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Evaluation of cleanup endpoint parameters for sandy beaches polluted with heavy fuel oil

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Evaluation of cleanup endpoint parameters for sandy beaches polluted with heavy fuel oil

G. Mansour¹, C. Sukhn², F. Al Ali³, B. Hatjian⁴ and N. Sabra⁵*

Abstract: This research work aims at evaluating the accuracy of a clean-up endpoint set by Lebanese authorities for sandy beaches following an oil spill that hit Lebanon in 2006. This was done by evaluating two factors affecting clean-up endpoint determination: background levels of Total Petroleum Hydrocarbons (TPH) in beaches, and TPH mobility. To determine background TPH levels, TPHs were analyzed in beaches that were not affected by the oil spill, and in beaches that were affected by the spill but were subject to subsequent treatment. The TPH concentrations of the two types of beaches were not statistically different, and averaged 249 mg kg⁻¹ of sand. To measure TPH mobility, leaching tests were performed on a stockpile of untreated contaminated sand (110 g of TPH/kg sand). The average TPH concentration in the sand leachate (19.9 mg L⁻¹ ± 7.04) slightly exceeded the national TPH standard of 20 mg L⁻¹. Since the mobility of pollution is associated with toxicity, the evaluation of the clean-up endpoint will depend essentially on the results of the TPH mobility tests. The maximum TPH concentration in treated sand should correspond to TPH levels below 20 mg L⁻¹ in leachate, as per the national standard. More research work, particularly sand clean-up tests and bioassays, is necessary to determine the maximum TPH concentration to be observed in sandy leachates, which would

ABOUT THE AUTHORS

Our collaborating authors are from different disciplines and have in common the interest of advancing the environmental situation in Lebanon through their research, analyses and/or publications. Georges Mansour is an analytical chemist and specializes in food safety and environmental engineering. Fatima al Ali is into supramolecular chemistry and environmental field work. Berj Hatjian is surveying the Risk Assessment of Environmental and Occupational Exposure to Chemical Carcinogens. Carol Sukhn is interested in following fate of toxicants in environmental and biological matrices. As for Nada Sabra, she is into remediation of contaminated sites. The authors are an example of collaboration between academia and the public sector in Lebanon and between scientists of complementary disciplines. We are hoping that this will initiate other third world countries with limited budget or funds to work on a similar scale when disaster such as oil spill hit their shores.

PUBLIC INTEREST STATEMENT

Following oil spills, the verification of the efficiency and limit of clean-up can be complicated and variable among countries since it is linked to the impact of the remaining oil residues on the local environment. In Lebanon, following an oil spill in 2006, authorities adopted a pollution threshold of 5 g of total petroleum hydrocarbons (TPHs) per kg sand for the treatment of oil contaminated sandy beaches. Despite the importance of this cleanup endpoint, it was necessary to evaluate it as each oil spill is unique and cannot be adopted from other countries. The evaluation was carried out in the present paper. It was based on the analysis of background TPH levels in beaches and on the study of the mobility of TPHs in sands. Bioxicity studies are still needed before finalizing the evaluation. This study would allow Lebanese authorities to optimize the treatment of remaining contaminated sands and to develop relevant cleanup standards.
enable us to determine the maximum total TPH concentration allowed in the sand, and thus decide on the accuracy of the clean-up endpoint of 5,000 mg TPH/kg sand.

**Subjects:** Environmental Sciences; Pollution Management; Coastal Management; Resource Management - Environmental Studies; Environmental Issues; Environment & Resources; Environmental Change & Pollution

**Keywords:** oil spill; heavy fuel oil; clean-up endpoint; sandy beach; shoreline; background TPH levels

1. **Introduction**
In 2006, the Lebanese coast was heavily polluted by an oil spill caused by war hostilities. An estimated weight of 15,000 tons of heavy fuel oil was released into the sea, contaminating approximately 150 km of the Lebanese coastline (Figure 1) in addition to some parts of the Syrian coast, making it the largest oil spill in the eastern Mediterranean Sea in recent years (Ministry of Environment-Republic of Lebanon, United Nations Development Program, ELARD, 2006; United Nations Environment Program, Experts Working Group for Lebanon, 2006). The released oil was categorized as an IFO 150 (number 6 fuel) also known as *bunk oil*, *bunker oil*, or *black liquor* (Khalaf, Nakhlé, Abboud-Abi Saab, Tronczynski, & Mouawad, 2006). This is a thick, black, tar-like liquid, consisting of a complex mix of hydrocarbons (Khalaf et al., 2006).

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**Figure 1.** Source and location of the oil spill that hit Lebanon in 2006 (Green Line Association, 2007).
The Lebanese Ministry of Environment treated the polluted beaches, aided by the international community. Driven by the south–north wave currents, the oil reached both rocky and sandy beaches. The affected beaches were situated north of Jiyeh village, the starting point of the spill, located in the south of Lebanon. While the treatment of rocky beaches with compressed water was relatively easy, the treatment of sandy beaches proved to be more difficult. The difficulty was not only limited to which remediation technology should be used, but also to which pollution threshold or clean-up endpoint that should be set. While several treatment technologies, such as sand washing and solidification/stabilization, have been used (Ministry of Environment-Republic of Lebanon, 2012), the following question remained unanswered: “How clean is clean?”.

