



Comparing different methods for assessing contaminant bioavailability during sediment remediation



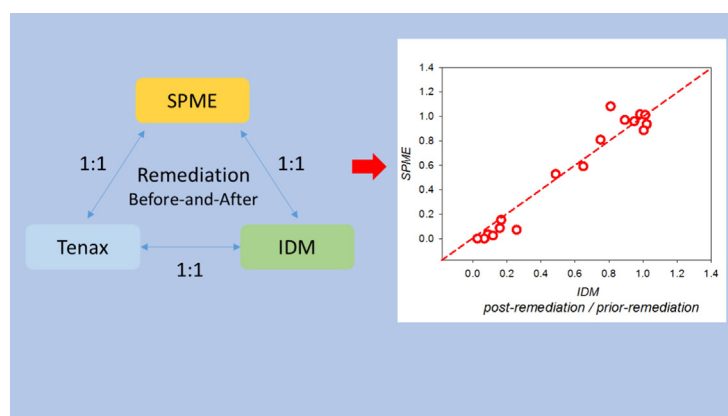
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HIGHLIGHTS

- SPME, Tenax desorption, and IDM methods predicted essentially the same degrees of changes in bioavailability in sediment remediation practices.
- Tenax desorption and IDM provide better sensitivity than SPME, while SPME is more appropriate for in situ application.

GRAPHICAL ABSTRACT



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ABSTRACT

Sediment contamination by persistent organic pollutants from historical episodes is widespread and remediation is often needed to clean up severely contaminated sites. Measuring contaminant bioavailability in a before-and-after manner lends to improved assessment of remediation effectiveness. However, a number of bioavailability measurement methods have been developed, posing a challenge in method selection for practitioners. In this study, three different bioavailability measurement methods, i.e., solid phase microextraction (SPME), Tenax desorption, and isotope dilution method (IDM), were compared in evaluating changes in bioavailability of DDT and its degradates in sediment following simulated remediation treatments. When compared to the unamended sediments, all three methods predicted essentially the same degrees of changes in bioavailability after amendment with activated carbon, charcoal or sand. After normalizing over the unamended control, measurements by different methods were linearly correlated with each other, with slopes close to 1. The same observation was further made with a Superfund site marine sediment. This finding suggests that different methods may be used in evaluating remediation efficiency. However, Tenax desorption or IDM consistently offered better sensitivity than SPME in detecting bioavailability changes. Results from this study highlight the value of considering bioavailability when evaluating remediation effectiveness and provide guidance on the selection of bioavailability measurement methods in such assessments.

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1. Introduction

Owing to their high affinity for solid particles, persistent organic pollutants (POPs) preferentially deposit onto the bed sediment in surface aquatic systems, where they are sorbed to the sediment phase. The sorption generally results in decreased bioaccumulation or toxicity to aquatic organisms (Luthy et al., 1997). Such contaminant sorption or sequestration may be intentionally enhanced by the addition of strong sorbents such as black carbon materials to achieve remediation purposes (Cornelissen et al., 2005; Ghosh et al., 2011; Koelmans et al., 2006; Millward et al., 2005). It is well known that due to sorption, the use of bulk chemical concentration may lead to substantial overexpression of the bioavailability or risk of sediment-borne POPs (Ehlers and Luthy, 2003), as a chemical must be in the freely dissolved form (C_{free}) to cross the cell membrane to be accumulated or exert toxicity. When the C_{free} is depleted, it is replenished by the readily desorbable pool, i.e., the bioaccessible fraction. Therefore, using bioavailability as an end-point is highly desirable for evaluating the effectiveness of remediation operations (Ehlers and Luthy, 2003; Semple et al., 2004).

A large number of bioavailability measurement methods have been introduced over the last two decades. These methods generally fall into two types, i.e., passive samplers that aim to determine C_{free} , and partial extraction methods that measure the readily desorbable fraction as an indicator of bioaccessibility (Reichenberg and Mayer, 2006). A number of passive samplers, such as polyethylene devices (PED), semi-permeable membrane devices (SPMD) and solid phase microextraction (SPME), have been used for measuring C_{free} of POPs in sediment (Fernandez et al., 2009; Ghosh et al., 2014; Greenberg et al., 2014; Hawthorne et al., 2009; Huckins et al., 1993; Lydy et al., 2014; Mayer et al., 2014). Various extraction or desorption-based methods have also been tested for predicting the bioaccessible fraction, which include mild solvent extraction (Kelsey et al., 1997; Khan et al., 2012; Tang and Alexander, 1999), Tenax-aided sequential desorption (Cornelissen et al., 1998, 2001), cyclodextrin extraction (Reid et al., 2000; Stokes et al., 2005) and the newly introduced isotope dilution method (IDM) (Delgado-Moreno and Gan, 2013; Jia et al., 2014). However, while these methods have been used with success in individual studies, few studies have compared their advantages or limitations for use in the evaluation of remediation performance (You et al., 2011). The lack of method comparison may have led to inconsistent results and difficulty to analyze data across different studies, while posing a significant challenge for practitioners who want to select the most suitable method.

