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# Effects of oil dispersants on settling of marine sediment particles and particle-facilitated distribution and transport of oil components

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## ABSTRACT

This work investigated effects of three model oil dispersants (Corexit EC9527A, Corexit EC9500A and SPC1000) on settling of fine sediment particles and particle-facilitated distribution and transport of oil components in sediment-seawater systems. All three dispersants enhanced settling of sediment particles. The nonionic surfactants (Tween 80 and Tween 85) play key roles in promoting particle aggregation. Yet, the effects varied with environmental factors (pH, salinity, DOM, and temperature). Strongest dispersant effect was observed at neutral or alkaline pH and in salinity range of 0–3.5 wt%. The presence of water accommodated oil and dispersed oil accelerated settling of the particles. Total petroleum hydrocarbons in the sediment phase were increased from 6.9% to 90.1% in the presence of Corexit EC9527A, and from 11.4% to 86.7% for PAHs. The information is useful for understanding roles of oil dispersants in formation of oil-sediment aggregates and in sediment-facilitated transport of oil and PAHs in marine eco-systems.

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## 1. Introduction

Oil dispersants are mixtures of surfactants and hydrocarbon-based solvents. Dispersants have been widely used to disintegrate spilled oil slicks into fine droplets, thereby facilitating dispersion of oil into the water column and accelerating the natural attenuation processes (Griffiths, 2012; Lessard and Demarco, 2000). Oil dispersants have been employed in several major oil spill accidents, and over years, this approach has evolved into a major countermeasure to mitigate the adverse effects of oil spills (Griffiths, 2012; Lessard and Demarco, 2000). For examples, ~12 t of Corexit EC9500A were applied in the *Sea Empress* spill in the UK in 1996; and ~11.5 t of Corexit EC9527A were sprayed in the pipeline spill offshore in Texas in 1998 (Lessard and Demarco, 2000). The single-largest application was carried out during the Deepwater Horizon (DwH) oil spill, where approximately 7800 t of Corexit EC9500A and EC9527A were applied on the surface (~5000 t) and deep-water (~2800 t) (Gong et al., 2014a; Kujawinski et al., 2011).

Researchers have studied adsorption of surfactants on minerals (Fuerstenau, 2002), but very few have addressed effects of oil dispersants on the settling behaviors of suspended particulate matter (SPM) or fine sediment particles in marine systems. Adsorption of dispersant components, e.g. surfactants, on the sediment particles may alter the surface potential of the particles, which can coagulate and destabilize

the particles and cause particle aggregation and enhance particle settling (Paria and Khilar, 2004).

Application of oil dispersant results in elevated concentrations of oil droplets in the water column, for example, the volatile aromatic hydrocarbons in an oil plume during the DwH oil spill reached 139 µg/L (Hazen et al., 2010). The high oil droplet concentration is favorable for the formation oil sediment aggregates (OSAs) (Gong et al., 2014a), which can not only alter the settling behavior, but also facilitate distribution and transport of important oil compounds that are associated with the aggregates. Guyomarch et al. (1999) found that the presence of the chemical dispersant, Inipol IP90, enhanced the formation of OSAs, and approximately 80% of oil was entrained in the OSAs, which subsequently settled down to the bottom of the tank during wave tank experiments. Recent work also showed that the presence of Corexit EC9500A promotes the formation of marine oil snow (MOS) and sorption of oil components onto MOS (Fu et al., 2014).