Clean-up endpoints are needed to set the standards for which treatment options are selected (American Petroleum Institute, 2016), and to determine the conditions beyond which increased treatment efforts may result in reduced return, delayed recovery, or even damage to the environment (American Petroleum Institute, 2016; Michel & Benggio, 1999; Oil Spill Response, 2011; Region 10 Regional Response Team & the Northwest Area Committee, 2017).

A number of papers and frameworks have discussed clean-up endpoints for sand (Michel & Benggio, 1999; National Oceanic & Atmospheric Administration, 2013; Owens & Sergy, 2003; Sergy & Owens, 2007). However, there are no unified standards or guidelines that can be applied universally for determining clean-up endpoints. Owens and Sergy (2003) stated that, “it is not feasible to have a single, detailed decision-making methodology for sand clean-up endpoints that is relatively simple and practical, yet comprehensive and universally applicable.” Since the circumstances of each spill are different, the endpoints must meet the specific conditions of each spill event; each oil spill has its own apposite standard. There may even be unique factors within a given shoreline segment that require a different approach to clean-up endpoint determination (Region 10 Regional Response Team & the Northwest Area Committee, 2017). Likewise, Sergy and Owens (2008) pointed out, that in a given oil spill, different criteria can apply to different segments of a given shoreline. In fact, clean-up endpoints can vary with the characteristics of the beach affected (Region 10 Regional Response Team & the Northwest Area Committee, 2017), like habitat use and geomorphology (Michel et al., 2013; Owens, Santner, et al., 2011) or the nature, character and extent of the oil spill (Michel et al., 2013). Clean-up endpoints determination is also influenced by the ecological risk or stress caused by the oil, the socioeconomic impact of the oil spill, the effects of clean-up operations of the beach ecosystem beyond a treatment level, the effects of increasingly more stringent treatment criteria, the extent of natural recovery and the roles and responsibilities of the decision makers (Fejes, Lindgreen, & Arbjork, 2005; Owens & Sergy, 2003). The dependence of clean-up endpoint decision on economic and political considerations has also been pointed out by Guénette, Aasnes, and Folium (1997) and Dicks, Parker, Purnell, and Santner (2002).

Based on the above, variations are observed in shoreline clean-up endpoints among countries. A study conducted by CEDRE Research Program on the Erika oil spill, which polluted the French coastal water with heavy fuel oil in 1999, specified a level of 1,000 ppm as the maximum oil concentration that can be present in the sand before it is deposited on the shore for final cleaning by wave action (surf washing) (Guerroué, Cariou, Poupon, & Xavier Merlin, 2003). In fact, sand with concentrations higher than 1,000 ppm were found to leach excessive oil and thus pollute the environment. Another study conducted by TOTAL (International Petroleum Company) on the Erika oil spill (Scherrer & Couvreur, 2003) concluded that if the TPHs concentration in treated sand is 2,500 ppm, the sand can be used as road or building material. However, if its TPH concentration is 5,000 ppm, then the sand should be land filled. The threshold values of 2,500 and 5,000 ppm were set by the French authorities following leaching tests performed on the oil-contaminated sand. TPHs were among the analytes monitored in the leaching tests. In Taiwan, the sand pollution standard set by the Environmental Protection Administration is set to 1,000 ppm of heavy fuel oil (Lin, 2016). In Sweden, an oil spill contaminated the most pristine white sandy beaches at Löderups Strandbad in the summer of 2003. Around 4,000 tons of beach sand were contaminated, and TPH levels in the sand ranged from 8% (80,000 mg kg⁻¹ sand) to 16% (160,000 mg kg⁻¹ sand) (Oil Pollution Services Limited, 2014). A program
was put in place to wash the sand without the use of chemicals, and the acceptable final limit for the TPHs in the washed sand was set to less than 200 mg kg\(^{-1}\) (personal communication, August 2017). In 2011, Owens, Santner, et al. (2011) conducted shoreline treatment operations for the Macondo oil spill in the Gulf of Mexico in the United States following the Deepwater Horizon accident that affected the region in 2010. The treatment operations were carried out until reaching “No Further Treatment” (NFT) conditions. The NFT guidelines were specific to the type of the shoreline or beaches affected and no unified clean-up endpoint concentration was used. For instance, in the Eastern States Sand Shorelines, the NFT guideline was “no visible oil above background levels” for oil-contaminated residential/amenity beaches while it was “<1% visible surface oil and oiled debris and no Surface Residue Balls (SRB) > 5 cm” for non-residential beaches. As part of the Deepwater Horizon catastrophe, the same authors conducted sand washing trials for the sand beaches of the Grand Isle LA area. Sand exiting the sand washing machine had a TPH concentration of less than 500 ppm. The sand was further treated by sediment relocation (Owens, Taylor, Graham, & Castle, 2011). No information was mentioned on the final TPH concentration after sediment relocation.

