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Effect of environmental factors on the effectiveness of ammoniated bagasse in wicking oil from contaminated wetlands

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Abstract

Ammoniated bagasse is a plant-derived organic sorbent that can be used for capturing oil and for supplying slow-release nutrients to oil-degrading microorganisms. We investigated the oil-wicking behavior of this sorbent under various conditions for its effectiveness in remediating oil-contaminated wetlands. Abiotic microcosms simulating a wetland environment were used to assess the influence of sand particle sizes (20×30 and 60×80 U.S. mesh), degrees of oil saturation (25% and 75%), water table levels (on top of the clean sand layer, oiled-sand layer, and sorbent layer), and the presence of sorbent. Results indicated that oil wicking favors higher oil contamination, larger sand particle size, and low water coverage. Water coverage was the predominant factor limiting the effectiveness of sorbent. The most plausible explanation for this limitation was that sorbent captured more water than oil at higher water coverage.

Key words: crude oil; wetlands; sorbent; remediation; sediment

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Introduction

Wetland ecosystems perform important functions by providing nurseries for fish, habitats for wildlife, and a rhizosphere for plants (Mitsch and Gosselink, 1986). These functions are put at grave risk when wetlands are contaminated with petroleum or petroleum products. Accidental discharges of petroleum in water bodies result in massive contamination to shoreline and wetland environments. Contamination in low energy environments is even more critical (e.g., marshes, mud flats, and subtidal areas) since they have been reported to persist for many years and cause severe damage to these sensitive ecosystems (Mendelssohn et al., 1990; Office of Technology Assessment, 1991; Webb et al., 1985).

Remediation of oil-impacted wetlands should be performed with caution, due to the fragile nature and the limited accessibility of such environments (Grace, 1999). Typical cleanup methods often cause more damage to plants, rhizosphere, and wildlife habitats than the spilled oil (Venosa et al., 2002). Furthermore, foot-traffic by humans can lead to deeper contaminant penetration into anoxic sediment, which limits subsequent restorations (Hoff and Shigenaka, 1993). *In situ* bioremediation is one of the most common and effective method to remediate oil spills along shorelines and wetlands, if cleanup activities are strictly controlled to minimize damage to ecosystems.

Biodegradation of hydrocarbons in nature is often limited by the nutrients needed for the growth of oil-degrading bacteria (de Laune et al., 1990). Thus, water-soluble fertilizer (e.g. nitrate salts, ammonium salts, or urea) is often used to compensate for nutrient deficiency in bioremediation sites. The main disadvantage of water-soluble fertilizers is that they are easily washed out by water during rain events and tidal or wave action (Zhu et al., 2004). To overcome this disadvantage, research has been performed to assess the benefits of slow-releasing fertilizers (Olivieri et al., 1976; Aarnio and Martikainen, 1995). Slow-releasing fertilizers are in solid form and consist of inorganic nutrients either coated with water-insoluble material, semi-permeable material, or degradation-controlled material.

To supply nutrients to oil-degrading microorganisms, researchers at Louisiana State University (LSU) suggested a different approach. They have tested the effectiveness of various natural organic sorbents with different nitrogen contents for wicking oil and stimulating biodegradation (de Silva, 1995). They observed that none of the naturally occurring materials have sufficient ability to promote biodegradation of hydrocarbons, due to either lack of longevity or nutrient deficiency. To overcome the perceived deficiencies, sugarcane bagasse, a waste product from sugar refining, was modified through an ammoniation reaction in the presence of ammonia and air at the temperatures of 80–200°C and at the pressure over 750 psi (Brieitenbeck

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and Kember, 2003).

The ammoniated bagasse was claimed to provide excellent sorbent properties in addition to serving as a source of slow-release nutrients (Breitenbeck and Grace, 1997). A field study by Grace (1999) showed that application of ammoniated bagasse on a contaminated marsh surface was more effective in promoting crude oil bioremediation than other treatments (peat moss, chicken feathers, and kenaf). Goodin and Hudnall (2001) performed a field study with ammoniated bagasse to remediate an oil-contaminated wetland after *in situ* burning. They demonstrated the usefulness of ammoniated bagasse for removal of petroleum residue and suggested using it immediately after an oil spill to avoid detrimental effects on the vegetation.

