data, respectively (i.e., the 25<sup>th</sup> and 975<sup>th</sup> metric estimate in ascending order for 2000 bootstrapped replicates). Metric estimates are reported as mean metric estimate (lower CI to upper CI).

The qualitative and quantitative analyses of remedy effectiveness were carried out in R (version 4.1.3)<sup>30</sup> using the RStudio IDE (Version 2022.02.0+443)<sup>31</sup> and the following packages: tidyverse,<sup>32</sup> here,<sup>33</sup> DescTools,<sup>34</sup> and ggpubr.<sup>35</sup> For the quantitative analyses, a remediated sample could be compared to multiple un-remediated sample types: baseline (prior to remediation), laboratory control (i.e., un-remediated sediment from same sample subject to same laboratory conditions), or a field control (nearby un-remediated contaminated sediment plot). Some articles contained multiple types of un-remediated samples (e.g., baseline and field control)—we used the same un-remediated samples in our analysis as the original authors used in their manuscripts.

## Results and Discussion

### Contaminated Sediment Sites

A total of 102 contaminated sites were found from the data sources previously described. Figure 1 summarizes the global distribution of contaminated sediment sites in which passive sampling was used to assess remedial effectiveness; the sites on the map include non-peer reviewed data, and the inset tables only include data from peer-reviewed sources. In Figure 1, results are described in terms of scale of study (i.e., laboratory or pilot/field), types of contaminants, and kinds of passive samplers. Interestingly, no studies from the southern hemisphere were found in the search, with the majority of investigations performed in North America, Western Europe, and East Asia.

For this subset of studies involving sediment remediation and passive sampling, Figure 2 and Figure 3 present the range of remedial approaches applied in North America, Europe and Asia, respectively: dredging, amendments, capping, and MNR. Both capping and amendments represent the most commonly applied remedies studied with passive sampling with them being applied evenly around the northern hemisphere. While dredging was once the most common form of remediation, monitoring of releases is often conducted only on the solid phase (e.g., sediment traps or turbidity monitoring) and passive sampling of the dissolved phase is performed less frequently. Despite the cost-effective appeal of MNR, this remedial approach is monitored with passive samplers the least. The discussion below is based on the data resulting from peer-reviewed studies included in Figure 1.

### Passive Sampling to Assess Remediation Effectiveness: Overview

The following section discusses the types of investigations using passive sampling to assess remediation effectiveness. The section is divided into (i) pre-remediation feasibility, (ii) during remediation, and (iii) post-remediation studies. The pre-remediation feasibility studies are sub-divided into (1) laboratory-based and (2) field-based studies and then further discussed by type of remediation approach (e.g., amendment, capping). All of the studies performed during remediation are field-based monitoring of dredging operations and are discussed by contaminant class including hydrophobic organic chemicals (HOCs) and toxic metals (TMs). Finally, the post-remediation studies are sub-divided into laboratory-based and field-based and are also discussed in terms of the contaminant class being remediated. For post-remediation studies, we included only sites where full-scale remediation was

undertaken; pilot- and demonstration-scale studies are discussed in the *Pre-remediation Feasibility* subsections.

To evaluate how passive sampling was used to assess remedy effectiveness, the tables in this section report the CoC and other background information (e.g., type of amendment), the PSD and the PSD endpoint being used, and the conventional metric(s) used to measure remedy effectiveness. For example, Table 2 lists an investigation by Chen et al.<sup>36</sup> in which PAHs and toxic metals were the CoCs in a sediment amended with magnetic activated carbon. The passive samplers included PDMS discs and DGT used to determine  $C_{\text{free}}$  while conventional metrics included conventional bulk sediment contaminant concentrations and bioaccumulation.

### Pre-Remediation Feasibility Studies: Laboratory-based Sediment Amendments

This category is the largest of the review, driven primarily by the number of researchers exploring sediment amendments for remediation. The following subsections provides an overview of how PSDs compared to conventional metrics for assessing remedial effectiveness (Table 2).

**Bioaccumulation**—Most studies that compared PSDs to bioaccumulation had separate deployments for the biota and PSDs. Generally, uptake into PSDs or PSD-derived  $C_{\text{free}}$  were determined in well-mixed, *ex-situ* experiments, whereas bioaccumulation was determined in static conditions with or without water renewals. Six studies co-deployed PSDs and biota: three each for HOCs and TMs. For HOCs, PSDs deployed in sediments during bioaccumulation studies showed nominally greater reductions in contaminant uptake compared to PSDs in the water column; one study observed reductions in PCB uptake into POM of 99% vs. 96%, in sediment versus water column deployments, respectively, at an activated carbon (AC) dose of 4.5%.<sup>37</sup> Reductions of bioaccumulation in organisms deployed in the water column tended to be similar in magnitude to the PSD deployed in the water column, and even if any differences were present, they were small.<sup>37, 38</sup> Further discussion is in the Supporting Information section.

Ten other studies compared bioaccumulation to *ex-situ* PSD accumulation, either biomimetically (e.g.,  $\text{ng g}^{-1}$ ) or calculated  $C_{\text{free}}$  (Table 2). These articles, focused mainly on HOCs, found reductions in bioaccumulation from amendments strongly correlated with reductions in PSD uptake. These findings highlight the reliability of *ex-situ* passive sampling estimates for bioavailability in bioaccumulation studies. Additionally, many of these studies influenced future research directions (see Supporting Information section for further discussion). More recent studies have largely focused on exploring alternative amendments,<sup>39, 40</sup> expanding the classes and life stages of organisms studied,<sup>41</sup> and investigating efficacy of amendments for reducing bioavailability of pesticides and PBDEs.<sup>40, 42</sup>

*Sediment, Water Column, and Porewater* Of laboratory-based pre-remediation studies investigating sediment amendments, ten analyzed bulk sediment samples of both control and remediated sediment, while porewater and water samples were collected in 9 and 2 studies, respectively. Sediments were generally analyzed for three reasons: (1) improving



the accuracy of calculations of BSAF for HOCs<sup>43</sup> and TMs,<sup>44, 45</sup> (2) fractionating of TMs in sediment,<sup>46–48</sup> and (3) monitoring reductions of HOCs through bioremediation.<sup>49–51</sup> Interestingly, in Hale et al., bioavailability of PAHs increased in inoculations containing PAH-degrading bacteria, presumably due to production of biosurfactants, while bulk sediment concentrations of PAHs did not significantly change.<sup>49</sup> However, when PCB-degrading bacteria were grown on AC that was subsequently added to PCB-contaminated sediments, bulk PCB concentrations decreased up to 78% while porewater  $C_{\text{free}}$  decreased even more (up to over 95%); these findings indicate that co-locating microbial degraders with a sorbing material is likely beneficial for decreasing bulk concentrations *in-situ*.<sup>51</sup>

Early studies analyzed porewater samples alongside PSDs (namely SPMDs) to verify reductions in porewater concentrations from sorbent addition were reflected by reductions in PSD uptake. Laboratory investigations found relatively good agreement among biota, porewater, and SPMD uptake for PCBs,<sup>52–56</sup> PAHs,<sup>53, 54</sup> and DDX.<sup>38, 57</sup> Since these studies, researchers have often opted to use PSD-derived  $C_{\text{free}}$  *in lieu* of collected porewater for HOCs. For TMs, a recent study analyzed porewater collected through Rhizon samplers and found amending the sediment with lanthanum-modified bentonite (LMB) and  $\text{Ca}(\text{NO}_3)_2$  reduced both dissolved and DGT-labile As by about 50%.<sup>48</sup>

**No Conventional Metrics**—Because early studies generally found similar reductions in porewater, biota, and PSD uptake, numerous more recent studies used PSDs as the sole metric to evaluate the effectiveness of sediment amendments. Generally, these studies employed PSDs in well-mixed *ex-situ* sampling to investigate how various characteristics of amendments affected relative uptake into the PSDs (i.e., biomimetic use) or the porewater  $C_{\text{free}}$ . However, a few studies deployed PSDs in static conditions. Researchers considered particle size,<sup>58–60</sup> dose,<sup>59–62</sup> carbon type,<sup>63–65</sup> mixing conditions,<sup>58–60, 66</sup> application method,<sup>60, 67, 68</sup> hydrodynamics,<sup>60</sup> and the effect of natural organic matter (NOM).<sup>63, 64</sup> The ease of deployment and analysis of PSDs relative to biota, along with documented good correlation with organism bioaccumulation,<sup>5, 6</sup> demonstrate the promise of PSDs a decision-making tool for laboratory-based amendment screening during pre-remediation feasibility studies.

### Pre-Remediation Feasibility Studies: Laboratory-based Capping

Far fewer studies investigated capping as a remedy in laboratory feasibility studies (Table 2). We note here that this review excluded studies that used PSDs as infinite sinks (e.g., a large passive sampler in the water column of a quiescent microcosm), which a few studies utilized.<sup>69, 70</sup>

Researchers have used both DGT (kinetic samplers) and *in-situ* dialysis samplers, or peepers (equilibrium samplers), to evaluate metal migration through caps in laboratory and/or mesocosms (see Supporting Information section for further discussion). Laboratory-based static column experiments showed concomitant reductions in DGT-labile and dissolved As in the porewater (from peepers) after capping the columns with aluminum sulfate<sup>71</sup> or LMB;<sup>72</sup> bulk sediment fractionation also indicated As became less mobile over time after capping with LMB.<sup>72</sup>

For HOCs, Eek et al.<sup>73</sup> used *ex-situ* POM sampling to determine  $C_{\text{free}}$  of PAHs to support modeling the results of a laboratory capping experiment. Lampert et al.<sup>74</sup> deployed PDMS fibers through sand caps of various thickness with simulated natural deposition of clean sediment on top. Using 1 cm resolution through 8 cm to 10 cm of cap and contaminated sediment, the authors showed significant reductions of PAH  $C_{\text{free}}$  in the bioturbation zone of the oligochaete worm *Ilyodrilus templetoni*. They also observed highly significant correlations between PDMS uptake and lipid-normalized body burdens. Rämö et al.<sup>75</sup> simulated resuspension of AC-amended thin layer caps (3 mm to 5 mm) in intact cores. SPMDs and DGTs were deployed for 60 d in the water columns (as infinite sinks) while equilibrium sampling PDMS fibers were deployed in the sediment. Powdered activated carbon (PAC)-amended thin layer caps significantly reduced fluxes of HOCs and metals in both static and simulated resuspension tests. PDMS-based  $C_{\text{free}}$  measurements showed no statistically significant differences among treatments, potentially due to analysis of large fiber segments relative to the thickness of cap (4 cm versus 0.5 cm).

### Pre-Remediation Feasibility Studies: Laboratory-based Multiple Remedies and Comparisons

Two studies used PSDs to compare multiple remedies, resulting in five separate peer-reviewed publications (Table 2). In the first study, researchers simulated MNR and AC-enhanced MNR of a DDX-contaminated sediment in stagnant column studies, with infinite-sink PSDs in the water column to measure fluxes and profiler PSDs in the sediment.<sup>76</sup> A follow-up article added column studies evaluating deposition of clean and contaminated sediment over thin AC caps. Without AC, bioturbation diluted any depositing sediment, causing no reduction in flux, bioaccumulation, or sediment LDPE uptake from controls. However, adding a thin layer of AC reduced sediment-to-water fluxes and sediment LDPE uptake, even with depositing contaminated sediment.<sup>77</sup>

In a similar study, bioaccumulation of PCBs in three different organisms (i.e., a bivalve, polychaete, and fish) was compared to co-deployed PSDs in the water column and sediment, and also to *ex-situ* PDMS-lined vials.<sup>78</sup> Sediments were mixed with AC,<sup>79</sup> capped with sand, or capped with a sand/AC mixture.<sup>80</sup> Furthermore, the flow-through mesocosms were occasionally dosed with either clean sediment inputs or sediment spiked with non-native PCBs, which acted as tracers to determine which sediment—bedded or deposited—was responsible for bioaccumulation. PCBs in polychaetes were the most linked to bed sediment, and while accumulation in fish and bivalves were also mostly from bedded PCBs, the two different capping sediment treatments had more influence in fish than the other organisms. These studies also found that PSDs in the water column were better indicators of bioaccumulation in fish and bivalves, whereas sediment-based PSDs correlated better with polychaetes. These results likely reflect the primary exposure media the organisms experienced; that is, the fish and filter feeding bivalves were exposed to contaminants in the water column of the laboratory system whereas the sediment-ingesting polychaetes interacted directly with the sediments. The mixed AC mesocosm reduced bioaccumulation the most, followed by the mixed sand/AC cap. A full description of this study (including more mesocosms) can be found in the final SERDP report.<sup>81</sup>

### Pre-Remediation Feasibility Studies: Field-based Sediment Amendments

Pilot projects using sediment amendments aimed to show this remedial technique could reduce bioaccumulation, toxicity, and water column concentrations of CoCs (Table 3). Additionally, they sometimes were used to refine remedy characteristics, such as delivery methods, that are difficult to simulate in the laboratory. A limited number of studies<sup>82–85</sup> compared changes in bulk sediment and/or water column concentrations of CoCs to changes in  $C_{\text{free}}$ , flux, and polymer uptake based on passive sampling. In two of those studies, benthic community composition was also evaluated versus passive sampling-based measures.<sup>82, 84</sup> (See Supporting Information section for further discussion).

A limited number of studies reported bulk sediment concentrations over time, but addition of 1.5% to 4% AC by weight did not significantly affect the sediment PCB concentration in pilot projects for up to 13 months,<sup>86</sup> 18 months,<sup>82</sup> and 33 months<sup>84</sup> after placement. The bulk PCBs concentrations in one study decreased significantly (30% to 52%) after 13 months of contact with AC amended with PCB-degrading bacteria.<sup>86</sup> Clearly, metrics other than bulk sediment concentrations are needed to evaluate the remedial efficacy of sediment amendments; these are discussed below.

Early studies targeted whether the reduction in bioavailability and correlation between bioaccumulation and passive samplers observed in laboratory studies translated to the field. Initially, researchers compared uptake to *in-situ* sediment SPMDs (and later, LDPE and POM) to co-deployed biota to determine if *in-situ* PSDs could serve as a proxy for bioaccumulation in native or introduced benthic organisms (see Supporting Information section for more discussion).

### Pre-Remediation Feasibility Studies: Field-based Capping

Compared to sediment amendment, fewer studies investigated capping as the only field-based remedy. Lampert et al.<sup>87</sup> had a few interesting findings from a capping demonstration in the estuarine Anacostia River (see Supporting Information section for more discussion). Cornelissen et al.<sup>88</sup> describes a large ( $3 \times 10000 \text{ m}^2$  and  $1 \times 40000 \text{ m}^2$  plots) field trial of passive and active thin-layer caps over PCDD/F-contaminated sediments in a deep (30 m and 100 m) Norwegian fjord. Researchers measured sediment-to-water fluxes using benthic chambers with SPMDs as infinite sinks. Additionally, they determined  $C_{\text{free}}$  in the water column 7 cm to 10 cm from the sediment surface with  $17 \mu\text{m}$  POM. Flux chambers and POM were deployed for four consecutive 6 month periods starting one month after capping (see Supporting Information section for more discussion).

Fernandez et al.<sup>89</sup> used *in-situ* PE, POM (both with PRCs), and PDMS fiber (without PRCs) samplers to determine whether a thin (between 4 cm and 16 cm) sand cap over DDX- and PCB-contaminated sediment off the coast of California (Palos Verdes) was reducing flux to the water column. A previous investigation with passive sampler deployments at 3 depths in the water column found higher DDX concentrations near the sediments than near the water surface along with horizontal migration of dissolved DDX down-current from heavily contaminated sediments.<sup>90</sup> Profiles of  $C_{\text{free}}$  within the capped sediment showed a sharp increase near the cap-water interface, indicating recontamination. Water column

concentrations directly above the sediment-water interface were similar among uncapped and capped sites, resulting in similar fluxes at the capped and nearby uncapped sites. While *in-situ* profiles indicated no reduction of flux, the authors did note that the fluxes were an order of magnitude lower than those calculated in an earlier study that utilized a one-carbon equilibrium partitioning (EqP) model to estimate the porewater concentrations. Additionally, porewater concentrations within the cap (roughly 2 cm below the sediment-water interface (SWI)) were about 50% of those observed at nearby uncapped sites. This finding suggested a cap could reduce both flux and bioavailability for benthos that burrow deeper than the contaminated layer at the SWI.

### Pre-Remediation Feasibility Studies: Field-Based MNR

Only one study was available for MNR. At Pallanza Bay (Verbano-Cusio-Ossola, Italy), Lin et al.<sup>91</sup> deployed LDPE samplers with 30 cm both above and below the sediment water interface at 2.5 cm resolution using a porewater probe designed for deep water deployments. LDPE was spiked with a PCB PRC and target analytes included DDT and its metabolites. Sediment traps and hydrodynamic equipment were deployed nearby with LDPE samplers attached to determine water column  $C_{free}$ . Interestingly, the PCB PRC, chosen because it has similar hydrophobicity to the most hydrophobic DDX, only released 10% to 20% from the LDPE. However, deployments of different lengths (60 d, 93 d and 130 d) did not indicate significant differences in DDX concentrations in the porewater or water column at the same locations, suggesting approximate equilibrium. Fickian modeling also suggested the fraction equilibrium ( $f_{eq}$ ) was greater than 80% for all DDX compounds. Adjustment for diffusivity and hydrophobicity resulted in  $f_{eq}$  of 20% to 90% depending on the DDX compound. Ultimately, the study found upward diffusive fluxes observed in the porewater would not overtake the sorption capacity of naturally deposited clean sediment. Consequently, it was concluded that Pallanza Bay was demonstrating adequate natural attenuation at the time of the study.

### Pre-Remediation Feasibility Studies: Field-Based Multiple Remedies and Comparisons

Two studies performed multiple remedy effectiveness comparisons. Sanders et al.<sup>92</sup> compared three AC addition methods in a PCB-contaminated oligohaline tidal marsh (Berry's Creek, NJ, USA): SediMite, granulated activated carbon (GAC), and GAC with an overlying thin layer (2 cm to 3 cm) of sand; the effective AC dosage was 5% over 10 cm depth. Monitoring of effectiveness included a pre-pilot baseline and six post-addition events up to 37 months after remedy implementation. Monitoring included determination of  $C_{free}$  with PRC-loaded LDPE and POM strips of different thicknesses (28 d deployment), *in-situ* bioaccumulation studies (top 5 cm to 8 cm, 14 d), *ex-situ* bioaccumulation studies (for one monitoring event), and native invertebrate bioaccumulation.<sup>92, 93</sup> SediMite, in which the active ingredient is PAC, reduced porewater  $C_{free}$  by over 90% in the top 2.5 cm, whereas the two GAC treatments showed temporal variability, with reductions of 34% to 86% versus pre-amendment  $C_{free}$ . Native biota showed similar reductions in each treatment versus an unamended plot whereas caged *in-situ* bioaccumulation showed higher reductions in the SediMite plot compared to the GAC plots relative to the unamended plot. However, this study did not investigate the effects of the finer PAC on toxicity or benthic community condition (e.g., abundance, richness) like Cornelissen et al.<sup>94</sup>

Cornelissen et al.<sup>94</sup> carried out a thin-layer active cap pilot in Trondheim Harbor (Norway) to determine which placement technique was optimal (i.e., AC only, mixed AC/clay layered AC/sand, sand only). Fluxes of PAHs from the caps were monitored for a year following placement with *in-situ* benthic chambers utilizing PDMS sheets as the infinite sink. Additionally, samplers holding 17  $\mu\text{m}$  POM passive samplers were deployed *in-situ* across the sediment-water interface for 60 d to 154 d (without PRCs) at 9 months after capping. Relative uptake in the POM samplers and calculated  $C_{free}$  were used as the metric. Regardless of addition method, AC reduced fluxes to the water column compared to the uncapped reference site, but none of the remedial treatments was superior. The POM samplers indicated reduced porewater concentrations coinciding with the depth of AC (0 cm to 5 cm deep) by about 50% but no differences in porewater below the AC or in the water column above the capping demonstration plots. However, a high effective AC dosage (up to 40% dry weight) resulted in statistically significant impacts to native benthic fauna abundance and richness, but the result was lessened in the clay/AC mixed cap.

