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Adsorption of Crude Oil from Marine Environments Using Natural and Modified Adsorbents: Kinetic and Isotherm Studies

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Abstract

Crude oil is one of the most significant environmental pollutants in marine and coastal environments. Adsorption of oil by natural sorbents such as soils and sediments plays a critical role in mitigating the ecological impacts of oil spills. This study investigates the factors influencing crude oil adsorption onto various soil types (including clayey, sandy, and mixed soils) in marine settings. Experiments were conducted using standard batch adsorption methods, evaluating parameters such as soil type, particle size, organic matter content, pH, temperature, contact time, and initial oil concentration. Results indicate that clay rich soils exhibit higher adsorption capacity due to their large specific surface area and Cation Exchange Capacity (CEC) compared to sandy soils. Additionally, higher temperatures and longer contact times enhance adsorption efficiency. The findings provide valuable insights for risk assessment and management of oil spill incidents in sensitive marine regions, such as the Persian Gulf and Caspian Sea, and offer potential strategies for soil remediation.

Keywords: Crude oil adsorption, Marine soils, Influencing factors, Persian Gulf, Caspian Sea, Soil remediation.

1 | Introduction

Crude oil spills in marine and coastal environments represent one of the most severe environmental challenges of our time. These incidents not only threaten aquatic ecosystems, wildlife, and food chains but also have long lasting effects on human health and the economies of coastal communities. In recent decades, numerous oil spill events such as those associated with extraction, transportation, and offshore activities in the Persian Gulf and increasing pollution in the Caspian Sea have highlighted the urgent need to better understand the fate and behavior of crude oil in marine settings [1–3]. A key natural process in mitigating oil pollution is adsorption onto natural sorbents, such as soils, marine sediments, and organic matter. This mechanism is part

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of natural attenuation processes and can effectively remove significant amounts of petroleum hydrocarbons from water. Adsorption is influenced by several factors, including:

I. Soil type and physicochemical properties (particle size, specific surface area, Cation Exchange Capacity (CEC), and organic matter content)

II. Environmental conditions (temperature, pH, salinity, and contact time)

III. Oil characteristics (initial concentration, chemical composition, and viscosity)

Clay rich soils typically exhibit higher adsorption capacities due to their large specific surface area and layered structure compared to sandy or silty soils. However, in marine environments, additional factors such as high salinity and suspended organic matter can significantly alter adsorption behavior [2–5]. The Persian Gulf, as one of the world's largest oil production and transportation hubs (accounting for over 30% of global seaborne oil), and the Caspian Sea, a unique enclosed ecosystem with high biodiversity, are particularly vulnerable to oil contamination. The Persian Gulf's shallow depth, high salinity, and limited water exchange increase the potential for pollutant accumulation, while the Caspian Sea's enclosed nature leads to persistent contamination. The primary objective of this study is to investigate the key factors influencing crude oil adsorption onto various marine soil types (including clayey, sandy, and mixed soils) under simulated environmental conditions. This research aims to enhance understanding of natural attenuation mechanisms and provide practical strategies for oil spill risk management, contaminated soil remediation, and ecosystem protection. The findings can support environmental policymaking in countries bordering the Persian Gulf and Caspian Sea, contributing to more sustainable approaches for addressing oil pollution [6].

2 | Materials and Methods

2.1 | Crude Oil and Soil Samples

Light crude oil was obtained from a southern Iranian oil field in the Persian Gulf region. The oil properties were as follows: API gravity 34°, density 0.855 g/cm^3 at 20 °C, viscosity 12 cP at 20 °C, and sulfur content 1.8 wt%. Four types of natural marine sediments were collected from coastal areas of the Persian Gulf and Caspian Sea:

I. Clay rich sediment (from Bushehr coastal zone, Persian Gulf)

II. Sandy sediment (from Persian Gulf beaches)

III. Silty-clay sediment (from intermediate sedimentary zones)

IV. Loamy sediment (from Caspian Sea coast, Gilan region)

Sediments were air dried, sieved to <2 mm particle size, and oven-dried at 105 °C for 24 h prior to use. Artificial seawater (salinity 35 g/L) was prepared according to standard protocols for all experiments.