In this study, three different methods were applied to measuring bioavailability of DDT derivatives in two spiked and one historically contaminated sediments following simulated remediation treatments by amending the sediment with activated carbon, charcoal or sand. Addition of black carbon materials has been often shown to effectively sequester or immobilize POPs in many bench-scale and pilot studies (Cho et al., 2009; Tomaszewski et al., 2007; Werner et al., 2010), while capping with gravel or sand has been used to physically separate the contaminated sediment from its overlaying water to minimize exposure (Hyun et al., 2006; Lampert et al., 2011; McDonough et al., 2007). Three methods, SPME, Tenax-aided desorption and IDM, were tested on the same unamended and amended sediments samples, or in a before-and-after fashion. Method performance and limitations were statistically compared and discussed. Information from this study may be used for improving data interpretation and guiding method selection in efforts to better assess progress or efficiency of remediation treatments.

2. Materials and methods

2.1. Chemicals, SPME fiber and Tenax

Standards of 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane (*p,p'*-DDT), 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene (*p,p'*-DDE), 1,1-dichloro-2,4-bis-(chlorophenyl)ethylene (*o,p'*-DDE), and internal

standard 2,2',3,3',4-pentachlorobiphenyl (PCB 82) were purchased from AccuStandard (New Haven, CT). Stable isotope labeled analogues *p,p'*-DDT- d_4 and *p,p'*-DDE- d_4 and surrogate *o,p'*-DDT- d_8 were purchased from C/D/N Isotopes (Pointe-Claire, Quebec, Canada), and ^{13}C -*o,p'*-DDE was purchased from Cambridge Isotope Laboratories (Tewksbury, MA). Thin fiber (430- μm diameter) coated with 35 μm polydimethylsiloxane (PDMS) was purchased from Polymicro Technologies (Phoenix, AZ) and used in the SPME analysis of C_{free} . The volume of PDMS polymer per fiber length was 51.1 $\mu\text{L}/\text{m}$. The PDMS fiber was pre-cleaned with Soxhlet extraction using ethyl acetate for 72 h and cut to 1-cm pieces with a razor blade before use (Jia et al., 2012). Tenax TA resin (60–80 mesh) was purchased from Scientific Instrument Services (Ringoes, NJ) and used in the Tenax-aided desorption measurement. All other solvents and chemicals used were of analytical or gas chromatography (GC) grade.

2.2. Amendment materials

Three types of amendment materials were used, including sand and two types of black carbon, i.e., activated carbon and charcoal. The activated carbon in fine granules was purchased from Calgon Carbon (Pittsburg, PA). The charcoal was derived from the combustion of macrocarpa in New Zealand at 400 °C (Jia and Gan, 2014). The sand was collected from San Diego Creek in Orange County, California, air-dried and stored at room temperature. All amendment materials were further ground in a mortar, and passed through a no. 100 mesh (≤ 0.15 mm) before use. The organic carbon (OC) contents, specific surface areas (SSA) and microporosity of the black carbon materials were determined using standard methods (Gustafsson et al., 1997). The SSA and micropore volume were 706.2 m^2/g and 162.2 cm^3/g for the activated carbon, and were 116.6 m^2/g and 42.4 cm^3/g for the charcoal as determined in the previous study (Jia and Gan, 2014).

2.3. Experiments with spiked sediments

The evaluation of method performance was first carried out using freshwater sediments spiked with *p,p'*-DDT and *p,p'*-DDE. Two river sediments with no detectable DDT residues were collected using a shovel from the surface layer (0–15 cm), including San Diego Creek sediment (SD, Orange County, CA) and Jordan Lake Reservoir sediment (JL, Chatham County, NC). The sediment samples were shipped to the laboratory in large plastic containers, and kept in a cold room (≤ 4 °C) before use. Both sediments were wet-sieved through a 2-mm mesh and stored at ≤ 4 °C before use. The sediment OC content was measured by combustion on a nitrogen-carbon analyzer (Thermo Finnigan, Woods Hole, MA) after removing the inorganic carbon with 1 M HCl. The black carbon (BC) content was determined by combusting an aliquot in a muffle furnace at 375 °C for 24 h, digesting with 1 M HCl, and then analyzing on the elemental analyzer (Gustafsson et al., 1997). The measured OC contents were 0.87% and 0.12% for the SD and JL sediments, respectively. The measured BC content was 0.11% for the SD sediment, and below the detection limit (LOD = 0.01%) for the JL sediment.

The sediments were spiked with *p,p'*-DDT and *p,p'*-DDE following the U.S. EPA guidelines (U.S. Environmental Protection Agency, 2001). Briefly, the stock solution with *p,p'*-DDT and *p,p'*-DDE in acetone was first applied to 5 g silica sand (≤ 0.15 mm) in a 125-mL wide-mouth glass jar. After the carrier solvent was evaporated in the fume hood, 50 g (dry weight equivalent) of sediment was added and an amendment material (sand, activated carbon, or charcoal) was incorporated into the sediment at 0 (unamended control), 1 and 5% (dry weight basis). The spiked sediments were mixed at 120 rpm on a shaker for 7 days and then aged for over 1 month before use. Aliquots of the incubated sediments were removed and analyzed for the bulk sediment concentration following solvent extraction. Details on the analysis of bulk sediment concentration (C_T) may be found elsewhere (Jia et al., 2014). Preliminary experiments showed that the recoveries of *p,p'*-

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