### Pre-Remediation Feasibility Studies: Field-Based Dredging

Dredging is the oldest method of sediment remediation, and because it is so established, the equipment mobilization and sediment disposal often make pilot dredging projects too costly except for optimization pilots in very large projects (e.g., Hudson River, NY, USA; Lower Passaic River, NJ, USA). Thus, only a single article has been published on a field-scale dredging test that used passive samplers as metrics to evaluate remedial efficacy. Yu et al.<sup>95</sup> collected sediment cores from Lake Taihu (Jiangsu, China), removed the top 25 cm from half of the cores, and placed all the cores back into the lake. After 11 months, the cores were removed from the lake and analyzed through 24 h DGT deployments and sequential bulk sediment extraction. While the absolute amount (i.e., bulk sediment concentration) of metals (Cu, Cd, Ni, and Zn) decreased after dredging, the fraction of exchangeable metals was higher, as were the labile concentrations near the surface, particularly for Cu and Zn. The authors attributed the increased lability to reduced sulfide content in dredged sediments and urged more study into the effect of dredging on the cycling and availability of metals in sediments.

### During Remediation

The following subsections briefly summarize hydrophobic organic contaminants (HOCs) and toxic metals measured with passive sampling during dredging events (Table 4) to assess remedy effectiveness. A detailed discussion of these studies is presented in the Supporting Information.

**Hydrophobic Organic Contaminants**—No targeted peer-reviewed studies in freshwater have been conducted, but circumstantial PSD evidence suggests that sediment disturbance from dredging can increase HOC concentrations. Investigations by Vrana et al.,<sup>96</sup> Allan et al.,<sup>97</sup> Sower and Anderson,<sup>98</sup> and Martinez et al.<sup>99</sup> discuss some of this evidence for PAHs and PCBs. It is important to note that none of these studies had other metrics (e.g., water samples, fish bioaccumulation) to compare with PSD results. For marine sediments, the data is more conclusive, as described Cornelissen et al.,<sup>100</sup> Schaanning et al.<sup>101</sup> and Joyce et al.,<sup>102</sup> dredging of contaminated sediment may result in

more substantial increases in water column  $C_{\text{free}}$ . Cornelissen et al.<sup>100</sup> and Schaanning et al.<sup>101</sup> used POM and SPMD, respectively, to determine PAH and PCB  $C_{\text{free}}$  in the water column around a CAD site near Oslo Harbor (Norway) both before and during deposition of dredged material into the CAD. The effects of dredging on PCB  $C_{\text{free}}$  were less pronounced including Joyce et al. investigation with LDPE at the New Bedford Harbor Superfund site (Massachusetts, USA) during dredging.<sup>100–102</sup> Finally, several studies (unrelated to remedial actions) including Schneider et al.,<sup>103</sup> Belles et al.,<sup>104</sup> and Allan et al.<sup>105</sup> have shown that resuspension of HOC-contaminated sediment can result in rapid and substantial desorption of the HOCs into the water column. Cantwell et al.<sup>106</sup> reported similar findings for several toxic metals.

**Toxic Metals**—As with HOCs, passive sampling data related to off-site release from dredging is a mix of targeted and circumstantial investigations, with only two studies specifically designed to determine the impacts of dredging on freely dissolved toxic metal concentrations in the water column. DGTs were the sole PSD used in all studies. Layglon et al.<sup>107</sup> carried out the most comprehensive study. From September 2013 to September 2017, the researchers collected monthly water column grab samples for dissolved and total Cu and Pb in Toulon Bay (France). ‘Dissolved’ was operationally defined as metals passing through a 0.45  $\mu\text{m}$  filter. Additionally, they deployed DGTs for one week every month during the study period to quantify DGT-labile Pb. In another wide-scale monitoring event, Schaanning et al.<sup>101</sup> used DGTs to monitor releases of dissolved toxic metals during dredged material disposal in a CAD near Oslo Harbor (Norway). DGTs were deployed at 14 different locations at depths of up to 65 m—sampling conditions that would make collection of composite dissolved and total samples logistically difficult. Another study found DGT-labile toxic metals were strongly influenced by dissolved oxygen (DO) concentrations, but the effect of dredging was unclear. Villanueva et al.<sup>108, 109</sup> deployed DGTs in the water column of the heavily-polluted, tidally-influenced Pasig River—which is entirely located within the Manila metropolitan area in the Philippines—to determine if the passive samplers could provide insight into seasonal metal variations. Finally, Vrana et al.<sup>96</sup> carried out similar monitoring in the upper Danube River to observe seasonal and spatial variation in total and dissolved toxic metal concentrations (in addition to PAH and PCB  $C_{\text{free}}$  noted above), and one location was immediately downstream of navigational dredging operations.

## Post-Remediation

Researchers and RPMs use PSDs far less often following full-scale remediation (i.e., after an early action, ROD activities, or similar) than during feasibility studies. Only 9 peer-reviewed journal articles included results from passive sampling campaigns after a site was remediated. The following sections introduces these investigations with more details provided in the Supporting Information including how results from *in-situ* and *ex-situ* PSDs compared to other metrics of remedy effectiveness (Table 5).

Ex-situ: Hydrophobic Organic Contaminants *Fetters et al.*<sup>110</sup> *comprehensively monitored an enhanced monitored natural recovery (EMNR) site contaminated with DDX using a combination of conventional metrics including 14 d in-situ bioaccumulation, bulk sediment cores, sediment traps, and ex-situ passive sampling. Samples were collected prior to*

placement of a thin (23 cm) sand cap and after 2 months, 14 months, and 25 months. In another ex-situ study, Li et al.<sup>111</sup> compared  $C_{\text{free}}$  results using PDMS discs in surface water and surface sediment to bulk concentrations in water, sediment, and native benthic and pelagic species to determine if environmental dredging following the construction of Yangshan port near Shanghai, China, successfully reduced PAH availability.

Ex-situ: Toxic Metals and Metalloids *Two studies have used DGTs in cores retrieved from field sites to assess how dredging affected the lability of toxic metals. The first was a research program to understand how dredging of Meiliang Bay in Lake Taihu, China, affected nutrient cycling and contaminant availability in the sediments. For metals and metalloids, they investigated Pb, Cd, Cu,<sup>112</sup> Co, Zn, Ni,<sup>113</sup> As, Se, Sb,<sup>114</sup> and Cr<sup>115</sup> collecting cores from the dredged portion and an un-dredged reference location in different seasons in 2016 and 2017, six years after dredging operations had ceased. In the other study, Parker et al.<sup>116</sup> used sediment probe DGTs in box cores to evaluate whether a marine disposal site for dredged material acted as a source of Pb, Cd, and Ni to the water column.*

**In-situ: Hydrophobic Organic Contaminants**—A major benefit of PSDs is that they can be used *in-situ* to determine whether a cap is performing as designed without severely damaging the integrity of the cap through coring. Thomas et al.<sup>117</sup> used PDMS-coated glass fibers loaded with PRCs in a modified Henry sampler to determine PCB  $C_{\text{free}}$  to a depth of 90 cm with 2 cm resolution at two Superfund sites in Washington and Tennessee (USA) with isolation (1 m to 1.5 m thick) caps. Minick and Anderson<sup>118</sup> used a rugged stainless-steel probe to encase PRC-loaded LDPE samplers and drive them into a sand and organoclay isolation cap topped with armor stone at the McCormick and Baxter Creosote Superfund Site within the larger Portland Harbor Superfund Site (Oregon, USA). Oen et al.<sup>119</sup> used benthic flux chambers equipped with SPMD as an infinite sink and also deployed POM samplers in the water column for 77 d to 86 d to investigate the long-term chemical stability of a capped CAD in Oslo Harbor, Norway. They also collected bulk sediment and overlying sediment from sediment traps prior to and after disposal and capping. In addition, they monitored native benthic macrofauna to assess how the CAD affected the native population. Schubauer-Berigan et al.<sup>120</sup> used SPMDs in three different designs—water column samplers, porewater samplers, and benthic flux chambers—to evaluate the efficacy of MNR remedy for PCBs at the Lake Hartwell Superfund Site (Pickens, South Carolina, USA). They also collected bulk sediments and composite water column samples. Patmont et al.<sup>121</sup> monitored the efficacy of a full-scale, 2 ha application of SediMite in a fresh to slightly brackish tidal lake in Dover, Delaware (USA). PRC-loaded POM and LPDE strips were deployed in the sediments and water column for one month at intervals before application and 1 month, 1 year, and 3 years after application. Researchers also collected sediments for laboratory bioaccumulation studies and collected native fishes to determine PCB bioaccumulation over time. Odetayo et al.<sup>122, 123</sup> set up 76  $\mu\text{m}$  thick, PRC-loaded POM passive air samplers near conventional high-volume air samplers (HVAS) for 42 d to 113 d to monitor emissions from a confined disposal facility housing sediment dredged from a nearby PCB-contaminated waterway. Samples were collected during and following dredged material disposal. Finally, a series of studies used LDPE PSDs in the water column, air, and

sediment porewater to investigate sources of PAHs at the Portland Harbor Superfund Site (Oregon, USA). The early studies did not use PRCs and thus did not determine  $C_{\text{free}}$ <sup>97, 98</sup> but were carried out before, during, and after early action remedies occurred at portions of the site. Later studies used PRCs and found the fluxes of PAHs to be from air to water at upstream locations but that the sediments within the site served as sources to both the water column and air for parent PAHs and oxidized PAHs.<sup>118, 124</sup>

**In-situ and Ex-situ: Hydrophobic Organic Contaminants**—Eek et al.<sup>125</sup> measured sediment-to-water fluxes of PAHs and PCBs in a dredged-and-capped area of Oslo Harbor (Norway) with benthic flux chambers and compared them to fluxes estimated from  $C_{\text{free}}$  determined through *in-situ* POM (water column) and *ex-situ* POM (sediment porewater) deployments. They found the flux chambers, fitted with infinite sinks of SPMD or silicon rubber, agreed well (largely within a factor of 2) with POM-derived fluxes at uncapped areas. Fluxes measured by the benthic chambers were reduced by 93% to 97% after capping with 20 cm of uncontaminated dredged clay, while comparison of  $C_{\text{free}}$  in the water and porewater indicated the flux direction was reversed—the clean cap was acting as a sink for PCBs and PAHs in the water column.

### Concurrence Analysis: Passive Sampling versus Conventional Metrics

Following careful review of the passive sampling data performed with matching conventional metrics as discussed above, the two types of concurrence analyses were performed. Of the 89 studies identified in this review, 35 had bioaccumulation data, 36 compared bulk sediments, 13 studied acute toxicity, 14 collected porewater data (not from peepers), and 14 took water column grab samples. All conventional chemical endpoints were included in the semi-quantitative analysis whereas the quantitative analysis focuses on comparing passive sampling-based endpoints (i.e., uptake and  $C_{\text{free}}$ ) only to bioaccumulation. Relative to contaminants with multiple structures (e.g., PCB congeners, PAHs), to make the dataset manageable, ‘totals’ of structures quantified by the investigators were calculated and used in the analyses described here.

**Semi-Quantitative Analysis**—In this analysis, for HOCs, there were 338 comparisons with the passive sampling endpoints agreeing with conventional metrics in 69% of cases (Figure 4a). When in disagreement, passive sampling generally identified the remedy as successful (i.e., reducing uptake, concentrations, or flux) (19% of comparisons) when conventional metrics suggested no change or an increase in an endpoint as a result of remediation. Approximately half of these disagreeing data points measured bioaccumulation as the conventional endpoint; tissue concentrations often decreased nominally in these cases, but the associated uncertainty in the measurements led to a finding of no significant change under hypothesis testing. The majority the remaining disagreeing data points with an increase or no change in the conventional endpoint measured bulk sediment concentration. In 10% of cases, passive sampling detected no difference or an increase from remediation while conventional endpoints decreased; much of these disagreements resulted from poor sampler resolution or media mismatch (e.g., passive pore water and conventional surface water).



Nearly identical agreement behavior was observed for the 24 comparisons involving toxic metals (Figure 4b) with 75% of cases agreeing. In 25% of comparisons, passive sampling detected decreases in contamination resulting from remediation while conventional endpoints found no significant effects, in these cases, all conventional samples were bulk sediment concentrations. Note that a study of dredging effects on toxic metal availability in Lake Taihu (Jiangsu province, China)<sup>112–115</sup> was not included in Figure 4b because of the comparisons were on a per-metal basis. A separate concurrence table for the Lake Taihu study is included and discussed in the SI (Figure S5). Overall, the semi-quantitative concurrence analysis found overwhelming agreement between passive sampling and conventional metrics for both HOCs and toxic metals. Further, when disagreements occurred, the passive sampling endpoints were found to be more sensitive in detecting significant remedy effectiveness.

**Quantitative Analysis**—From this analysis, there were a total of 211 viable comparisons to bioaccumulation data including 60, 130, and 21 cases based on passive sampling uptake (biomimetic), porewater  $C_{\text{free}}$ , and surface water  $C_{\text{free}}$  metrics, respectively. For this discussion, we focus on the use of Lin's CCC and the factor of two summary statistics (and their respective confidence intervals (CIs)) to quantitatively assess 'good' agreement between bioaccumulation and passive sampling endpoints. For example, Lin's CCC was 0.50 (with lower and upper CIs 0.39 to 0.60) and 61% (55% to 68%) of the data were within a factor of two of each other for the full passive sampler – bioaccumulation comparative dataset (Figure 5a). Similarly, Lin's CCC was 0.70 (0.56 to 0.82) for biomimetic uptake (Figure 5c) while the same metric was 0.44 (0.30 to 0.57) for  $C_{\text{free}}$  (Figure 5b). Finally, 77% (65% to 87%) of comparisons (Figure 5c) for biomimetic uptake into the passive samplers were within a factor of two while only 56% (48% to 64%) of comparisons of porewater and surface water  $C_{\text{free}}$  values (Figure 5b) fell in the same range.

Based on the overall quantitative results presented in Figure 5, we investigated various factors that were potential sources of disagreement between remedial effectiveness measured by PSDs and bioaccumulation. In performing this analysis, factors that were relatively 'easily' manipulated when performing passive sampling and bioaccumulation deployments were emphasized. For example, Figure 6 contains data from laboratory studies where the PSDs were within the same exposure chamber as the biota (Figure 6a) and where PSDs and biota were in separate chambers (Figure 6b). Placing PSDs and biota in the same vessel during the bioaccumulation experiment resulted in better agreement than separating them. For example, the Lin's CCC was 0.89 (0.82 to 0.94) and 0.68 (0.56 to 0.79) for exposures performed in the same chambers versus different chambers. In addition, 79% (67% to 92%) versus 72% (61% to 82%) of the PSD and bioaccumulation data were within a factor of two for the same chamber versus different chamber comparison. This finding is likely because of two reasons: (1) when together, PSDs and biota are exposed to the same environmental conditions (e.g., CoC concentrations in various media, hydrodynamics), and (2) many separated PSD deployments were mixed *ex-situ* deployments in which the sediments were homogenized on a roller mill, better approximating equilibrium conditions than a static bioaccumulation test. This part of the analysis demonstrates that as the PSDs

and biota are deployed under different conditions, the agreement between them regarding remedial efficacy becomes worse.

We speculated the agreement between PSD and bioaccumulation datasets collected in the field might be worse than laboratory-based studies (Figure 6a, b) because of the relative difficulty in co-locating PSDs with biota in the field. Many field studies deployed the Sediment Ecotoxicity Assessment Ring (SEA Ring), or previous iterations of the device, which uses *in-situ* cores to contain both the biota and PSDs.<sup>126</sup> When these types of devices are used or native biota are collected near the passive samplers, the percentage of data points within a factor of two between biota and PSDs was lower (50% with 35% to 65% CIs) (Figure 6c) than that of either type of laboratory deployment (79% and 72% (Figure 6a,b)). Further, the Lin's CCC also showed less agreement (0.39 (0.17 to 0.60)) (Figure 6c) compared to the laboratory data when the PSDs and biota were next to each other (0.89 (0.82 to 0.94)) or when they were separated under laboratory conditions (0.68 (0.56 to 0.79)) (Figure 6a,b). When bioaccumulation studies were performed *in-situ* and the passive samplers were used on associated sediments in the laboratory, the relationship between PSDs and bioaccumulation was quite poor, showing no agreement (Figure 6d): Lin's CCC of -0.14 (-0.33 to -0.01) and only 37% (16% to 58%) of PSD and bioaccumulation data were within a factor of two. Biota exposed under field conditions (and not caged) are free to roam (especially organisms like pelagic fish) and may have a home range larger than the remediated area, resulting in exposure to potentially un-remediated contaminated sediment. Conversely, the *in-situ* or *ex-situ* deployed PSD are stationary with a relatively 'stable' exposure. Thus, this analysis quantitatively confirms concerns with data variability and uncertainty when using bioaccumulation from native or mobile organisms as a remedial metric for remedies of sediments within larger contaminated sites.

The choice of species used for biomonitoring remedy effectiveness also affected the degree of agreement between PSD and bioaccumulation remedy metrics. Figure 7 shows the magnitude of agreement between PSDs and bivalves, oligochaetes and polychaetes (worms), fish, and other biota (e.g., amphipods or insects). Bivalves and PSDs showed very similar behavior when used to monitor remedy effectiveness: Lin's CCC was 0.82 (0.71 to 0.91) and 87 % (78 % to 95 %) of the data points fell within a factor of two indicating the two metrics were very comparable (Figure 7a). Oligochaetes and polychaetes showed relatively good agreement with PSDs: Lin's CCC was 0.61 (0.46 to 0.74) and the factor of two inclusion was 58% (47% to 68%) (Figure 7b). In contrast, the fish (Figure 7c) and other biota (Figure 7d) showed poor agreement with Lin's CCCs of 0.19 and 0.15, respectively, and datasets for which only 42% and 47%, respectively, were within a factor of two. The disagreement with fish is again likely because of their expanded home range in the field compared to the relatively sessile bivalves and worms resulting in the fish experiencing a more varied exposure to contaminants (i.e., remediated and unremediated areas). Similarly, the disagreement with other biota may be from differing mechanisms or rates of contaminant uptake in PSDs and those classes of biota. In addition, the bioaccumulation methods used for bivalves and worms are very well established compared to methods used for many other species which may also result in greater data variability.