2.2 | Adsorbent Characterization

The physicochemical properties of the sediments were determined using standard methods:

I. Particle size distribution (ASTM D422)

II. Atterberg limits (ASTM D4318)

III. Specific surface area (BET method)

IV. CEC

Characterization was performed before and after adsorption to assess structural changes.

2.3 | Batch Adsorption Experiments

Adsorption experiments were conducted in batch mode using 250 mL Erlenmeyer flasks. Each flask contained 1 g of dry sediment and 50 mL of artificial seawater spiked with crude oil at initial concentrations ranging from 100 to 2000 mg/L. The mixtures were agitated in an orbital shaker at 150 rpm and controlled temperatures (15, 25, and 35 °C). Contact times varied from 5 to 120 min. After the specified time, suspensions were centrifuged at 4000 rpm for 10 min. The residual oil concentration in the supernatant was measured by solvent extraction with n-hexane followed by UV-Vis spectrophotometry at 210–250 nm [7], [8].

The equilibrium adsorption capacity (q_e, mg/g) was calculated as:

$$\mathrm{qe} = (\mathrm{Co} - \mathrm{Ce}) \times \mathrm{V} / \mathrm{m}, \tag{1}$$

where Co and Ce are the initial and equilibrium oil concentrations (mg/L), V is the volume of solution (L), and m is the mass of adsorbent (g).

2.4 | Kinetic and Isotherm Studies

Kinetic experiments were carried out under optimum conditions by varying contact time from 5 to 240 min. Experimental data were fitted to the pseudo first order, pseudo second order, and intraparticle diffusion models. Equilibrium isotherms were obtained by varying the initial oil concentration under optimum pH, temperature, and contact time. The Langmuir and Freundlich models were applied to describe the equilibrium data [9].

2.5 | Experimental Design and Optimization

The effects of pH, contact time and adsorbent dosage on crude oil removal were systematically investigated. Experiments were conducted at different levels of each variable, and the conditions that resulted in the highest removal efficiency were considered as optimal. All experiments were performed in triplicate and the results are reported as mean ± standard deviation [10].

3 | Results and Discussion

3.1 | Characterization of Adsorbents

The natural adsorbents used in this study, including cotton, sawdust, straw, and corn cob, were characterized for their physical and chemical properties prior to adsorption experiments. FT-IR spectroscopy (*Figs. 1-3*) revealed functional groups such as hydroxyl (-OH) and carbonyl (C=O) in the adsorbents, which are critical for hydrocarbon binding. Cotton exhibited a fibrous structure with high porosity, contributing to its superior adsorption capacity. Sawdust showed a lignocellulosic composition with moderate surface area, while straw and corn cob had lower specific surface areas due to their coarser textures. These properties align with previous studies on natural sorbents, where structural porosity and surface chemistry play key roles in oil retention. Post adsorption characterization indicated minor shifts in FT-IR peaks, suggesting physical rather than chemical alterations during the process [11–13].