Clean-up endpoints are usually developed based on clean-up objectives, which can be: (1) to minimize exposure hazards affecting human health; (2) to speed up the recovery of affected areas if possible; and (3) to reduce the threat of additional or prolonged impact on natural resources (Michel & Benggio, 1999). These objectives lead to the development of clean-up strategies that do not cause more harm to the environment than the remaining oil, whether visible or buried (Michel & Benggio, 1999). This point has also been referred at by Guénette et al. (1997).

In addition to the human health and ecological criteria, the determination of the clean-up endpoints may also be based on the odor characteristics of the sand or its TPH level (Michel & Benggio, 1999; Mulhare & Therrien, 1997; Region 10 Regional Response Team & the Northwest Area Committee, 2017). In this regard, the clean-up endpoint would be reached when all visible oil has been removed (Michel & Benggio, 1999; National Oceanic & Atmospheric Administration, 2013; Sergy & Owens, 2007; Shigenaka, 2011). This goal is often difficult to achieve, particularly if there is already a background rate of oil deposition (Michel & Benggio, 1999; National Oceanic & Atmospheric Administration, 2013). In fact, removal of all visible oil is usually applicable when the oil can be easily removed and in a way that does not delay the recovery of the impacted area (Fejes et al., 2005). When oil removal is difficult or may delay the recovery of the affected area, the elimination or withdrawal of all visible oil can have an impact on the environment that is more negative than the impact of having residual TPHs below background levels (Fejes et al., 2005). In this case, a more appropriate endpoint would be visible oil, but no more than the background amount (Michel & Benggio, 1999). Such a clean-up endpoint would be more realistic from an operational and environmental point of view and has been pointed out by other authors such as National Oceanic and Atmospheric Administration (2013) and Owens, Santner, et al. (2011) and Oil Spill Response (2011). Background data can serve as a benchmark and be one of the parameters for determining clean-up endpoints (Oil Spill Response, 2011).

Clean-up endpoints can also be determined based on other parameters, such as the mobility of the remaining oil in the treated sand, and its eco-toxicity (Australian Maritime Safety Authority, 2015; Sergy & Owens, 2007, 2008). The remaining oil residues in the sand should not be mobile nor cause toxicity (Australian Maritime Safety Authority, 2015), and hence should not leach out into near shore waters with concentrations above the pre-spill background level (Sergy & Owens, 2007).

In 2006, the clean-up endpoint for oil-contaminated sands was set to 5,000 ppm by the Lebanese authorities, based on a literature review of related environmental catastrophes (personal communication). While this clean-up endpoint was crucial, it was still necessary to evaluate it through scientific research, since each oil spill has its own specificities.

A more thorough study of the clean-up endpoint of 5,000 ppm is also important due to the presence of a certain quantity of sand (around 2,400 m\(^3\)), that is still untreated and stored in containers.
When the Lebanese authorities will treat this contaminated sand in the future, it would be imperative to know if the sand treatment methods should bring down the sand TPH levels to 5,000 ppm, or to a lower value.

To address this issue, it is necessary to determine the background levels of TPHs in the beaches. The oil level in the contaminated sands may then be lowered to the background levels. No data exist so far on the background levels of TPHs in sandy beaches in Lebanon. The review of the clean-up endpoint value of 5,000 ppm would allow generating data on the background concentrations of TPHs in Lebanese beaches. On the other hand, it is necessary to study the mobility of the oil as well since mobility is an important parameter for clean-up endpoint determination as stated earlier. If the oil is barely mobile or not mobile, it may not be necessary for the clean-up methods to reduce the oil levels to background levels, a matter that affects both clean-up time and cost.

The main objective of this research work is to evaluate the clean-up endpoint of 5,000 ppm set by the Lebanese authorities in 2006 for oil contaminated beach sand. In this regard, the background level of TPHs was first quantified in sandy beaches along the Lebanese coastline. Sampling covered both the sandy beaches that were affected by the 2006 oil spill and subsequently treated by the Lebanese Ministry of Environment, and beaches that were not affected by the oil spill. The TPH levels of these two types of beaches were then compared. Then, the mobility of TPHs in the contaminated sand that is still untreated has been quantified using leaching tests and the TPH levels in the leachates were subsequently compared with the national standard level.

Based on the output of these two objectives, it would be possible to determine if the clean-up of the polluted sand needs to comply with the pollution threshold of 5,000 ppm set by the Lebanese authorities in 2006, or with some other value.

The present research work is the first scientific study on shoreline clean-up endpoints in Lebanon, and is important because each oil spill is unique and requires a tailored approach to clean-up endpoint determination. Not only is it the first study to determine background TPH levels in sandy beaches in the country, but it also responds to a national need for developing pollution standards for solid environmental matrices including sand. It will further contribute to the scarce scientific research conducted so far on clean-up endpoints.

2. Materials and methods

2.1. Sand Sampling

To determine the background levels of TPHs in the sandy beaches, sand samples of pure sand were collected from nine Lebanese beaches between April 17 and 2 May 2012. No rain was recorded during the sampling period. The average temperature during the sampling events, carried out on sunny days, was 25.3°C ± 2.95 at 5–20 cm depth. As illustrated in Figure 1, six beaches were affected by the spill and were subject to subsequent treatment by the Ministry of Environment. These were the beaches of Damour, Ramlet Al Bayda, Jbeil, Chekka, Tripoli, and Al Aarida. The remaining three beaches (beaches of Nakoura, Tyr and Jounieh) were not affected by the spill and are considered reference sites for the oil-contaminated beaches.

