



Treatment characteristics of various sediment components spiked with 2-chlorobiphenyl using reactive activated carbon

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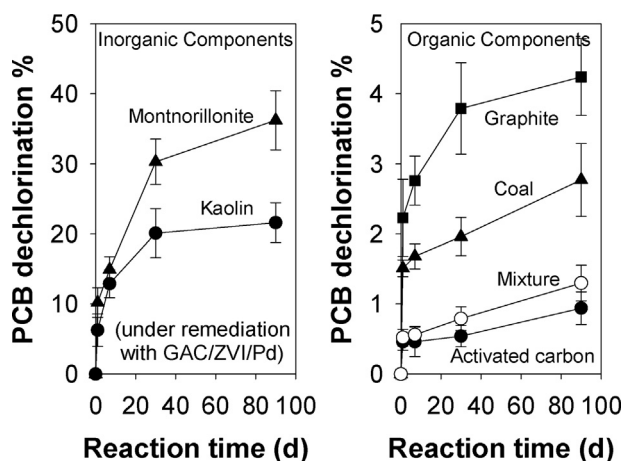
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HIGHLIGHTS

- Various solid organic and inorganic sediment components were tested.
- Fate and transport of 2-CIBP spiked to the sediment components was fully traced.
- Role of the various components in adsorbing and desorbing 2-CIBP was studied.
- Performance of the most advanced remediation material, RAC was evaluated.
- 2-CIBP spiking was correlated with 2-CIBP desorption and 2-CIBP dechlorination.

GRAPHICAL ABSTRACT



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ABSTRACT

Previously, the concept of reactive activated carbon (RAC), where the porous structure of activated carbon (AC) is impregnated with palladized zerovalent iron, has been proposed to be effective to adsorb and dechlorinate polychlorinated biphenyls (PCBs). To explain the low dechlorination of PCBs bound to actual aquatic sediments under remediation with RAC, this study investigated the role of various solid organic and inorganic sediment components in adsorbing and desorbing PCBs. Detailed fate and transport mechanism of 2-chlorinated biphenyl (2-CIBP) spiked to sediment components, including kaolin, montmorillonite (MMT), coal, graphite, AC, and their mixture, was revealed. Adsorption and holding capability of sediment components toward 2-CIBP strongly influenced amount of spiked 2-CIBP, amount of desorbed 2-CIBP, overall dechlorination of 2-CIBP to biphenyl (BP), and eventual partitioning of 2-CIBP and BP to water, sediment component, and RAC. Order of the amount of spiked 2-CIBP to sediment components after drying, following AC > mixture > coal > graphite > kaolin > MMT, was in agreements (in opposite direction) with order of the amount of desorbed 2-CIBP and order of overall 2-CIBP dechlorination. Substantial role of organic components in aquatic sediments for holding 2-CIBP and thus preventing it from dechlorination on RAC was proven.

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1. Introduction

Contaminated aqueous sediments are of great concern in the United States [1,2]. Particular interest is given to polychlorinated

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biphenyls (PCBs) due to their chemical persistence, environmental toxicity, and hydrophobic property [3–5]. Capping contaminated sites with clean sand layer has been proposed to isolate sites from direct contact [6]. Sand layer can be further amended with activated carbon (AC) or AC layer can also be added on the top of sand layer, which significantly reduces PCBs level in water [7–10]. Since AC-based approaches do not decompose PCBs, electrochemical dechlorination of PCBs to relatively biodegradable biphenyl (BP) has been also studied using reactive metal particles such as zerovalent iron (ZVI) [6,11,12]. The studies mostly tested aqueous phase PCBs to quickly investigate kinetics and mechanisms of the dechlorination reaction. Treatment of real aquatic sediments contaminated with PCBs using such reactive metals has been rarely reported most probably because PCBs in sediments would be much less available for the reaction at metal surface. PCBs are strongly bound to sediment solids, particularly organic components, and thus they are hard to desorb into water [13].

In order to facilitate desorption of PCBs in sediments to water and then reaction of the desorbed PCBs onto metal surface, the concept of reactive activated carbon (RAC) has been proposed, where the porous structure of granular activated carbon (GAC) is impregnated with palladized ZVI nanoparticles [14]. RAC was able to promptly adsorb PCBs available in water, effectively dechlorinate them to eventually BP, and completely sequester PCBs and reaction intermediates [15–17]. The RAC strategy or similar approaches seemed to work synergistically for removal of many organic and inorganic chemicals and metals, while taking advantages of the two remediation approaches, physical adsorption and chemical dechlorination [18–22]. As one of the most highly contaminated sites with PCBs, Waukegan Harbor sediment (WHS), IL has attracted significant attention [23,24]. Eventually, treatment of WHS by using the RAC strategy was recently reported [25]. The study experimentally revealed desorption of PCBs from WHS, their adsorption and subsequent dechlorination onto RAC, and ensuing partitioning of PCBs and intermediates into water, WHS, and RAC. Although the presence of RAC facilitated PCBs desorption from WHS, the slow desorption of PCBs limited overall performance, resulting in five-order of magnitude lower dechlorination when compared with treatment of aqueous PCBs [16,25].

