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Assessment of Cleanup Needs of Oiled Sandy Beaches: Lessons from the *Prestige* Oil Spill

ANA M. BERNABEU,*,† DANIEL REY,† BELÉN RUBIO,† FEDERICO VILAS,† CARMEN DOMÍNGUEZ,‡ JOSEP M. BAYONA,‡ AND JOAN ALBAIGÉS‡

GEOMA, Marine and Environmental Geology Group, Department of Marine Geosciences, Universidad de Vigo, 36310 Vigo, Spain, and Department of Environmental Chemistry, CID-CSIC, Jordi Girona, 18-26, 08034 Barcelona, Spain

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Surveys of the oiled sandy beaches along the northern Atlantic coast of Spain, 2–5 years after the *Prestige* oil spill of November 2002, have provided new evidence regarding buried fuel and its behavior. The persistence and depth of burial of oil, and the capacity of the beach for natural regeneration, depend on beach morphodynamics, which drive a sequence of physicochemical processes that reduce subsurface tar balls to highly divided oil forms while also allowing appreciable weathering despite burial. These findings prompted reassessment of current spill evaluation strategies. A protocol is proposed that combines the modeling of beach morphodynamics, an environmentally friendly coring survey, and well-calibrated hydrocarbon analysis.

Introduction

The costs of cleanup following coastal pollution by oil spills can be huge. In 2007, the International Oil Pollution Compensation Funds paid out 31,872,606 euros to cover spill treatment activities, 25% of all compensation outlay (*1*), and some 80–90% of cleanup costs derive from shoreline activities. On average, shoreline cleanup costs range from $5 to $111 per square meter, depending on the intensity of oiling (*2, 3*). To maximize the efficiency of cleanup investment, the continuous evaluation of spill assessment methods and response decision strategies with a view to optimal deployment of remediation technologies should be as much a priority as the improvement of remediation technologies themselves.

The efficiency of cleanup efforts depends to a considerable extent on the swift and accurate estimation of the extent of oiling (*4, 5*). On beaches, assessment targets both surface and subsurface oil. Surface oil can reliably be detected by aerial survey and direct inspection on foot; efficient cleanup strategies for this type of pollution, such as manual or mechanical removal and relocation of polluted sediment, are well-established and regularly employed. Assessment of subsurface oiling is, or should be, more complex. The currently common practice of estimating the affected area by digging pits and trenches no more than about 50 cm deep—the rationale for which is limitation of the environmental impact of the evaluation procedure itself—ignores deeper deposits. This can not only result in underestimation of the extent of pollution, but also deprives the evaluator of information on the fate of the stranded oil.

Two basic mechanisms of subsurface pollution of beaches by oil are described in the literature: penetration and burial (*6, 7*). The former consists in the downward percolation of oil to a depth of about 40 cm, depending on sand or sediment texture and oil type. This mechanism, which results in low-level subsurface oiling, is typical on beaches composed of gravel or coarse sand; it was the main mechanism of subsurface pollution in the *Exxon Valdez* spill (*8*), and since then most of the studies, i.e., see Owens et al. (*9*), have assumed that it has also been the main mechanism of subsurface pollution of coarse beaches in other spills, and cleanup methods have been designed accordingly.

The second mechanism of subsurface pollution is the burial of oil by the action of beach dynamics (*7*), which can result in intense oiling at depths of up to several meters. This was the main subsurface pollution mechanism after the *Prestige* oil spill (POS) of November 2002, which affected more than 500 sandy beaches along the Atlantic coast of Spain (*10*). In general, cleanup manuals (*11, 12*) underestimate the importance of deeply buried oil in this kind of beaches, particularly where it is unlikely that the oil would surface during the tourist season. However, deep oil is very persistent because degradation slows down with increasing depth (*13–15*), and in some cases may result in significant long-term toxicity (*16–18*).