The effectiveness of this method depends on various environmental factors. Gandee (2007) demonstrated that sorbents, including ammoniated bagasse, have limited effectiveness in wicking oil from the sediment at low levels of oil contamination (< 50 g oil/kg sediment). This limited effectiveness may be attributed to the sorption of the oil to the sediment surfaces and the limited availability of free oil. Using microcosm studies, de Silva (1995) observed that oil wicking from a saline marsh soil was more difficult than that from beach sand. Furthermore, personal communications with LSU researchers revealed that the position of the water table had a major bearing on the effectiveness of oil wicking as observed from field studies. Oil wicking experiments performed on highly saturated sediment gave inconsistent results due to the interfacial tension between water and oil. Sediment properties and degree of saturation can affect wicking by decreasing the interconnected capillary forces drawing the oil to the surface.

These observed challenges in oil wicking motivated a systematic study to evaluate the effectiveness of oil wicking under various simulated wetlands conditions. The objective of the study is to perform controlled microcosm experiments to determine the effect of various conditions on the effectiveness of oil wicking as a potential biostimulation method. Specifically, microcosm conditions included the effect of sand particle size, position of the water table, and the degree of oil contamination. This study provides useful information on the use of ammoniated bagasse for successful bioremediation of wetlands impacted by oil spills.

1 Materials and methods

1.1 Materials

Coarse grain sand (20 × 30 U.S. mesh) was purchased from Fisher Scientific (USA), and fine grain sand (60 × 80 U.S. mesh) was obtained by sieving bulk sand purchased from Acros Chemical (USA). Both sand specimens were washed (with 1 mol/L nitric acid) and dried in the oven to remove impurities. Ammoniated bagasse was obtained from LSU (Breitenbeck, personal communication) and used as a wicking agent. It was dried in a 105°C oven overnight to remove moisture and kill microorganisms prior to use. Distilled water, supplemented with 5 mg/L sodium azide as a disabler of aerobic microorganisms, was added to achieve the desired water table level. Bonny light crude oil, previously weathered by aeration overnight, was obtained from the US EPA and mixed with sand to simulate an oil-contaminated environment. Weathered oil was selected to simulate natural weathering expected to occur by the time spilled oil reaches a wetland environment.

1.2 Methods

A total of 24 different microcosm conditions were constructed in duplicates to simulate saturated wetland environments. Variables tested included 2 sand particle sizes, 2 levels of oil contamination (25% and 75% of saturation), 3 levels of water table height (the top of the clean sand layer, top of the oiled-sand layer, and top of a 2-cm thick sorbent layer), and 2 levels of sorbent (presence and absence). The microcosms were enclosed in closed glass cylinders (10 cm in diameter and 10 cm in height). Each microcosm was layered in the following sequence (from the bottom to the top): a clean sand layer, an oiled-sand layer, and in most cases a layer of sorbent. The microcosms were first filled with water, and sand was gently poured into the water to insure absence of air bubbles. A stainless steel screen (100 × 100 mesh) was placed on top of the clean sand layer as a separator. Atop this layer, an oil-sand mixture was compacted to a depth of 1 cm. Sorbent was placed on top of the oil-contaminated layer to a depth of 2 cm. Distilled water was subsequently added to the microcosms to accomplish the desired coverage, as shown in Fig. 1. Parafilm and aluminum foil were used to seal the tops of the microcosms, and the microcosms were kept in a dark room to avoid the confounding effects of photodegradation. Oil wicking was allowed to proceed for 3 months. The initial sampling was performed on day zero on the clean sand, sorbent, and oil-sand mixture. These analyses were used to calculate the initial amount of oil in each layer of the microcosms. After three months, the final sampling event took place. Each layer of the microcosm was separated and sampled in triplicate. Samples were

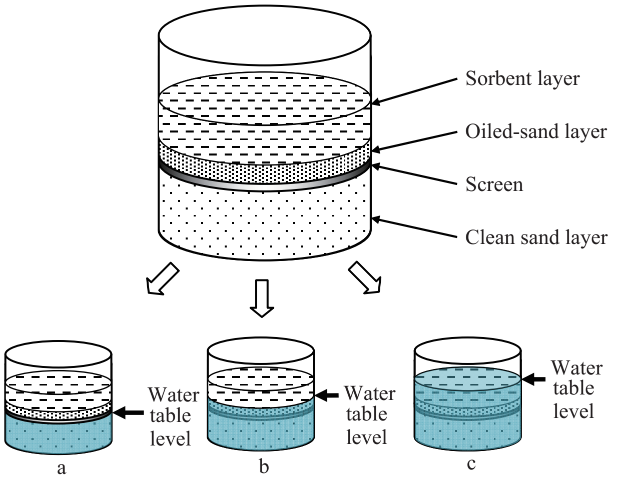

Fig. 1 Schematic of microcosms according to the position of water table located on top of the clean sand layer (a), the oiled-sand layer (b), and the sorbent-layer (c).

extracted with dichloromethane (DCM) using a Soxhlet extraction apparatus and separatory funnels. Extraction efficiency was checked by spiking surrogate compounds before extraction. In addition, blank extraction (DCM with surrogate spike) was included on a batch of extractions to check contamination during extraction. Samples with surrogate recovery of 75%–125% were taken. Extracts were concentrated in an evaporator (Turbovap, Zymark) and stored at 4°C in a refrigerator for analysis.

