



# Remediation of hydrocarbon polluted soils using 2.45 GHz frequency-heating: Influence of operating power and soil texture on soil temperature profiles and contaminant removal kinetics



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## ABSTRACT

2.45 GHz frequency heating treatment of diesel-contaminated soils was applied to investigate the effects of operating power and soil texture on soil temperature profiles and contaminant removal kinetics. Four soil size aggregate fractions, corresponding to medium (200–350  $\mu\text{m}$ ) and fine (75–200  $\mu\text{m}$ ) sand, silt (10–75  $\mu\text{m}$ ) and clay (<4  $\mu\text{m}$ ) were artificially contaminated with diesel fuel and remediated with *lab-scale* apparatus for different lengths of time. Results suggest that soil texture significantly influences the contaminant removal kinetics and that thermodynamic and dielectric properties of soils have a major effect on the final temperature reachable during the heating processes. Minimal contaminant residual concentrations (<400  $\text{mg kg}^{-1}$ ) were achievable only for sandy soils, for which the highest contaminant removals (>90%) were reached. The power low kinetic model showed a quite good correlation ( $r^2 > 0.976$ ) with the experimental data for residual concentration at all operating powers and for all soils investigated. Contaminant rate decay ( $k$ ) variation observed for the different soils indicates that contaminant removal processes strictly depend both on thermal properties and specific surface area of soil regulating the adsorption–desorption phenomena related to the decontamination treatment. Especially for sandy soils, microwave heat treatment could be a better choice for remediation of diesel polluted soils compared to several other biological, chemical–physical or conventional thermal treatments due to its removal efficiency.

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## 1. Introduction

Hydrocarbon products including gasoline, diesel or lubricants can be released through accidents, managed spills, or as unintended by-products of industrial, commercial or private actions, causing local and diffuse pollution in the environment (Pinedo et al., 2013).

Among hydrocarbons, petroleum-derived diesel is composed of approximately 75% saturated hydrocarbons (primarily paraffins), and 25% aromatic hydrocarbons (including naphthalenes and alkylbenzenes), and it represents a permanent source of soil pollution (Pazos et al., 2011). In the search for effective and economically competitive diesel-polluted soil remediation strategies, several options have been proposed including chemical–physical or biologic treatments (Do et al., 2009; Dong et al., 2013; Fernández et al., 2011; Khalladi et al., 2009; Mena et al., 2014; Pazos et al., 2011; Tomei et al., 2013; Tsai et al., 2010). However, these treatments may be ineffective, too expensive or lengthy (Chien, 2012). Ex-situ conventional thermal desorption was also successfully applied to remedy contaminated soil (dela Cruz et al., 2014; Falciglia et al., 2011b; Li et al., 2014; Thuan and Chang,

2012), however it may be expensive due to the energy costs (Falciglia et al., 2013).

Compared to other remediation methods, microwave (MW) ( $f = 2.45$  GHz) heating remediation has advantages including simplicity, safety, flexibility and cost-effectiveness since it offers the potential to significantly reduce treatment times, risk of contamination and costs due to the direct interaction of microwaves with the soil and its ability to overcome heat and mass transfer limitations. In conventional heating systems, the energy is transferred through conduction, convection and radiation, while in MW heating, energy is supplied directly to soil by molecular interaction with the electromagnetic field generated. Therefore, the components of the soil are heated individually and instantaneously, overcoming limits imposed by material heat transfer properties (Pereira et al., 2014).

Previous studies (Falciglia et al., 2013; Gomes et al., 2013; Huang et al., 2011; Li et al., 2009; Lin et al., 2010; Liu and Yu, 2006; Pereira et al., 2014; Robinson et al., 2009, 2012; Yuan et al., 2006) have shown that MW remediation has the potential to remove polar and semi-polar organic pollutants from soil.

The key factor of the remediation process is represented by the mechanism due to a partial dissipation of the electromagnetic field energy ( $f = 0.3$ –300 GHz) and its conversion into heat, avoiding the limitations of conductive heating phenomena of conventional thermal

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desorption treatment. Therefore, the majority of the absorbed microwave power is converted to heat and the rate of heat generated ( $\Delta T \Delta t^{-1}$  [ $^{\circ}\text{C min}^{-1}$ ]) is given by the following equation (Clark et al., 2000):

$$\frac{\Delta T}{\Delta t} = \frac{P}{c_p \cdot \rho} = \frac{\omega \cdot \epsilon_0 \cdot \epsilon'' |E|^2}{c_p \cdot \rho} \quad (1)$$

where:

- $P$  is power absorbed per unit volume ( $\text{W m}^{-3}$ );
- $\omega$  is the angular frequency ( $\omega = 2\pi f$ ,  $f$  microwave frequency [Hz]);
- $\epsilon_0$  is the permittivity of free space ( $8.85 \cdot 10^{-12} \text{ F m}^{-1}$ );
- $\epsilon''$  is the dielectric loss factor (–);
- $E$  is the internal electric field ( $\text{V m}^{-1}$ );
- $c_p$  is the heat capacity of the medium ( $\text{KJ kg}^{-1} \text{ }^{\circ}\text{C}^{-1}$ );
- $\rho$  is the density of the medium ( $\text{kg m}^{-3}$ ).

$\epsilon'$  denotes the electric energy storage capacity of the medium, while  $\epsilon''$  denotes the ability of the medium to convert electromagnetic energy into heat due to the dielectric polarization of the particles in an alternating electric field.  $c_p$  is defined as the amount of heat per unit mass of the medium required to raise the temperature by  $1^{\circ}\text{C}$ .