This paper describes new findings from follow-up of POS-affected beaches that suggest ways to improve current spill assessment strategies for sandy beaches. Specifically, we envisage that a combination of morphodynamics modeling, environmental-friendly core-based beach sampling, and well-calibrated hydrocarbon analyses would allow both estimation of the likelihood of oil burial during the early stages of an oil spill, and evaluation of the persistence of buried oil.

Materials and Methods

Fieldwork. We studied eight Spanish beaches oiled by the POS (Figure 1): three in the Basque Country (N Spain; Bakio, Laida, and Laga, all sampled in February 2005) and five on the west coast of Galicia (NW Spain; Nemiña, O Rostro, Arnela, Mar de Fora, and Figueiras, variously sampled in January 2004, March 2005, June 2006, and May 2007). The most affected beach examined in January 2004 (*7*), Nemiña, was sampled yearly.

Sampling was performed by extracting suction cores 5 cm in diameter along profiles representative of the targeted beaches (Figure S1). Generally, three cores were taken from each profile, one each from the low, mid, and high intertidal zones. Samples were taken at ebb spring tide to ensure the widest possible coverage of the intertidal zone. In all, 66 cores varying in length from 0.70 to 3.74 m were taken. The general characteristics of beaches and cores are described in Table S1.

Core Extraction Device. A conventional suction corer, of the kind commonly used in sedimentological studies (*19, 20*), was modified for optimal sand recovery in the intertidal zone of beaches: in order to allow the recovery of coarser grain size, increase the amount of sample per cm (and hence the vertical resolution of analyses), shorten sampling time (and

* Corresponding author phone: +34 986 812005; fax: +34 986 812556; e-mail:bernabeu@uvigo.es.

† GEOMA, Marine and Environmental Geology Group, Department of Marine Geosciences, Universidad de Vigo.

‡ Department of Environmental Chemistry, CID-CSIC.

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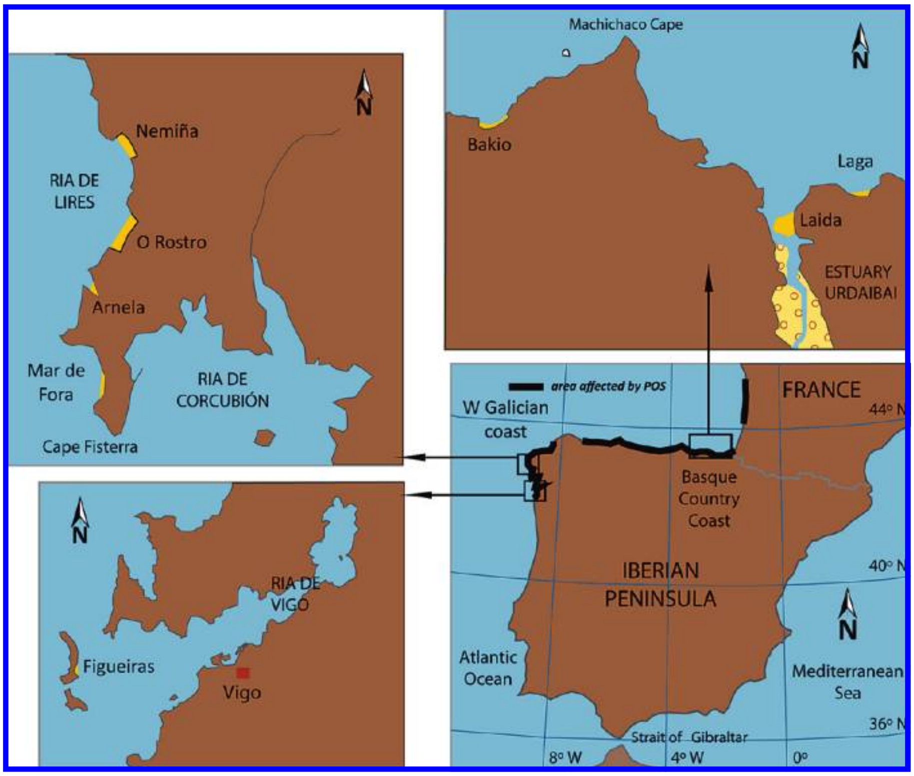

