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Development and Application of a Mathematical Model for the Assessment of the Treatability of Radionuclides Polluted Soils by Microwave Heating Stabilisation: Preliminary Results

Pietro P. Falciglia[a,*], Salvatore Cannata[b,c], Giuseppe Urso[b,c], Pietro Scandura[a], Antonio Triglia[c], Stefano Romano[c,d], Federico G. A. Vagliasindi[a]

[a]Department of Civil and Environmental Engineering, University of Catania. Viale A. Doria, 6 - 95125 Catania, Italy
[b]Department of Environmental Science, Informatics and Statisics, Cà Foscari University of Venice. Santa Marta, Dorsoduro 2137, 30121 Venice, Italy
[c]Laboratori Nazionali del Sud - INFN, Via S. Sofia, 62 - 95125 Catania, Italy
[d]Department of Physics and Astronomy, University of Catania. Viale A. Doria, 6 - 95125 Catania, Italy
ppfalci@dica.unict.it

Radionuclides polluted soils represent a major concern for environment and their remediation or stabilization are a key factor for human health. In recent years, treatments by MW heating have attracted great attention in environmental field, since it represents a novel and optimal approach for removal or stabilisation of organic pollutants and/or heavy metals. Therefore, in situ MW heating could represent an optimal choice for radionuclides polluted soils stabilisation. A huge effort has been made by several scientists in order to demonstrate MW applicability by means of lab-scale experiments, but very limited results were shown by pilot or full-scale activities, making unclear the limits and therefore the real applicability of in situ MW interventations. This lack makes essential the development of specific tools to plan full-scale MW treatments, in particular when large polluted soils volumes are involved.
In this work a mathematical model was developed and applied to understand the effects of a long period MW radiation on the variability of several physical soil parameters. For simulating, a treatment time of 1 month, a maximum investigated distance from the microwave source (antenna) of 4 m and a power of 4 kw for the microwave generation at 2.45 GHz were applied.
Preliminary results showed that in situ MW remediation of radionuclides polluted sandy soil is applicable for a limited distance of about 50 cm from the antenna and for a remediation time of 30 days. Powers higher than 4 kW or remediation time longer than 1 month should be investigated in order to successfully treat radionuclides polluted soils.

1. Introduction

Soil contamination caused by radionuclides is a serious problem worldwide and it currently represents a living matter (Antovic et al., 2012). Radionuclides are introduced in the environment following nuclear power plant accident or nuclear, military and scientific activity, resulting in both soils and groundwater pollution (Hu et al., 2010). Among radionuclides, isotopes able to generate gamma (γ)-radiation have high radiotoxicity and they are of major concern in the environment (Falciglia et al., 2012). Indeed, γ-radiation is composed of photons, which have neither mass nor electric charge, penetrates much further through matter than either α or β radiation, being thus biologically hazardous. Therefore, the remediation or the stabilisation of radionuclides polluted soils represent a key factor for human health.
Limited environmental restoration methods for radioactively contaminated sites have been recently studied: electrokinetic decontamination (Kim et al., 2011), soil washing (Lozano et al., 2011), phytoextraction (Mihalík et al., 2012), or cement based stabilisation/solidification (Falciglia et al., 2012).

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However, these treatments may be prohibitively costly, time consuming or environmentally unsustainable such as disposal in landfills if large areas of land are involved.

Thermal treatment was shown to be a very effective technique to remedy polluted soil (Falciglia et al., 2011-a) and in situ stabilisation by heating could represent an optimal choice due to its efficiency and costs. In particular, in recent years, treatments by MW heating have attracted great attention in environmental field, because it represents a novel and optimal approach for removal or stabilisation of organic pollutants and/or heavy metals.

MW heating was successfully applied to remove organic contaminants from soil matrix: PCBs (Huang et al., 2011; Liu et al., 2008), HCB (Yuan et al.,2006), TCE (Kawala and Atamanczuk, 1998), PAHs (Robinson et al., 2009), antibiotics (Lin et al., 2010) or crude oil (Li et al., 2009).

The stabilisation and immobilization of metal ions in soil and metal sludge through the microwave treatment are also reported (Hsieh et al., 2007; Jothiramalingam et al., 2010; Tai and Jou, 1999). This demonstrates that MW is efficient in inhibiting the leaching of metal ions from soil or sludge. In particular, Abramovitch et al. (2003) proposed MW energy for the in situ remediation of soils contaminated with toxic metal ions. Therefore, in situ MW heating could represent an optimal choice to stabilise radionuclides polluted soils, however its applicability has never been investigated. Furthermore very limited *lab*- or *pilot-scale* researches on MW have been carried out. Moreover an in situ MW application results in a changing of soil parameters (i.e.: internal electric field, temperature) very different from that achievable by means of a typical *bench-scale* experiment (Chien, 2012). This lack makes unclear the limits and therefore the real applicability of an in situ MW treatment.

In this work a mathematical model was developed and applied to understand the effects of a long period MW radiation on the variability of several soil physical parameters. Results will allow to define the real applicability of an in situ MW treatment for the remediation of radionuclides polluted soil.