In each of the nine beaches, samples were collected from two horizontal lines parallel to the shore and from one vertical line perpendicular to the shore as illustrated in Figure 2. One of the horizontal lines was 2 meters away from the sea and was continuously hit by waves. It is referred to as, “near the sea line.” The area between this line and the sea line represents the active area of the beach, since it is always hit by waves. The second horizontal line was near the dry land, at two-thirds of the width of the beach, starting from the sea line. It is referred to as, “near the dry land line,” as it is not hit by waves. The area between this line and the “near the sea line” represents the inactive area of
the beach, as it is not reached by waves. Finally, the vertical line was perpendicular to the shore, and included both the active and inactive areas.

As the Lebanese sandy beaches are no longer pristine (Ministry of Environment-Republic of Lebanon, United Nations Development Program, ECODIT, 2010), a sand sample was also collected from a clean sandy private beach in Brazil, using the same sampling procedures as for the Lebanese beaches. This sample was used as a control beach sample for TPH levels. As indicated in the following section, the sand sample from Brazil will be analyzed using the same extraction and analytical methods used for the sand samples collected from the Lebanese beaches. This makes it a control sample that can be reliably compared with the Lebanese beaches.

The distance between the sampling points on each of the two horizontal lines was 20 m for long beaches (beaches with a coastline length of more than 100 m) and 10 m for short coastline beaches (less than 100 m). The distance between sampling points on the vertical line was around 5 m for wide beaches (>30 m), and 2.5 m for narrow beaches. All the samples were collected at 20 cm depth. In each beach, the Global Positioning System (GPS) coordinates, the time of sampling, the weather details, the temperature, the waves action, the presence/absence of litter on the beach, human activity, and the surrounding structures were recorded.

For each sampling line on a given beach, sand samples were collected using augers. Tap water and n-hexane were used to clean augers before and after use (at the beginning and at the end of the sampling from each beach) to avoid cross contamination of the collected samples. Sand samples from each sampling line were then mixed and composited. From each beach, every composited sand sample was divided into four quarters after thorough mixing. Three quarters were discarded, and around 1 kg was collected from the fourth quarter for analysis. Three composite samples corresponding to the three sampling lines were thus obtained per beach, and a total of 27 composite samples were obtained from the nine studied beaches.

For the experiment assessing of the mobility of TPHs in the oil-contaminated sand samples, TPH mobility was measured in polluted untreated sand samples, and compared to the reference and control sands. Untreated sand samples were collected from polluted sand stored by the Lebanese
Ministry of Environment in containers since 2006, awaiting further treatment. Forty kilograms of sand were randomly collected from the polluted sand stocks, mixed, and divided into four quarters. Around 1 kg was collected from one quarter for TPH analysis.

### 2.2. Laboratory analyses

To determine background levels of TPHs in Lebanese beaches, the sand samples collected from different beaches were analyzed for the TPH as per the US EPA method 9071B (United States Environmental Protection Agency, 1998). The method uses n-hexane as the extraction solvent in a solid–liquid extraction apparatus, the Soxhlet apparatus (Gerhardt Analytical Systems), to yield n-hexane extractable material (HEM). The same protocol was used to determine TPHs in both the untreated polluted sand and the reference/control sand.

Representative portions of sand samples (10 g) were mixed with the equivalent amount of anhydrous sodium sulfate (≥99% purity, Sigma-Aldrich) in order to remove traces of water from the sand. Next, the HEM was extracted with n-hexane (85% purity, Sigma-Aldrich) using the Soxhlet apparatus for four hours (cycling rate of 20 cycles/hour) at a temperature of 70°C. The n-hexane extract was then evaporated using a rotary evaporator, which was adjusted to complete the evaporation in less than 30 min. The HEM was desiccated and weighed until the weight stabilized. The HEM content was considered as TPH content. The limit of quantification (LOQ) was then determined. The TPH LOQ in sand samples was 100 mg kg⁻¹ dry sand.

Basic quality control measures were adopted for TPH determination. A blank test was done to check traceable presence of HEM in the thimble, in the anhydrous sodium sulfate, and in the solvent used in the extraction of the sand samples. HEM levels were determined first in the blank samples and in the reference and control sands. For this, three replicates were performed to determine the HEM levels in the Lebanese beaches, where one composited sampling line corresponded with one replicate. Given their heterogeneity, seven replicates were used to assess the HEM levels in the untreated polluted sand. Two sand samples, one from a beach not affected by the oil spill (Tyr) and one from a beach affected by the oil spill (Byblos), were spiked in triplicates with a known concentration of Petroleum oil collected during the 2006 spill floating on the sea. This was done to determine the recovery percentage of the extraction. The TPH recovery percentage of the sand spiked with oil was found to be equal to 64%. For all experiments, the HEM concentrations were reported after treatment with the extraction recovery percentage. HEM data were expressed as mg per kg (ppm) dry weight of the analyzed sand. Sand samples with TPH levels above the limit of quantification were characterized using gas chromatography–mass spectrometry analysis (Agilent 6890-5973).