The concentration and mass of target compounds in each sample were quantified using a GC-MS (HP-5890 series II equipped with mass selective detector, Hewlet Packard, USA). The following GC-MS conditions were used for the analysis. The oven temperature was programmed to increase from 45 to 200°C at 4°C/min and then to increase from 200 to 310°C (held for 10 min) at 10°C/min. The temperature of the inlet and detector were 290 and 320°C, respectively. Chromatographic separations of target compounds were achieved with a capillary column (SPB-5, Supelco, USA). Ultra high purity helium (99.999% pure) was used as the carrier gas at a flow rate of 1 mL/min, and the mass selective detector was operated in selective ion monitoring (SIM) mode. GC-MS results were quantified with calibration curves maintained within 80%–120% of check standards. Mass balances on each microcosm were determined as the sum of alkanes (*n*C10–*n*C35); pristane, phytane, and hopane; 2-, 3-, and 4-ring PAHs; and pyrogenic PAHs containing 5 and 6 rings. Mass closure (M_c, %) and effectiveness (E, %) of wicking (either by sorbent or overlying water) were assessed by comparing the mass of total target compounds in all layers ($M_{T,t}$) and in the sorbent layer ($M_{S,t}$) or in the overlying water layer ($M_{W,t}$) after three months to the initial mass ($M_{T,0}$). The formulas used are given below:

$$M_c = \frac{M_{T,t}}{M_{T,0}} \times 100\% \quad (1)$$

$$E = \frac{M_{S,t} \text{ or } M_{W,t}}{M_{T,0}} \times 100\% \quad (2)$$

2 Results and discussion

2.1 Presence of sorbent

To avoid erroneous conclusions, the degree of closure on the mass of recovered oil at the end of the three-month experiment must be consistently high. After the three-month wicking periods mass closures in the range of 90%–97% were obtained for the various microcosms. Losses in target compounds can be attributed to volatilization during the experiment, sampling, and storage, with the majority of lost oil constituents observed for the more volatile alkanes (*n*C10–*n*C15). Relatively higher mass closure was observed for the experiments with the higher level of oil saturation, smaller grain size, and higher water coverage, although differences among treatments were not statistically significant ($p > 0.05$). Oil wicking test results observed for the different microcosm conditions are shown in Fig. 2. Each figure summarizes results relative to mass closure and wicking effectiveness for two levels of oil contamination (25% and 75% oil saturation) and two levels of sand particle size (coarse sand and fine sand) at a specific water table position (on top of the clean sand layer (Fig. 2a); on top of the oiled-sand layer (Fig. 2b); on top of the sorbent layer (Fig. 2c). The differences in mass closures are negligible compared to the differences in effectiveness among the different experimental conditions.