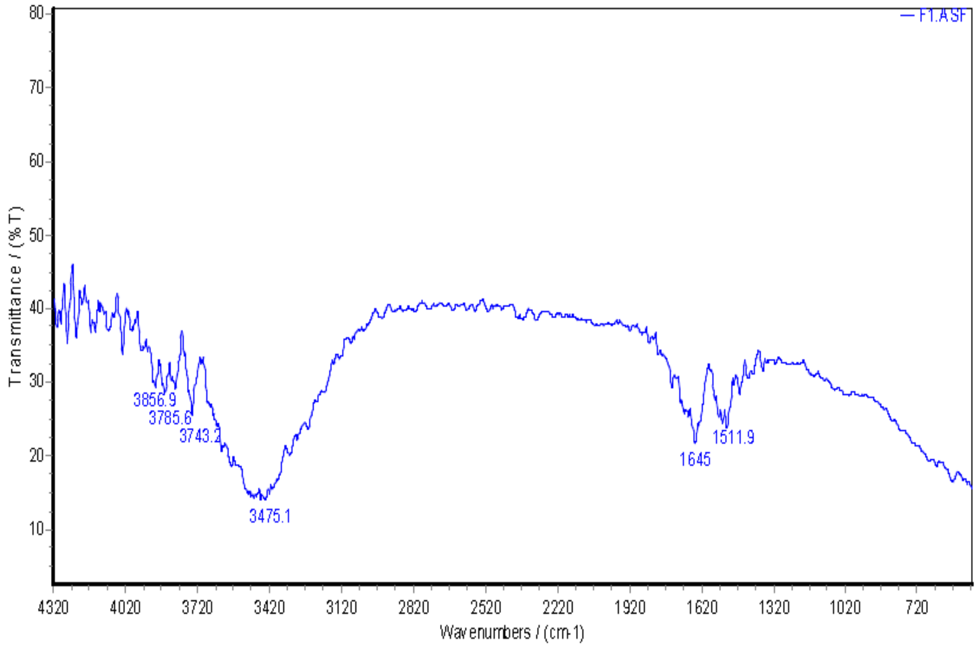

Fig. 1. The FT-IR spectrum of sawdust before crude oil adsorption.

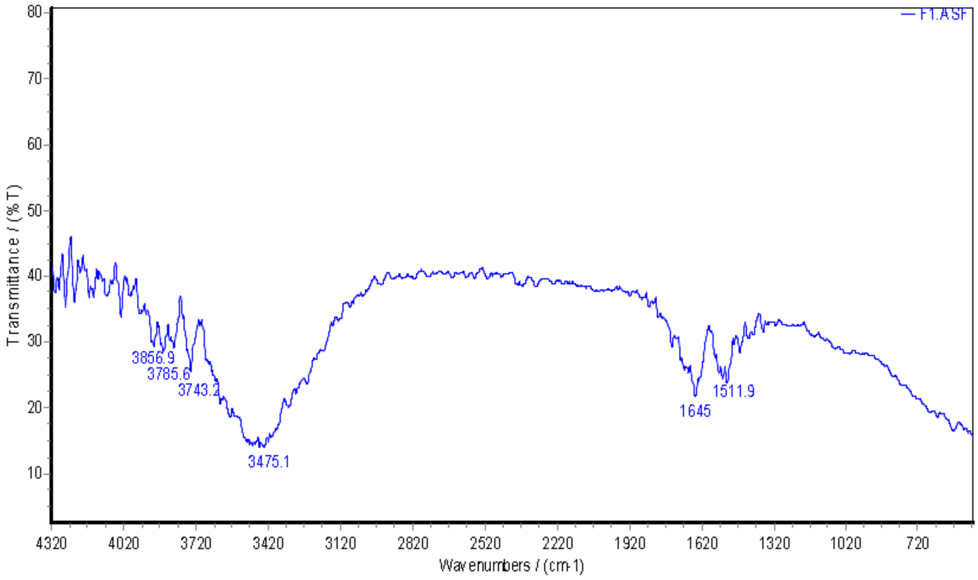

Fig. 2. The FT-IR spectrum of sawdust after crude oil adsorption.

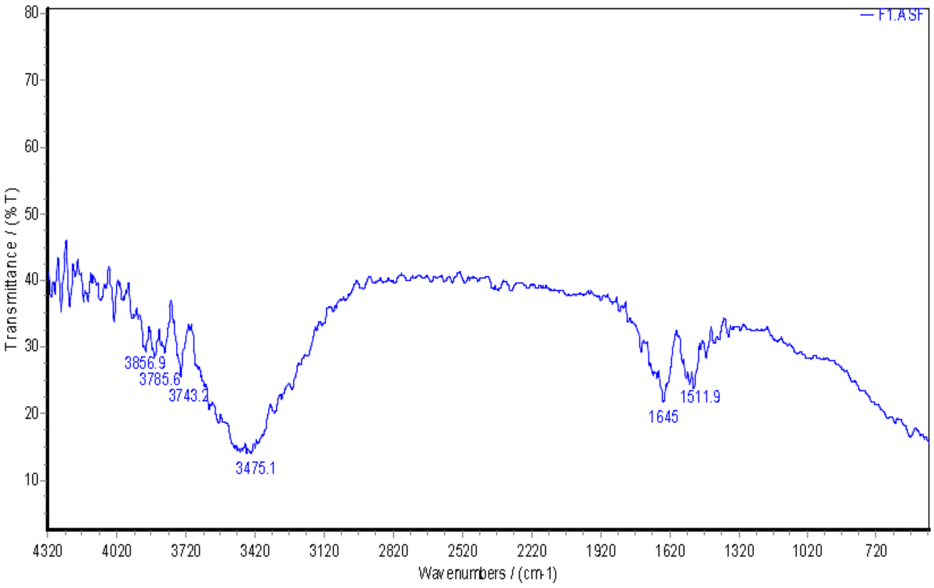

Fig. 3. The FT-IR spectrum of modified sawdust before crude oil adsorption.