To assess TPH mobility, samples from the contaminated untreated sand and from reference and control sands were subjected to the toxicity characteristics leaching procedure (TCLP) as per the USEPA method 1311 (United States Environmental Protection Agency, 1992). One hundred grams of sand samples were mixed with two liters of acidified water, and agitated on the magnetic stirrer for 18 h at room temperature. The extraction fluid used was a function of the alkalinity of the sand. If the pH of a mixture composed of 5 g of sand and 96.5 ml of reagent water was less than 5, the sand is to be extracted with an extraction fluid having a pH of 4.93 ± 0.05 and composed of glacial acetic acid, sodium hydroxide, and reagent water. However, if the pH of the sand and water mixture is greater than 5, the sand is to be extracted with an extraction fluid having a pH of 2.88 ± 0.05 and constituted of glacial acetic acid and reagent water. In the present research work, the pH of the contaminated untreated sand and of the reference and control sands was greater than 5. Accordingly, each of these sands was extracted with the extraction fluid characterized by the pH of 4.93 ± 0.05 and composed of glacial acetic acid (ACS reagent, ≥99.7%, Sigma-Aldrich), sodium hydroxide (ACS reagent, ≥98%, pellets, Sigma Aldrich), and reagent water. Following the extraction procedure, the solid and liquid phases were separated by filtration, and the liquid phase or the leachate was considered the TCLP extract; its HEM could then be quantified. Three replicates were performed to obtain the leachate of the control sand as well as that of the polluted sand. Four replicates were used for the reference sand. The HEM content of the leachate obtained from the US EPA 1311 method was...
determined using the USEPA method 1664 (United States Environmental Protection Agency, National Service Center for Environmental Publication, 1999). A one liter sample was acidified to pH < 2 and serially extracted three times with n-hexane in a separatory funnel. The solvent extracts from the three serial extractions were combined, and dried over anhydrous sodium sulphate. The solvent was evaporated to dryness, and the HEM was dessicated and weighed.

2.3. Statistical analyses
SPSS statistical software (version 20, IBM compatible) was used to determine if there were statistical significant differences between the HEM levels of the different beaches (affected and not affected beaches). The non-parametric Kruskal–Wallis test was used to compare the HEM content of the beaches at a significance level of 95%. For statistical purposes, levels below the limit of quantification (100 mg kg⁻¹ dry sand) have been reported at half value (50 mg kg⁻¹ dry sand).

3. Results and discussion
3.1. Determination of the background levels of TPH in the Lebanese beaches
Beaches were divided into two groups: beaches affected by the oil spill, and beaches not affected by the oil spill. As indicated in Table 1, the average concentration of HEM was 275 ± 278 in the beaches affected by the oil spill and 196 ± 83 mg kg⁻¹ in those not affected by the spill.

The Kruskal–Wallis test did not show a significant difference between the two beach categories (α = 0.05, p > 0.05, n = 27). The average HEM concentration of all beaches from both categories (249 mg kg⁻¹ dry sand) can be considered as reflecting the current background levels of TPHs in the Lebanese sandy beaches.

The observed HEM levels in beaches with HEM concentrations above the LOQ may be due to pollution sources that are not of petroleum origin. In fact, two of the sites that were above 100 mg kg⁻¹ dry sand (Tyr and Byblos) underwent gas chromatography–mass spectrometry analysis. The chromatographic output revealed the presence of fatty acids of vegetable source rather than heavy fuel oil components.

<table>
<thead>
<tr>
<th>Beach name</th>
<th>Beach category</th>
<th>Sampling line*</th>
<th>Average ± SD per beach</th>
<th>Average ± SD per beach category</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Near the sea</td>
<td>Near the dry land</td>
<td>Vertical line</td>
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<tr>
<td>Brazilian beach</td>
<td>Control beach</td>
<td>50</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>Abdeh</td>
<td>Lebanese Beaches hit by the oil spill</td>
<td>50</td>
<td>50</td>
<td>50</td>
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<tr>
<td>Tripoli</td>
<td></td>
<td>958</td>
<td>654</td>
<td>803</td>
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<tr>
<td>Chekka</td>
<td>Byblos</td>
<td>50</td>
<td>591</td>
<td>595</td>
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<td></td>
<td></td>
<td>281</td>
<td>264</td>
<td>50</td>
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<td></td>
<td>Ramlet al Bayda</td>
<td>50</td>
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<td></td>
<td>Damour</td>
<td>400</td>
<td>50</td>
<td>50</td>
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<tr>
<td>Jounieh</td>
<td>Lebanese Beaches not hit by the oil spill</td>
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<td>50</td>
<td>50</td>
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<tr>
<td>Tyr</td>
<td></td>
<td>323</td>
<td>50</td>
<td>124</td>
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<tr>
<td>Mansouri</td>
<td></td>
<td>50</td>
<td>376</td>
<td>445</td>
</tr>
</tbody>
</table>

*Sampling lines at beaches as explained in the Methods section.
†SD is standard deviation.
It is difficult to compare the determined background HEM level with international standards or guidelines, since these guidelines only refer to marine waters and sediments. For instance, in Germany, according to Galgani, Ellerbrake, Fries, and Goreux (2011), no information is available on the toxicity or the presence of organic trace contaminants in natural beaches, a situation which calls for action according to the authors of the study.

