



Environmental Life Cycle Assessment of marine sediment decontamination by citric acid enhanced-microwave heating



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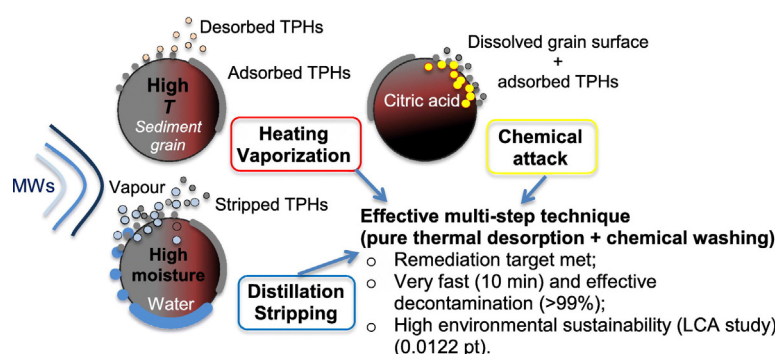
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HIGHLIGHTS

- A citric acid enhanced-MW treatment of TPH-contaminated sediment was investigated.
- The environmental sustainability of the treatment through a LCA study was assessed.
- Citric acid/MW resulted in a very rapid and effective (>99%) decontamination.
- Results revealed pure thermal desorption and chemical washing as removal mechanisms.
- The LCA classified MW technology as the most effective sustainable alternative.

GRAPHICAL ABSTRACT



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ABSTRACT

The potential ability of microwave heating (MWH) for the remediation of marine sediments affected by severe hydrocarbon (HC) contamination was investigated. Decontamination effectiveness and environmental sustainability through a comparative Life Cycle Assessment (LCA) were addressed. Main results revealed that the application of a 650-W MWH treatment resulted in a rapid (15 min) HC removal. A citric acid (CA) dose of 0.1 M led to enhanced-HC removals of 76.9, 96.5 and 99.7% after 5, 10 and 15 min of MW irradiation, respectively. The increase in CA dose to 0.2 M resulted in a shorter successful remediation time of 10 min. The exponential kinetic model adopted showed a good correlation with the experimental data with R^2 values in the 0.913–0.987 range. The nature of the MW treatment was shown to differently influence the HC fraction concentration after the irradiation process. Achieved HC removals in such a short remediation time are hardly possible by other clean-up techniques, making the studied treatment a potential excellent choice. Removal mechanisms, which allowed the enhanced-MWH to operate as a highly effective multi-step technique (pure thermal desorption + chemical washing), undoubtedly represent a key factor in the whole remediation process. The LCA highlighted that the MW technology is the most environmentally sustainable alternative for sediment decontamination applications, with a total damage, which was 75.74% lower than that associated with the EK (0.0503 pt).

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1. Introduction

The release of petroleum-based products into marine environment has led to a serious contamination of marine sediment worldwide (Zhang et al., 2015). Petroleum hydrocarbons (HCs) are recalcitrant pollutants and can be present in marine sediments as a consequence of industrial discharges, accidental spills and shipping activities, adversely affecting the health of aquatic life and humans. The Mediterranean Sea is one of the most congested basins in terms of oil tanker traffic, hosting about 20% of the global traffic, and this made Mediterranean industrial coasts and harbours exposed to severe HC contaminations (Barbato et al., 2016; Iannelli et al., 2012). One of the most polluted petrochemical coastal European site is Augusta Bay (Southern Italy, Mediterranean Sea) (D'Alessandro et al., 2016). Marine sediments of Augusta Bay have been documented as highly impacted by HC contamination with concentrations much higher than the regulatory limits (ICRAM, 2008). The possibility to successfully remedy HC-contaminated marine sediments is an essential issue in terms of human safety and social sustainability. However, the characteristics of marine sediments, among which low permeability, high salt, organic and water contents hinder their remediation using conventional chemico-physical remediation technologies such as electrokinetic (EK) (Pazos et al., 2013; Rozas and Castellote, 2015). Furthermore, despite bioremediation and phytoremediation being considered as safe, cost effective and eco-friendly treatments, their successful application is generally critical (low contaminant removal rate or very long remediation times), especially if high HC amounts need to be removed (Abbasian et al., 2016). Therefore, the development of rapid and effective techniques aimed at successfully decontaminating high HC-polluted marine sediments is strongly desirable.