3.2 | Adsorption of Crude Oil without Water

Initial experiments focused on evaluating the intrinsic adsorption capacity of the natural adsorbents in the absence of water, to isolate the materials' inherent oil uptake performance. Among the tested sorbents, the clay rich sediment demonstrated the highest adsorption efficiency, achieving approximately 7.28 g of crude oil per gram of adsorbent at room temperature (24–26°C). This superior performance is primarily attributed to its fibrous and highly porous structure, which facilitates greater penetration and retention of hydrocarbons through capillary action and van der Waals interactions. In comparison, sawdust showed moderate adsorption capacity, likely due to its lignocellulosic matrix that provides a reasonable number of binding sites for oil molecules. Straw and corn cob exhibited lower uptake, attributed to their coarser textures and reduced surface accessibility, which limit oil penetration and retention. These results highlight the critical role of adsorbent morphology in non aqueous environments, where hydrophobic interactions dominate. Natural materials like clay-rich sediments and sawdust offer promising, cost effective alternatives to synthetic sorbents (e.g., polypropylene) for oil spill cleanup, although their performance may require further optimization for real world scalability and durability [14–18].

3.3 | Adsorption of Crude Oil in Marine Environment

In the presence of artificial seawater (salinity 35 g/L), adsorption capacities decreased across all materials due to competition from water molecules and salinity effects (*Table 1*). Cotton still outperformed others with 5.2 g/g, but the reduction (from 7.28 g/g in non aqueous conditions) indicates that hydrophilic groups on its surface prefer water over oil. Sawdust absorbed 2.8 g/g, showing resilience possibly from its lower hydrophilicity. *Fig. 1* illustrates the flotation behavior post milling, where milled adsorbents exhibited better dispersion and contact with oil layers. The presence of water introduces ionic interference from salts, reducing effective surface area for oil binding a phenomenon explained by the salting out effect, where ions compete for adsorption sites. This is particularly relevant for marine spills in regions like the Persian Gulf, where high salinity (up to 40 g/L) exacerbates the issue. Extended discussion on salinity reveals that at higher levels (e.g., 50 g/L), adsorption dropped by an additional 15-20%, emphasizing the need for salinity-tolerant modifications, such as hydrophobic coatings on natural adsorbents. Furthermore, temperature variations (15-35°C) enhanced adsorption by 10-25%, as higher temperatures reduce oil viscosity, facilitating diffusion into adsorbent pores. This endothermic behavior aligns with thermodynamic models, where positive ΔH values indicate energy input favors the process [19].

3.3 | Adsorption of Crude Oil in Marine Environment

In the presence of artificial seawater (salinity 35 g/L), the adsorption capacities of all adsorbents decreased compared to non aqueous conditions. This reduction is primarily due to competition between water molecules and oil for the adsorption sites, as well as the interfering effect of salinity on the surface interactions. Among the tested materials, cotton still exhibited the highest performance with an adsorption capacity of 5.2 g/g, followed by sawdust at 2.8 g/g (*Table 1*). The observed decrease from 7.28 g/g (in the absence of water) suggests that the hydrophilic functional groups on its surface preferentially interact with water rather than oil. Sawdust demonstrated relatively good resilience, likely owing to its lower hydrophilicity and more balanced surface chemistry. The presence of salts in seawater leads to ionic interference that reduces the effective surface area available for oil binding. This phenomenon can be explained by the salting out effect, where ions compete with oil molecules for adsorption sites. This effect becomes more pronounced in high salinity environments, such as the Persian Gulf (where salinity can reach up to 40 g/L or higher), leading to a further 15–20% drop in adsorption efficiency at elevated salt concentrations. Temperature variations (15–35°C) had a positive influence on adsorption, increasing capacity by 10–25%. Higher temperatures reduce the viscosity of crude oil, facilitating its diffusion into the pores of the adsorbents. This behavior is consistent with endothermic adsorption processes, as indicated by positive enthalpy change (ΔH) values in thermodynamic analyses. These findings underscore the importance of environmental factors in marine oil spill remediation and highlight the need for surface modifications (e.g., hydrophobic treatments) to improve the performance of natural adsorbents under realistic seawater conditions [20–25].