FIGURE 1. Coasts affected by the POS, with details showing the locations of the beaches studied.

hence increase the number of cores taken), and reach the full depth of the active beach profile, the pushing head was simplified and the core section was widened to enable the extraction of PVC-encapsulated sand cores up to 4 m long and 5 cm in diameter (see Figure S2 for full details). This beach-optimized suction corer (BOSC) is easily handled by three operators and reduces sample collection time to approximately 30 min per core, including extraction, cutting into standard 100 cm long sections, sealing, and marking.

Laboratory Characterization and Analysis. Once in the laboratory, cores were split in half, examined visually (note being taken of tar balls and gray sand horizons), and sampled for analysis at 10 cm intervals and in any gray sand horizons. Alternate samples were saved for textural and geochemical analysis. Grain size was measured by dry sieving. Elemental analysis by X-ray fluorescence determined both major and trace elements. Mineralogy was determined by X-ray diffractometry of representative cores from each beach. Total organic carbon (TOC) and $CaCO_3$ content were determined with a LECO analyzer (St. Joseph, MI). Selected samples were also examined by scanning electron microscopy (SEM) to inspect the oil coatings. The degree of analytical detail pursued varied according to beach morphodynamics, observations at the time of sample collection, and the visual appearance of the cores.

Hydrocarbon Analysis. Samples (6−7 g wet weight) were spiked with surrogates (naphthalene-d_8, anthracene-d_{10}, pyrene-d_{10}, benzo[a]pyrene-d_{12}, and benz[a]anthracene-d_{12}) and freeze-dried for 24 h. Water content was determined by weighing before and after drying. The freeze-dried samples were extracted with 1:1 hexane/acetone in a pressurized solvent extraction apparatus from Applied Separations (Allentown, PA): the sample was packed in an 11 mL stainless steel extraction cell between 1 g of Hydromatrix (bottom) and 4 g of washed sea sand (top) and was extracted in three 10 min cycles at 100 °C and 140 bar. The extraction cell was then flushed under pressure with solvent (5 s) and nitrogen (3 min). The recovered extracts were carefully evaporated to near-dryness and were then separated into aliphatic and aromatic fractions by adsorption chromatography on a 45 × 1.2 cm glass column packed with 1 g of anhydrous sodium sulfate (top), 6 g of neutral alumina (middle; activated at 400 °C, deactivated with 5% water), and 6 g of silica gel (bottom; activated at 120 °C, deactivated with 5% water). Aliphatic hydrocarbons were eluted with 20 mL of *n*-hexane, and polycyclic aromatic hydrocarbons were eluted with 50 mL of 80:20 *n*-hexane/dichloromethane. Each of these fractions was analyzed in a TRACE GC/MS apparatus (from Thermo-Finnigan, Manchester, UK) using the following: splitless injection (hexane, 280 °C; the purge valve was activated 50 s after the injection); helium carrier gas (1.2 mL/min); a 30 m × 0.25 mm i.d. column with a 0.25 μm coating of ZB-5MS as stationary phase (from Phenomenex Zebron, Torrance, CA); a column temperature program consisting of 1 min at 60 °C, a 10 °C/min ramp up to 200 °C, a 4.8 °C/min ramp up to 320 °C, and 10 min at 320 °C; transfer line and ion source temperatures of 250 and 200 °C, respectively; EI mode MS at 450 eV; data acquisition in full-scan mode from 50 to 350 amu at 10 scan/s, with 6 min of solvent delay; and data processing by Thermo-Finnigan X-calibur software.