The DwH oil spill gushed 4.1 million barrels (653,000 m<sup>3</sup>) of crude oil into the Gulf of Mexico ecosystems (Allan et al., 2012), of which  $2.1 \times 10^7$  kg of polycyclic aromatic hydrocarbons (PAHs) was released (Reddy et al., 2012). During the spill, the concentration of PAHs near the wellhead reached as high as 0.189 mg/L (Allan et al., 2012; Diercks et al., 2010). As PAHs can strongly interact with suspended sediment particles, a good portion of PAHs can be adsorbed on, or incorporated in, the OSAs. Consequently, the formation of OSAs and subsequent particle settling/transport in the marine environment can alter the distribution, transport and environmental fate of PAHs and other important oil

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components (Bouloubassi et al., 2006; Zuijgeest and Huettel, 2012). For instance, adsorption of PAHs onto sediment particles may mitigate the peak concentration of PAHs in the water column by transferring significant amounts of PAHs into the sediment phase; on the other hand, the particle-facilitated transport process can facilitate transport of PAHs to the sea bottom or surface or nearshore areas. In addition, as PAHs persist much longer in the sediment phase than at the sea surface (Harayama et al., 1999), sediment adsorption may prolong the life time of PAHs in the ecosystems. Recent studies have shown that oil dispersants can enhance sorption of PAHs by sediment and induce sorption hysteresis (Gong et al., 2014b; Zhao et al., 2015). While a wealth of information on the adsorption of oil components onto sediments has been reported (Guo et al., 2007; Tremblay et al., 2005; Zhao et al., 2015), little is known about effects of dispersants or dispersed oil on the transport of fine sediment particles and how such particle-facilitated oil transport affects the fluxes and fate of oil components (Allan et al., 2012). As oil spills often occur in, or can reach, nearshore and estuarine zones, where the concentration of suspended sediment particles is usually high (NRC, 2005), the oil-particle interactions may play even more profound roles in these more ecologically sensitive areas.

Environmental conditions, such as pH, salinity, temperature, and dissolved organic matter (DOM) are known to affect settling of suspended particles. Kretzschmar et al., (1998) investigated influence of pH and humic acid on the coagulation of kaolinite by the dynamic light scattering technique, and observed that at pH < 5.8 the interparticle attractive forces predominate the suspension system, resulting in destabilization of the particles. Humic acid was reported to render a suspension system more stable due to the combination of electrostatic and steric stabilization effects (Heil and Sposito, 1993a,b; Kretzschmar et al., 1993). However, knowledge has been lacking on the effects of such environmental parameters in the presence of oil dispersants, especially, how oil dispersants affect the settling properties of suspended particles under various environmental conditions.

The overall goal of this study was to systematically investigate effects of three model oil dispersants (Corexit EC9527A, Corexit EC9500A, and SPC1000) on the settling behaviors of sediment particles and how such dispersant-particle interactions affect distribution and transport of key oil components. The specific objectives were to: 1) determine the settling kinetics of representative sediment particles in the presence of the model dispersants; 2) elucidate the mechanisms underlying the particle-dispersant interactions; 3) test the dispersant effects under various environmental conditions, such as pH, salinity, DOM, and temperature; and 4) explore how dispersant-facilitated particle sedimentation affects distribution and transport of important oil components in sediment-water systems.

## 2. Materials and methods

### 2.1. Materials

The following chemicals (analytical grade or higher) were used as received: humic acid (Fluka Chemie, Switzerland), sodium hydroxide (NaOH) and NaN<sub>3</sub> (Fisher Scientific, Fair lawn, NJ, USA), hydrochloric acid (HCl, 36.5–38.0%, Mallinckrodt Chemical, St. Louis, MO, USA), phenanthrene and pyrene (98%, Alfa Aesar). The dispersants Corexit EC9500A and Corexit EC9527A were obtained per courtesy of Nalco Company (Naperville, IL, USA), and SPC1000 was purchased from U.S. Polychemical Corporate (Chestnut Ridge, NY, USA). The key compositions of the Corexit dispersants, such as Tween 85, Tween 80, Dioctyl sulfosuccinate sodium salt (DOSS) and 2-butoxyethanol, were purchased from VWR Corporate (Radnor, PA, USA). A standard reagent of 16 EPA listed PAHs, and a standard of *n*-alkanes mixtures (C9–C40), Pristane and Phytane, and two internal standards (fluorine-*d*<sub>10</sub> for PAHs and 5  $\alpha$ -androstane for *n*-alkanes) were acquired from Supelco (Bellefonte, PA, USA).