Another choice available when using PSDs for HOCs is the passive sampler material. Early adopters of PSDs used multi-phase SPMDs, but single-phase polymeric samplers (e.g., POM, LDPE, PDMS or silicone rubber) have become more common, with LDPE and PDMS the most common and favored materials.<sup>127</sup> PDMS agreed the best with bioaccumulation with a Lin's CCC of 0.79 (0.69 to 0.86) and 76 % (66% to 86%) of data points falling within a factor of two of each other (Figure 8a). LDPE and SPMD both performed relatively well (Figure 8b, d) and exhibited similar degrees of agreement with Lin's CCCs of 0.53 and 0.62, respectively, and 61% and 75%, respectively, of datapoints falling within a factor of two of each other. Potentially, the similar responses are due to the identical diffusive layers used in both LDPEs and SPMDs. We found poor agreement when POM was used in conjunction with bioaccumulation to investigate remedy effectiveness (e.g., Lin's CCC of only -0.05 and a low 34% of datapoints within a factor of two of the 1:1 line), lending weight to the PSD user-community moving away from POM for this application (Figure 8c). For toxic metals, DGTs and bioaccumulation were within a factor of two of each other for 80% (60% to 100%) of data points but had a low Lin's CCC of only 0.14. The limited sample size (n = 15) likely contributed to the disparate statistical outcomes for DGTs (Figure 8e); more studies in this area are warranted.

Interestingly, all pilot- or field-scale studies had at least one point of comparison that fell outside the factor of two of each other metric range, reflecting the greater uncertainty caused by *in-situ* processes. Authors of two of those studies collected native biota that were noted to have a larger home range than the remediated area.<sup>93, 121</sup> In these cases, passive sampling endpoints, specifically porewater-based  $C_{free}$ , offer a more local data source better at identifying the effectiveness of the remedy. Other sources of disagreement between passive sampling and conventional endpoints were less clear. Species-specific effects were possible, with one study noting difficulty removing fine AC particles from the surface of a gastropod, potentially causing extraction of AC along with the biota tissues.<sup>128</sup> Many disagreeing points were found in amendment studies with relatively high (i.e., > 2% dry weight) doses of AC. Although it is not entirely clear why these disagreements occur, the next section discusses a possible explanation involving the use of  $f$  to compare passive sampler uptake and bioaccumulation when the remediation is very effective. Other points of disagreement may have been caused by *ex-situ* passive sampling without prior sediment-amendment equilibrium. Depending on the desired accuracy and species, the use of fate and transport models to translate passive sampling  $C_{free}$ s to specific exposure concentrations may address some of these disagreements. However, despite the limited number of cases where agreement did not occur, like the semi-quantitative analysis, we found a very good prevalence of agreement between the conventional and passive sampling-based metrics despite taking a more quantitative approach and focusing on a single conventional metric (i.e., bioaccumulation). Both type of concurrence analysis, semi-quantitative and quantitative, confirm using passive sampling as an alternate or surrogate metric is viable and should continue to be pursued.

Finally, we recognize that using a relative range of agreement, while often applied in comparing passive sampling data to bioaccumulation, penalizes remedies that were extremely effective in reducing passive sampling uptake or bioaccumulation. For example, a range of  $0.5 < f_{\text{passive sampling}} < 2$  signifies a factor of two for a remedy that did not

affect passive sampler uptake. For a very effective remedy, passive sampler uptake might be reduced by 99% while bioaccumulation could be reduced by 96% giving  $f_{\text{passive}}/f_{\text{bioaccum}} = 0.01/0.04 = 0.25$ , which is outside of the acceptable range. Conversely, using an absolute range would favor agreement in very effective remedies, but make agreement more difficult to achieve for less effective remedies. For example, for a 20% absolute agreement value, if one metric is not detectable, the other metric could have  $f$  up to 0.2 to agree, whereas  $0.8 < f_{\text{passive}}/f_{\text{bioaccum}} < 1.2$  for an ineffective remedy is a much smaller range than the relative range discussed previously. For both of these examples, the resulting ranges are plotted in Figures S6 to S9 for illustration based on the data originally shown in Figures 5 to 8. In addition, alternatively, Figure S10 presents the data shown in Figure 5 as simply percent (%) reduction (not as  $f$ ). Clearly, more investigation is needed to determine the optimal way to compare, express and present the relationship between conventional and passive sampling-based metrics.

## Implications and Recommendations

This investigation found that PSDs have been used as tools to evaluate remedial efficacy at contaminated sediment sites for the last 25 years. Their applications are largely focused on pre-remediation feasibility studies with environmental managers using PSDs after full-scale remediation; that is, following establishment of the agreement defining long-term monitoring (i.e., the Record of Decision (ROD) in the United States), in fewer than 10% of the contaminated sites identified in this review. The relative lack of use after full-scale clean-ups may be part of the technology maturation process—feasibility studies have often been conducted by academics or government and private research institutions evaluating novel methods before standardization and commercialization. Long-term monitoring is usually conducted by environmental consulting companies using established and standardized methods. The good agreement found in our semi-quantitative and quantitative concurrence analyses between PSD-based and conventional metrics lends weight for adding PSDs to the ‘toolkit’ for long-term monitoring of remediated sediment sites. In addition, although not the objective of this investigation, other researchers are welcomed to evaluate this dataset to simply assess how often remediation was effective (regardless of the metrics used) at cleaning-up contaminated sediment sites. To help bridge the use-gap between pre-remedial and post-remedial investigations, the following recommendations are proposed:

1. This investigation demonstrates many studies have already compared passive sampling and conventional ecological metrics to evaluate remediation efficacy. However, on a site-specific-basis, we would argue bioaccumulation and toxicity testing should continue to be performed alongside PSDs to gain confidence in using PSD as a major long-term monitoring tool. For example, if native mussels are being collected or reference area mussels deployed to determine bioaccumulation, deploying PSDs for a period immediately prior to mussel collection or in parallel with deployed organisms would build site-specific mussel-PSD correlations that could be used during long-term biomonitoring. If *ex-situ* bioaccumulation or toxicity tests are planned, we suggest ensuring the sediment-water-amendment (if present) equilibrium status is maintained for both the PSD and biota tests. For example, if an amended sediment is collected for

bioaccumulation tests, consider placing a PRC-loaded PSD statically in the same exposure chamber as the biota. If that is not feasible and agitated *ex-situ* PSD sampling is planned (e.g., 28 d shaking or rolling), contemplate performing the same treatment to the sediment prior to use in the bioaccumulation test to ensure similar sediment-water-amendment equilibrium status.

2. When possible, use sessile organisms to monitor remedy effectiveness, particularly for sites within a large contaminated area. Sessile organisms, like PSDs, better reflect local conditions. While reducing bioaccumulation of native species (e.g., sport or culturally-important fish) are common remedial goals, few peer-reviewed articles collected these organisms while also deploying passive samplers. We recommend more long-term monitoring programs include passive samplers along with native organisms as passive sampling technology matures.
3. Although PSDs have been used sparingly to monitor dredging, users of PSDs must be cautious when choosing what type of PSD to monitor dredging. Values for  $C_{\text{free}}$  may vary greatly during removal and disposal operations, while they likely stabilize after work is completed. When  $C_{\text{free}}$  values are variable, integrative samplers (i.e., those with large capacities and negligible release, like POCIS and DGT) provide better estimates of the time-weighted average (TWA)  $C_{\text{free}}$  if the sampling rate is known (which may be challenging to determine). The TWA  $C_{\text{free}}$  can be used to estimate total dissolved mass release from dredging to complement estimates of contaminant release associated with particles. However, the TWA  $C_{\text{free}}$  may be much less than the peak concentration, which would be used to predict toxic effects to nearby biota; determining peak concentrations are not yet achievable using passive sampling technology.
4. There remains some disagreement in the literature regarding the effects of highly adsorbing amendments (e.g., activated carbon) on the interpretation of passive sampler PRC loss. Some studies have found these sediment amendments can cause PRC molecules to release from the PSDs faster than the target molecules are accumulated by the passive samplers,<sup>121,136</sup> while others have found no artifactual effect. Further study of potential anisotropic effects and whether they are significant are needed to increase confidence in *in-situ*, PRC-corrected analyses for long-term monitoring at sites utilizing strongly adsorbing amendments as part of the remedy.

As discussed in the *Concurrence Analysis*, we lack a robust metric for comparing the effectiveness of remedies across sample endpoints, locations, and remedy type. As RPMs and environmental managers move toward utilizing PSDs at more sites, we request collection of more data comparing various passive sampling-based and conventional remedial endpoints (e.g., bioaccumulation). Ideally, analysis of this growing dataset of comparisons will lend itself to developing such a metric. For example, and as noted earlier, the development of mechanistic fate and transport models using PSD-based  $C_{\text{free}}$ s could provide more accurate estimates of exposure than current correlative relationships. Similarly, a future area of research for a subset of this dataset, or similar

datasets, is to compare measured bioaccumulation to modelled bioaccumulation. Hypothetically, predicted values based on using  $C_{\text{free}}$  to model bioaccumulation would be compared to measured bioaccumulation in the concurrence analysis. This assessment would address the magnitude of the uncertainty associated with interpreting and applying modelled bioaccumulation versus measured  $C_{\text{free}}$ .

5. For a range of reasons, as illustrated in this review, the use of passive sampling at contaminated sediments sites for evaluating remediation effectiveness is increasing. Reasons include (i) the greater sensitivity and lower levels of detection provided by PSDs resulting from the accumulation of target contaminants or CoCs compared to conventional grab sampling to (ii) the ability to compare PSD data directly to organism bioavailability in terms of exposure using  $C_{\text{free}}$  or bioaccumulation via passive sampler uptake. More practically, PSDs can be less expensive to deploy and chemically analyze than conventional metrics such as large volumes of water, sediments or tissues. In addition, in the cases of sediments and tissues, passive sampling polymers are often much simpler to ‘clean-up’ following target contaminant extraction. These very same characteristics also result in PSD data often being less variable than comparable conventional measures. While regulatory organizations globally continue to support the use of passive sampling for managing contaminated sediment sites,<sup>8, 10, 13</sup> a major impediment to their adoption more widely has been limited commercial chemical laboratory capacity. However, recent studies like those performed by Jonker et al.<sup>9, 11</sup> and Lotufo et al.<sup>12</sup> provide definitive scientific guidance for environmental regulators, including RPMs, and commercial laboratory managers to have confidence in the performance of PSDs for assessing remedial effectiveness at contaminated sediment sites.

## Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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## References

1. Gschwend PM; MacFarlane JK; Reible DD; Lu X; Hawthorne SB; Nakles DV; Thompson T Comparison of Polymeric Samplers for Accurately Assessing PCBs in Pore Waters. *Environ. Toxicol. Chem* 2011, 30 (6), 1288–1296. DOI: 10.1002/etc.510 [PubMed: 21360577]
2. Mayer P; Parkerton TF; Adams RG; Cargill JG; Gan J; Gouin T; Gschwend PM; Hawthorne SB; Helm P; Witt G; You J; Escher BI Passive sampling methods for contaminated sediments: Scientific rationale supporting use of freely dissolved concentrations. *Integr. Environ. Assess. Manage* 2014, 10 (2), 197–209. DOI: 10.1002/ieam.1508

3. Leslie HA; ter Laak TL; Busser FJM; Kraak MHS; Hermens JLM Bioconcentration of Organic Chemicals: Is a Solid-Phase Microextraction Fiber a Good Surrogate for Biota? *Environ. Sci. Technol* 2002, 36 (24), 5399–5404. DOI: 10.1021/es0257016 [PubMed: 12521167]
4. Friedman CL; Burgess RM; Perron MM; Cantwell MG; Ho KT; Lohmann R Comparing Polychaete and Polyethylene Uptake to Assess Sediment Resuspension Effects on PCB Bioavailability. *Environ. Sci. Technol* 2009, 43 (8), 2865–2870. DOI: 10.1021/es803695n [PubMed: 19475963]
5. Joyce AS; Portis LM; Parks AN; Burgess RM Evaluating the Relationship between Equilibrium Passive Sampler Uptake and Aquatic Organism Bioaccumulation. *Environ. Sci. Technol* 2016, 50 (21), 11437–11451. DOI: 10.1021/acs.est.6b03273 [PubMed: 27680295]
6. Schmidt SN; Burgess RM Evaluating Polymeric Sampling as a Tool for Predicting the Bioaccumulation of Polychlorinated Biphenyls by Fish and Shellfish. *Environ. Sci. Technol* 2020, 54 (16), 9729–9741. DOI: 10.1021/acs.est.9b07292 [PubMed: 32585088]
7. Ghosh U; Kane Driscoll S; Burgess RM; Jonker MTO; Reible D; Gobas F; Choi Y; Apitz SE; Maruya KA; Gala WR; Mortimer M; Beegan C Passive sampling methods for contaminated sediments: Practical guidance for selection, calibration, and implementation. *Integr. Environ. Assess. Manage* 2014, 10 (2), 210–223. DOI: 10.1002/ieam.1507
8. U.S. EPA/SERDP/ESTCP, Laboratory, Field, and Analytical Procedures for Using Passive Sampling in the Evaluation of Contaminated Sediments: User's Manual; Office of Research and Development: Washington, D. C., 20460, 2017.
9. Jonker MTO; Burgess RM; Ghosh U; Gschwend PM; Hale SE; Lohmann R; Lydy MJ; Maruya KA; Reible D; Smedes F Ex situ determination of freely dissolved concentrations of hydrophobic organic chemicals in sediments and soils: basis for interpreting toxicity and assessing bioavailability, risks and remediation necessity. *Nat. Protoc* 2020, 15 (5), 1800–1828. DOI: 10.1038/s41596-020-0311-y [PubMed: 32313252]
10. Booij K; Robinson CD; Burgess RM; Mayer P; Roberts CA; Ahrens L; Allan IJ; Brant J; Jones L; Kraus UR; Larsen MM; Lepom P; Petersen J; Pröfrock D; Roose P; Schäfer S; Smedes F; Tixier C; Vorkamp K; Whitehouse P Passive Sampling in Regulatory Chemical Monitoring of Nonpolar Organic Compounds in the Aquatic Environment. *Environ. Sci. Technol* 2016, 50 (1), 3–17. DOI: 10.1021/acs.est.5b04050 [PubMed: 26619247]
11. Jonker MTO; van der Heijden SA; Adelman D; Apell JN; Burgess RM; Choi Y; Fernandez LA; Flavetta GM; Ghosh U; Gschwend PM; Hale SE; Jalalizadeh M; Khairy M; Lampi MA; Lao W; Lohmann R; Lydy MJ; Maruya KA; Nutile SA; Oen AMP; Rakowska MI; Reible D; Rusina TP; Smedes F; Wu Y Advancing the Use of Passive Sampling in Risk Assessment and Management of Sediments Contaminated with Hydrophobic Organic Chemicals: Results of an International Ex Situ Passive Sampling Interlaboratory Comparison. *Environ. Sci. Technol* 2018, 52 (6), 3574–3582. DOI: 10.1021/acs.est.7b05752 [PubMed: 29488382]
12. Lotufo GR; Michalsen MM; Reible DD; Gschwend PM; Ghosh U; Kennedy AJ; Kerns KM; Rakowska MI; Odetayo A; MacFarlane JK; Yan S; Bokare M Interlaboratory Study of Polyethylene and Polydimethylsiloxane Polymeric Samplers for Ex Situ Measurement of Freely Dissolved Hydrophobic Organic Compounds in Sediment Porewater. *Environ. Toxicol. Chem* 2022, 41 (8), 1885–1902. DOI: 10.1002/etc.5356 [PubMed: 35512673]
13. U.S. EPA, Sediment Assessment and Monitoring Sheet (SAMS) #3: Guidelines for Using Passive Samplers to Monitor Organic Contaminants at Superfund Sediment Sites; Office of Solid Waste and Emergency Response: Washington, D. C., 20460, 2012.
14. U.S. EPA, Contaminated Sediment Remediation Guidance for Hazardous Waste Sites; Office of Solid Waste and Emergency Response: Washington, D. C., 20460, 2005.
15. Linkov I; Burmistrov D; Cura J; Bridges TS Risk-Based Management of Contaminated Sediments: Consideration of Spatial and Temporal Patterns in Exposure Modeling. *Environ. Sci. Technol* 2002, 36 (2), 238–246. DOI: 10.1021/es010721d [PubMed: 11827058]
16. Kvasnicka J; Stylianou KS; Nguyen VK; Huang L; Chiu WA; Burton AGA; Semrau J; Jolliet O Human Health Benefits from Fish Consumption vs. Risks from Inhalation Exposures Associated with Contaminated Sediment Remediation: Dredging of the Hudson River. *Environ. Health Perspect* 2019, 127 (12), 127004. DOI: 10.1289/EHP5034 [PubMed: 31834828]
17. Apitz SE; Davis JW; Finkelstein K; Hohreiter DW; Hoke R; Jensen RH; Jersak J; Kirtay VJ; Mack EE; Magar VS; Moore D; Reible D; Stahl RG Jr. Assessing and managing contaminated

- sediments: Part I, developing an effective investigation and risk evaluation strategy. *Integr. Environ. Assess. Manage* 2005, 1 (1), 2–8. DOI: 10.1897/ieam\_2004a-002.1
18. Apitz SE; Davis JW; Finkelstein K; Hohreiter DW; Hoke R; Jensen RH; Jersak J; Kirtay VJ; Mack EE; Magar VS; Moore D; Reible D; Stahl RG Jr. Assessing and managing contaminated sediments: Part II, evaluating risk and monitoring sediment remedy effectiveness. *Integr. Environ. Assess. Manage* 2005, 1 (1), e1–e14. DOI: 10.1897/IEAM\_2004a-002e.1
  19. Gustavson KE; Burton GA; Francingues NR Jr.; Reible DD; Vorhees DJ; Wolfe JR Evaluating the Effectiveness of Contaminated-Sediment Dredging. *Environ. Sci. Technol* 2008, 42 (14), 5042–5047. DOI: 10.1021/es087185a [PubMed: 18754345]
  20. Bergen BJ; Nelson WG; Mackay J; Dickerson D; Jayaraman S Environmental Monitoring Of Remedial Dredging At The New Bedford Harbor, Ma, Superfund Site. *Environ. Monit. Assess* 2005, 111 (1), 257–275. DOI: 10.1007/s10661-005-8223-4 [PubMed: 16311831]
  21. Nelson WG; Bergen BJ The New Bedford Harbor Superfund site long-term monitoring program (1993–2009). *Environ. Monit. Assess* 2012, 184 (12), 7531–7550. DOI: 10.1007/s10661-012-2517-0 [PubMed: 22367364]
  22. National Research Council, Toxicity Testing in the 21st Century: A Vision and a Strategy. The National Academies Press: Washington, DC, 2007.
  23. U.S. EPA, Introduction to Contaminated Sediments; Office of Science and Technology: Washington, DC, 20460, 1999.
  24. Zeman AJ; Patterson TS Preliminary results of demonstration capping project in Hamilton Harbour. *Water Qual. Res. J. Can* 1997, 32 (2), 439–452. DOI: 10.2166/wqrj.1997.028
  25. Azcue JM; Zeman AJ; Mudroch A; Rosa F; Patterson T Assessment of sediment and porewater after one year of subaqueous capping of contaminated sediments in Hamilton Harbour, Canada. *Water Sci. Technol* 1998, 37 (6–7), 323–329. DOI: 10.1016/S0273-1223(98)00214-5
  26. WebPlotDigitizer: Version 4.6; Rohatgi Ankit, 2022. <https://automeris.io/WebPlotDigitizer/index.html> (accessed 2022-12-22)
  27. Lin LI-K A Concordance Correlation Coefficient to Evaluate Reproducibility. *Biometrics* 1989, 45 (1), 255–268. DOI: 10.2307/2532051 [PubMed: 2720055]
  28. Lin LI-K Correction: A note on the concordance correlation coefficient. *Biometrics* 2000, 56 (1), 324–325. URL: <http://www.jstor.org/stable/2677159>
  29. Rousselet GA; Pernet CR; Wilcox RR The Percentile Bootstrap: A Primer With Step-by-Step Instructions in R. *Adv. Meth. Pract. Psychol. Sci* 2021, 4 (1), 2515245920911881. DOI: 10.1177/2515245920911881
  30. R: A language and environment for statistical computing; R Core Team: Vienna, Austria, 2022. <https://www.R-project.org/>
  31. RStudio: Integrated Development for R; RStudio, PBC: Boston, MA, 2022. <http://www.rstudio.com/>
  32. Wickham H; Averick M; Bryan J; Chang W; McGowan L. D. a.; François R; Golemund G; Hayes A; Henry L; Hester J; Kuhn M; Pedersen TL; Miller E; Bache SM; Müller K; Ooms J; Robinson D; Seidel DP; Spinu V; Takahashi K; Vaughan D; Wilke C; Woo K; Yutani H Welcome to the tidyverse. *J. Open Source Softw.* 2019, 4 (43), 1686. DOI: 10.21105/joss.01686
  33. Müller K here: A Simpler Way to Find Your Files. 2020. R package version 1.0.1 <https://CRAN.R-project.org/package=here>
  34. Signorell A et al. DescTools: Tools for descriptive statistics. 2022. R package version 0.99.47 <https://CRAN.R-project.org/package=DescTools>
  35. Kassambra A ggpubr: ‘ggplot2’ Based Publication Ready Plots. 2022. R package version 0.5.0 <https://CRAN.R-project.org/package=ggpubr>
  36. Chen Y; Yu W; Zheng R; Li J-Y; Zhang L; Wang Q; Yin J; Jin L Magnetic activated carbon (MAC) mitigates contaminant bioavailability in farm pond sediment and dietary risks in aquaculture products. *Sci. Total Environ* 2020, 736, 139185. DOI: 10.1016/j.scitotenv.2020.139185 [PubMed: 32485365]
  37. Fadaei H; Watson A; Place A; Connolly J; Ghosh U Effect of PCB Bioavailability Changes in Sediments on Bioaccumulation in Fish. *Environ. Sci. Technol* 2015, 49 (20), 12405–12413. DOI: 10.1021/acs.est.5b03107 [PubMed: 26402889]