Hydrodynamic and Morphodynamic Modeling. Simulated deep water wave climate data (wave height, period, and direction), generated using the WAM model, were obtained from the WANA database of the Spanish Ministry of Works Oceanographic Data Bank for WANA points 1042072 (43° N, −9.5 °W; Costa da Morte, Galicia), 1044069 (42.25 °N, −9 °W; Islas Cíes, Galicia), and 1070074 (43.5 °N, −2.5 °W, Bizkaia, Basque Country). Wave propagation from deep water to breaking point was modeled using MOPLA, the beach morphodynamics module of the coastal modeling system SMC (*21*), which is based on the REFDIF model (*22*).

Results

Sediment Characteristics. All the sediments studied consisted of medium-coarse sand (0.3−0.554 mm). Beach sands were moderately well sorted at Nemiña, O Rostro, Figueiras, and Laida, but mostly poorly sorted at the other beaches

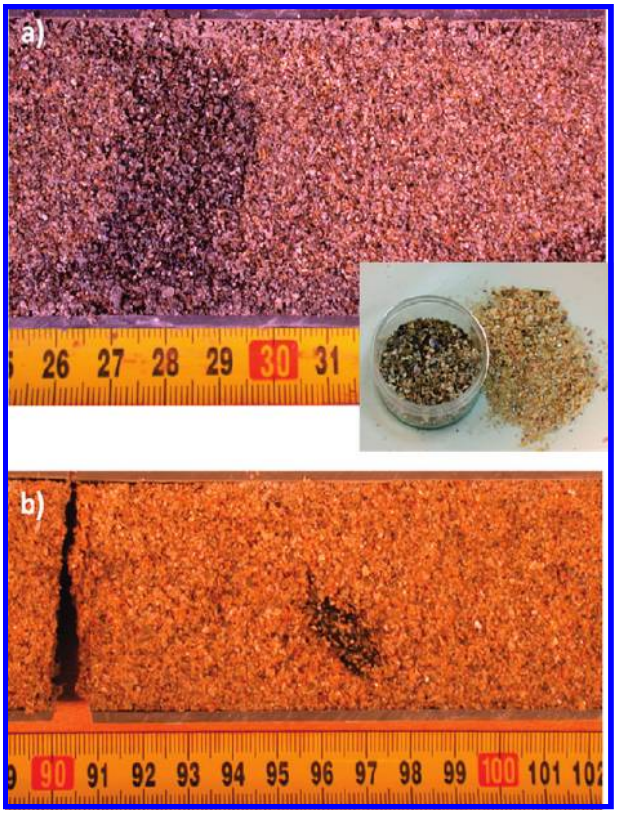

FIGURE 2. Segments of cores from (a) the low tidal zone of the northern part of Nemiña Beach, and (b) the low tidal zone of the southern part of O Rostro Beach. Both cores were collected in 2006, almost 4 years after the POS.

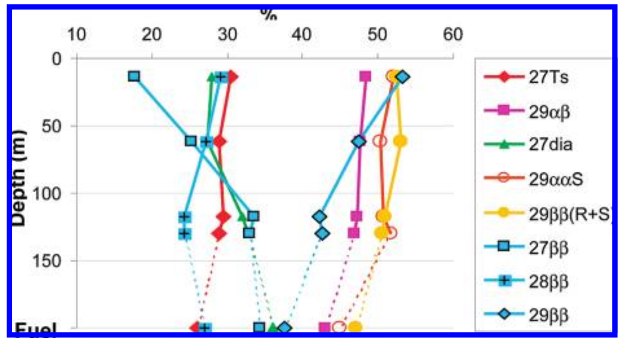

FIGURE 3. Vertical profiles of the sterane indices 27−29$\beta\beta$ and 29$\alpha\alpha$, and of the triterpane indices 27Ts and 29$\alpha\beta$, at Figueiras Beach. Indices are defined in Table S5.