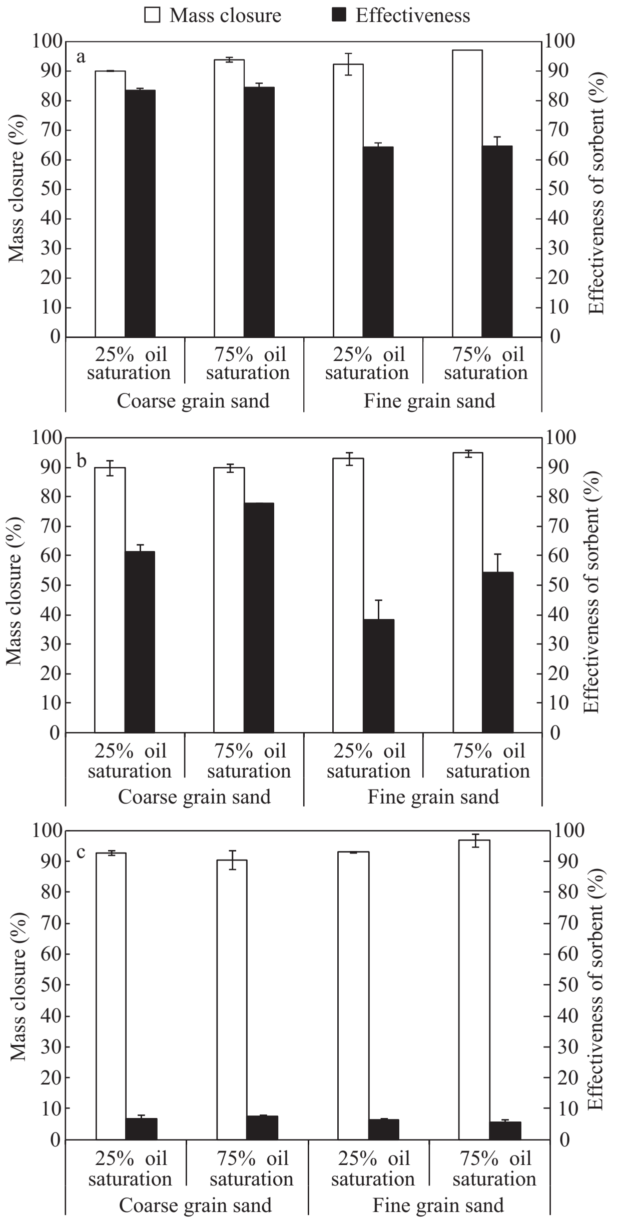

Fig. 2 Mass closure and effectiveness of sorbent in wicking oil under different sand particle sizes and oil contaminations levels when water table is located on top of the clean sand layer (a), the oiled sand layer (b), and the sorbent layer (c).

2.1.1 Effect of sand particle sizes

Figure 2 shows the effectiveness of the sorbent for different particle sizes. Effectiveness of sorbent is approximately 85% for coarse sand and 65% for the fine sand, when the water table is located on top of the clean sand layer (Fig. 2a). The effectiveness is approximately 60% (for the lower oil saturation) and 80% (for the higher oil saturation) for coarse sand, and about 40% (for the lower oil saturation) and 55% (for the higher oil saturation) for fine sand when the water table is on top of the oiled-sand layer (Fig. 2b). However, oil wicking effectiveness decreases to below 10% regardless of sand particle size and oil saturation degree when the water level covers the sorbent layer. The results suggest that sorption favours wicking oil from larger grained sand. The larger void spaces between the grains of coarse sand lead to higher oil wicking effectiveness. Similar findings were reported in the literature. Inagaki et al. (2004) used alumina powder in different particle sizes (175–713 μm) to explain the effect of sand particle size on capillary oil-wicking by a sorbent (exfoliated graphite). According to that study, oil wicking (cm) from coarse-grained sand was higher than that from fine sands.

2.1.2 Effect of oil contamination degrees

The two different degrees of oil contamination (25% and 75% oil saturation) do not impact wicking effectiveness when the added water covers either the clean sand layer or the sorbent layer as shown in Fig. 2a and c. However, considerable wicking effectiveness differences were observed for the two levels of oil contamination when water covers the oiled-sand layer in Fig. 2b. For coarse sand, effectiveness is approximately 60% (at 25% oil saturation) and increases to approximately 80% (at 75% oil saturation). For fine sand, effectiveness is about 40% (at 25% oil saturation) and about 55% (at 75% oil saturation). These results show that higher oil saturation results in higher oil wicking effectiveness when the water table is on top of the oiled-sand layer. In oil exploration studies, multiphase transports (oil and water) in porous media are often described in terms of relative permeability. High oil saturation contributes to a high relative permeability in a porous medium (e.g., sand) saturated with oil and water (Ollivier and Magot, 2005). Thus, oil can be removed more effectively from the environment when the degree of oil saturation is high and this removal may be enhanced by a pressure difference (e.g., capillary wicking).

2.1.3 Effect of water table levels

From Fig. 2, oil wicking effectiveness of sorbent for coarse sand is about 85% when the water table is on top of the clean sand layer, 60%–80% when the water level is on top of the oiled-sand layer, and less than 10% when it is on top of the sorbent layer. For fine sand, the corresponding degrees of effectiveness are about 65%, 40%–55%, and less than 10%. The results show that sorption effectiveness decreases as water level increases, with a marked decrease when water covers the sorbent layer. Water sorption by sorbent or the disruption of the interconnected oil-network by the water table under highly saturated conditions likely results in a low capillary force for wicking the oil. The effect of water table levels on oil wicking has rarely been reported in the literature. Vegetable fibers (e.g., kenaf and cotton) often exhibit poor buoyancy (i.e., sinking) and poor oil sorption capacity when they are presoaked with water during oil sorption tests in aqueous medium (Lee et al., 1999; Deschamp et al., 2003; Annunciado et al., 2005). The poor wicking abilities of hydrophilic vegetable fibers may be attributable to the adsorption of moisture.