38. Tomaszewski JE; McLeod PB; Luthy RG Measuring and modeling reduction of DDT availability to the water column and mussels following activated carbon amendment of contaminated sediment. *Water Res.* 2008, 42 (16), 4348–4356. DOI: 10.1016/j.watres.2008.07.016 [PubMed: 18723202]
39. Gomez-Eyles JL; Ghosh U Enhanced biochars can match activated carbon performance in sediments with high native bioavailability and low final porewater PCB concentrations. *Chemosphere* 2018, 203, 179–187. DOI: 10.1016/j.chemosphere.2018.03.132 [PubMed: 29614411]
40. Chen Y; Yu W; Jin L; Wang Q; Yin J; Lin J; Li J-Y Stabilization of hydrophobic organic contaminants in sediments by natural zeolites: bioavailability-based assessment of efficacy using equilibrium passive sampling. *J. Soils Sediments* 2019, 19 (12), 3898–3907. DOI: 10.1007/s11368-019-02381-9
41. Nybom I; Abel S; Waissi G; Väänänen K; Mäenpää K; Leppänen MT; Kukkonen JVK; Akkanen J Effects of Activated Carbon on PCB Bioaccumulation and Biological Responses of *Chironomus riparius* in Full Life Cycle Test. *Environ. Sci. Technol* 2016, 50 (10), 5252–5260. DOI: 10.1021/acs.est.6b00991 [PubMed: 27100921]
42. Bao L-J; Jia F; Crago J; Zeng EY; Schlenk D; Gan J Assessing bioavailability of DDT and metabolites in marine sediments using solid-phase microextraction with performance reference compounds. *Environ. Toxicol. Chem* 2013, 32 (9), 1946–1953. DOI: 10.1002/etc.2275 [PubMed: 23661411]
43. Parks AN; Chandler GT; Portis LM; Sullivan JC; Perron MM; Cantwell MG; Burgess RM; Ho KT; Ferguson PL Effects of single-walled carbon nanotubes on the bioavailability of PCBs in field-contaminated sediments. *Nanotoxicology* 2014, 8 (Suppl. 1), 111–117. DOI: 10.3109/17435390.2013.858794 [PubMed: 24266834]
44. Li J; Zhang C; Lin J; Yin J; Xu J; Chen Y Evaluating the bioavailability of heavy metals in natural-zeolite-amended aquatic sediments using thin-film diffusive gradients. *Aquac. Fish* 2018, 3 (3), 122–128. DOI: 10.1016/j.aaf.2018.05.003
45. Bailon MX; Park M; Solis KL; Na Y; Chaudhary DK; Kim S; Hong Y Reduction in mercury bioavailability to Asian clams (*Corbicula fluminea*) and changes in bacterial communities in sediments with activated carbon amendment. *Chemosphere* 2022, 291 (1), 132700. DOI: 10.1016/j.chemosphere.2021.132700 [PubMed: 34710454]
46. Zhang C; Shan B; Zhu Y; Tang W Remediation effectiveness of *Phyllostachys pubescens* biochar in reducing the bioavailability and bioaccumulation of metals in sediments. *Environ. Pollut* 2018, 242 (B), 1768–1776. DOI: 10.1016/j.envpol.2018.07.091 [PubMed: 30072221]
47. Gao M; Sun Q; Wang J; Ding S Investigation of the combined use of capping and oxidizing agents in the immobilization of arsenic in sediments. *Sci. Total Environ* 2021, 782, 146930. DOI: 10.1016/j.scitotenv.2021.146930 [PubMed: 33848870]
48. Lin J; Fu Z; Yao J; Wei X; Wang D; Ning D; Chen M Behavior of iron and other heavy metals in passivated sediments and the coupling effect on phosphorus. *The Sci. Total Environ* 2022, 808, 152151. DOI: 10.1016/j.scitotenv.2021.152151 [PubMed: 34875323]
49. Hale SE; Meynet P; Davenport RJ; Jones DM; Werner D Changes in polycyclic aromatic hydrocarbon availability in River Tyne sediment following bioremediation treatments or activated carbon amendment. *Water Res.* 2010, 44 (15), 4529–4536. DOI: 10.1016/j.watres.2010.06.027 [PubMed: 20630559]
50. Han Z; Sani B; Akkanen J; Abel S; Nybom I; Karapanagioti HK; Werner D A critical evaluation of magnetic activated carbon's potential for the remediation of sediment impacted by polycyclic aromatic hydrocarbons. *J. Hazard. Mater* 2015, 286, 41–47. DOI: 10.1016/j.jhazmat.2014.12.030 [PubMed: 25550081]
51. Payne RB; Ghosh U; May HD; Marshall CW; Sowers KR Mesocosm Studies on the Efficacy of Bioamended Activated Carbon for Treating PCB-Impacted Sediment. *Environ. Sci. Technol* 2017, 51 (18), 10691–10699. DOI: 10.1021/acs.est.7b01935 [PubMed: 28809549]
52. Millward RN; Bridges TS; Ghosh U; Zimmerman JR; Luthy RG Addition of activated carbon to sediments to reduce PCB bioaccumulation by a polychaete (*Neanthes arenaceodentata*) and an amphipod (*Leptocheirus plumulosus*). *Environ. Sci. Technol* 2005, 39 (8), 2880–2887. DOI: 10.1021/es048768x [PubMed: 15884389]

53. Zimmerman JR; Ghosh U; Millward RN; Bridges TS; Luthy RG Addition of carbon sorbents to reduce PCB and PAH bioavailability in marine sediments: Physicochemical tests. *Environ. Sci. Technol* 2004, 38 (20), 5458–5464. DOI: 10.1021/es034992v [PubMed: 15543751]
54. Zimmerman JR; Werner D; Ghosh U; Millward RN; Bridges TS; Luthy RG Effects of dose and particle size on activated carbon treatment to sequester polychlorinated biphenyls and polycyclic aromatic hydrocarbons in marine sediments. *Environ. Toxicol. Chem* 2005, 24 (7), 1594–1601. DOI: 10.1897/04-368R.1 [PubMed: 16050574]
55. Werner D; Higgins CP; Luthy RG The sequestration of PCBs in Lake Hartwell sediment with activated carbon. *Water Res.* 2005, 39 (10), 2105–2113. DOI: 10.1016/j.watres.2005.03.019 [PubMed: 15922398]
56. McLeod PB; Luoma SN; Luthy RG Biodynamic modeling of PCB uptake by *Macoma balthica* and *Corbicula fluminea* from sediment amended with activated carbon. *Environ. Sci. Technol* 2008, 42 (2), 484–490. DOI: 10.1021/es070139a [PubMed: 18284151]
57. Tomaszewski JE; Werner D; Luthy RG Activated carbon amendment as a treatment for residual DDT in sediment from a superfund site in San Francisco Bay, Richmond, California, USA. *Environ. Toxicol. Chem* 2007, 26 (10), 2143–2150. DOI: 10.1897/07-179R.1 [PubMed: 17867891]
58. Hale SE; Werner D Modeling the Mass Transfer of Hydrophobic Organic Pollutants in Briefly and Continuously Mixed Sediment after Amendment with Activated Carbon. *Environ. Sci. Technol* 2010, 44 (9), 3381–3387. DOI: 10.1021/es903582n [PubMed: 20392086]
59. Choi Y; Cho Y-M; Gala WR; Luthy RG Measurement and Modeling of Activated Carbon Performance for the Sequestration of Parent- and Alkylated-Polycyclic Aromatic Hydrocarbons in Petroleum-Impacted Sediments. *Environ. Sci. Technol* 2013, 47 (2), 1024–1032. DOI: 10.1021/es303770c [PubMed: 23240641]
60. Choi Y; Cho Y-M; Luthy RG In Situ Sequestration of Hydrophobic Organic Contaminants in Sediments under Stagnant Contact with Activated Carbon. 1. Column Studies. *Environ. Sci. Technol* 2014, 48 (3), 1835–1842. DOI: 10.1021/es403335g [PubMed: 24083415]
61. Rakowska MI; Kupryianchuk D; Grotenhuis T; Rijnaarts HHM; Koelmans AA Extraction of sediment-associated polycyclic aromatic hydrocarbons with granular activated carbon. *Environ. Toxicol. Chem* 2013, 32 (2), 304–311. DOI: 10.1002/etc.2066 [PubMed: 23147869]
62. Jia F; Liao C; Xue J; Taylor A; Gan J Comparing different methods for assessing contaminant bioavailability during sediment remediation. *Sci. Total Environ* 2016, 573, 270–277. DOI: 10.1016/j.scitotenv.2016.08.097 [PubMed: 27570195]
63. Amstaetter K; Eek E; Cornelissen G Sorption of PAHs and PCBs to activated carbon: Coal versus biomass-based quality. *Chemosphere* 2012, 87 (5), 573–578. DOI: 10.1016/j.chemosphere.2012.01.007 [PubMed: 22297197]
64. Choi Y; Wu Y; Sani B; Luthy RG; Werner D; Kim E Performance of retrievable activated carbons to treat sediment contaminated with polycyclic aromatic hydrocarbons. *J. Hazard. Mater* 2016, 320, 359–367. DOI: 10.1016/j.jhazmat.2016.08.047 [PubMed: 27566929]
65. Gustafsson Å; Hale S; Cornelissen G; Sjöholm E; Gunnarsson JS Activated carbon from kraft lignin: A sorbent for in situ remediation of contaminated sediments. *Environ. Technol. Innovation* 2017, 7, 160–168. DOI: 10.1016/j.eti.2016.11.001
66. Allan IJ; Raffard V; Kringstad A; Naes K Assessment of marine sediment remediation efficiency with SPME-based passive sampling measurement. *Sci. Total Environ* 2021, 756, 143854. DOI: 10.1016/j.scitotenv.2020.143854 [PubMed: 33279202]
67. Redell CJ; Elmore AC; Burken JG; Stringer RD Waterjet injection of powdered activated carbon for sediment remediation. *J. Soils Sediments* 2011, 11 (6), 1115–1124. DOI: 10.1007/s11368-011-0392-x
68. Stringer RD; Burken JG; Elmore AC; Reible DD Using in situ solid phase microextraction (SPME) for depth profiling in sediments treated with activated carbon. *J. Soils Sediments* 2014, 14 (5), 1013–1020. DOI: 10.1007/s11368-014-0857-9
69. Schaanning M; Breyholtz B; Skei J Experimental results on effects of capping on fluxes of persistent organic pollutants (POPs) from historically contaminated sediments. *Mar. Chem* 2006, 102 (1–2) 46–59. DOI: 10.1016/j.marchem.2005.10.027

70. Josefsson S; Schaanning M; Samuelsson GS; Gunnarsson JS; Olofsson I; Eek E; Wiberg K Capping Efficiency of Various Carbonaceous and Mineral Materials for In Situ Remediation of Polychlorinated Dibenzo-p-dioxin and Dibenzofuran Contaminated Marine Sediments: Sediment-to-Water Fluxes and Bioaccumulation in Boxcosm Tests. *Environ. Sci. Technol* 2012, 46 (6), 3343–3351. DOI: 10.1021/es203528v [PubMed: 22339559]
71. Lin J; Sun Q; Ding S; Wang D; Wang Y; Tsang DCW First observation of labile arsenic stratification in aluminum sulfate-amended sediments using high resolution Zr-oxide DGT. *Sci. Total Environ* 2017, 609, 304–310. DOI: 10.1016/j.scitotenv.2017.07.165 [PubMed: 28753505]
72. Cui J; Wang D; Lin J; Wang Y; Ren M; Yang Y; Shi P New application of lanthanum-modified bentonite (Phoslock) for immobilization of arsenic in sediments. *Environ. Sci. Pollut. Res* 2020, 28, 2052–2062. DOI: 10.1007/s11356-020-10565-x
73. Eek E; Cornelissen G; Kibsgaard A; Breedveld GD Diffusion of PAH and PCB from contaminated sediments with and without mineral capping; measurement and modelling. *Chemosphere* 2008, 71 (9), 1629–1638. DOI: 10.1016/j.chemosphere.2008.01.051 [PubMed: 18336865]
74. Lampert DJ; Sarchet WV; Reible DD Assessing the Effectiveness of Thin-Layer Sand Caps for Contaminated Sediment Management through Passive Sampling. *Environ. Sci. Technol* 2011, 45 (19), 8437–8443. DOI: 10.1021/es200406a [PubMed: 21846112]
75. Rämö R; Bonaglia S; Nybom I; Kreutzer A; Witt G; Sobek A; Gunnarsson JS Sediment Remediation Using Activated Carbon: Effects of Sorbent Particle Size and Resuspension on Sequestration of Metals and Organic Contaminants. *Environ. Toxicol. Chem* 2022, 41 (4), 1096–1110. DOI: 10.1002/etc.5292 [PubMed: 35040192]
76. Lin D; Cho Y-M; Werner D; Luthy RG Bioturbation Delays Attenuation of DDT by Clean Sediment Cap but Promotes Sequestration by Thin-Layered Activated Carbon. *Environ. Sci. Technol* 2014, 48 (2), 1175–1183. DOI: 10.1021/es404108h [PubMed: 24359108]
77. Lin D; Cho Y-M; Tommerdahl JP; Werner D; Luthy RG Bioturbation facilitates DDT sequestration by activated carbon against recontamination by sediment deposition. *Environ. Toxicol. Chem* 2018, 37 (7), 2013–2021. DOI: 10.1002/etc.4128 [PubMed: 29521428]
78. Schmidt SN; Wang AP; Gidley PT; Wooley AH; Lotufo GR; Burgess RM; Ghosh U; Fernandez LA; Mayer P Cross Validation of Two Partitioning-Based Sampling Approaches in Mesocosms Containing PCB Contaminated Field Sediment, Biota, and Activated Carbon Amendment. *Environ. Sci. Technol* 2017, 51 (17), 9996–10004. DOI: 10.1021/acs.est.7b01909 [PubMed: 28766940]
79. Gidley PT; Kennedy AJ; Lotufo GR; Wooley AH; Melby NL; Ghosh U; Burgess RM; Mayer P; Fernandez LA; Schmidt SN; Wang AP; Bridges TS; Ruiz CE Bioaccumulation in Functionally Different Species: Ongoing Input of PCBs with Sediment Deposition to Activated Carbon Remediated Bed Sediments. *Environ. Toxicol. Chem* 2019, 38 (10), 2326–2336. DOI: 10.1002/etc.4526 [PubMed: 31233239]
80. Gidley PT; Lotufo GR; Kennedy AJ; Melby NL; Wooley AH; Laber CH; Burgess RM; Ruiz CE; Bridges TS Effect of Activated Carbon in Thin Sand Caps Challenged with Ongoing PCB Inputs from Sediment Deposition: PCB Uptake in Clams (*Mercenaria mercenaria*) and Passive Samplers. *Arch. Environ. Contam. Toxicol* 2021, 82 (1), 95–104. DOI: 10.1007/s00244-021-00894-4 [PubMed: 34669000]
81. Bridges TS; Gidley PT; Lotufo GR; Kennedy AJ; Ruiz CE; Melby NL; Ballentine ML; Wooley A; Laber C; Ghosh U; Burgess RM; Fernandez LA; Wang A; Mayer P; Schmidt S Quantitative Thermodynamic Exposure Assessment (QTEA) Supporting Resilient Contaminated Sediment Site Restoration; Final Report for ER-2431; SERDP: Alexandria, VA, 22350, 2022.
82. Cho Y-M; Ghosh U; Kennedy AJ; Grossman A; Ray G; Tomaszewski JE; Smithenry DW; Bridges TS; Luthy RG Field Application of Activated Carbon Amendment for In-Situ Stabilization of Polychlorinated Biphenyls in Marine Sediment. *Environ. Sci. Technol* 2009, 43 (10), 3815–3823. DOI: 10.1021/es802931c [PubMed: 19544893]
83. Beckingham B; Ghosh U Polyoxymethylene passive samplers to monitor changes in bioavailability and flux of PCBs after activated carbon amendment to sediment in the field. *Chemosphere* 2013, 91 (10), 1401–1407. DOI: 10.1016/j.chemosphere.2012.12.074 [PubMed: 23415491]