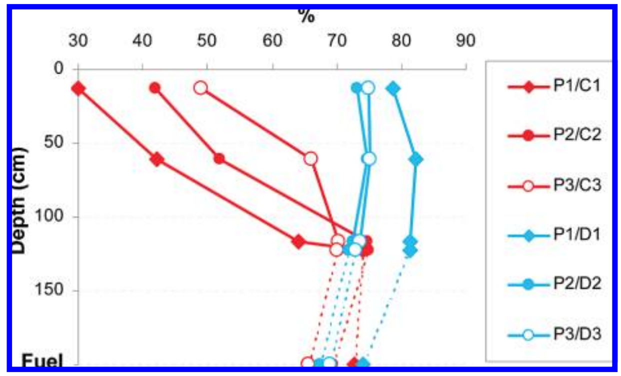

FIGURE 4. Vertical profiles of polycylic aromatic hydrocarbon ratios at Figueiras Beach. P1−P3, alkylphenanthrenes; D1-D3, dibenzothiophenes; C1−C3, chrysenes (see Table S5).

(Table S2). $CaCO_3$ content ranged from 19.7% to 62.0%. TOC content was relatively low (0.2−0.6%), as expected for sandy beaches.

Subsurface Oil. *Visual Inspection.* All Galician beaches presented buried oil with a discontinuous depth distribution (Table S3). Oil exhibited distinct morphologies, from tar balls to grain coatings (Figure 2). At Nemiña, oil was found in 10 of the 11 cores taken in 2004 (most abundantly in cores from the central and northern areas of the beach), and persisted in 2005, 2006, and 2007.

Depth of burial was variable. Isolated tar balls were found at depths of between 4 cm (indicative of a short-term process) and more than 1 m; tarball-rich levels were usually associated with high concentrations of small tar-balls (mm). The longest core obtained (3.74 m) had significant amounts of oil-coated sand below 3 m of apparently clean sand. Oil-coated sand layers up to 1 m thick were common.

The annual monitoring at Nemiña showed changes in buried oil morphology. The predominant oil form evolved from tar balls of several centimeters in diameter, to tar balls smaller than 1 mm, to coatings; while variations in groundwater level spread the oil vertically, giving rise to gray horizons of coated sand in association with significant compositional changes (see below). At Figueiras, apparently similar processes led to tar balls being found at depths of up to 1.30 m, and to the entire sand column presenting millimetric tar balls at various concentrations, while oil coatings were mainly restricted to the lower half of the cores. By contrast, no significant morphological changes were detected between samplings at O Rostro (2004 and 2006), where the predominant forms on both occasions were tar balls of different sizes; this persistent presence of tar balls, and the marked absence of oil coatings, suggest the recent reburial of oil deposited periodically as a result of the seasonal cycle of the beach, in accordance with the model proposed by Bernabeu et al. (*7*).

In the cores from the Basque beaches, no oil was detected either visually or analytically when they were sampled in 2005.

Chemical Analysis. Hydrocarbon concentrations were highest in the lowest levels of the oil-coated sand layers (15 mg/kg for the 32 PAHs at Figueiras and 1 mg/kg at Nemiña), and no more than about 0.05 mg/kg at depths 1 m above these levels (Table S4). Detailed analysis of the sterane and triterpane biomarkers most commonly used for oil fingerprinting and weathering assessment (Table S5) (*23, 24*) showed that as depth decreased, the C_{29} sterane 29$\beta\beta$ accounted for an increasing proportion of 27$\beta\beta$ + 28$\beta\beta$ + 29$\beta\beta$ at the expense of its C_{27} homologue, whereas the triterpane ratio %29$\alpha\beta$ and other indices based on aliphatic biomarkers remained relatively unaltered (Figure 3). This is what is generally observed in field bioremediation studies (*25*), and is thus indicative of biodegradation, which will likely have been facilitated by the supply of oxygen, degrading microorganisms, and nutrients by the water flowing through the sand column (*26*). Among the PAH-based indices that have been put forward for use in oil fingerprinting and weathering assessment (*27*), the C_1−C_3 alkylphenanthrene/alkylchrysene ratios fell markedly with decreasing depth (probably due to the leaching of the more soluble compounds and/or biodegradation), while the corresponding alkylphenanthrene/alkyldibenzothiophene ratios remained almost unchanged (Figure 4); these trends are again consistent with those observed in previous studies (*25, 28*), and indicate appreciable weathering despite the burial of the oil.