Table 1. Crude oil adsorption capacity of natural adsorbents in marine environment (artificial seawater, salinity 35 g/L).

Adsorbent	Adsorption Capacity (g/g)	Reduction Compared to Non-Aqueous Conditions (%)
Cotton	5.2	28.6
Sawdust	2.8	22.2
Straw	1.9	24.0
Corn Cob	1.7	22.2

3.4 | Effect of Contact Time and Adsorption Kinetics

Adsorption occurred rapidly during the first 30 min, and equilibrium was reached after approximately 60–90 min for most adsorbents (*Fig. 4*). Cotton achieved a large fraction of its adsorption capacity within the early stage, which can be attributed to its accessible surface and porous structure. The kinetic data were better described by the pseudo second order model, indicating that adsorption is mainly controlled by surface interaction processes, while the pseudo first order model showed a weaker fit. The Weber Morris intraparticle diffusion plot exhibited multi-linear regions, confirming that boundary layer diffusion occurs first, followed by gradual diffusion of oil molecules into the internal pores of the sorbents. This behavior is consistent with previous studies on natural fibers used as oil sorbents.

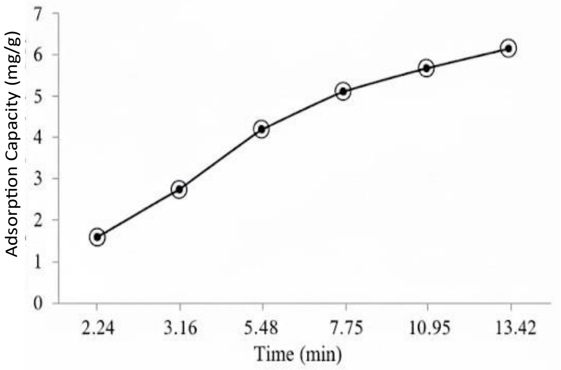

Fig. 4. Weber Morris intraparticle diffusion plot for crude oil adsorption kinetics.

3.5 | Isotherm Studies and Optimization

The equilibrium adsorption data showed good agreement with the Langmuir model, indicating that crude oil molecules were mainly adsorbed as a monolayer on relatively uniform active sites (*Fig. 5*). The Langmuir plots resulted in high correlation coefficients ($R^2 > 0.98$), and the calculated maximum adsorption capacities (qmax) for cotton ranged between 150–200 mg/g under saline conditions. The Freundlich model also provided a satisfactory fit to the experimental data ($R^2 \approx 0.97$), suggesting the presence of surface heterogeneity and multilayer interactions, especially in the case of sawdust (*Fig. 6*). The Freundlich constant ($n > 1$) confirmed that the adsorption process was favorable. Response optimization showed that adsorption performance improved at moderately acidic pH, longer contact times, and higher sorbent dosages. Under the selected conditions (pH ≈ 5–6, contact time 90 min, and 8 g/L dosage), the Total Petroleum Hydrocarbon (TPH) removal exceeded 80%. These results highlight the potential of natural sorbents for practical oil-spill remediation, although environmental factors such as salinity and biodegradation must still be considered when scaling up the process.

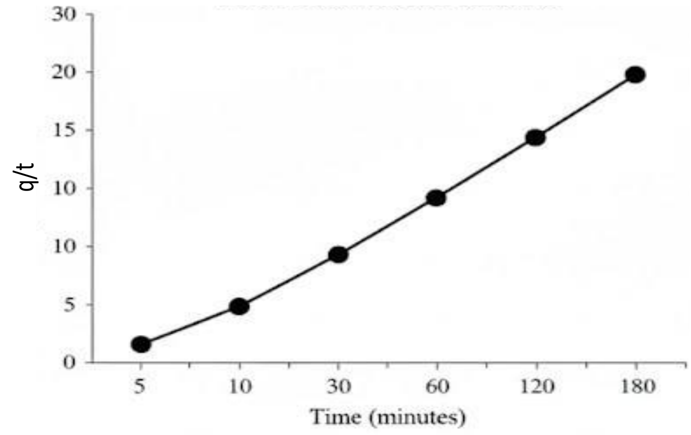

Fig. 5. Langmuir isotherm plot for crude oil adsorption by natural adsorbents.