The dielectric and thermodynamic properties, and therefore the features of the polluted soil to be treated, are also important parameters in determining the penetration depth ( $D_p$ ), that represents the ability of the electromagnetic waves to penetrate into the medium.  $D_p$  is given by the following expression (Acierno et al., 2004):

$$D_p = \frac{\lambda_0}{2\pi} \cdot \frac{\sqrt{\epsilon'}}{\epsilon''} \quad (2)$$

where  $\lambda_0$  is the wavelength of the radiation in the free space (m) and  $\epsilon'$  is the dielectric constant (–).

For a full-scale application of MW remediation, a schematic design of a microwave heating system has recently been reported by Ha and Choi (2010), Barba et al. (2011) or Chien (2012).

Based on the above considerations, it is clear that dielectric and thermodynamic parameters of soils, in a wide range for different soil textures (i.e., sand, silt or clay), strongly influence the MW propagation phenomena and the soil temperature increasing and hence the effectiveness of the remediation system.

In addition, it has been shown that the characteristics of soil influence contaminant–soil interaction, adsorption phenomena, and contaminant removal efficiency during the decontamination processes due to the soil texture and consequently the specific surface area influences the interaction of the compounds with the soil and the availability and the remediation of hydrocarbon pollutants (Amellal et al., 2001; Gu et al., 2012; Huang et al., 2011; Lee et al., 2002).

Therefore, knowing the influence of the polluted soil nature and MW treatment operating parameters on soil temperature achievable and contaminant removal kinetics is essential for the applicability of the heating treatment.

In the present work, MW thermal desorption treatment of diesel-polluted soils with different soil textures was studied using an experimental bench-scale apparatus. The main goals were: (i) to assess the influence of power treatment, treatment time and soil texture on the soil temperature profiles, on the diesel residual contamination and thus on the percentage removal; (ii) to fit the contaminant residual concentration data with kinetic model equation in order to calculate the desorption parameters useful for studying the desorption process and for predicting the response of the decontamination treatment of different soils.

## 2. Materials and methods

### 2.1. Soil contamination procedure

Since soil surface area (which increases with decreasing particle diameter) influences significantly the contaminant soil interaction in adsorption and thermal desorption processes, model soils, free of anthropogenic contamination, with a wide range of specific surface areas were selected for the experiments. Specifically, four soil size aggregate fractions, corresponding to medium (200–350  $\mu\text{m}$ ) (MS), fine (75–200  $\mu\text{m}$ ) silica sand (FS), silt as silica flour (10–75  $\mu\text{m}$ ) (S) and clay as kaolin (<4  $\mu\text{m}$ ) (C) (Che.Mi.Fil. s.r.l., Verona, Italy) were used. Soils' properties are given in Table 1. Selected soils were artificially contaminated using commercial diesel fuel (Table 2) following the method reported in Falciglia et al. (2011b). After the contamination procedure, the soil samples were analysed by n-hexane ( $\text{C}_6\text{H}_{14}$ , purity 99% – Merck KGaA, Germany) extraction and subsequently gas chromatography for contaminant content before MW treatment. Contamination procedure was carried out in triplicates and mean and standard deviation values of adsorbed contaminant concentration ( $C_0$ ) were calculated.

### 2.2. Experimental apparatus and procedures

A MW thermal treatment was simulated using a bench-scale apparatus carried out by modifying a 2.45 GHz domestic microwave oven (Panasonic NN-GD458W Inverter – maximum power of 1000 W) (Fig. 1).

For the experiments, the selected contaminated soil samples (20 g) were placed into a quartz reactor (h 100 mm, internal  $\varnothing$  80 mm) in the centre-section of the oven cavity, where it was irradiated. The gas outlet section of the quartz reactor was connected to a condensing system, to a VOC capture system (activated carbon filters, Norit Italia S.p.A. – Ravenna, Italy), and then to a vacuum pump to remove the contaminated gas from the reactor.

Soil samples were treated for a time of either 5, 10, 18, 30 or 60 min, using applied power ( $P$ ) values, obtained from oven regulation, ranging from 250 to 1000 W. After a desired residence time the soil temperature was measured with a sheltered type-k thermocouple axially inserted up to the middle of the soil sample.

After treatment, soil samples were removed from the apparatus, cooled at room temperature ( $20^{\circ}\text{C}$ ) and stored in a dark room at  $4^{\circ}\text{C}$  prior to analysing. The thermal treatment procedure was carried out in triplicates and mean values of contaminant residual concentrations ( $C$ ) as a function of the treatment time were obtained for each selected power and soil.

For the calculation of  $C_0$  and  $C$  values, for each 20 g sample, a 2 g subsample was analysed for hydrocarbon concentration. The subsample was mixed with n-hexane in a Soxhlet extractor for 6 h. 5 mL of effluent was mixed with 2 mL of n-hexane in a separate funnel, stirred for 2 min, then left at rest for separation. The supernatant phase was mixed with internal standard (ISM-560 Ultra Scientific, USA) and analysed by gas chromatography.

Due to their high proportion in diesel fuel, n-alkane compounds ( $\text{C}_{10}$ – $\text{C}_{25}$ ) were chosen as representative components (Falciglia et al., 2011b; Khaladi et al., 2009), and their total concentration in spiked and treated soil samples was taken as that of diesel fuel and expressed as  $\text{mg/kg}_{\text{soil}}$ .

The concentration of n-alkanes in soil samples was measured by gas chromatography (GC) (Agilent Technologies 6890N) equipped with a mass spectrometer (Agilent Technologies 5975) using the US-EPA 8270-C method. A capillary column (HP-5, 30 m length  $\times$  0.32 mm ID  $\times$  0.25  $\mu\text{m}$  film thickness) was used. The GC was operated with a helium-carrier-gas flow rate of  $1.5 \text{ mL min}^{-1}$  and an oven temperature programme starting at  $40^{\circ}\text{C}$  (held for 4 min) and increasing at a rate of

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