2.2 Absence of sorbent

The amount of oil displaced by the overlying water layer when no sorbent is applied and the associated material balance on the oil are summarized in Table 1. In the absence of sorbent, most of the oil is retained in the oiled-sand layer. However, a significant amount of oil (about 30%–50%) is detected in the overlying water layer of the microcosms charged with fine sand, while a negligible amount (< 1%) was detected in the overlying water layer of the microcosms charged with coarse sand. The possible reason for this difference is that fine sand particles are light enough to be carried by the buoyant force of oil bubbles while coarse sand particles are not.

2.3 Statistical analysis

A linear regression model was applied to fit experimental data for each water table level using Minitab software.

$$y = a_0 + a_1 x_1 + a_2 x_2 \tag{3}$$

where, a_0 is the intercept, a_1 and x_1 are the coefficient and the variable for the effect of oil contamination level, and a_2 and x_2 are the coefficient and the variable for the effect of sand particle size, responses (y) and variables (x_1 and x_2) were divided by appropriate scale factors to generate dimensionless numbers ranging from 0 to 100. Analyses of variance (ANOVA) were performed to test null

Table 1 Mass closure and effectiveness of wicking by the overlying water layer when sorbent is absent under different conditions

Water table level	Sand particle	Oil saturation (%)	M_c (%)		E (%)	
			Average	Error	Average	Error
On the top of clean sand layer	Coarse	25	93.92	4.04	N.A.	N.A.
		75	91.06	2.33	N.A.	N.A.
	Fine	25	93.12	0.83	N.A.	N.A.
		75	92.94	0.86	N.A.	N.A.
On the top of contaminated sand layer	Coarse	25	93.98	3.13	N.A.	N.A.
		75	92.06	2.03	N.A.	N.A.
	Fine	25	90.47	2.33	N.A.	N.A.
		75	94.77	2.49	N.A.	N.A.
On 2 cm-high overlying water layer	Coarse	25	96.92	2.33	< 1	< 1
		75	90.39	2.50	< 1	< 1
	Fine	25	91.00	0.86	33.74	0.88
		75	89.80	4.08	53.22	4.42

M_c: mass closure; E: effectiveness of wicking by overlying water layer; N.A.: data are not available.

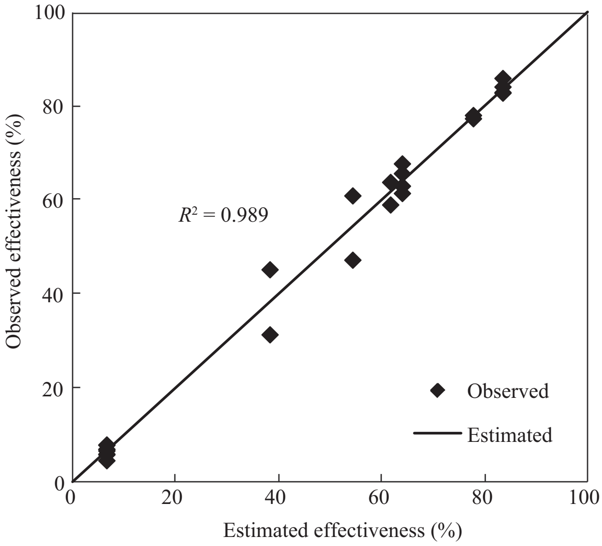

Fig. 3 Effectiveness of sorbent estimated by model equations in comparison to experimental data.

hypothesis (variables do not significantly affect response). When water covered the clean sand layer, sand particle size significantly affected sorption effectiveness ($p < 0.05$), while the degree of oil contamination did not ($p > 0.05$). When the water table covered the oiled-sand layer, both oil contamination levels ($p < 0.05$) and sand particle sizes ($p < 0.05$) significantly affected sorption effectiveness. When the water table covered the sorbent layer, none of the factors influenced effectiveness ($p > 0.05$). ANOVA test results suggest that different model equations should be established for different water table levels. Thus, hierarchical linear regressions for each water table level were performed as summarized in Table 2. The parameters for selected models were typed in bold font. Effectiveness of sorbent estimated by model equations under different water table levels were compared with experimental data as shown in Fig. 3. Experimental data fit well with model equations ($R^2 = 0.989$) and deviations from experimental data are within error ranges.