Increasing attention has recently been given to microwave heating (MWH) as very effective remediation technique for the treatment of hazardous wastes (Al-harashseh et al., 2014; Pereira et al., 2014) or the remediation of organic-contaminated soils (Buttress et al., 2016) and aquifers (Falciglia et al., 2016b). The MWH remediation principle is based on transformation of the MW energy adsorbed by the irradiated medium into heat, due to the dielectric properties of the medium. The penetration of the generated MW electric field (E_d , $V\ m^{-1}$) into the medium depends on the incident electric field (E_0), which in turn is proportional to the power (P) applied at the MW generator. E_d as a function of the distance from the MW source (d, m) can be expressed by Eq. 1 (Falciglia et al., 2015):

$$E_d = E_0 \cdot e^{-\frac{d}{D_p}} \quad (1)$$

where D_p (m) is the penetration depth, which depends on the dielectric properties of the medium, namely ε' (–) and ε'' (–), which are the real (dielectric constant) and the imaginary part (dielectric loss factor) of the complex permittivity, respectively. D_p is defined as the distance from the emission point at which the electric field drops to 0.37 from its value at the emission point. It is clear that the higher the electric power dissipated into the medium, the higher the heat (\dot{Q}) generated per unit of soil volume during the irradiation (Falciglia et al., 2015):

$$\dot{Q} = \frac{1}{2} \omega \varepsilon_0 \left| E_{max}^2 \right| = \omega \varepsilon_0 \left| E^2 \right| \quad (2)$$

where ω is the angular frequency ($\omega = 2\pi f$), ε_0 is the permittivity of free space ($8.85 \cdot 10^{-12}\ F\ m^{-1}$), E_{max} is the electromagnetic field peak value ($V\ m^{-1}$) and E is electromagnetic field effective value ($V\ m^{-1}$). In the case of apolar contaminants, such as HCs, the achieved increase in temperature allows the separation, through desorption, of the adsorbed contaminants. Main MWH advantages are the possibility to achieve high temperatures and contaminant removals in short remediation times, homogeneous heating and high flexibility (De Guidi et al.,

2016; Robinson et al., 2012), due to the direct interaction between the MWs and the medium (Buttress et al., 2016; Tyagi and Lo, 2013). Contrary to the high water amount and salinity jointly with chemico-physical properties of sediments negatively affecting the final result of conventional techniques, in MWH applications these can represent main advantages. Because MWH performance is greatly affected by the dielectric properties of the irradiated medium, the mineralogical composition of the sediments in the Augusta Bay area (Di Leonardo et al., 2014) and the high salinity can play a key role in an effective conversion of the MW energy adsorbed, resulting in a rapid and large increase in sediment temperature (Lasne et al., 2008; Li et al., 2014). In addition, in the presence of a high moisture, another important contaminant removal mechanism can be the vapour stripping by water distillation (Falciglia et al., 2016a). Furthermore, this further would allow the use of additional chemicals, which, acting as extraction agents in conventional “washing” treatments, could largely enhance the MWH effectiveness. The possibility to combine a high biodegradability with a high extraction potential would also make the enhanced MWH an effective eco-friendly remedial alternative. Citric acid (CA) is a natural low molecular weight organic acid and, due to its chelating ability and acidity can be successfully used in enhanced-remediation treatments (Ma et al., 2015).

In this context, both technical feasibility and environmental sustainability are critical elements and their investigation is strongly desired. Over the years, Life Cycle Assessment (LCA) has been proven as a valid tool for environmental assessments in a huge number of applications, also through comparison of different solutions (De Benedetto and Klemes, 2009; Jeswani et al., 2010). For this reason, LCA was used in this study to compare treatment options for sediment decontamination. LCA has been improved during the past three decades and so has become more systematic and robust for both identification and quantification of the potential environmental impacts associated with a product in its life cycle (Jeswani et al., 2010). Currently, LCA is used in product/process selection, design and optimisation, and can be adopted in combination with simulation techniques and design tools for companies to be fully aware of the environmental consequences of their actions, both on- and off-site (Compagno et al., 2014). Hence, it can be considered as an invaluable tool for: environmental assessment and improvement of materials, fuels and energies, production technologies, waste treatment scenarios; and for identification of strategies towards societies and economies where material and energy commodities, by-products and wastes are produced, consumed and managed in more responsible and sustainable manners. LCA is standardised by the ISO 14040-44 (ISO, 2006a, 2006b) as a methodology aimed at evaluating the environmental burdens associated with a product, process or activity by identifying and quantifying the material and energy flows throughout the system, as well as at identifying and evaluating opportunities for improvements.

In this study, the potentiality of the MWH technique also enhanced using citric acid for the remediation of heavily hydrocarbon-contaminated salty sediments from Augusta Bay was assessed at the lab-scale. LCA was also applied to investigate the environmental sustainability of the MW remediation treatment compared to an alternative decontamination method. The results are expected to provide an effective and eco-friendly technique in future engineering applications by means of meeting the severe Italian law limit required in remediation activities of sediments contaminated with hydrocarbons.

2. Materials and methods

2.1. Sediment sampling, characterisation and preparation procedures

For the experiments, several samples of superficial bottom sediments were dredged from a highly impacted zone of Augusta Bay (Sicily, Southern Italy), in front of the petrochemical plants in May 2016. After the collecting procedures, the sediment samples were stored at a temperature of $-4\ ^\circ C$, then slowly air-dried for 72 h, sieved at 2 mm

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