Wave Climate and Beach Response. Comparison of the affected beaches in Galicia, which showed persistent oil, with those of the Basque Country, which were clean, clearly showed that the fate of deep-buried subsurface oil depended mainly on erosion and accretion processes.

During winter 2002−2003, wave heights were unusually high off the NW Galician coast, and the most frequent wave

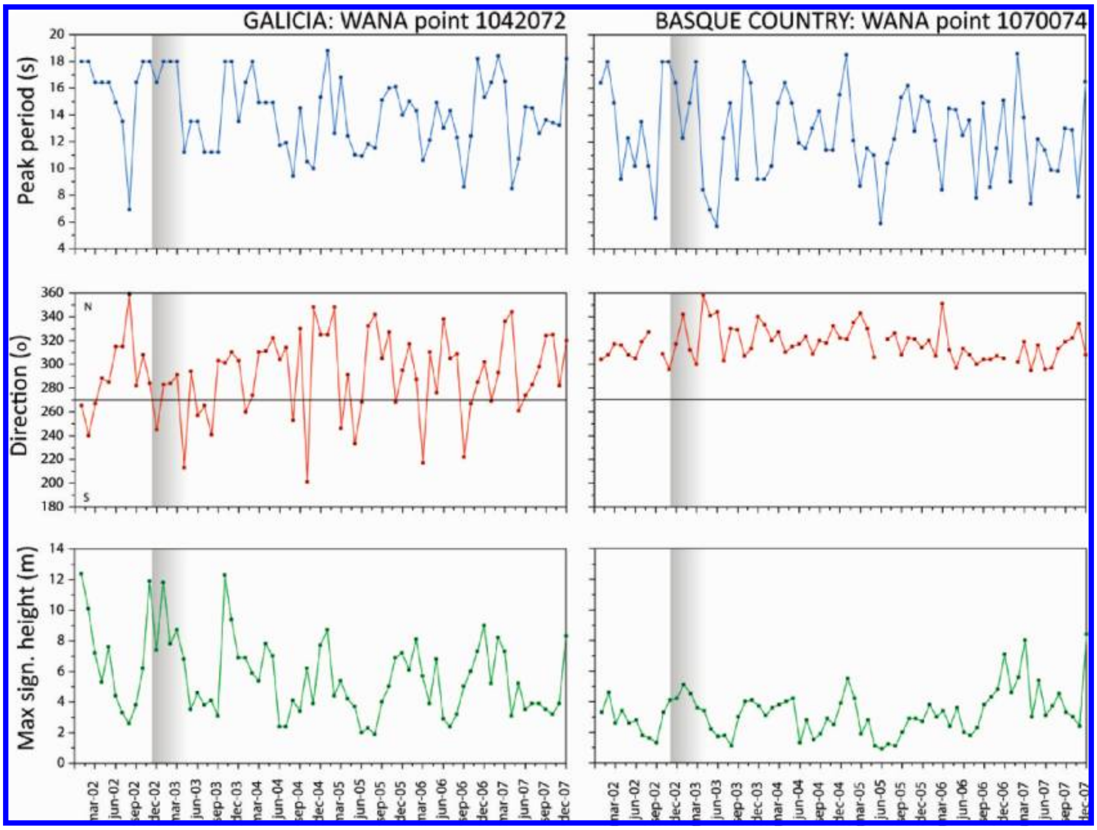

FIGURE 5. Simulated monthly time series of peak period, direction of wave incidence in deep water, and maximum significant wave height, at WANA points 1042072 (off the Galician coast) and 1070074 (off the Basque coast). The gray band shows the period during which oil arrived on the coast in successive strandings.