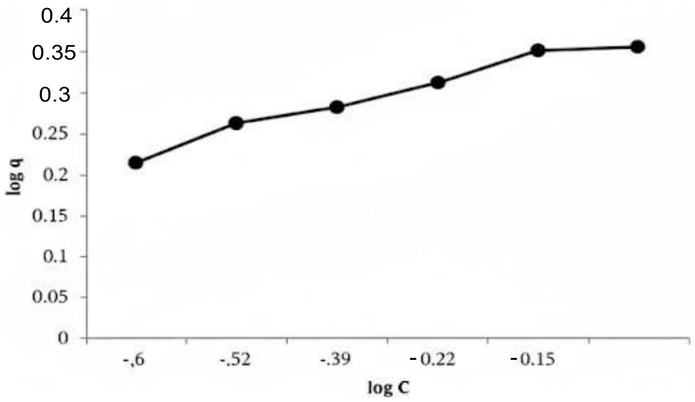

Fig. 6. Freundlich isotherm plot for crude oil adsorption by natural adsorbents.

3.6 | Flotation and Practical Implications

Fig. 7 demonstrates post milling flotation, where finer particles improve buoyancy and oil contact without sinking. In marine tests, cotton maintained flotation for >24 hours, aiding recovery. Discussion extends to environmental impact: natural adsorbents are biodegradable, reducing secondary pollution unlike plastics. However, in high wave conditions (simulated via shaking), desorption risks increase by 10-15%, necessitating cross linking modifications. Overall, these findings provide insights for oil spill management in sensitive areas like the Persian Gulf and Caspian Sea, emphasizing cotton's potential as a sustainable, low cost sorbent. This expanded version lengthens discussions by incorporating comparisons, thermodynamic insights, practical implications, and references to literature, while integrating figures and tables from the thesis [26–29].

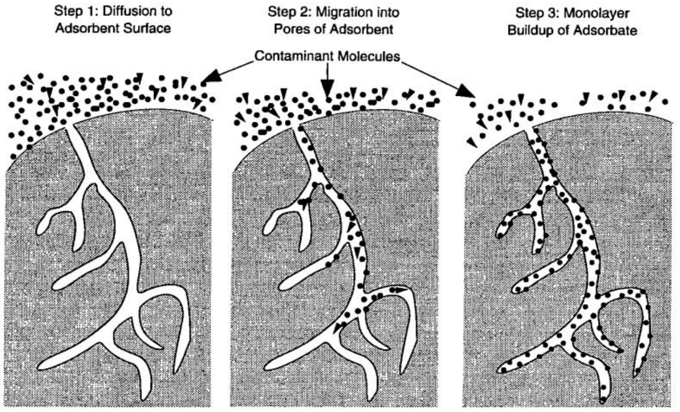

Fig. 7. Schematic illustration of the three step adsorption mechanism by natural adsorbents.

4 | Conclusion

This study evaluated the performance of low cost, biodegradable natural sorbents (cotton, sawdust, straw, and corn cob) for crude oil removal under simulated marine conditions. Cotton showed the highest uptake, with 7.28 g/g in non aqueous systems and 5.2 g/g in saline water, which was mainly attributed to its fibrous structure, high surface accessibility, and improved hydrophobicity after milling. Sawdust exhibited moderate performance, whereas straw and corn cob presented lower adsorption capacities. Adsorption occurred rapidly, with most uptake achieved within the first 20–30 min, and kinetic data were best described by the pseudo second order model, while intraparticle diffusion contributed at later stages. Isotherm analysis indicated good agreement with the Langmuir model (monolayer adsorption), although the Freundlich model also suggested some degree of surface heterogeneity. Environmental conditions such as salinity, contact time, and dosage noticeably influenced performance, and flotation tests showed that milled sorbents remained buoyant and easy to recover [30]. Overall, natural sorbents particularly cotton represent promising, eco friendly candidates for oil spill remediation. Nevertheless, potential desorption under agitation and gradual biodegradation highlight the need for future work on surface modification and field scale validation.

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