In order to find scientific reasons for the slow desorption process and thus low dechlorination yield for WHS and presumably for many other aquatic sediments potentially under remediation with such reactive media, this current study investigated the role of various solid sediment components (herein, sediment or component) in adsorbing and desorbing PCBs in the presence of RAC. Considering sediments are a heterogeneous mixture of various solid organic and inorganic components, kaolin, montmorillonite (MMT), coal, graphite, GAC, and even their mixture were selected as sediment components, spiked with 2-chlorinated biphenyl (2-CIBP), and treated using RAC. A systematic lab-scale test was designed in manner of direct mixing and compartment configuration to reveal the fate and transport of 2-CIBP in the treatment environment [25]. Eventually, the capability of sediment components to adsorb and hold 2-CIBP during spiking process was correlated with the desorption trend of 2-CIBP to water and RAC and with its dechlorination onto RAC.

2. Experimental

2.1. RAC synthesis

Detailed synthesis procedure of RAC was reported previously [14]. Briefly, 2–3 mm size mesoporous GAC (HD3000, Norit Americas Inc.) was used as base material for RAC. The surface area of GAC at 574 m²/g was significantly decreased to 358 m²/g after in-situ

placement of ZVI/Pd particles via incipient wetness impregnation. The porous structure of GAC was well occupied with Fe at around 14.4% and Pd at around 0.7%. Synthesis and handling of RAC was performed in anaerobic glove box (oxygen <0.05 mg/L and hydrogen at 3.5%).

2.2. Sediment components and PCB spiking

Initially four different solid components were selected; two inorganic components (kaolin and MMT) and two organic components (coal and graphite). GAC used as base material for RAC was also chosen as organic component because it might be interesting to investigate partitioning behavior of 2-CIBP spiked to GAC in the presence of RAC. Such organic components are known to exhibit stronger chemical affinity for PCBs than inorganic components [13]. Lastly, mixture of the sediment components was also prepared. As a result, total 6 individual components were tested in this study. Among the components, mixture was the closest to actual sediments. Kaolin (Sigma-Aldrich), MMT (Sigma-Aldrich), coal (BCR reference, CRM 460), graphite (EC100, Graphite Sales, Inc.) and GAC (HD3000, Norit America, Inc.) were further purified with deionized water several times to remove any impurities. Their properties were characterized including surface area using Tristar 3000 porosimetry analyzer (Micromeritics) and organic carbon content using TOC analyzer with SSM-5000A solid module (Shimadzu).

To spike sediment components with 2-CIBP, 500 mg/L of 2-CIBP solution in acetone (Sigma-Aldrich) was prepared and then 2.4 mL of 2-CIBP solution was transferred to 12 g of each sediment component or 2.5 mL of 2-CIBP solution was transferred to 12.5 g of mixture (2.5 g of each component). Then, 40 mL of acetone was additionally added to mix sediment component and 2-CIBP under rigorous tumbling at 30 rpm for 15 h. After spiking, slurry was dried in an oven at 58 °C for 6 h to remove acetone. Theoretical solid phase concentration of 2-CIBP was 100 mg/kg as target concentration, assuming all 2-CIBP added was left on sediment component without loss. It should be noted that 2-CIBP was transformed only to BP by dechlorination reaction on RAC, which makes it easy to interpret experimental result.

2.3. Sacrificial batch test

Most of experimental procedures, from sample preparation to sample analysis, were done in anaerobic glove box or within air-tight bottles. All experiments were based on sacrificial batch. Experimental set up to determine fate and transport of spiked 2-CIBP was categorized into control, direct mixing, and compartment configuration, as precisely described elsewhere [25]. To investigate desorption of spiked 2-CIBP from sediment component to water in the absence of RAC, 0.9 g of sediment component was directly mixed with 22 mL of water (control). Batch reactors were disturbed at 150 rpm in rotary shaker. 2-CIBP would partition into water and sediment component. Additional set was built with RAC, where 0.6 g of RAC was directly mixed with the aqueous sediment component (direct mixing).

Separation of RAC from sediment component, which is not feasible in direct mixing configuration, is essential for understanding partitioning behavior of spiked 2-CIBP between the two heterogeneous materials. To address the need, RAC was physically separated from sediment component (compartment configuration). RAC compartment made of 2 mL vial (Supelco) was filled with 0.6 g of RAC followed by water, and capped with filter paper (Whatman). RAC vial was then inserted to main reactor containing water and sediment component. The filter allows 2-CIBP and BP to move freely between the two compartments but it prevents sediment component and RAC from mixing. As a result, it was possible to

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