84. Kirtay V; Conder J; Rosen G; Magar V; Grover M; Arblaster J; Fetters K; Chadwick B Performance of an in situ activated carbon treatment to reduce PCB availability in an active harbor. *Environ. Toxicol. Chem* 2018, 37 (6), 1767–1777. DOI: 10.1002/etc.4121 [PubMed: 29480553]
85. Cho Y-M; Smithenry DW; Ghosh U; Kennedy AJ; Millward RN; Bridges TS; Luthy RG Field methods for amending marine sediment with activated carbon and assessing treatment effectiveness. *Mar. Environ. Res* 2007, 64 (5), 541–555. DOI: 10.1016/j.marenvres.2007.04.006 [PubMed: 17570482]
86. Payne RB; Ghosh U; May HD; Marshall CW; Sowers KR A Pilot-Scale Field Study: In Situ Treatment of PCB-Impacted Sediments with Bioamended Activated Carbon. *Environ. Sci. Technol* 2019, 53 (5), 2626–2634. DOI: 10.1021/acs.est.8b05019 [PubMed: 30698958]
87. Lampert DJ; Lu X; Reible DD Long-term PAH monitoring results from the Anacostia River active capping demonstration using polydimethylsiloxane (PDMS) fibers. *Environ. Sci.: Processes Impacts* 2013, 15 (3), 554–562. DOI: 10.1039/C3EM30826J
88. Cornelissen G; Amstaetter K; Hauge A; Schaanning M; Beylich B; Gunnarsson JS; Breedveld GD; Oen AMP; Eek E Large-Scale Field Study on Thin-Layer Capping of Marine PCDD/F-Contaminated Sediments in Grenlandfjords, Norway: Physicochemical Effects. *Environ. Sci. Technol* 2012, 46 (21), 12030–12037. DOI: 10.1021/es302431u [PubMed: 23046183]
89. Fernandez LA; Lao W; Maruya KA; Burgess RM Calculating the Diffusive Flux of Persistent Organic Pollutants between Sediments and the Water Column on the Palos Verdes Shelf Superfund Site Using Polymeric Passive Samplers. *Environ. Sci. Technol* 2014, 48 (7), 3925–3934. DOI: 10.1021/es404475c [PubMed: 24564763]
90. Fernandez LA; Lao W; Maruya KA; White C; Burgess RM Passive Sampling to Measure Baseline Dissolved Persistent Organic Pollutant Concentrations in the Water Column of the Palos Verdes Shelf Superfund Site. *Environ. Sci. Technol* 2012, 46 (21), 11937–11947. DOI: 10.1021/es302139y [PubMed: 23062073]
91. Lin D; Cho Y-M; Oen A; Eek E; Tommerdahl JP; Luthy RG Toolset for assessment of natural recovery from legacy contaminated sediment: Case study of Pallanza Bay, Lake Maggiore, Italy. *Water Res* 2017, 121, 109–119. DOI: 10.1016/j.watres.2017.05.024 [PubMed: 28525783]
92. Sanders JP; Andrade NA; Ghosh U Evaluation of passive sampling polymers and nonequilibrium adjustment methods in a multiyear surveillance of sediment porewater PCBs. *Environ. Toxicol. Chem* 2018, 37 (9), 2487–2495. DOI: 10.1002/etc.4223 [PubMed: 29978585]
93. Sanders JP; Andrade NA; Menzie CA; Amos CB; Gilmour CC; Henry EA; Brown SS; Ghosh U Persistent reductions in the bioavailability of PCBs at a tidally inundated *Phragmites australis* marsh amended with activated carbon. *Environ. Toxicol. Chem* 2018, 37 (9), 2496–2505. DOI: 10.1002/etc.4186 [PubMed: 29870109]
94. Cornelissen G; Kruså ME; Breedveld GD; Eek E; Oen AMP; Arp HPH; Raymond C; Samuelsson G; Hedman JE; Stokland Ø; Gunnarsson JS Remediation of contaminated marine sediment using thin-layer capping with activated carbon—a field experiment in Trondheim harbor, Norway. *Environ. Sci. Technol* 2011, 45 (14), 6110–6116. DOI: 10.1021/es2011397 [PubMed: 21671651]
95. Yu J; Chen Q; Zhang J; Zhong J; Fan C; Hu L; Shi W; Yu W; Zhang Y In situ simulation of thin-layer dredging effects on sediment metal release across the sediment-water interface. *Sci. Total Environ* 2019, 658, 501–509. DOI: 10.1016/j.scitotenv.2018.12.226 [PubMed: 30579207]
96. Vrana B; Kluárová V; Benická E; Abou-Mrad N; Amdany R; Horáková S; Draxler A; Humer F; Gans O Passive sampling: An effective method for monitoring seasonal and spatial variability of dissolved hydrophobic organic contaminants and metals in the Danube river. *Environ. Pollut* 2014, 184, 101–112. DOI: 10.1016/j.envpol.2013.08.018 [PubMed: 24047546]
97. Allan SE; Sower GJ; Anderson KA Estimating risk at a Superfund site using passive sampling devices as biological surrogates in human health risk models. *Chemosphere* 2011, 85 (6), 920–927. DOI: 10.1016/j.chemosphere.2011.06.051 [PubMed: 21741671]
98. Sower GJ; Anderson KA Spatial and Temporal Variation of Freely Dissolved Polycyclic Aromatic Hydrocarbons in an Urban River Undergoing Superfund Remediation. *Environ. Sci. Technol* 2008, 42 (24), 9065–9071. DOI: 10.1021/es801286z [PubMed: 19174872]
99. Martinez A; Awad AM; Herkert NJ; Hornbuckle KC Determination of PCB fluxes from Indiana Harbor and Ship Canal using dual-deployed air and water passive samplers. *Environ. Pollut* 2019, 244, 469–476. DOI: 10.1016/j.envpol.2018.10.048 [PubMed: 30366294]

100. Cornelissen G; Arp HPH; Pettersen A; Hauge A; Breedveld GD Assessing PAH and PCB emissions from the relocation of harbour sediments using equilibrium passive samplers. *Chemosphere* 2008, 72 (10), 1581–1587. DOI: 10.1016/j.chemosphere.2008.04.041 [PubMed: 18514764]
101. Schaanning MT; Harman C; Staalstrøm A Release of dissolved trace metals and organic contaminants during deep water disposal of contaminated sediments from Oslo harbour, Norway. *J. Soils Sediments* 2011, 11 (8), 1477–1489. DOI: 10.1007/s11368-011-0436-2
102. Joyce AS; Fernandez LA; Burgess RM In Situ Investigation of Performance Reference Compound-Based Estimates of PCB Equilibrated Passive Sampler Concentrations and  $C_{free}$  in the Marine Water Column. *Environ. Toxicol. Chem* 2020, 39 (6), 1165–1173. DOI: 10.1002/etc.4714 [PubMed: 32187698]
103. Schneider AR; Porter ET; Baker JE Polychlorinated Biphenyl Release from Resuspended Hudson River Sediment. *Environ. Sci. Technol* 2007, 41 (4), 1097–1103. DOI: 10.1021/es0607584 [PubMed: 17593705]
104. Belles A; Mamindy-Pajany Y; Alary C Simulation of aromatic polycyclic hydrocarbons remobilization from a river sediment using laboratory experiments supported by passive sampling techniques. *Environ. Sci. Pollut. Res* 2016, 23 (3), 2426–2436. DOI: 10.1007/s11356-015-5462-y
105. Allan IJ; Nilsson HC; Tjensvoll I; Bradshaw C; Næs K PCDD/F release during benthic trawler-induced sediment resuspension. *Environ. Toxicol. Chem* 2012, 31 (12), 2780–2787. DOI: 10.1002/etc.1999 [PubMed: 22936523]
106. Cantwell MG; Burgess RM; Kester DR Release and Phase Partitioning of Metals from Anoxic Estuarine Sediments during Periods of Simulated Resuspension. *Environ. Sci. Technol* 2002, 36 (24), 5328–5334. DOI: 10.1021/es0115058 [PubMed: 12521157]
107. Layglon N; Misson B; Durieu G; Coclet C; D’Onofrio S; Duc Huy D; François D; Mullot J-U; Mounier S; Lenoble V; Omanovi D; Garnier C Long-term monitoring emphasizes impacts of the dredging on dissolved Cu and Pb contamination along with ultraplankton distribution and structure in Toulon Bay (NW Mediterranean Sea, France). *Mar. Pollut. Bull* 2020, 156, 111196. DOI: 10.1016/j.marpolbul.2020.111196 [PubMed: 32510358]
108. Villanueva JD; Coustumer PL; Huneau F; Motelica-Heino M; Perez TR; Materum R; Espaldon MVO; Stoll S Assessment of Trace Metals during Episodic Events using DGT Passive Sampler: A Proposal for Water Management Enhancement. *Water Resour. Manag* 2013, 27 (12), 4163–4181. DOI: 10.1007/s11269-013-0401-5
109. Villanueva JD; Le Coustumer P; Denis A; Abuyan R; Huneau F; Motelica-heino M; Peyraube N; Celle-jeanton H; Perez TR; Espaldon MV Trends of labile trace metals in tropical urban water under highly contrasted weather conditions. *Environ. Sci. Pollut. Res. International* 2015, 22 (18), 13842–13857. DOI: 10.1007/s11356-015-4835-6
110. Fetters K; Rosen G; Kirtay V; Chadwick B; Conder J; Sacks Victoria P; Grover M; Magar V Demonstration and validation of enhanced monitored natural recovery at a pesticide-contaminated sediment site. *J. Soils Sediments* 2020, 20 (1), 204–219. DOI: 10.1007/s11368-019-02386-4
111. Li J-Y; Yu W; Yin J; Chen Y; Wang Q; Jin L Reduced bioavailability and ecological risks of polycyclic aromatic hydrocarbons in Yangshan port of East China Sea: Remediation effectiveness in the transition from construction to operation. *Sci. Total Environ* 2019, 687, 679–686. DOI: 10.1016/j.scitotenv.2019.06.040 [PubMed: 31220721]
112. Chen M; Ding S; Gao S; Fu Z; Tang W; Wu Y; Gong M; Wang D; Wang Y Efficacy of dredging engineering as a means to remove heavy metals from lake sediments. *Sci. Total Environ* 2019, 665, 181–190. DOI: 10.1016/j.scitotenv.2019.02.057 [PubMed: 30772548]
113. Chen M; Ding S; Gao S; Xu S; Yang C; Wu Y; Gong M; Wang D; Wang Y Long-term effects of sediment dredging on controlling cobalt, zinc, and nickel contamination determined by chemical fractionation and passive sampling. *Chemosphere* 2019, 220, 476–485. DOI: 10.1016/j.chemosphere.2018.12.138 [PubMed: 30594799]
114. Sun Q; Ding S; Chen M; Gao S; Lu G; Wu Y; Gong M; Wang D; Wang Y Long-term effectiveness of sediment dredging on controlling the contamination of arsenic, selenium, and antimony. *Environ. Pollut* 2019, 245, 725–734. DOI: 10.1016/j.envpol.2018.11.050 [PubMed: 30500752]

115. Gao S; Xing X; Ding S; Fan X The Long-Term Effects of Dredging on Chromium Pollution in the Sediment of Meiliang Bay, Lake Taihu, China. *Water* 2021, 13 (3), 327. DOI: 10.3390/w13030327
116. Parker R; Bolam T; Barry J; Mason C; Kröger S; Warford L; Silburn B; Sivyer D; Birchenough S; Mayes A; Fones GR The application of Diffusive Gradients in Thin Films (DGT) for improved understanding of metal behaviour at marine disposal sites. *Sci. Total Environ* 2017, 575, 1074–1086. DOI: 10.1016/j.scitotenv.2016.09.183 [PubMed: 27693155]
117. Thomas C; Lampert D; Reible D; Janssen EML; Luthy RG Remedy performance monitoring at contaminated sediment sites using profiling solid phase microextraction (SPME) polydimethylsiloxane (PDMS) fibers. *Environ. Sci.: Processes Impacts* 2014, 16 (3), 445–452. DOI: 10.1039/C3EM00695F
118. Minick DJ; Anderson KA Diffusive Flux of Paha across Sediment-Water and Water-Air Interfaces at Urban Superfund Sites. *Environ. Toxicol. Chem* 2017, 36 (9), 2281–2289. DOI: 10.1002/etc.3785 [PubMed: 28262984]
119. Oen AMP; Pettersen A; Eek E; Glette T; Brooks L; Breedveld GD Monitoring Chemical and Biological Recovery at a Confined Aquatic Disposal Site, Oslofjord, Norway. *Environ. Toxicol. Chem* 2017, 36 (9), 2552–2559. DOI: 10.1002/etc.3794 [PubMed: 28294397]
120. Schubauer-Berigan JP; Foote EA; Magar VS Using SPMDs to Assess Natural Recovery of PCB-contaminated Sediments in Lake Hartwell, SC: I. A Field Test of New In-Situ Deployment Methods. *Soil Sediment Contam.* 2012, 21 (1), 82–100. DOI: 10.1080/15320383.2012.636777
121. Patmont E; Jalalizadeh M; Bokare M; Needham T; Vance J; Greene R; Cargill J; Ghosh U Full-Scale Application of Activated Carbon to Reduce Pollutant Bioavailability in a 5-Acre Lake. *J. Environ. Eng* 2020, 146 (5), 04020024. DOI: 10.1061/(ASCE)EE.1943-7870.0001667
122. Odetayo AA; Reible DD; Acevedo-Mackey D; Price C; Thai L Development of polyoxymethylene passive sampler for assessing air concentrations of PCBs at a confined disposal facility (CDF). *Environ. Pollut* 2020, 265 (Part A), 114720. DOI: 10.1016/j.envpol.2020.114720 [PubMed: 32473506]
123. Odetayo AA; Reible DD; Acevedo-Mackey D; Price C; Thai L Application of polyoxymethylene passive air sampler to monitor hydrophobic organics in air around a confined disposal facility. *Chemosphere* 2021, 263, 127827. DOI: 10.1016/j.chemosphere.2020.127827 [PubMed: 32835966]
124. Tidwell LG; Paulik LB; Anderson KA Air-water exchange of PAHs and OPAHs at a superfund mega-site. *Sci. Total Environ* 2017, 603, 676–686. DOI: 10.1016/j.scitotenv.2017.01.185 [PubMed: 28372820]
125. Eek E; Cornelissen G; Breedveld GD Field Measurement of Diffusional Mass Transfer of HOCs at the Sediment-Water Interface. *Environ. Sci. Technol* 2010, 44 (17), 6752–6759. DOI: 10.1021/es100818w [PubMed: 20704281]
126. Rosen G; Chadwick DB; Colvin MA; Stransky C; Burton A; Radford J; Bailey H; Cibor A; Grover M; Greenberg M Demonstration and Commercialization of the Sediment Ecosystem Assessment Protocol; Final Report for ER-201130; SERDP: Alexandria, VA, 22350, 2017.
127. Rusina TP; Smedes F; Klanova J; Booij K; Holoubek I Polymer selection for passive sampling: A comparison of critical properties. *Chemosphere* 2007, 68 (7), 1344–1351. DOI: 10.1016/j.chemosphere.2007.01.025 [PubMed: 17331562]
128. Cornelissen G; Breedveld GD; Naes K; Oen AMP; Ruus A Bioaccumulation of native polycyclic aromatic hydrocarbons from sediment by a polychaete and a gastropod: Freely dissolved concentrations and activated carbon amendment. *Environ. Toxicol. Chem* 2006, 25 (9), 2349–2355. DOI: 10.1897/06-026R.1 [PubMed: 16986789]
129. Lin J; Fu Z; Ding S; Ren M; Gao S Laboratory investigation on calcium nitrate induced coupling reactions between nitrogen, phosphorus, sulfur, and metals in contaminated sediments. *Environ. Sci. Pollut. Res* 2021, 28 (20), 25866–25877. DOI: 10.1007/s11356-021-12441-8
130. Han Z; Abel S; Akkanen J; Werner D Evaluation of strategies to minimize ecotoxic side-effects of sorbent-based sediment remediation. *J. Chem. Technol. Biotechnol* 2017, 92 (8), 1938–1942. DOI: 10.1002/jctb.5224 [PubMed: 28781403]

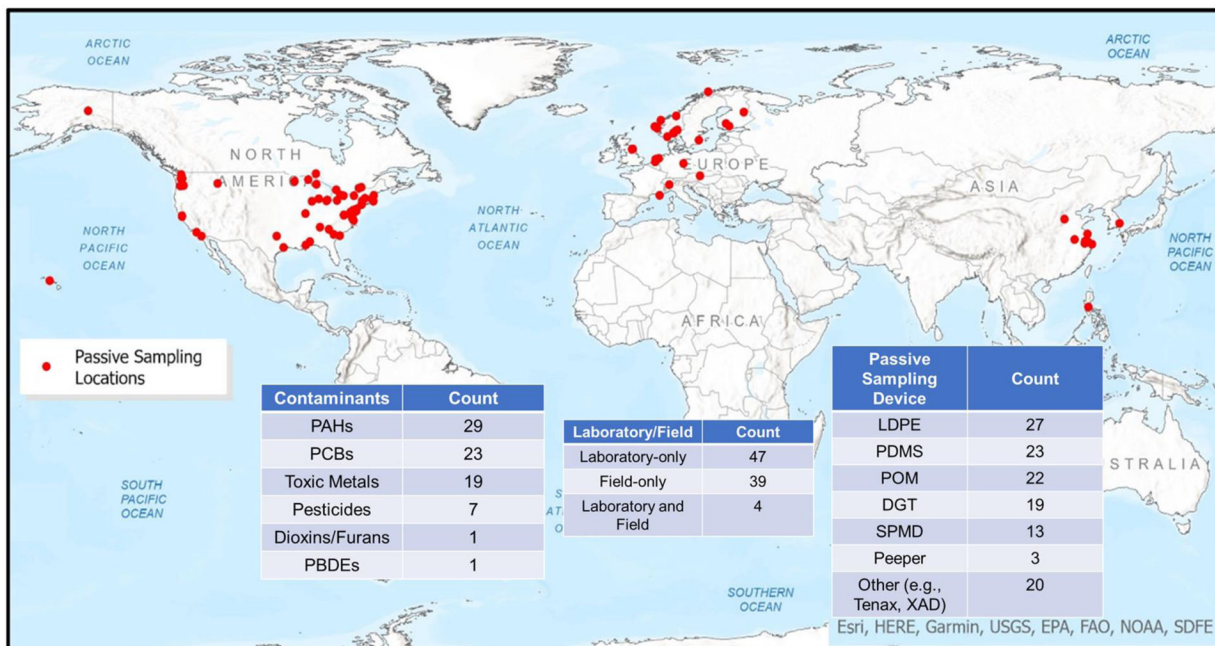
131. Kupryianchyk D; Reichman EP; Rakowska MI; Peeters ETHM; Grotenhuis JTC; Koelmans AA Ecotoxicological Effects of Activated Carbon Amendments on Macroinvertebrates in Nonpolluted and Polluted Sediments. *Environ. Sci. Technol* 2011, 45 (19), 8567–8574. DOI: 10.1021/es2014538 [PubMed: 21846106]
132. Cornelissen G; Breedveld GD; Kalaitzidis S; Christanis K; Kibsgaard A; Oen AMP Strong sorption of native PAHs to pyrogenic and unburned carbonaceous geosorbents in sediments. *Environ. Sci. Technol* 2006, 40 (4), 1197–1203. DOI: 10.1021/es0520722 [PubMed: 16572775]
133. Nybom I; Waissi-Leinonen G; Mäenpää K; Leppänen MT; Kukkonen JVK; Werner D; Akkanen J Effects of activated carbon ageing in three PCB contaminated sediments: Sorption efficiency and secondary effects on *Lumbriculus variegatus*. *Water Res.* 2015, 85, 413–421. DOI: 10.1016/j.watres.2015.08.044 [PubMed: 26364225]
134. Sun X; Ghosh U The effect of activated carbon on partitioning, desorption, and biouptake of native polychlorinated biphenyls in four freshwater sediments. *Environ. Toxicol. Chem* 2008, 27 (11), 2287–95. DOI: 10.1897/08-020.1 [PubMed: 18517308]
135. Thompson JM; Hsieh C-H; Hoelen TP; Weston DP; Luthy RG Measuring and Modeling Organochlorine Pesticide Response to Activated Carbon Amendment in Tidal Sediment Mesocosms. *Environ. Sci. Technol* 2016, 50 (9), 4769–4777. DOI: 10.1021/acs.est.5b05669 [PubMed: 27040592]
136. Janssen EML; Oen AMP; Luoma SN; Luthy RG Assessment of Field-Related Influences on Polychlorinated Biphenyl Exposures and Sorbent Amendment Using Polychaete Bioassays and Passive Sampler Measurements. *Environ. Toxicol. Chem* 2011, 30 (1), 173–180. DOI: 10.1002/etc.367 [PubMed: 20872900]
137. Yan S; Rakowska M; Shen X; Himmer T; Irvine C; Zajac-Fay R; Eby J; Janda D; Ohannessian S; Reible DD Bioavailability assessment in activated carbon treated coastal sediment with in situ and ex situ porewater measurements. *Water Res.* 2020, 185, 116259–116259. DOI: 10.1016/j.watres.2020.116259 [PubMed: 32798889]
138. Cho Y-M; Werner D; Choi Y; Luthy RG Long-term monitoring and modeling of the mass transfer of polychlorinated biphenyls in sediment following pilot-scale *in-situ* amendment with activated carbon. *J. Contam. Hydrol* 2012, 129–130, 25–37. DOI: 10.1016/j.jconhyd.2011.09.009
139. Tomaszewski JE; Luthy RG Field deployment of polyethylene devices to measure PCB concentrations in pore water of contaminated sediment. *Environ. Sci. Technol* 2008, 42 (16), 6086–6091. DOI: 10.1021/es800582a [PubMed: 18767670]
140. Oen AMP; Janssen EML; Cornelissen G; Breedveld GD; Eek E; Luthy RG In Situ Measurement of PCB Pore Water Concentration Profiles in Activated Carbon-Amended Sediment Using Passive Samplers. *Environ. Sci. Technol* 2011, 45 (9), 4053–4059. DOI: 10.1021/es200174v [PubMed: 21473574]
141. Rakowska MI; Kupryianchyk D; Koelmans AA; Grotenhuis T; Rijnaarts HHM Equilibrium and kinetic modeling of contaminant immobilization by activated carbon amended to sediments in the field. *Water Res.* 2014, 67, 96–104. DOI: 10.1016/j.watres.2014.07.046 [PubMed: 25262554]
142. Kupryianchyk D; Noori A; Rakowska MI; Grotenhuis JTC; Koelmans AA Bioturbation and Dissolved Organic Matter Enhance Contaminant Fluxes from Sediment Treated with Powdered and Granular Activated Carbon. *Environ. Sci. Technol* 2013, 47 (10), 5092–5100. DOI: 10.1021/es3040297 [PubMed: 23590290]
143. Kupryianchyk D; Rakowska MI; Roessink I; Reichman EP; Grotenhuis JTC; Koelmans AA In situ Treatment with Activated Carbon Reduces Bioaccumulation in Aquatic Food Chains. *Environ. Sci. Technol* 2013, 47 (9), 4563–4571. DOI: 10.1021/es305265x [PubMed: 23544454]
144. Knox AS; Paller MH; Roberts J Active capping technology—New approaches for in situ remediation of contaminated sediments. *Remediation* 2012, 22 (2), 93–117. DOI: 10.1002/rem.21313
145. Bailon MX; Park M; Choi Y-G; Reible D; Hong Y The application of DGTs for assessing the effectiveness of in situ management of Hg and heavy metal contaminated sediment. *Membr. Water Treat* 2020, 11 (1), 11–23. DOI: 10.12989/mwt.2020.11.1.011
146. Jacobs PH A new rechargeable dialysis pore water sampler for monitoring sub-aqueous in-situ sediment caps. *Water Res.* 2002, 36 (12), 3121–3129. DOI: 10.1016/S0043-1354(01)00542-5 [PubMed: 12171411]