There is a paucity of research in Lebanon about hydrocarbons pollution of sandy beaches. No studies addressing the monitoring of hydrocarbons in Lebanese sandy beaches have been reported by national research centers. With reference to the 2006 oil spill, research papers tackled the monitoring of organic pollution (mainly polycyclic aromatic hydrocarbons-PAHs) and inorganic pollution in fish, mollusks, phytoplankton, chlorophyll-a, and meiofauna, as well as remote sensing monitoring (Abi Saab et al., 2008; Ministry of Environment-Republic of Lebanon, 2012; Nakhle et al., 2008). To date, no background studies exist on hydrocarbons levels in sandy beaches in Lebanon.

Since no studies were conducted on oil pollution levels in sandy beaches in Lebanon prior the oil spill incident, the obtained average background TPH value of all studied beaches (249 mg kg⁻¹ dry sand) can be considered the background TPH value for future studies and standardization in Lebanon, especially that it covered most of the sandy beaches along the Lebanese coast.

The identification of background concentrations of pollutants prior to oil spills has been noted by Loh, Yim, Ha, An, and Kim (2017) as one of the most important actions that need to be undertaken by authorities to better study the effects of an oil spill.

The present work is in line with the recommendations by Guénette et al. (1997), who suggest revisiting old spill sites to evaluate the effectiveness of clean-up methods and to monitor long-term fate and recovery of oil spills.

The revisiting of an old spill site was also carried out by Kim et al. (2017). The authors monitored PAH levels in beach sediments in Korea from 2007 to 2015 following the Hebei crude oil spill that affected the shoreline of Taean peninsula in the country. As in the present study, no background levels of oil were available for the sediments before the spill. The authors followed an approach that was similar to the one adopted in this research work as they compared post spill PAH concentrations with the PAH levels of other non-polluted beaches (regional coasts). Seven and a half years after the oil spill, the PAH concentrations in the oil spill affected sediments returned to regional background levels. Likewise, in the present study, six years after the oil spill in Lebanon, the TPH concentrations of the beaches that were affected by the oil spill were found to be similar to those of the beaches that were not affected by the oil spill.

In the absence of Lebanese sand pollution standards, a comparison was made with the few existing sand clean-up endpoints. The recently determined TPH background level in the sampled Lebanese beaches (249 mg kg⁻¹ dry sand) is below the threshold of 1,000 ppm of oil set by the Taiwanese authorities (Lin, 2016) and is lower than the preliminary cut-off value of 500 ppm value that was used for treating the sand in Grand Isle LA in the Gulf of Mexico (Owens, Taylor, et al., 2011). The Lebanese beaches may be considered relatively clean as far as TPH levels are concerned. Confirmation is however needed as clean-up endpoints are oil spill specific (Owens & Sergy, 2003; Region 10 Regional Response Team & the Northwest Area Committee, 2017).

Should the nine Lebanese beaches turn out to be not polluted with TPHs, one question arises: is it possible to use the currently determined TPH background level in the nine beaches as a clean-up endpoint to remediate the untreated polluted sand that is still stockpiled in containers in Lebanon awaiting further treatment? The evaluation of the other clean-up endpoint parameter, namely the mobility of TPHs in the sand sample that is still untreated, is required to answer this question. The TPH levels of the untreated contaminated sand and the mobility of TPHs in the sand samples will
thus be determined to better decide on the clean-up endpoint that should be used when cleaning the contaminated sand.

3.2. Determining HEM mobility in the untreated oil-contaminated sand samples using leaching tests

The mobility of oil from beach sand has been discussed by Kim et al. (2017). According to the authors, oil residues adsorbed on sediments can be mobilized into the water column, particularly from sand sediments.

According to Figure 3, the average HEM concentration in the leachate of the untreated polluted sand (19.9 mg L\(^{-1}\) ± 5.75) is higher than the leachate TPH levels of the control sand (3.67 ± 3.35), and of the nine beaches that have been used for determining the TPH background levels (4.35 mg L\(^{-1}\) ± 3.34).

Taking into account the positive standard deviation, the TPH level in the contaminated sand leachate estimated by the HEM exceeds the maximum TPH concentration (20 mg L\(^{-1}\)) allowed in liquid waste that can be discharged into the sea, as specified by the Ministerial Decision 8/1 issued by the Lebanese Ministry of Environment in 2001 (Ministry of Environment-Republic of Lebanon, 2001). On the contrary, the TPH level in the leachate of the nine studied beaches (4.35 mg L\(^{-1}\)) is far below the national standard. This fact confirms the no pollution status of the nine beaches as suggested in the previous section.

The analysis of the untreated stockpiled polluted sand samples revealed that they were highly contaminated. Their average HEM level was 110,000 mg kg\(^{-1}\) ± 7,760. This level was 22 times higher than the clean-up endpoint of 5,000 mg kg\(^{-1}\) sand set by the Lebanese Ministry of Environment and 110 times above the level set by the Taiwanese authority (1,000 mg kg\(^{-1}\) sand).