directions were likewise unusual, with no waves arriving from the north and an increase in the number of those arriving from the southwest, which are frequently associated with storm events (Figure 5). The massive arrival of the *Prestige* oil took place during a storm event of exceptional intensity ($Hs_{max} = 9.34$ m, $Ts_{max} = 15.5$ s; return period 11 years). These conditions subjected the NW Galician beaches to intense erosion (*29*), with the result that the oil was stranded at exceptionally low topographic levels. However, during the following weeks the sea became progressively smoother, with significant wave height and period adopting typical winter values (mean Hs = 2.4 m, mean Ts = 8.56 s) and approximately 20% of waves arriving from the north. This activated the accretional phase of the morphodynamic cycle of the beaches, burying the oil under more than 2 m of clean sand in the intertidal zone; and this exceptional burial depth favored long oil residence times, because these layers were hardly affected by seasonal intertidal sand movements during the less extreme wave climate conditions of the following years (Figure 5).

In contrast to the Galician situation, wave climate parameters 800 km away on the Basque beaches were typical of an average winter when the *Prestige* oil began to arrive there in December 2002, with mean Hs = 1.5 m and mean Ts = 13 s. The erosion being suffered by the oiled beaches was no more severe than in a typical winter, and in the following years the usual morphodynamic cycle of these beaches subjected the stranded oil to repeated offshore exposure and onshore burial (*30*), abrading and degrading it physically and biogeochemically.

Discussion

Sampling of sandy beaches on the northwest Spanish coast between 2002 and 2007 showed the persistence of buried oil 5 years after the POS. Oil was buried at depths of up to 4 m, well below the standard 0.5 m pits that are generally used to estimate the occurrence of buried oil in beaches. An unknown fraction of subsurface oil thus escaped detection by traditional spill assessment methods—which accordingly underestimated the total volume of stranded oil—and was initially ignored by cleanup operations. The long-term persistence of buried oil in beaches may have important environmental consequences resulting from its gradual release into seawater to the possible detriment of biological resources.

Oil Burial and Beach Morphodynamics. A sandy beach is a dynamic sedimentary environment that responds to long- and short-term fluctuations in wave climate with characteristic and predictable onshore−offshore oscillations involving thousands to millions of cubic meters of sediment. On the annual time scale, this results in the intertidal zone shifting landward in winter and seaward in summer, forcing a particular point in the beach profile to move several meters vertically and horizontally (Figure 6).

Landward shifts in the beach profile are exacerbated by storm events, which maximize erosion; and since marine oil spills are frequently associated with storm events, oil stranding tends to occur when nearby beaches are in their most erosive extreme. During the subsequent milder-weather periods, the eroded sediment deposited in the subtidal zone is returned by waves to the intertidal zone, where it buries stranded oil. The fate of buried oil thus depends on the hydrodynamic and morphodynamic conditions of the sea and beach during oiling, and on relevant wave return periods, especially for beaches where the combination of high wave energy and high tide levels results in the seasonal transport of large volumes of sand up and down the profile (*31*).