147. Jacobs PH Monitoring of subaqueous depots with active barrier systems for contaminated dredged material using dialysis samplers and DGT probes. *J. Soils Sediments* 2003, 3 (2), 100–107. DOI: 10.1007/BF02991075
148. Brennan AA; Johnson NW The Utility of Solid-Phase Microextraction in Evaluating Polycyclic Aromatic Hydrocarbon Bioavailability During Habitat Restoration with Dredged Material at Moderately Contaminated Sites. *Integr. Environ. Assess. Manage* 2018, 14 (2), 212–223. DOI: 10.1002/ieam.1994
149. Samuelsson GS; Hedman JE; Kruså ME; Gunnarsson JS; Cornelissen G Capping in situ with activated carbon in Trondheim harbor (Norway) reduces bioaccumulation of PCBs and PAHs in marine sediment fauna. *Mar. Environ. Res* 2015, 109, 103–112. DOI: 10.1016/j.marenvres.2015.06.003 [PubMed: 26121661]
150. Oen AMP; Beckingham B; Ghosh U; Kruså ME; Luthy RG; Hartnik T; Henriksen T; Cornelissen G Sorption of Organic Compounds to Fresh and Field-Aged Activated Carbons in Soils and Sediments. *Environ. Sci. Technol* 2012, 46 (2), 810–817. DOI: 10.1021/es202814e [PubMed: 22128748]
151. Allan SE; Smith BW; Tanguay RL; Anderson KA Bridging environmental mixtures and toxic effects. *Environ. Toxicol. Chem* 2012, 31 (12), 2877–2887. DOI: 10.1002/etc.2018 [PubMed: 23001962]
152. Smith AV; Shen X; Garza-Rubalcava U; Gardiner W; Reible D In Situ Passive Sampling to Monitor Long Term Cap Effectiveness at a Tidally Influenced Shoreline. *Toxics* 2022, 10 (3), 106. DOI: 10.3390/toxics10030106 [PubMed: 35324732]
153. Reible DD, Ed. Processes, Assessment and Remediation of Contaminated Sediments; SERDP ESTCP Environmental Remediation Technology; Springer New York: New York, NY, 2014; Vol. 6; pp 1–12. DOI: 10.1007/978-1-4614-6726-7\_1
154. Burgess RM; Scott KJ The Significance of In-Place\* Contaminated Marine Sediments on the Water Column: Processes and Effects. In *Sediment Toxicity Assessment*, Burton GA, Ed.; CRC Press: Boca Raton: 1992; Chapter 7, pp 129–165.
155. Hilber I; Bucheli TD Activated Carbon Amendment to Remediate Contaminated Sediments and Soils: A Review. *Global NEST J* 2010, 12 (3), 305–317. DOI: 10.30955/gnj.000723
156. Rakowska MI; Kupryianchik D; Harmsen J; Grotenhuis T; Koelmans AA In situ remediation of contaminated sediments using carbonaceous materials. *Environ. Toxicol. Chem* 2012, 31 (4), 693–704. DOI: 10.1002/etc.1763 [PubMed: 22389227]
157. Patmont CR; Ghosh U; LaRosa P; Menzie CA; Luthy RG; Greenberg MS; Cornelissen G; Eek E; Collins J; Hull J; Hjarland T; Glaza E; Bleiler J; Quadrini J In Situ Sediment Treatment Using Activated Carbon: A Demonstrated Sediment Cleanup Technology. *Integr. Environ. Assess. Manage* 2015, 11 (2), 195–207. DOI: 10.1002/ieam.1589
158. Labianca C; De Gisi S; Todaro F; Notarnicola M; Bortone I A review of the in-situ capping amendments and modeling approaches for the remediation of contaminated marine sediments. *Sci. Total Environ* 2022, 806 (Part 3), 151257. DOI: 10.1016/j.scitotenv.2021.151257 [PubMed: 34710404]
159. Meric D; Sheahan TC; Alshawabkeh A; Shine J A Consolidation and Contaminant Transport Device for Assessing Reactive Mat Effectiveness for Subaqueous Sediment Remediation. *Geotech. Test. J* 2010, 33 (6), 423–433. DOI: 10.1520/GTJ102880
160. Meric D; Barbuto SM; Alshawabkeh AN; Shine JP; Sheahan TC Effect of reactive core mat application on bioavailability of hydrophobic organic compounds. *Sci. Total Environ* 2012, 423, 168–175. DOI: 10.1016/j.scitotenv.2012.01.042 [PubMed: 22386995]
161. Ghosh U; Luthy RG; Cornelissen G; Werner D; Menzie CA In-situ Sorbent Amendments: A New Direction in Contaminated Sediment Management. *Environ. Sci. Technol* 2011, 45 (4), 1163–1168. DOI: 10.1021/es102694h [PubMed: 21247210]
162. U.S. EPA, Use of Amendments for In Situ Remediation at Superfund Sediment Sites; Office of Solid Waste and Emergency Response: Washington, D. C., 20460, 2013.
163. National Research Council, Sediment Dredging at Superfund Megsites: Assessing the Effectiveness. The National Academies Press: Washington, DC, 2007; p 316.

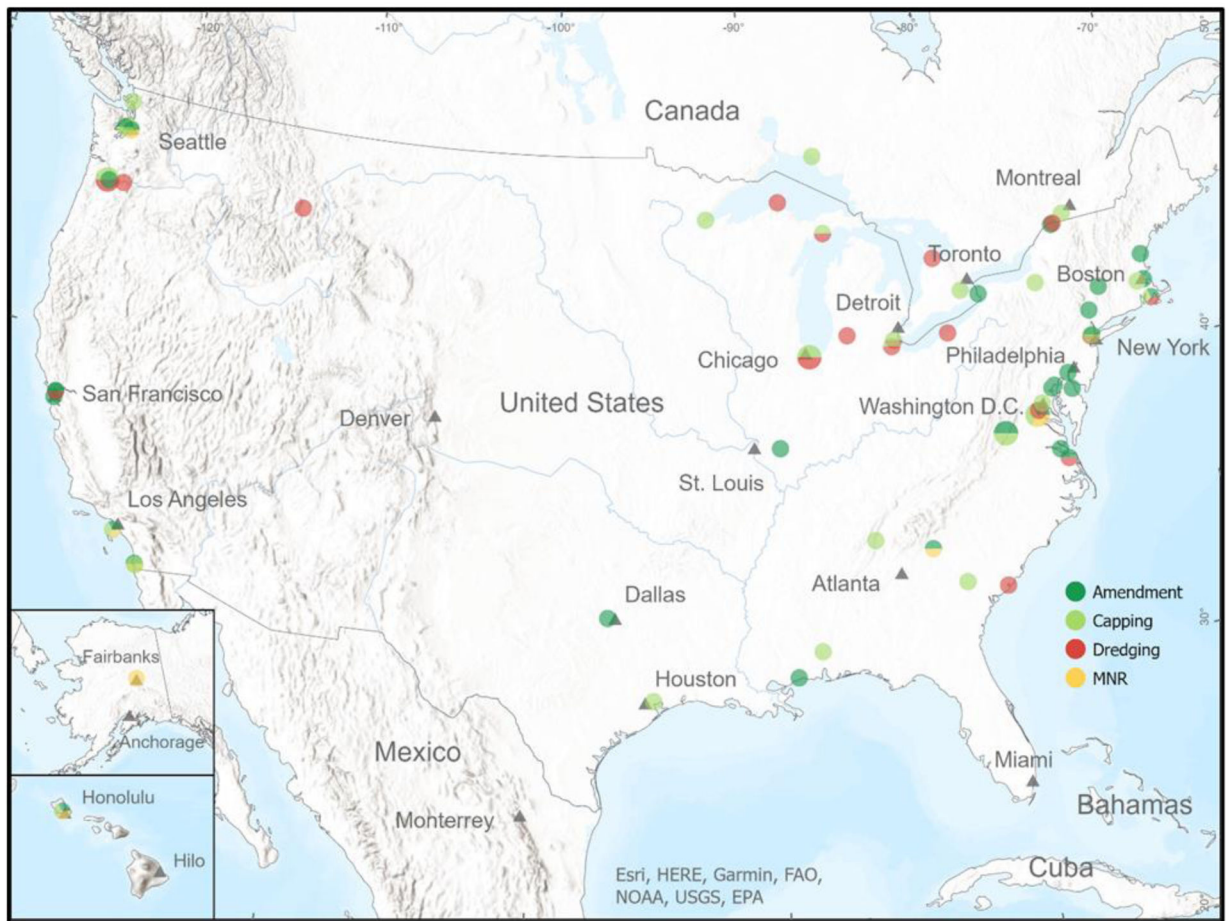


164. Magar VS; Wenning RJ The Role of Monitored Natural Recovery in Sediment Remediation. *Integr. Environ. Assess. Manage* 2006, 2 (1), 66–74. DOI: 10.1002/ieam.5630020112
165. Bridges TS; Gustavson KE; Schroeder P; Ells SJ; Hayes D; Nadeau SC; Palermo MR; Patmont C Dredging processes and remedy effectiveness: Relationship to the 4 Rs of environmental dredging. *Integr. Environ. Assess. Manage* 2010, 6 (4), 619–630. DOI: 10.1002/ieam.71
166. Merritt KA; Conder J; Kirtay V; Chadwick DB; Magar V Review of Thin-Layer Placement Applications to Enhance Natural Recovery of Contaminated Sediment. *Integr. Environ. Assess. Manage* 2010, 6 (4), 749–760. DOI: 10.1002/ieam.76
167. Huckins JN; Booij K; Petty JD *Monitors of Organic Chemicals in the Environment: Semipermeable Membrane Devices*; Springer US; Boston, MA, 2006; pp 29–43.
168. Peijnenburg WJGM; Teasdale PR; Reible D; Mondon J; Bennett WW; Campbell PGC Passive sampling methods for contaminated sediments: State of the science for metals. *Integr. Environ. Assess. Manage* 2014, 10 (2), 179–196. DOI: 10.1002/ieam.1502
169. Burton GA; Johnston EL Assessing contaminated sediments in the context of multiple stressors. *Environ. Toxicol. Chem* 2010, 29 (12), 2625–2643. DOI: 10.1002/etc.332 [PubMed: 20931605]
170. Mills GA; Vrana B; Allan I; Alvarez DA; Huckins JN; Greenwood R Trends in monitoring pharmaceuticals and personal-care products in the aquatic environment by use of passive sampling devices. *Anal. Bioanal. Chem* 2007, 387 (4), 1153–1157. DOI: 10.1007/s00216-006-0773-y [PubMed: 17077970]
171. Lissalde S; Charriau A; Poulier G; Mazzella N; Buzier R; Guibaud G Overview of the Chemcatcher® for the passive sampling of various pollutants in aquatic environments Part B: Field handling and environmental applications for the monitoring of pollutants and their biological effects. *Talanta* 2016, 148, 572–582. DOI: 10.1016/j.talanta.2015.06.076 [PubMed: 26653486]
172. Sultana T; Murray C; Ehsanul Hoque M; Metcalfe CD Monitoring contaminants of emerging concern from tertiary wastewater treatment plants using passive sampling modelled with performance reference compounds. *Environ. Monit. Assess* 2016, 189 (1), 1. DOI: 10.1007/s10661-016-5706-4 [PubMed: 27909849]
173. Booij K; Smedes F An Improved Method for Estimating in Situ Sampling Rates of Nonpolar Passive Samplers. *Environ. Sci. Technol* 2010, 44 (17), 6789–6794. DOI: 10.1021/es101321v [PubMed: 20701278]
174. Fernandez LA; Harvey CF; Gschwend PM Using Performance Reference Compounds in Polyethylene Passive Samplers to Deduce Sediment Porewater Concentrations for Numerous Target Chemicals. *Environ. Sci. Technol* 2009, 43 (23), 8888–8894. DOI: 10.1021/es901877a [PubMed: 19943662]
175. Apell JN; Gschwend PM Validating the Use of Performance Reference Compounds in Passive Samplers to Assess Porewater Concentrations in Sediment Beds. *Environ. Sci. Technol* 2014, 48 (17), 10301–10307. DOI: 10.1021/es502694g [PubMed: 25093866]
176. Joyce AS; Burgess RM Using performance reference compounds to compare mass transfer calibration methodologies in passive samplers deployed in the water column. *Environ. Toxicol. Chem* 2018, 37 (8), 2089–2097. DOI: 10.1002/etc.4167 [PubMed: 29744915]
177. Lao WJ; Maruya KA; Tsukada D An exponential model based new approach for correcting aqueous concentrations of hydrophobic organic chemicals measured by polyethylene passive samplers. *Sci. Total Environ* 2019, 646, 11–18. DOI: 10.1016/j.scitotenv.2018.07.192 [PubMed: 30041043]
178. Shen X; Reible D An analytical model for the fate and transport of performance reference compounds and target compounds around cylindrical passive samplers. *Chemosphere* 2019, 232, 489–495. DOI: 10.1016/j.chemosphere.2019.05.198 [PubMed: 31174007]
179. Apell JN; Shull DH; Hoyt AM; Gschwend PM Investigating the Effect of Bioirrigation on In Situ Porewater Concentrations and Fluxes of Polychlorinated Biphenyls Using Passive Samplers. *Environ. Sci. Technol* 2018, 52 (8), 4565–4573. DOI: 10.1021/acs.est.7b05809 [PubMed: 29578337]

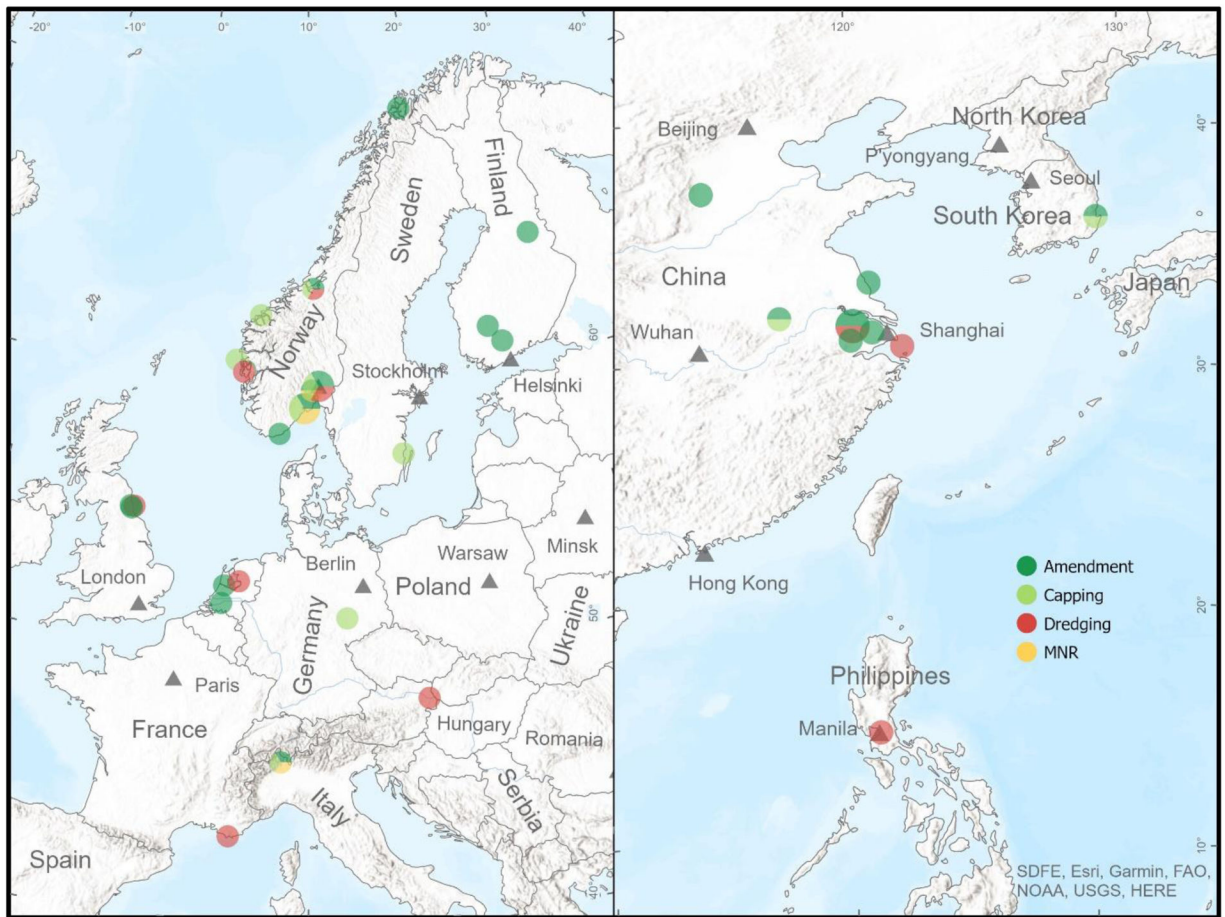
180. Apell JN; Gschwend PM In situ passive sampling of sediments in the Lower Duwamish Waterway Superfund site: Replicability, comparison with ex situ measurements, and use of data. *Environ. Pollut* 2016, 218, 95–101. DOI: 10.1016/j.envpol.2016.08.023 [PubMed: 27552042]
181. Schaanning MT; Beylich B; Gunnarsson JS; Eek E Long-term effects of thin layer capping in the Grenland fjords, Norway: Reduced uptake of dioxins in passive samplers and sediment-dwelling organisms. *Chemosphere* 2021, 264 (Part 2), 128544. DOI: 10.1016/j.chemosphere.2020.128544 [PubMed: 33065324]
182. Samuelsson GS; Raymond C; Agrenius S; Schaanning M; Cornelissen G; Gunnarsson JS Response of marine benthic fauna to thin-layer capping with activated carbon in a large-scale field experiment in the Grenland fjords, Norway. *Environ. Sci. Pollut. Res* 2017, 24 (16), 14218–14233. DOI: 10.1007/s11356-017-8851-6
183. Trannum HC; Raymond C; Næss R; Borgersen G; Gunnarsson JS; Schaanning MT Long-term response of marine benthic fauna to thin-layer capping with powdered activated carbon in the Grenland fjords, Norway. *Sci. Total Environ* 2021, 776, 145971. DOI: 10.1016/j.scitotenv.2021.145971 [PubMed: 33652322]
184. Nagy AS; Szabó J; Vass I Occurrence and distribution of polycyclic aromatic hydrocarbons in surface water and sediments of the Danube River and its tributaries, Hungary. *J. Environ. Sci. Health., Part A* 2014, 49 (10), 1134–1141. DOI: 10.1080/10934529.2014.897155