The intrinsic effect of the water table levels is evidenced by the comparison of intercepts in the linear equations for different water levels. As water table level increased, the intercept decreased significantly from 54.9 to 6.6. These results suggested that the position of water table is a predominant factor limiting sorption effectiveness. Sand particle size influenced effectiveness of the sorbent similarly when water covered either the clean sand layer (0.291) or the oiled-sand layer (0.349). The effect of oil saturation became significant (0.321) only when the oiled-sand layer was covered by water. When water covered the oiled-sand layer, the water table would confer a buoyancy effect on the oil. In this case, a high degree of oil contamination in the saturated condition would drive more oil to the interface between the oiled-sand layer and the sorbent layer. This conclusion was supported by the experimental results in oil displacement tests by water without sorbent. A higher percentage of oil (about 50%) was displaced by water under higher oil saturation conditions than that observed under lower oil saturation conditions (about 30%). To summarize, the results demonstrate that the effect of increased water table level inhibits sorption effectiveness, while the effect of increased oil contamination and sand particle size significantly increase sorption effectiveness. The position of the water table is the most dominant factor influencing oil-wicking effectiveness of the bagasse sorbent, followed by sand particle size and degree of oil contamination.

2.4 Moisture sorption and effectiveness

Figure 4 shows the amount of sorbate (oil and water) in the sorbent layer at final sampling in comparison with the amount of oil applied at time zero with regard to different microcosm conditions at 25% oil saturation. The amount of sorbate increases significantly when the sorbent layer is covered by water, compared to when the oiled-sand layer is covered and when the clean sand layer is covered. Furthermore, the amount of sorbate exceeds that of the initially applied oil. Thus, water sorption significantly contributes to an increase in the weight of the sorbent layer. As stated earlier, more saturated environments result in less oil wicking effectiveness, likely due to more water sorption. To support this idea, moisture sorption by the bagasse under each condition was estimated by optimizing the mass balances of each layer (a clean sand layer, an oiled sand layer, and a sorbent layer) and material balances (sand, oil, water, and sorbent) using Microsoft Excel Solver. The following assumptions were made in the calculations: (1) oil is composed of GC-resolvable hydrocarbons and non-GC-resolvable hydrocarbons. (2)

Table 2 Hierarchical regression parameters for effectiveness of sorbent under different water levels

Water table level	Coefficient	Models			
		$y = a_0$	$y = a_0 + a_1x_1$	$y = a_0 + a_2x_2$	$y = a_0 + a_1x_1 + a_2x_2$
On top of clean sand layer	a_0	–	73.6	**54.9**	54.3
	a_1	–	0.012	–	0.012
	a_2	–	–	**0.291**	0.291
	R^2	–	0.1	96.2	96.3
On top of oiled-sand layer	a_0	–	41.9	34.7.	**18.7**
	a_1	–	0.321	–	**0.321**
	a_2	–	–	0.349	**0.349**
	R^2	–	28.5	60.4	88.9
On top of sorbent layer	a_0	**6.6**	6.77	5.54	5.68
	a_1	–	–0.0028	–	–0.0028
	a_2	–	–	0.0164	0.0164
	R^2	–	0.4	27.6	28.0

Bold numbers were used as coefficients of models to predict effectiveness of sorbent in different water table levels.

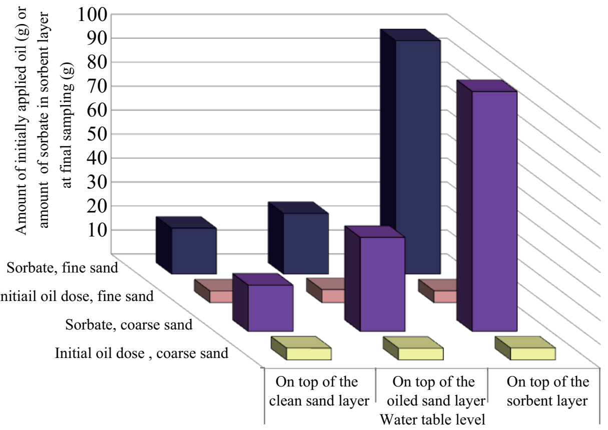

Fig. 4 Amount of oil applied in oiled-sand layer at time zero and the amount of sorbate (oil and water) recovered in sorbent layer at final sampling from microcosms constructed with 25% oil saturation, different sand particle sizes, and different water table levels.