Despite its very high HEM level, the untreated polluted sand has only leached 19.9 ± 5.75 mg L\(^{-1}\) HEM. This limited level of HEM mobility can be explained by the physio-chemical properties of the heavy fuel oil, particularly its richness in poorly water soluble constituents, such as heavy polycyclic aromatic hydrocarbons (PAH) and asphaltenes (Khalaf et al., 2006). The concentration of hydrocarbons that can be leached in water from heavy fuel oils is affected by the carbon number distribution of the heavy fuel oil, the composition of the hydrocarbons in the heavy fuel oil, and by the oil to water ratio (American Petroleum Institute, 2012). The limited TPH mobility can also be explained by the aging of the oil (Centro para la Prevencion y lucha contra la Contaminacion Maritima y del Litoral, Ministeria de la Presidencia, 2006) as with time, fuel weathering occurs.

**Figure 3.** Average HEM levels (mg L\(^{-1}\)) in the leachates of the polluted sand, in control sand and in sands used for determining background TPH levels. The maximum allowed TPH level of 20 mg/L is set by national legislation namely Ministerial Decision 8/1-2001 issued by the Lebanese Ministry of Environment (Ministry of Environment-Republic of Lebanon, 2001).

**Note:** Error bars refer to the standard deviation.
transformations in petroleum hydrocarbons have been reported to increase the hydrophobicity of the oil residues (Centro para la Prevencion y lucha contra la Contaminacion Maritima y del Litoral, Ministerio de la Presidencia, 2006). In the same respect, oil aging complicates the treatment of oil-contaminated rocky areas if clean-up is not carried out within the first three to six months of the oil spill (Owens, Santner, et al., 2011).

As the leachate of the untreated beach sands had TPH levels exceeding the national standard of 20 mg L\(^{-1}\), the treatment of this sand is necessary, especially that the sand may leach TPHs under wave or rain actions.

### 3.3. Evaluating the suitability of the clean-up endpoint of 5,000 mg TPH/kg sand

As indicated in the introduction, the present research work aimed at evaluating the suitability of the clean-up endpoint set by Lebanese authorities (5,000 mg TPH/kg sand). This has been carried out by studying two factors that affect clean-up endpoint determination: TPH mobility; and background TPH levels, which reflect total, rather than mobile, TPH levels.

The analysis of the polluted sand revealed a very high concentration of total TPHs (110,000 mg kg\(^{-1}\) dry sand), exceeding the background levels (249 mg kg\(^{-1}\) dry sand) by approximately 440 times. The analysis also showed a limited mobility of the TPHs in the polluted sand, since the TPH concentration in the leachate of the polluted sand (19.9 ± 5.75 mg L\(^{-1}\)) was slightly higher than the national standard of 20 mg/L. This value was found to be much higher than the TPH concentration observed in the leachates of the sand samples that were used to determine the background levels of TPH in beaches (4.35 mg L\(^{-1}\)).

Given the above results, a question arises when the polluted sand will be treated: Which clean-up parameter should be considered for the treatment? Should it be the TPH mobility in the sand expressed as mg TPH/L of leachate, or the background total TPH concentration expressed as mg TPH/kg sand? How does each scenario reflect on the suitability of the clean-up endpoint of 5,000 mg of TPH/kg sand set by the Lebanese authorities?

The review of the existing literature detailed earlier indicates that both parameters are important. However, as the mobility of pollution is associated with increased bioavailability and ecotoxicity (Agbo, 2013; Weyman, Rufli, Weltje, Salinas, & Hamitou, 2012), TPH mobility has a higher importance than the background total TPH levels. According to Kim et al. (2017), fuel oil compounds that leach from beach sediments into water are highly bioavailable to marine organisms and can present significant toxicity risks to such organisms.

When considering TPH mobility for clean-up endpoint determination, the polluted stockpiled sand samples will need to be treated in a way that reduces their leachate TPH concentration (19.9 mg L\(^{-1}\) ± 5.75) to below the national standard of 20 mg L\(^{-1}\). At this level, it is necessary to know if the treatment should be stopped once the leachate TPH levels are slightly below the Lebanese standard of 20 mg L\(^{-1}\), or when they reach a lower value, such as the TPH level found in the leachate of the sampled beaches (4.35 mg L\(^{-1}\)).

The answer to this question will require conducting bioassays, as the TPH levels in the leachates need to be below toxicity thresholds for Biota (Sergy & Owens, 2007, 2008). During the treatment of shorelines affected by oil spills, clean-up may be terminated when shorelines no longer release oil in concentrations that harm sensitive habitats or the fauna (Fejes et al., 2005; National Oceanic & Atmospheric Administration, 2013; Owens, Santner, et al., 2011; Sergy & Owens, 2008). Besides evaluating the impact of the oil spill on ecosystems and human health, the use of bioassays for monitoring the efficacy of remediation has been pointed by Martínez-Gómez et al. (2010) and Zhu, Venosa, Suidan, and Lee (2004). Greco, Corrà, Garaventa, Chelossi, and Faimali (2006) have also used bioassays to screen new dispersants for remediating oil spills in the Mediterranean region.
Once bioassays are conducted, it will be possible to determine to which extent the TPH concentration of the leachate needs to be lowered. It will then be possible to determine the total sand TPH concentration that corresponds with the leachate TPH level.