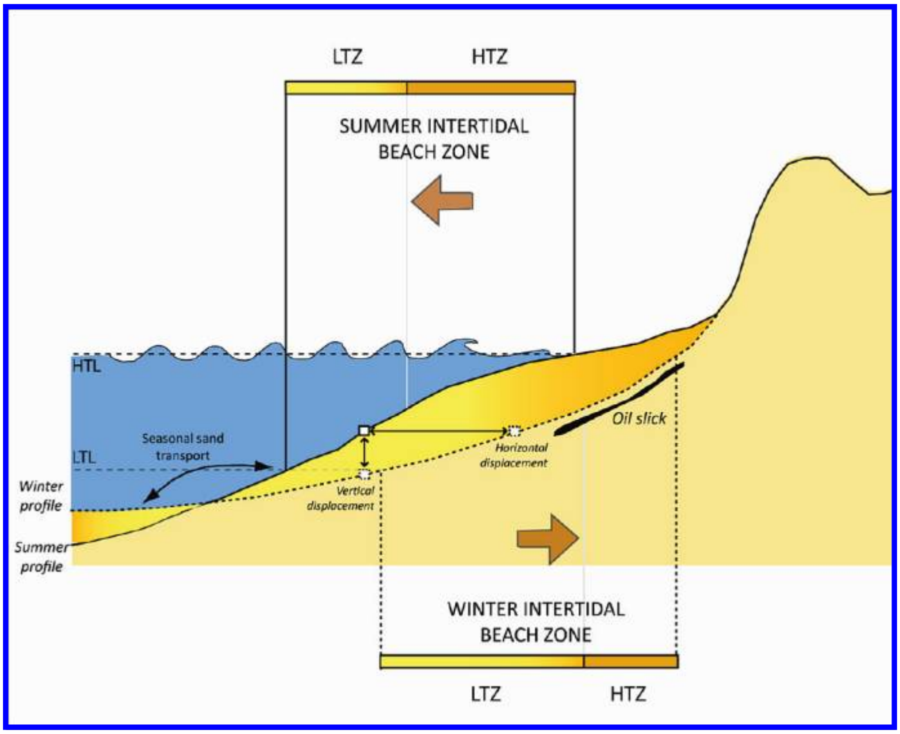

FIGURE 6. Scheme of seasonal beach profile variations in response to wave climate. A point located in the low tidal zone (LTZ) in summer could be located in the subtidal zone during winter.

Although beach morphodynamics have to some extent been taken into account in shoreline treatment protocols for over 30 years, their spatial scale and special role in highly dynamic sandy beaches have not been duly reflected in cleanup strategies (*32*), possibly because access to deep sediment is difficult and highly disruptive. However, our results show that the morphodynamic behavior of sandy beaches has a key influence in the early stages of their oiling, determining both the initial depth of oil burial (which may reach several meters) and the extent of oiling. Information on beach morphodynamics is thus critical for discriminating between beaches that will undergo natural cleaning and beaches that will need the responder's intervention.

We propose that beach morphodynamics, as a fundamental factor for evaluation of the scale of the cleanup challenge, be integrated at the very beginning of spill assessment, in a simple albeit indirect fashion, through the analysis of wave climate time series. This should provide a sufficient indication of the probable depth of oil burial as to orient shoreline treatment priorities: a simple comparison of wave conditions during a spill with the average conditions in the area should indicate the likelihood of short- or medium-term natural recovery of a beach.

Assessment of Buried Oil. Current methods of sandy beach sampling fail to afford an adequate basis for estimation of the extent of oiling because they are unable to reach oil buried below several meters of sand. With our modified suction corer (BOSC), it is quite simple to sample depths that are at present disregarded. It thus enables a comprehensive three-dimensional map of oiling intensity to be constructed in a nondisruptive, environmentally friendly fashion at beaches where it is desired to verify the predictions of the proposed wave climate analysis, or where more detailed information is required.

Further development of this approach may incorporate a preliminary beach survey by in situ measuring equipment, such as a geoprobe system holding a laser-induced fluorescence spectroscopy device, for the direct determination of PAHs (*33*). Of course, this would not rule out sampling to "ground truth" but would reduce the total number of collected cores.

To sum up, our findings bear clear implications for improving the assessment, and hence the remedial treatment, of sandy beaches oiled under certain weather conditions. Analysis of wave climate simulations in the very early stages of a spill should allow rough estimation of the probability of finding subsurface oil below the standard depth of 0.5 m, and of whether the natural degradation of stranded oil will be enhanced by the seasonal movement of beach sediment in the short-to-medium term, or remain dependent on physical and chemical degradation within the sand column due to the location of much of the oil in immobile sand layers. The latter situation implies the longer-term persistence of oil in the environment, and leads to the formation of oil microparticles that are swept away by groundwater, making emulsioned oil more available to local organisms. The consequences of this process deserve further investigation.

Acknowledgments

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Supporting Information Available

Additional figures and tables describing the sampling sites, and analytical results. This information is available free of charge via the Internet at http://pubs.acs.org.

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