Estimated mass of non-GC resolvable hydrocarbon and water are equal to or greater than zero. (3) Mass closures for each layers and material balance of non-GC resolvable hydrocarbon are assumed to be 100%. (4) Material balance of water is equal or less than 100%. This balance is chosen as a target equation to be optimized. (5) Three different initial estimates (100% oil removal, 50% oil removal, and 0% oil removal from the oiled sand layer) were made.

Different initial guesses converged to an optimized solution in each case with a reasonable material balance (96%–100%) on water. Estimated solutions are expressed as estimated moisture content (C_{est}, %) as Eq. (3):

$$C_{\mathrm{est}} = \frac{m_{\mathrm{cal}}}{m_{\mathrm{st}}} \times 100\% \tag{4}$$

where, m_{cal} (g) is the calculated mass of moisture; m_{st} (g) is the mass of wet sorbent after 3 months.

Estimated moisture contents and the corresponding effectiveness of sorbent are illustrated in Fig. 5. Oil-wicking effectiveness of sorbent tends to decrease as moisture content increases at a fixed sand particle size. Effectiveness reaches maximum when estimated moisture sorption is below 40%, while effectiveness falls below 10% when moisture sorption is about 80%. Effectiveness of sorbent from coarse sand is higher than from fine sand at a fixed moisture sorption level. Higher oil saturation promotes oil sorption and results in less water sorption. Overall, limitations of sorbent effectiveness by water sorption are evidenced in this analysis. Effectiveness of oil wicking was severely limited when ammoniated bagasse had approximately 80% of estimated moisture content, which was not observed by the LSU researchers because they assumed that ammoniated bagasse was saturated when it held 1 g water/g of dry sorbent (50% moisture content). Accordingly, methods to obtain a higher oil wicking effectiveness by reducing moisture sorption are suggested: (1) use of a more hydrophobic sorbent to capture more oil and less water in saturated environments, and (2) use of sorbent to remediate inter-tidal wetlands where the water level changes periodically.

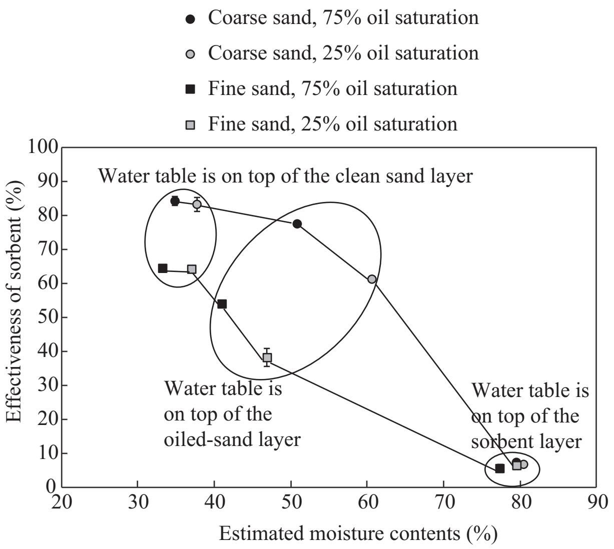

Fig. 5 Estimated moisture content vs. effectiveness of sorbent under different microcosm conditions.

3 Conclusions

This study provides new information on the use of ammoniated bagasse, which was not reported by de Silva (1995) and Breitenbeck and Grace (1997), who first suggested the use of ammoniated bagasse in remediation of oil-contaminated wetlands. In this study, we proposed that oil wicking is the most important step to determine effectiveness of this remediation process. The results indicate that the effectiveness of oil wicking, a possible biostimulation method, increased with higher oil contamination, larger sand particle size, and lower water coverage. Oil wicking was severely limited when water covered the sorbent layer. Water coverage was the most dominant factor limiting the effectiveness of wicking. Higher water coverage resulted in higher moisture sorption, which played a significant role in limiting effectiveness. The results suggested that higher moisture adsorption by ammoniated bagasse at higher water coverage caused limitation in oil wicking, and this was verified by the estimation of moisture content.

The implications of these findings are important for the proper use of this sorbent for the remediation of oil-contaminated wetlands. This cleanup method would be suitable for treating oil spills in sensitive wetland ecosystems where lack of oxygen and nutrients would limit natural attenuation. The effectiveness of this technique would greatly increase in contaminated environments with high oil contamination, large sand particle size, and low tides. Once this sorbent is applied on the surface of contaminated wetlands, it will immediately reduce the impact on the ecosystem and prevent further contamination. If the oil is successfully wicked out of the sediment to the surface where oxygen is available, successful restoration would likely be achieved through rapid aerobic biodegradation of hydrocarbons without nutrient deficiency problems. Furthermore, application of this technique can be accomplished with minimal damage to ecosystems since the addition of sorbent is possible without significant trampling, leaving no permanent residue.

