



Investigation into the feasibility of black carbon for remediation of nonylphenol polluted sediment through desorption kinetics after different order spiking



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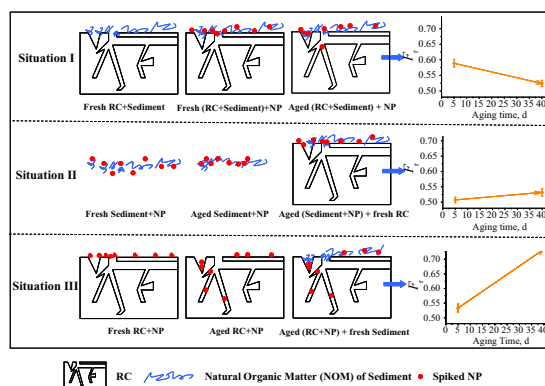
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HIGHLIGHTS

- A practical method was proposed for properly assessing the validity of a sorbent.
- NP inter-phase transport between RC and sediment is faster than desorption.
- With a short contact time, NP may be adsorbed on exterior surface sites of RC.
- With a longer time, NP diffusion into the RC inter-pores reduced release risk.
- RC is effective for sorbing and segregating of NP in aquatic environment.

GRAPHICAL ABSTRACT



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ABSTRACT

The binding order of sorbent, sediment and organic compounds, as well as binding time is important factors determining the potential success of sorbent amendment, which should be considered when the practicability of sorbents was assessed. But until now, relevant research was rare. In this study, desorption in three practical conditions were simulated, by three mixing spiking orders among nonylphenol (NP), rice straw black carbon (RC) and sediment (the order of mixing spiking is (RC + Sediment) + NP, (Sediment + NP) + RC and (RC + NP) + Sediment, for situation I, II and III, respectively), to discuss the feasibility of using RC to remedy NP pollution. Results demonstrated that amendment of RC into sediment decreased desorption fractions of NP, and increased the resistant desorption fraction (F_r), implying strong affinity of NP to RC and efficient sequestration by RC. No significant differences were observed for desorption among the three fresh situations, meaning NP may be adsorbed on RC exterior surface sites and inter-phase diffusion is faster than desorption. However, F_r for three aged situations was in the order: situation I < II < III, due to NP diffusion into the inter-pores or irreversible sorption sites of RC, reducing the releasing risk of NP. Regardless of time, F_r of three situations were all > 0.5, suggesting RC is an effective sorbent for remedying NP pollution in the aquatic environment. Overall, we proposed a practical and analytical method for properly assessing the validity of a sorbent.

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1. Introduction

Sorption studies are typically used to investigate sorbent performance with hydrophobic organic compounds (HOCs). However, for sorbents with high affinity for HOCs, desorption is irreversible; thus, using the sorption ability and capacity alone is insufficient for proper evaluation of sorbent efficiency when it is applied in organic pollution control. Desorption should also be considered when assessing the contaminant release risk examining sorbent utility. Currently, desorption techniques are increasingly and widely applied to feasible studies of sorbents such as Tenax TA, XAD resin and hydroxypropyl- β -cyclodextrin (Xu et al., 2008; Rhodes et al., 2010; Sandhu and Gu, 2013). These porous polymers can act as a sink for HOCs that are desorbing from sorbents where the aqueous concentration remains at zero (Pignatello, 1990) and where the desorption distribution and binding limits can be realized (You et al., 2007). Thus, consecutive desorption events aided by these porous polymers represent a good method for simulating the process of contaminant release in a real water environment. Moreover, the rapid-, slow- and very slow-desorbing fractions can be predicted (van Noort et al., 2003; Jonker et al., 2005; You et al., 2007).

In previous studies, the desorption process was usually conducted following such order: mixing the sorbents (e.g., black carbon [BC]) and sediment first, then spiking the mixture with HOCs. However, the order that (BC + Sediment) + HOCs (situation I) only represents one condition in which HOCs are desorbed from BC-inclusive sediment that is polluted by HOCs. This situation simulates acute pollution control using BC and only represents one case among the multiple complex conditions found in the environment. Other two situations should also be considered: 1) (Sediment + HOCs) + BC (situation II), where sediment is spiked with HOCs first and mixed with the sorbent BC, and then desorption occurs, which may support a process whereby BC is applied to aged HOCs for pollution control in sediment; and 2) (BC + HOCs) + Sediment (situation III), where the sorbent BC is spiked with HOCs and then mixed with sediment. Situation III may simulate HOC release when BC is used for water pollution control and settles into the sediment after BC adsorbs HOCs in the overlying water.

