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Effects of oil dispersant on solubilization, sorption and desorption of polycyclic aromatic hydrocarbons in sediment–seawater systems

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ABSTRACT

This work investigated effects of a prototype oil dispersant on solubilization, sorption and desorption of three model PAHs in sediment–seawater systems. Increasing dispersant dosage linearly enhanced solubility for all PAHs. Conversely, the dispersant enhanced the sediment uptake of the PAHs, and induced significant desorption hysteresis. Such contrasting effects (adsolubilization vs. solubilization) of dispersant were found dependent of the dispersant concentration and PAH hydrophobicity. The dual-mode models adequately simulated the sorption kinetics and isotherms, and quantified dispersant-enhanced PAH uptake. Sorption of naphthalene and 1-methylnaphthalene by sediment positively correlated with uptake of the dispersant, while sorption of pyrene dropped sharply when the dispersant exceeded its critical micelle concentration (CMC). The deepwater conditions diminished the dispersant effects on solubilization, but enhanced uptake of the PAHs, albeit sorption of the dispersant was lowered. The information may aid in understanding roles of dispersants on distribution, fate and transport of petroleum PAHs in marine systems.

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1. Introduction

Oil spill accidents have been a major challenge to the petroleum industry (Wang and Fingas, 1997). Oil spill often causes serious ecological consequences. For example, oil components such as polycyclic aromatic hydrocarbons (PAHs) can persist in the environment and pose long-term threat to the environmental and ecological health.

The 2010 Deepwater Horizon (DWH) oil spill gushed an estimated 7.94×10^8 – 1.11×10^9 L of crude oil into the Gulf of Mexico (Reddy et al., 2012; Sammarco et al., 2013). To mitigate the environmental impacts, approximately 8.21×10^6 L of chemical dispersants (Corexit EC9500A and Corexit 9527A) was applied (Kujawinski et al., 2011).

Crude oil is a mixture of more than 17,000 hydrocarbon compounds with varying volatility, solubility and toxicity (Sammarco et al., 2013). The light molecular weight hydrocarbons are relatively more soluble in water, such as PAHs, monoaromatic hydrocarbons, phenols and nitrogen- and sulfur-containing heterocyclic compounds, which are often of priority concern due to their high mobility and toxicity (Gong et al., 2014a).

PAHs are important oil components, and can also result from incomplete combustion of fuels. PAHs represent a major environmental concern associated with oil spill, discharge and seepage (Nam et al., 2008). Alkylated PAHs are more toxic, and are often present at higher concentrations, than the parent PAHs. EPA (2003) has regulated 34 PAHs, including 18 parent-PAHs and 16 groups of alkylated PAHs (Choi et al., 2012). The DWH oil contained about 3.9% of PAHs by weight and a total of 2.1×10^4 tons of PAHs was released into the water column (Reddy et al., 2012). During and after the DWH oil spill, elevated concentrations of PAHs and alkylated PAHs were detected, including naphthalene, 1-methylnaphthalene, phenanthrene, pyrene, chrysene, and benzo(a)pyrene in the Gulf of Mexico. During the accident, up to 0.13 mg/L of naphthalene and 0.34 mg/L of pyrene were detected on the sea surface near the site, and 1.22 mg/L and 0.02 mg/L, respectively, near the wellhead. The alkylated naphthalene concentration reached 0.71 mg/L in the surface water and 32.1 mg/L near the wellhead (Diercks et al., 2010).

In marine eco-systems, PAHs undergo various physical and chemical processes, including solubilization, chemical and biological transformation, sorption to sediments and suspended particulate matter (SPM), and dispersion in the water column. Interactions with sediment particles or SPM can alter the fate and transport of PAHs. For example, association of PAHs with SPM can facilitate formation of marine oil snow and result in

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vertical transport of PAHs, and sediment-sorbed PAHs are more resistant to biotic or abiotic degradation (Yamada et al., 2003). In addition, the horizontal SPM movement with the ocean current may spread PAHs to the open sea (Tremblay et al., 2005).

Sediment organic matter (SOM) often plays a governing role in sorption of hydrophobic hydrocarbons including PAHs (White and Pignatello, 1999; Zhao et al., 2001). PAHs are subject to strong sorption on SOM. While SOM in estuarine sediments is largely of terrestrial character, organic matter in deep-sea sediments and marine suspended matter is more characteristic of marine organic matter. In addition, marine sediments may contain a larger fraction of refractory SOM and a significant amount of black carbon (BC), with BC/OC% values ranging from $15 \pm 2\%$ to $21 \pm 6\%$ in abyssal sediments and up to $50 \pm 40\%$ in coastal sediments (Coppola et al., 2014). Temperature and salinity can also affect the sorption rate and extent. For example, temperature inversely affect the PAH uptake due to the exothermic nature of the sorption (Tremblay et al., 2005; Zhang et al., 2009).

Oil dispersants are designed to disperse oil in the water column, enhance biodegradation and mitigate the impacts on near-shore ecosystems (Kujawinski et al., 2011). The key components in the dispersants include three nonionic surfactants (Span 80, Tween 80, and Tween 85), and one anionic surfactant dioctyl sodium sulfosuccinate (DOSS) (Thibodeaux et al., 2011). Corexit EC9527A contains 48 wt.% of the non-ionic surfactants and 35 wt.% of the anionic surfactant and 17% of solvents (Scelfo and Tjeerdema, 1991). Corexit 9500A shares the same surfactants as Corexit 9527A, but it does not include 2-butoxy ethanol as a solvent.