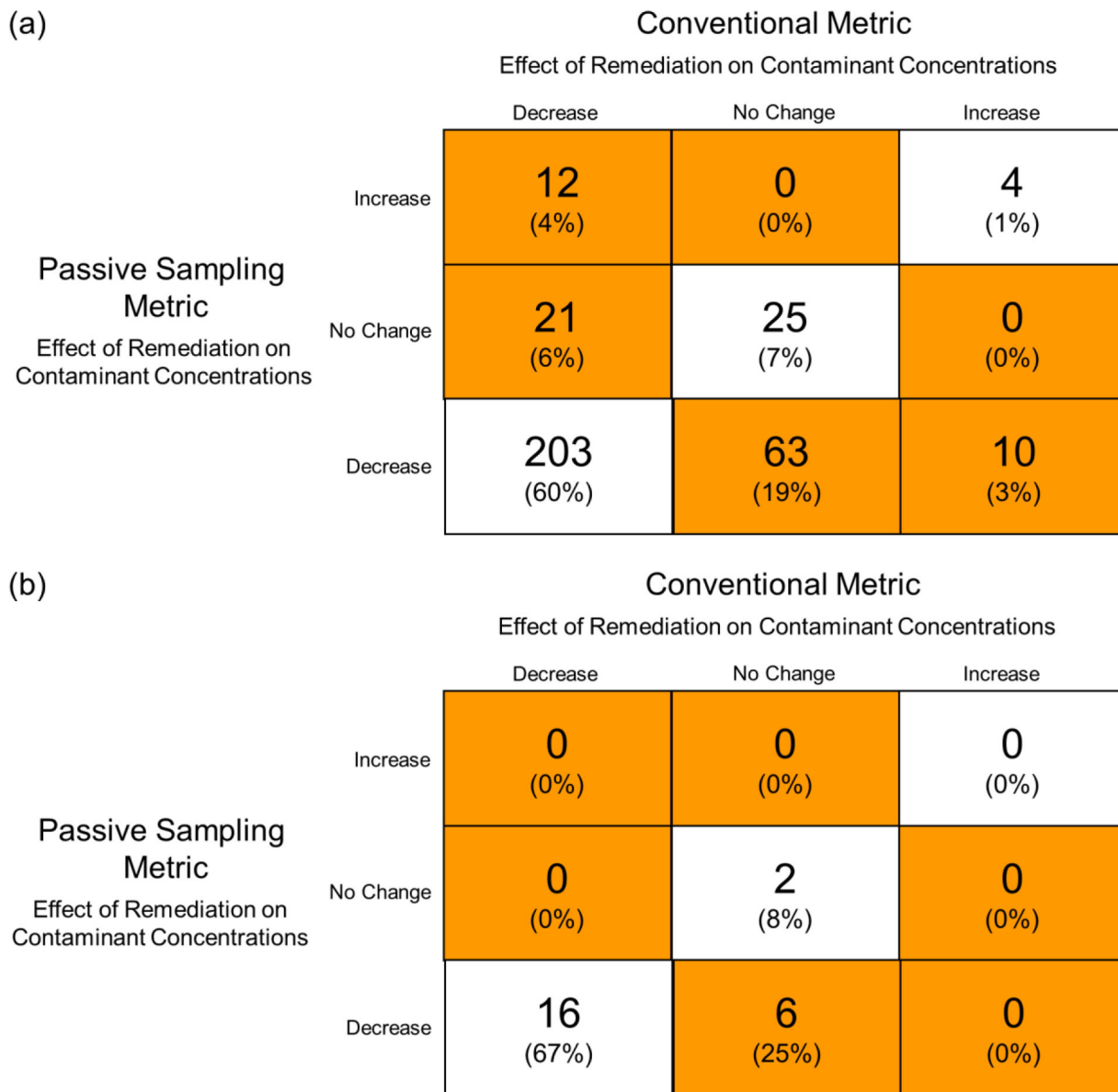
**Figure 1:** Locations of contaminated sediment sites using passive sampling devices (peer-reviewed and non-peer-reviewed) with summary count statistics (count is # of studies) from the peer-reviewed literature for types of contaminants, deployment location, and kinds of passive sampling devices. Details of search results for non-peer-reviewed studies are included as a file in the Supplementary Information.



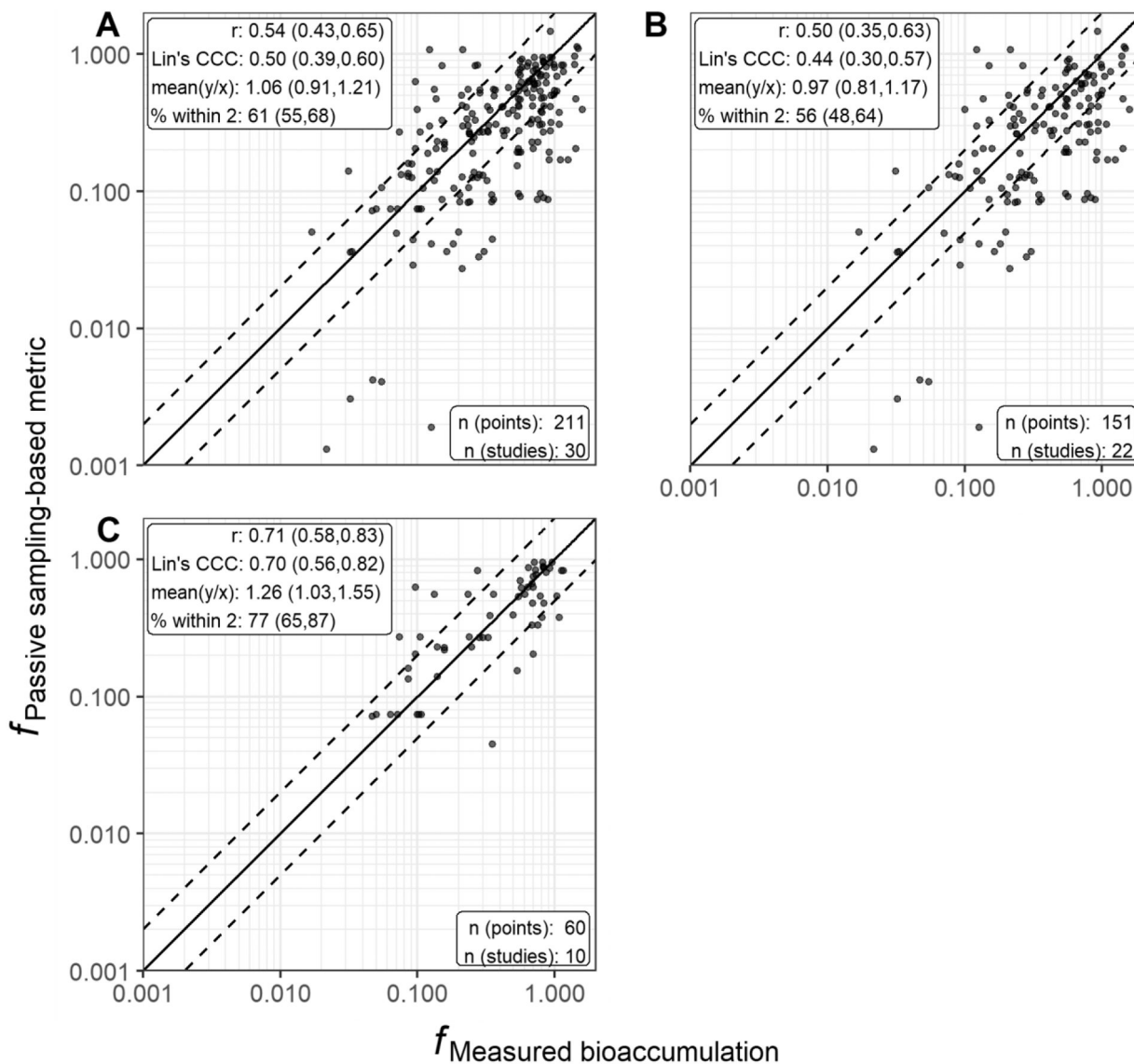
**Figure 2:**  
 Types of remediation or remedial approaches investigated at each site in North America. Larger circles indicate a site with investigations at multiple sub-sites. For example, Indiana Harbor and Shipping Canal and Grand Calumet River near Chicago, IL, are both part of the Grand Calumet River Area of Concern.



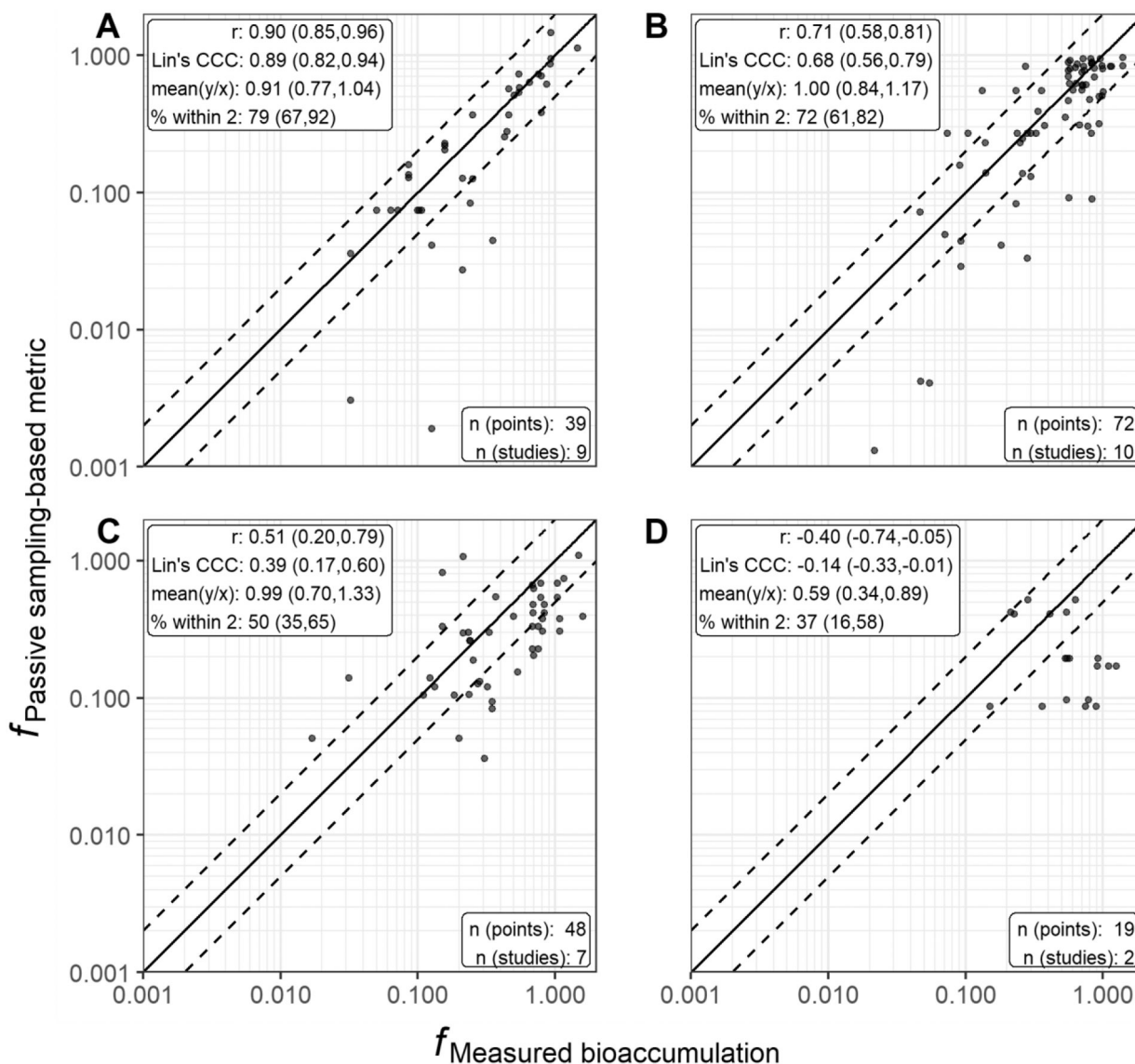
**Figure 3:**  
Types of remediation or remedial approaches investigated at each site in Europe and Asia.  
Larger circles indicate a site with investigations at multiple sub-sites.



**Figure 4:** Semi-quantitative concurrence analysis based on reported significance of remedial efficacy for (a) hydrophobic organic compounds (HOCs, n = 338 comparisons) and (b) toxic metals (n = 24 comparisons). Values are the number comparisons occurring in each concurrence category. All passive sampling (e.g.,  $C_{free}$ , uptake and flux) and conventional (e.g., bulk sediment, water grab, bioaccumulation, acute toxicity) endpoints are included in the comparison. White boxes reflect the number of studies where the passive sampling and conventional metrics agreed. Only total contaminant concentrations (e.g., total PCBs, not congeners, or homologues) were considered.

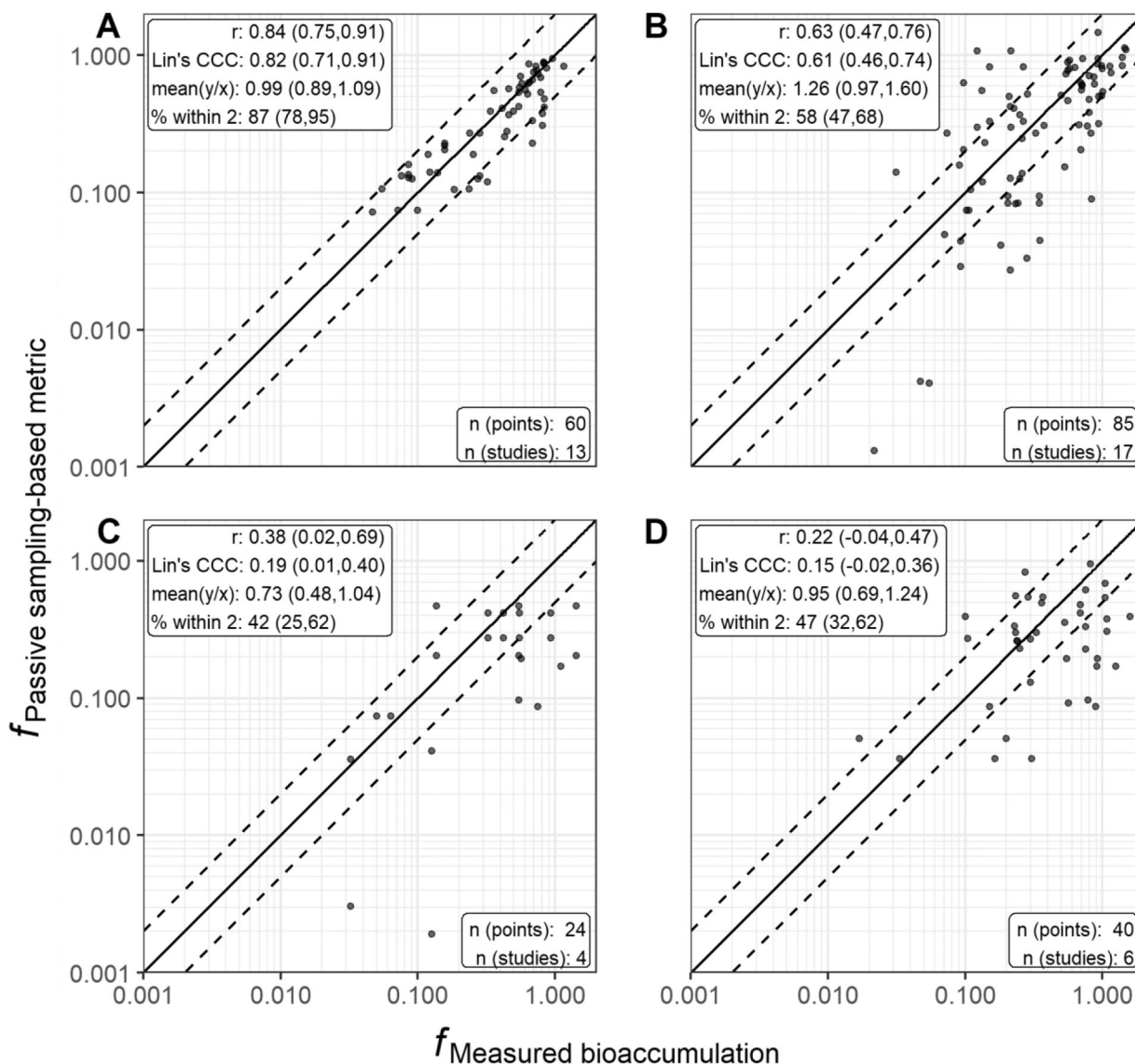


**Figure 5:** Comparison of remedial effectiveness ( $f$  is the ratio of post-remedy value to the un-remediated value) and related summary statistical analyses of the relationships between passive sampling-based measurements of bioaccumulation and measured bioaccumulation contaminants in the (a) full dataset, (b)  $C_{\text{free}}$  from passive samplers, and (c) biomimetic passive samplers. Summary statistics include means and the corresponding lower and upper 95% confidence intervals based on bootstrapping estimates. Solid diagonal line is the 1:1 (perfect agreement) and the dashed lines signify a factor of two off the 1:1 line.