A surrogate Louisiana Sweet Crude (LSC) oil was obtained by courtesy of BP (Houston, TX, USA). This oil is considered physically, chemically, and toxicologically similar to the Macondo Well crude oil in the Mississippi Canyon Block 252. The artificial oil weathering method and preparation of water accommodated oil (WAO) and dispersed WAO (DWAO) are described in Section S1, Supplementary Material (SM).

Seawater was collected from the Grand Bay area, AL (N30.37873, W88.30679). The seawater sample was filtered with 0.22  $\mu$ m pore size membrane and preserved at 4 °C with NaN<sub>3</sub> of 200 mg/L to inhibit microbial activities. The salient features of the seawater were as follows: pH = 7.9, DOM = 0.77 mg/L (as total organic carbon (TOC)), and salinity = 3.15 wt%. The PAHs in the seawater were not detected. Three model sediments, referred to as GB, OS and EIWR were collected, respectively, from Grand Bay, AL (N30.37873, W88.30679), the Ocean Springs National Seashore Park, MS (N30.39772, W88.79175), and the Elmer's Island Wildlife Refuge (EIWR), LA (N29.17764, W90.07401). The sediment samples were collected at the depth of 0–60 cm, thoroughly mixed and wet-sieved through 2 mm openings to remove the plant debris and large objects, and then air-dried for 1 week. The dried aggregates were then crushed using ceramic mortar and pestle, and sieved through 0.84 mm openings, and the subsamples (i.e. particles of  $\leq 0.84$  mm) were oven-dried for 6 h at 80 °C, which also inactivated the sediment microorganisms. The salient physical and chemical properties of the sediments were tested by the Soil Testing Laboratory at Auburn University, the details on the analytical methods have been described elsewhere (Gong et al., 2012). Table 1 gives the physicochemical characteristics of the sediment samples.

### 2.2. Effects of dispersants on sediment settling

The settling rate of the sediment particles was tested in 480-mL amber bottles with PTFE lined caps. First, each bottle was filled with 300 mL seawater, and then a known mass (1.2 to 4.2 g, depending on the sediment type) of a sediment was added into the seawater (the sediment mass was adjusted to achieve an equal initial turbidity of  $180 \pm 24$  NTU for the suspensions). The sediment-seawater suspensions were then thoroughly mixed by hand shaking. Upon addition of a known concentration (up to 20 mg/L) of a dispersant, the bottles were placed on an orbital shaker (Excella E5 Platform Shaker, New Brunswick scientific, NJ, USA) operated at 200 rpm for 12 h, and then at 50 rpm for 10 min to allow complete contacts between the dispersant molecules and sediment particles. The fully mixed suspensions were then allowed to stand still to initiate quiescent gravity settling of the particles. At predetermined times, samples (4 mL each) were taken from the center (6 cm from the surface) of the suspensions, and then measured for the turbidity change over time. It should be noted that it took about 15 s to obtain a stable turbidity reading. In addition to the three dispersants, the effects of key individual dispersant components, including three surfactants (Tween 80, Tween 85, DOSS) and one solvent (2-butoxyethanol) were also tested to assess their individual effects on the settling performance. In all cases, control tests were carried out without dispersant but under otherwise identical conditions. To assure data quality, all experiments were conducted in duplicate. In all figures, data were plotted as mean of duplicates and error bars were calculated as standard deviation to indicate data reproducibility.

Gordon (1970) showed that the suspended particles are numerically most abundant at the size (diameter) of <7  $\mu$ m in the ocean. For the sediment particles involved, it takes ~40 min for the 7- $\mu$ m particles to settle from surface to below the sampling point (~6 cm) (See Section 2.3). Therefore, the 40-min sedimentation efficiency ( $R_{40}$ ) was used to compare settling rates of particles under various conditions.

$$R_{40} = \frac{\text{Turbidity at 40 min}(T_{40})}{\text{Initial Turbidity}(T_0)} \quad (1)$$

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