Many investigations suggested that the sorption capacities of sorbents varied under different conditions (Werner et al., 2006; Fojut and Young, 2011; Ghosh et al., 2011; Xiao et al., 2012). For example, sorption capacities differ between fresh and aged sorbents in sediment. When sorbents are used to control pollution in sediment, the sorption capacity of sorbents and the levels of HOCs desorption can change depending on the contact time between the sorbents and sediment. Upon sediment exposure, the binding of HOCs to sorbent particles may change due to the following two processes. First, HOCs diffuse into the inter-pore or irreversible sorption sites of the sorbent, which most likely leads to gradually greater sorption by the sorbent over time and lower HOCs desorption. Second, sorption sites may be clogged and/or blocked through binding with colloids, natural organic matter (NOM) or other organic contaminants, which leads to lower sorbent sorption for new HOCs (Pignatello et al., 2006), but would not influence sorption for previously sorbed HOCs. Consequently, the sorption capacity of sorbents changes with time and water quality conditions when it was applied in organic pollution controls, due to the complexity of real water environments. Hence, multiple perspectives should be considered when investigating the practicality of sorbents, and the binding order between the sorbent, sediment and HOCs, as well as the binding time are important for determining the potential success of sorbent use (Oen et al., 2012). However, few studies have focused on HOC desorption kinetics from sorbents under different conditions.

In the present study, a typical endocrine disruptor and persistent pollutant, nonylphenol (NP), was used as a representative HOC. Substantial ecotoxicological data have demonstrated that NP induces significant estrogenic effects, is toxic and accumulates in biological systems. We studied Tenax desorption kinetics for NP from a rice straw black carbon (RC)-sediment system with different aging times and using three different spiking orders – namely, (RC + Sediment) + NP, (Sediment + NP) + RC and (RC + NP) + Sediment, representing situations I, II and III, respectively. The objective of this research was not only to investigate the feasibility of using RC for NP pollution control but also to explore the mechanism of aging and sorption as well as the effect of different operating conditions on NP desorption using an RC-sediment system.

2. Materials and methods

2.1. Chemicals and materials

NP with a purity of >99% was purchased from Aladdin (China) and prepared to generate a concentrated stock solution with acetonitrile. Tenax TA (60–80 mesh) was obtained from Supelco (USA). Tenax TA was activated or regenerated through ultrasonic washing with methanol, acetone and hexane in that order (each solvent was used 3 times, and 10 mL solvent was used for 1 g Tenax TA).

The sediment was obtained from the Qiantang River, Hangzhou, Zhejiang province, China using a clam sampler. The water content of the sediment was 55.01%, the pH was 7.14, the cation exchange capacity (CEC) was 10.94 cmol/kg, and it was composed of 7.12% sand, 13.84% silt and 79.04% clay. The TOC and BC levels in the sediment were 0.964% and 0.37%, respectively; for more detail, see Lou et al. (2011).

RC was prepared from air-dried rice straw collected from the Hua-jia-chi Campus of Zhejiang University in China. The rice straw was burned on a stainless steel plate in an open field under uncontrolled conditions on a still and sunny afternoon (Lou et al., 2011). To generate purified RC, the sample was treated in 2 M HCl and 1 M: 1 M HCl-HF solutions, washed with distilled water, and oven-dried overnight at 105 °C. RC was finely ground (<250 μ m particle size), and its properties is referenced in our previous studies (Lou et al., 2011).

2.2. Spiking and aged methods of NP on RC and sediment

RC and sediment were spiked with NP at 4000 mg/Kg and 20 mg/Kg (dry weight), respectively, by adding acetonitrile solutions containing NP (the final ratios were 20 μ L of 1000 mg/Kg NP stock solution/5 mg of dry RC, and 10 μ L of 200 mg/Kg NP stock solution/100 mg of dry sediment). After the carrier solvent was allowed to evaporate for approximately 1 h in a fume hood until dry, the treated RC and sediment samples were capped and shaken at 80 rpm in the dark at room temperature for 5 days and 40 days, respectively.

2.3. Mixed spiking methods of NP on RC-sediment system

Three different mixed spiking processes of NP on RC-sediment system were performed to produce three different mixed sorbents with a NP concentration of 20 mg/Kg (dry weight).

Situation I: First, 5 mg RC was mixed with 95 mg sediment, and the mixed sorbent was immediately spiked with 10 μ L 200 mg/L NP solution. Next, the spiked samples were capped and shaken at 80 rpm in the dark at room temperature for 5 days and 40 days, respectively.

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