If this total TPH concentration is close to the clean-up endpoint of 5,000 mg/kg set by the Lebanese authorities, this clean-up endpoint may be considered acceptable.

The resulting clean-up endpoint value will be characteristic of the Lebanese oil spill. The clean-up endpoints determined for other oil spills or set by authorities in other countries cannot be compared to the current Lebanese clean-up endpoint or the one that would result after conducting the bioassay experiments because each oil spill is unique and has its own specific clean-up endpoint (Michel et al., 2013; Owens, Santner, et al., 2011; Owens & Sergy, 2003; Region 10 Regional Response Team & the Northwest Area Committee, 2017; Sergy & Owens, 2007).

There is a paucity of scientific publications tackling scientific experiments for clean-up endpoint determination. So far, existing publications present guidelines for determining and measuring clean-up endpoint in shorelines affected by oil spills rather than presenting results of research experiments (Australian Maritime Safety Authority, 2015; Fejes et al., 2005; Michel et al., 2013; National Oceanic & Atmospheric Administration, 2013; Oil Spill Response, 2011; Owens & Sergy, 2003; Sergy & Owens, 2007, 2008).

Guidelines for deciding on clean-up endpoints include scientific guidelines like the need to account for the type of the shore/beach, the characteristics of the oil and the ecological impact of the oil spill (Fejes et al., 2005; Owens & Sergy, 2003; Sergy & Owens, 2007). The decision-making process of a particular endpoint involves as well social or political guidelines like the socioeconomic impact of the oil spill and the point of view of stakeholders (Fejes et al., 2005; Guénette et al., 1997; Owens & Sergy, 2003). As a scientific article, the present research work is concerned mainly with the scientific guidelines of clean-up endpoint determination and it is in line with these guidelines. It is also in line with the guidelines set for measuring whether a given clean-up endpoint is reached. These include analytical methods which comprise the chemical analyses of pollutants (Fejes et al., 2005; Guénette et al., 1997; National Oceanic & Atmospheric Administration, 2013; Owens & Sergy, 2003; Sergy & Owens, 2007) as has been done in the present study as well as toxicological studies as is forecasted in the future.

4. Conclusion and recommendations
The suitability of the clean-up endpoint set by the Lebanese authorities for oil-contaminated sandy beaches (5,000 mg TPH/kg sand) was assessed by studying two factors affecting clean-up endpoint determination: background levels of TPH in sandy beaches, and TPH mobility in contaminated untreated sand samples.

TPH concentrations were determined in sandy beaches that were affected by the 2006 oil spill and that were cleaned later on, and in sandy beaches not affected by the oil spill. No significant difference was observed in the TPH levels between the two types of beaches. The average TPH concentration in this study was 249 mg TPH/kg sand. This value can be considered the current background concentration of TPHs in the sandy beaches. It is much lower than the average TPH concentration observed in the contaminated untreated sand samples (110,000 mg kg⁻¹).

To measure TPH mobility, the sand samples were subject to leaching tests, and TPH levels were determined in the resulting leachates. Despite its elevated total TPH concentration, the polluted sand was characterized by a limited TPH mobility, as the TPH concentration in its leachate (19.9 mg L⁻¹ ± 5.75) was slightly higher than the national standard of 20 mg/L. This limited mobility is most probably associated with the hydrophobic nature of heavy fuel oil, and with the age of pollution.
As pollution mobility is associated with ecotoxicity, the evaluation of the clean-up endpoint value set by Lebanese authorities (5,000 mg TPH/kg) will depend essentially on the results of the TPH mobility experiments. The maximum TPH concentration in the sand to be treated should correspond to leachate TPH levels below 20 mg/L.

The present study gave a first analysis of the factors affecting clean-up endpoint determination. However, more research work, particularly sand clean-up tests and bioassays, are needed before it is possible to determine the maximum TPH concentration to be observed in the sand leachates (slightly less than 20 mg/L?, or much lower than 20 mg/L?). This would enable the determination of the maximum total TPH concentration allowed in the sand, and thus an evaluation of the accuracy of the clean-up endpoint of 5,000 mg total TPH/kg sand set by Lebanese authorities in 2006.

This research work revisited old oil spill sites along the Lebanese coast, and allowed for their preliminary assessment after several years of treatment. It also provided important elements for evaluating the clean-up endpoint set by Lebanese authorities following the oil spill crisis in 2006.

The research work is in line with the work of Steiner (2006), who highlighted the need for governments to conduct research projects to monitor recovery and restoration, as this will enable a better management of oil spills in the future. It is also concurrent with the work of Guénette et al. (1997), who recommended the revisiting of old sites affected by oil spills to evaluate the effectiveness of the clean-up methods, and monitor the fate and recovery of the oil-contaminated shorelines in the long term.

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Competing Interests
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Cover image
Sandy beach polluted with heavy fuel oil in Lebanon during the 2006 oil spill catastrophe.

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