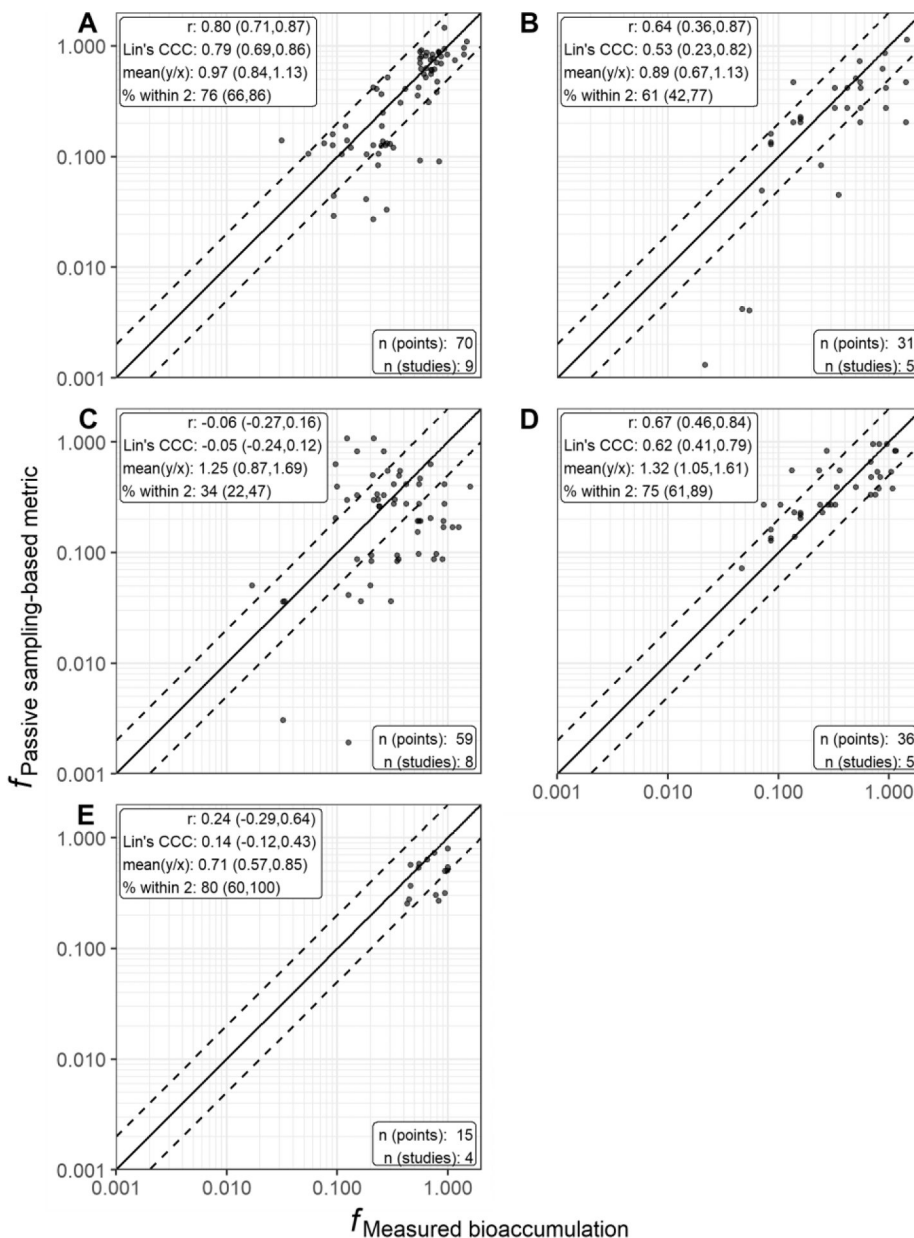


**Figure 6:** Comparison of remedial effectiveness ( $f$  is the ratio of post-remedy value to the un-remediated value) and related summary statistical analyses of the relationships between passive sampling-based measurements of bioaccumulation and measured bioaccumulation contaminants where (a) the PSDs and biota were in the same vessel during laboratory bioaccumulation tests, (b) the PSDs and biota were deployed in separate vessels in the laboratory, (c) PSDs and biota were deployed together or nearby in the field, and (d) biota were collected in the field and PSDs were used ex-situ. Summary statistics include means and the corresponding lower and upper 95% confidence intervals based on bootstrapping estimates. Solid diagonal line is the 1:1 (perfect agreement) and the dashed lines signify a factor of two off the 1:1 line.





**Figure 7:** Comparison of remedial effectiveness ( $f$  is the ratio of post-remedy value to the un-remediated value) and related summary statistical analyses of the relationships between passive sampling-based measurements of bioaccumulation and measured bioaccumulation performed with: (a) bivalves, (b) oligochaetes and polychaetes, (c) fish, and (d) other organisms (e.g., amphipods). Summary statistics include means and the corresponding lower and upper 95% confidence intervals based on bootstrapping estimates. Solid diagonal line is the 1:1 (perfect agreement) and the dashed lines signify a factor of two off the 1:1 line.



**Figure 8:** Comparison of remedial effectiveness ( $f$  is the ratio of post-remedy value to the un-remediated value) and related summary statistical analyses of the relationships between passive sampling-based measurements of bioaccumulation and measured bioaccumulation performed with: (a) PDMS, (b) LDPE, (c) POM, (d) SPMD, and (e) DGT. Summary statistics include means and the corresponding lower and upper 95% confidence intervals based on bootstrapping estimates. Solid diagonal line is the 1:1 (perfect agreement) and the dashed lines signify a factor of two off the 1:1 line.

**Table 1:**

Summary of variables found in the literature search applied in this review to determine relationship between passive sampling and conventional metrics to evaluate remediation efficacy. Variables not included are also listed.

Relevance to Review	Contaminants	Conventional Metrics	Passive Sampling Metrics	
			PSD	Endpoints
Included	Toxic metals (TM), Antimony, Arsenic, Chromium, Polycyclic aromatic hydrocarbons (PAHs), Polychlorinated biphenyls (PCBs), Dichloro-diphenyltrichloroethane and primary degradation products (DDXs), Other organochlorine pesticides (OCPs), Pyrethroids, Polychlorinated dibenzo-dioxins/furans (PCDD/F), Polybrominated diphenyl ethers (PBDEs)	Bulk sediments, Water column, Porewater, Toxicity, Bioaccumulation, Benthic community condition, High volume air sampling, Sediment-water flux	DGT, Peepers, LDPE, PDMS fiber, PDMS-lined jars, POM, Silicone rubber, SPMD, Polyacrylate fiber	Biomimetic, $C_{free}$ , Flux, Site-specific partition coefficients ( $SS-K_p$ ), Air concentrations
Not included	Hexachlorobenzene (HCB), Octachlorostyrene (OCS)	Gut fluid extraction	Tenax, XAD, Infinite-sink applications of polymeric PSDs	Desorption kinetics

**Table 2:**

Summary of laboratory-based feasibility studies using passive sampling devices involving amendments, capping and multiple remedies investigated in this review.

Contaminants	Study Type & Details	PSDs	Metric of Remedial Effectiveness		References
			Passive Sampling	Conventional	
<i>Laboratory Amendment Studies</i>					
	Amendment Materials				
As	LMB, CaNO <sub>3</sub>	DGT	C <sub>free</sub>	Sediment, Water column, Porewater concentrations	47
DDX	AC	LDPE, SPMD	C <sub>free</sub> , Flux, Biomimetic	Sediment, Water column, Porewater concentrations	38
DDX	AC	PDMS Fiber	C <sub>free</sub>	–	62
DDX	AC, Sand	PDMS Fiber	C <sub>free</sub>	Bioaccumulation	42
DDX	AC	SPMD	Biomimetic	Porewater concentrations	57
Hg	AC	DGT	C <sub>free</sub>	Bioaccumulation, Bacterial community analysis, Sediment concentrations	45
TM	Biochar	DGT	C <sub>free</sub>	Bioaccumulation, Toxicity, Sediment concentrations	46
TM	CaNO <sub>3</sub>	DGT	C <sub>free</sub>	–	129
TM	LMB, Polyaluminum chloride	DGT	C <sub>free</sub>	Sediment concentrations	48
TM	Zeolite	DGT	C <sub>free</sub> , Biomimetic	Bioaccumulation, Sediment concentrations	44
TM, PAH	Magnetic AC	DGT, PDMS Disk	C <sub>free</sub>	Bioaccumulation	36
OCP PAH Pyrethroids PBDE	Natural zeolite	PDMS Disk	C <sub>free</sub>	Bioaccumulation	40
PAH	AC	LDPE	Biomimetic, Partition Coefficients	–	59
PAH	AC	LDPE	Biomimetic	–	60
PAH	AC	PDMS Fiber	C <sub>free</sub>	–	67, 68
PAH	AC, Sand	PDMS Fiber	Biomimetic	–	66
PAH	AC	PE	Biomimetic, Partition Coefficients	–	58
PAH	AC, Biochar, Magnetic AC, Magnetic biochar	PE	Biomimetic, Partition Coefficients	Toxicity, Sediment concentrations	50
PAH	AC, Biostimulant	PE	Biomimetic	Microbial community analysis, Sediment concentrations	49
PAH	Magnetic AC, Magnetic biochar	PE	Biomimetic	Toxicity	130
PAH	Magnetic AC, AC cloth, AC	PE	Biomimetic	–	64

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PAH	AC	POM	$C_{free}$ , Partition Coefficients	–	61	
PAH	AC	POM	$C_{free}$	Toxicity	131	
PAH	AC	POM	$C_{free}$	Bioaccumulation	128, 132	
PAH, PCB	AC, Anthracite	PDMS Fiber, PDMS Sheet	$C_{free}$ , Biomimetic	–	66	
PAH, PCB	AC from lignin, coal, and coconut, AC	POM	$C_{free}$ , Partition Coefficients	–	63, 65	
PCB	AC	LDPE	Biomimetic	–	60	
PCB	AC	PDMS-coated vial	$C_{free}$	Bioaccumulation, Toxicity	41	
PCB	AC	PDMS-coated vial	$C_{free}$	Bioaccumulation, Sediment concentrations, Toxicity	133	
PCB	Single-walled Carbon Nanotubes	PE	$C_{free}$	Bioaccumulation, Sediment concentration, Toxicity	43	
PCB	AC	POM	$C_{free}$	Bioaccumulation	134	
PCB	AC	POM	$C_{free}$ , Flux	Bioaccumulation	37	
PCB	Bioamended AC (SediMite), AC (SediMite)	POM	$C_{free}$	Bacterial community analysis, Sediment concentrations	51	
PCB	AC	SPMD	Biomimetic	Bioaccumulation Porewater concentrations	56	
PCB	AC	SPMD	Biomimetic	Porewater concentrations	55	
PCB, PAH	AC, Biochar, Organoclay PM-199, Coke	PE	$C_{free}$	Bioaccumulation	39	
PCB, PAH	AC	SPMD	Biomimetic	Bioaccumulation Porewater concentrations	54	
PCB, PAH	Coke, AC	SPMD	Biomimetic	Toxicity, Bioaccumulation, Porewater concentrations	52, 53	
<b>Laboratory Capping Studies</b>						
Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
				Passive sampling	Conventional	
	Capping Material			Passive sampling	Conventional	
As	Aluminum hydroxide		DGT	$C_{free}$ , Flux	Sediment, Water column, Porewater concentrations	71
As	Phoslock (La-modified bentonite)		DGT	$C_{free}$ , Flux	Sediment, Water column, Porewater concentrations	72
PAH	Sand/Clean Sediment		PDMS Fiber	$C_{free}$	Bioaccumulation, Sediment concentrations	74
PAH	Limestone, Gneiss		POM	$C_{free}$	–	125
PCB, PAH, TM	AC/Clay, Clay		PDMS fiber, DGT	$C_{free}$	Benthic community analysis, Sediment concentrations	75

<b>Laboratory Multiple Remedy Studies</b>						
Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
	Remedy	Remedy Materials		Passive Sampling	Conventional	
DDX	Amendment, Capping, MNR/ Capping	AC, AC/Clean sediment, AC/ Contaminated	PE	Biomimetic	Bioaccumulation, Sediment concentrations	76, 77

Laboratory Multiple Remedy Studies						
Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
	Remedy	Remedy Materials		Passive Sampling	Conventional	
		sediment, Clean sediment				
PCB	Amendment, Capping	AC, Sand	PDMS-coated vial, LDPE	C <sub>free</sub> , Biomimetic	Bioaccumulation, Sediment concentration	78-80

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**Table 3:**

Summary of pilot- and field-based feasibility studies using passive sampling devices involving amendments, capping and multiple remedies investigated in this review. Also see Table S3.

Contaminants	Study Type & Details	PSDs	Metric of Remedial Effectiveness		References	
			Passive Sampling	Conventional		
<b>Field Amendment Studies</b>						
	Amendment					
OCP	AC	PE	Biomimetic	–	135	
PCB	AquaGate+PAC	PDMS fiber	C <sub>free</sub>	Bioaccumulation, Benthic community analysis, Sediment concentrations	136	
PCB	AquaGate+PAC, SediMite	PDMS fiber	C <sub>free</sub>	Bioaccumulation	137	
PCB	AC	PDMS fiber, PE, SPMD, POM	C <sub>free</sub> , Biomimetic	Bioaccumulation Sediment, Water column, Porewater concentrations	82, 117, 136, 138	
PCB	Bioamended AC (SediMite), AC (SediMite)	PE	C <sub>free</sub>	Bacterial community analysis, Sediment concentrations	86	
PCB	AC	PE, POM, SPMD	C <sub>free</sub> , Biomimetic	Bioaccumulation, Benthic community analysis, Sediment, Water column, Porewater concentrations	82, 139, 140	
PCB	AC	POM	C <sub>free</sub> , Flux, Biomimetic	Bioaccumulation, Sediment, Water column, Porewater concentrations	83	
PCB, PAH	AC	POM, Polyacrylate fiber	C <sub>free</sub> , Partition Coefficients	Bioaccumulation, Toxicity, Sediment concentrations	141–143	
<b>Field Capping Studies</b>						
Contaminants	Study Type & Details	PSDs	Metric of Remedial Effectiveness		References	
	Capping Material		Passive Sampling	Conventional		
TM	Apatite/Sand	DGT	C <sub>free</sub>	Toxicity; Water column, Porewater concentrations	144	
TM	Zeolite, AC/zeolite, AC/sand, Zeolite/sand	DGT	C <sub>free</sub>	–	145	
TM	Sand/Phillipsite Zeolite	Dialysis sampler	C <sub>free</sub>	–	146, 147	
TM	Sand	Peeper	C <sub>free</sub> , Flux	Sediment concentrations	24, 25	
PAH	Aquablok and Sand, Apatite and Sand, Coke mat and Sand, Sand	PDMS Fiber	C <sub>free</sub>	Bioaccumulation, Sediment concentrations	87	
PAH	Dredged sediment	PDMS Fiber	C <sub>free</sub>	Bioaccumulation; Water column, Sediment concentrations; Toxicity	148	
<b>Field Multiple Remedy Studies</b>						
Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
	Remedies	Remedy Materials		Passive Sampling	Conventional	
PAH, PCB	Amendment, Capping	AC, Clay/AC, Sand/AC, Sand	POM	C <sub>free</sub>	Bioaccumulation, Sediment concentrations, Benthic community analysis	94, 149

Field Multiple Remedy Studies						
Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
	Remedies	Remedy Materials		Passive Sampling	Conventional	
PCB	Amendment, Capping	Sediment, AC, Sand/AC, Sand	PE, POM	C <sub>free</sub>	Bioaccumulation, Sediment concentrations	92, 93
PCB, DDX	Capping, MNR	Sand	PDMS Fiber, POM, PE	C <sub>free</sub> , Flux	Porewater concentrations	89, 90
PCDD/F	Amendment, Capping	AC/Dredged Clay, Dredged Clay, Limestone, AC	POM	C <sub>free</sub>	Sediment concentrations	88

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Summary of dredging studies using passive sampling devices at contaminated sediment sites investigated in this review.

Table 4:

Application of PSD	Sampled Media	Contaminants	PSDs	Metric of Remedial Effectiveness		References
				Passive Sampling	Conventional	
During Dredging	Water Column	PAH	LDPE	Biomimetic	-	97
		PAH	SPMD	C <sub>free</sub>	-	98
		PAH/PCB	SPMD	C <sub>free</sub>	Turbidity	96, 101
		PAH/PCB	POM	C <sub>free</sub>	Turbidity	100
		PCB	LDPE	C <sub>free</sub> , Flux	Water column concentrations (dissolved)	99, 102
		Toxic metals	DGT	C <sub>free</sub>	Turbidity, Water column concentrations (total and dissolved)	96, 101, 107-109
Transport or Disposal	Air	PCB	PUF	C <sub>free</sub> , Flux	-	99
		PAH/PCB	SPMD	C <sub>free</sub>	Turbidity	100, 101
		Toxic metals	DGT	C <sub>free</sub>	Turbidity, Water column concentrations (total and dissolved)	101, 107
		PAH	PDMS	C <sub>free</sub> , Flux	Water column concentrations (total), Bioaccumulation, Sediment concentrations	111 *
		Toxic metals	DGT	C <sub>free</sub>	Water column concentrations (total and dissolved)	107
		Toxic metals	DGT/Peeper	C <sub>free</sub> , DGT Flux	Sediment concentrations	95, 112-114
Disposal Site	Water Column	PAH	PDMS	C <sub>free</sub> , Flux	Water column concentrations (total), Bioaccumulation, Sediment concentrations	111 *
		Toxic metals	DGT/Peeper	C <sub>free</sub> , DGT Flux	Sediment concentrations	95, 112-114
		PAH/PCB	POM	C <sub>free</sub>	Flux, sediment traps	119
		Toxic metals	DGT	C <sub>free</sub>	Water column concentrations (total and dissolved)	107
		PAH/PCB	SPMD	C <sub>free</sub>	Benthic community analysis, Sediment concentrations	119
		Toxic metals	DGT	DGT Flux	Sediment concentrations, Flux	116
General Site Monitoring	Water Column	Toxic metals	DGT	DGT flux	Sediment concentrations	116
		PAH/PCB	POM	C <sub>free</sub>	High-volume air sampler	122, 123
		PAH	LDPE	Biomimetic	Flux	97, 124
		PAH	SPMD	C <sub>free</sub>	-	98
		PCB	LDPE	C <sub>free</sub> , Flux	-	99

Application of PSD	Sampled Media	Contaminants	PSDs	Metric of Remedial Effectiveness		References
				Passive Sampling	Conventional	
	Air	PCB	PUF	$C_{\text{pass}}$ , Flux	–	99

\* Indicates *ex-situ* sampling.

**Table 5:**

Summary of post-remediation studies using passive sampling devices at contaminated sediment sites investigated in this review.

Contaminants	Study Type & Details		PSDs	Metric of Remedial Effectiveness		References
				Passive Sampling	Conventional	
	Remedy	Remedy Materials				
PCB	Amendment	AC	POM	C <sub>free</sub> , Partition Coefficients	Sediment concentration	150
PAH	Capping	Organoclay/Sand/Armoring, Sand/Armoring	LDPE	C <sub>free</sub> , Biomimetic	Toxicity	151
PAH	Capping	Organoclay/Sand/Armoring, Sand/Armoring	LDPE	C <sub>free</sub> , Flux	–	118
PAH	Capping	Sand/gravel	PDMS Fiber	C <sub>free</sub>	–	152
PAH	Capping	Sand/sediment, Aquablok	PDMS Fiber	C <sub>free</sub>	–	117
PAH	Capping	Organoclay/Sand/Armoring, Sand/Armoring	SPMD	C <sub>free</sub>	–	98
PAH, PCB	Capping	Dredged sediment	POM	C <sub>free</sub>	Sediment concentrations	125
PAH	Capping, Dredging	Sand/Organoclay	LDPE	C <sub>free</sub> , Flux	–	124

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