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References

Aarnio T, Martikainen P J, 1995. Mineralization of C and N and nitrification in scots pine forest soil treated with nitrogen fertilizers containing different proportions of urea and its slow-releasing derivative, ureaformaldehyde. *Soil Biology and Biochemistry*, 27(10): 1325–1331.

Annunciado T R, Sydenstricker T H D, Amico S C, 2005. Experimental investigation of various vegetable fibers as sorbent materials for oil spills. *Marine Pollution Bulletin*, 50(11): 1340–1346.

Breitenbeck G A, Grace B, 1997. Use of ammoniated cellulosic materials for remediation of oil-contaminated wetlands. Michael Holiday & Associates. Louisiana Applied Oil Spill Research and Development Program, OSRADP Technical Report Series, 96–001.

Brietenbeck G A, Kember K, 2003. Process for ammoniating cellulosic materials. US Patent 6548659.

de Laune R D, Gambrell R P, Pardue J H, Patrick W H, 1990. Fate of petroleum hydrocarbons and toxic organics in Louisiana coastal environments. *Estuaries and Coasts*, 13(1): 72–80.

de Silva A P, 1995. Ammoniation of cellulosic materials for use in fertilization and oil spill remediation. Ph.D. Thesis. University of Louisiana. Baton Rouge, LA.

Deschamp G, Caruel H, Borredon M -E, Bonin C, Vignoles C, 2003. Oil removal from water by selective sorption on hydrophobic cotton fibers. 1. Study of sorption properties and comparison with other cotton fiber-based sorbents. *Environmental Science and Technology*, 37(5): 1013–1015.

Gandee J, 2007. Testing the ability of sorbents to wick oil from subsurface wetland to simulate aerobic biodegradation. M.S. Thesis, University of Cincinnati, Cincinnati, OH.

Goodin D A, Hudnall W H, 2001. Use of ammoniated bagasse to remediate petroleum contaminated wetland and forest soils. In: 8th International Petroleum Environmental Conference. Houston, TX.

Grace B, 1999. Use of N-rich materials for the *in situ* remediation of wetlands contaminated with crude oil. M.S. Thesis, Louisiana State University, Baton Rouge, LA.

Hoff R Z, Shigenaka G, 1993. Salt marsh recovery from a crude oil spill: vegetation, oil weathering, and response. In: Proceedings of the 1993 International Oil Spill Conference, American Petroleum Institute, Washington. 307–311.

Inagaki M, Kawahara A, Konno H, 2004. Recovery of heavy oil from contaminated sand by using exfoliated graphite. *Desalination*, 170(1): 77–82.

Mendelssohn I A, Hester M W, Sasser C, Fischel M, 1990. The effect of a Louisiana crude oil discharge from a pipeline break on the vegetation of a southeast Louisiana brackish marsh. *Oil & Chemical Pollution*, 7(1): 1–15.

Mitsch W J, Gosselink J G, 1986. Wetlands. Van Nostrand Reinhold Company Inc., New York. 393–414.

Office of Technology Assessment, 1991. Bioremediation for Marine Oil Spills. Congress of the United States, Office of Technological Assessment, Congressional Board of the 102nd Congress, Washington, DC.

Olivieri R, Bacchin P, Robertiello A, Oddo N, Degen L, Tonolo A, 1976. Microbial degradation of oil spills enhanced by a slow-release fertilizer. *Applied Environmental Microbiology*, 31(5): 629–634.

Ollivier B, Magot M, 2005. Petroleum Microbiology. American Society for Microbiology Press, Washington, DC.

Venosa A D, Lee K, Suidan M T, Garcia Blanco S, Cobanli S, Moteleb M et al., 2002. Bioremediation and biorestoration of a crude oil contaminated freshwater wetland on the St. Lawrence River. *Bioremediation Journal*, 6(3): 261.

Webb J W, Alexander K S, Winters J K, 1985. Effects of autumn application of oil on Spartina alterniflora in Texas salt marsh. *Environmental Pollution*, 38(4): 321–337.

Zhu X, Venosa A D, Suidan M T, 2004. Literature review on the use of commrecial bioremediation agents for cleanup of oil-contaminated eustarine environments. Report under a contract with Office of Research and Development, U.S. Environmental Protection Agency. http://www.epa.gov/oem/docs/oil/edu/litreviewbiormd.pdf.

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