While surfactants are known to affect sorption of PAHs by oil and sediment, little is known on the effects of oil dispersants. In water–sediment systems, surfactant molecules can exist as micelles, monomers, hemimicelles and admicelles, depending on the surfactant concentration. Partitioning of oil or PAHs into surfactant micelles or monomers can increase the apparent solubility, thereby dispersing or solubilizing more oil/PAHs in the water column. On the other hand, the surfactants can also be adsorbed on the particles to form hemimicelles or admicelles, providing additional capacity to adsorb PAHs (Fig. S1 in Supplementary Materials, SM) (Ko et al., 1998). Gong et al. (2014b) reported that Corexit 9500A enhanced uptake of phenanthrene onto a sandy loam sediment.

The presence of oil or dispersed oil may alter the distribution of PAHs in sediment–water systems. Walter et al. (2000) investigated distribution of 9 PAHs in an oil-contaminated soil system and observed that more PAHs were distributed in the adsorbed oil and liquid oil phases than in the aqueous phase. However, there has been little information available on the effects of oil dispersants and dispersed oil.

During the DWH oil spill, $\sim 2.91 \times 10^6$ L Corexit EC9500A was applied at the 1500 m deep wellhead (Kujawinski et al., 2011). The deepwater conditions are characterized by high pressure (~ 16 MPa), low temperature (~ 4 °C), little or no light, constant salinity (3.5%), and sluggish currents (< 0.25 knots) (Glover and Smith, 2003). However, there has been no information available on sorption/desorption behaviors of oil dispersant and PAHs under such conditions.

The overall goal of this study was to investigate effects of a model oil dispersant (Corexit EC9500A) on the solubilization, sorption and desorption of two model parent PAHs (naphthalene and pyrene) and one alkylated PAH (1-methylnaphthalene) in sediment–seawater systems. The specific objectives were to: (1) investigate effects of the dispersant on solubilization of the PAHs, (2) examine sediment sorption and desorption behaviors of the dispersant under surface water and deepwater conditions, (3) determine the effects of the dispersant and dispersed oil on sediment sorption and desorption of the PAHs and elucidate the underlying

mechanisms, and (4) test the PAH–sediment interactions under deepwater conditions in the presence of the dispersant.

2. Materials and methods

2.1. Chemicals

All chemicals were of analytical grade or higher. Naphthalene, 1-methylnaphthalene, and pyrene were purchased from Alfa Aesar (Ward Hill, MA, USA). Table S1 in SM summarizes salient physico-chemical properties of these PAHs. ^{14}C -radiolabeled naphthalene (specific activity: 52 mCi/mmol) and pyrene (60 mCi/mmol) were purchased from Moravex Biochemicals Inc. (Brea, CA, USA). Sodium azide (NaN_3) and sodium chloride (NaCl) were obtained from Fisher Scientific (Fair lawn, NJ, USA). Ecoscint cocktail was purchased from National diagnostics (Atlanta, GA, USA). Corexit EC9500A was acquired from Nalco Company (Naperville, IL, USA). Table S2 in SM shows the compositions of this dispersant. A surrogate Louisiana sweet crude oil was obtained by courtesy of the BP Company (Houston, TX, USA).

2.2. Seawater and sediment samples

Seawater was collected at Grand Bay, AL, USA (N30.38/W88.31). Before use, the seawater was filtered through a membrane ($0.45 \mu\text{m}$) to remove suspended solids, and then sterilized through autoclaving at 121 °C for 35 min. The salient properties of the treated seawater include: pH = 8.88, dissolved organic carbon (DOC) = 0.43 mg/L, salinity = 3.2%, Cl^- = 18.55 g/L, NO_3^- = 2.55 g/L and SO_4^{2-} = 4.25 g/L. To inhibit microbial activities during the experiments, 200 mg/L of sodium azide (an aerobic metabolic inhibitor) was added in the seawater.

Two marine sediments, a loamy sand and a sandy loam, were sampled at N30.37926/W88.30684 and N30.37873/W88.30679, respectively. The sediments were wet-sieved with seawater to obtain a fraction of 75–840 μm and then air-dried for 7 days. The dried aggregates were further oven-dried for 6 h at 80 °C before use. Sediment analyses were performed by the Soil Testing Laboratory at Auburn University, and the method details have been described elsewhere (Gong et al., 2012). Table S3 gives salient properties of the sediments.

2.3. Dispersant-enhanced solubilization of PAHs

The apparent solubility of PAHs in seawater was measured in the presence various concentrations of the dispersant based on the procedure by Zhao et al. (2005). Individual solutions of a super-saturated PAH were prepared by mixing 0.1 g of a PAH with 43 mL of seawater containing 0–200 mg/L of the dispersant. Upon mixing for 3 days, the vials were centrifuged at 3000 rpm (1509 g-force) for 10 min and 10 mL of the supernatants were carefully sampled and diluted with 1.0 mL of methanol. And the PAH concentration was then quantified via HPLC (HP Series 1100, Hewlett Packard, CA, USA) equipped with a UV detector and a Zorbax SB-C18 column (150×468 mm). The mobile phase consisted of 80% acetonitrile, 19.9% water, and 0.1% phosphoric acid. The operating flow rate was set at 1.0 mL/min at 40 °C with an optimal wavelength of 254 nm.

2.4. Sorption of Corexit EC9500A by sediment

Batch experiments were conducted to construct sorption/desorption isotherms of the dispersant with the sandy loam. Eight grams of the sediment were mixed with 42 mL seawater in the presence of 5–6000 mg/L of the dispersant. The mixtures were

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