2. MW heating theory

MW are a separate band of electromagnetic radiation with frequencies in the range of 300 MHz to 300 GHz. The key factor of the remediation process is represented by the mechanism due to a partial dissipation of the electromagnetic field energy and its conversion into heat necessary for the thermal desorption of the contaminants. In fact, the internal temperature distribution of a material, such as the soil, using conventional heating, is limited by its thermal conductivity, whereas in the case of MW radiation, the alternating electromagnetic field induces the rotation of the dipoles of water and other polar substances present in the soil. The intermolecular friction results in the generation of heat. Moreover, MW are absorbed by materials with a high dielectric loss factor (absorbing), while passing through the low loss (transparent) material, resulting in a selective, uniform and rapid heating. Therefore, heating times can be significantly reduced compared with those required when using conventional heating methods.

The majority of the absorbed microwave power is converted to heat within the materials and the rate of heat generated ($\Delta T\ \Delta t^{-1}$ [°C min^{-1}]) depends directly on the frequency of the applied electromagnetic field and on the dielectric properties of the treated medium:

$$\frac{\Delta T}{\Delta t} = \frac{P}{c_p \cdot \rho} = \frac{\omega \cdot \varepsilon_0 \cdot \varepsilon'' |E|^2}{c_p \cdot \rho} \tag{1}$$

where ω is the angular frequency ($\omega = 2\pi f$, f microwave frequency [Hz]); ε_0 is the permittivity of free space ($8.85 \cdot 10^{-12}$ F m^{-1}), ε' and ε'' are the real part (dielectric constant) and the imaginary parts (dielectric loss factor) of the complex permittivity, respectively; E is the magnitude of the internal electric field (V m^{-1}); c_p is the heat capacity of the medium (kJ kg^{-1} °C^{-1}) and ρ is its density (kg m^{-3}).

The real part of the complex permittivity ε' denotes the electric energy storage capacity of the medium, while the imaginary parts of the complex permittivity ε'' can be considered as the ability of the medium to convert electromagnetic energy into heat due to the dielectric polarization of the particles in an alternating electric field. The parameters ε' and ε'' are temperature dependent. The dielectric properties are also important parameters in determining the internal electric field (E_z) variation generated by MW penetration (being E_0 the intensity of the incident electric field):

$$E_z = E_0 \cdot e^{-\frac{z}{D_p}} \tag{2}$$

where D_p is the penetration depth (m), that represents the ability of the electromagnetic waves to penetrate into the medium. In particular, D_p is defined as the distance from the emission point at which the power (P) drops to 0.37 from its value at the emission point. For low loss dielectric materials such as soil, D_p is given by the following expression:

$$D_p = \frac{\lambda_0}{2\pi} \cdot \frac{\sqrt{\varepsilon'}}{\varepsilon''} \quad (3)$$

where λ_0 is the wavelength of the radiation in the free space (*m*). On the other hands, different types of soil significantly influence the processes of heating propagation (conductivity and dielectric) (Falciglia et al., 2011b). Knowing the temporal variability of the above reported quantity represents a key factor in evaluating the MW heating applicability in different cases (i.e.: type of soil, soil moisture, contaminant).

3. Modeling

MW process has been modelled with coupled mono-dimensional transient equations of energy taking into account the interaction between electromagnetic field and matter (soil). We assume that the temperature and the electromagnetic fields are axisymmetric with respect to the axis of the hole in which the antenna is inserted. This make appropriate the use of a polar coordinate systems to study the evolution of the evolution of the physical quantities. More specifically, the evolution of soil temperature (T) with time (t) and distance (d) from the antenna was modeled by numerically solving the heat equation, in polar coordinates, with a source term, reported below (Barba et al., 2012):

$$\rho c_p \frac{\partial}{\partial t} T = -\frac{\partial^2}{\partial d^2} T + \dot{Q} \quad (4)$$

$$\dot{Q} = \frac{1}{2} \omega \epsilon_0 \varepsilon'' E^2 \quad (5)$$

where k is the thermal conductivity, Q is the term derived by by Eq (1) – Eq (3) that takes into account the increasing of T due to the MW energy dissipation. Other terms take into account the thermal conduction phenomena.

Equation 4 has been solved numerically by finite differences approximations in an annulus of external radius R_e and internal radius R_i. The latter is equal to the radius of the hole which is much smaller than R_e. The value of the external radius is chosen large enough such that no appreciable temperature variations are observed at $d=R_e$. Boundary conditions are imposed by assuming the vanishing of the derivative of the temperature with respect to d both at $d=R_e$ and at $d=R_i$. The boundary condition at $d=R_i$ is justified by the assumed axisymmetric, while that at $d=R_e$ by the fact that R_e is large, as previously explained.

Modeling was performed considering a polluted sandy soil as irradiation medium and the schematic of Figure 1 shows a sketch of the simulated experiment. The maximum investigated distance (d) was of 4.00 m and the maximum remediation time investigated was 30 d. The value of incident electric field E_0 used was 1,000 V m^{-1} (Magnetron power, P = 4 kW). Other parameters used for the simulation are given in Table 1. Mathematical model results allow to predict variation of T, D_p, E, ε' and ε'' with time (t) and distance (d).

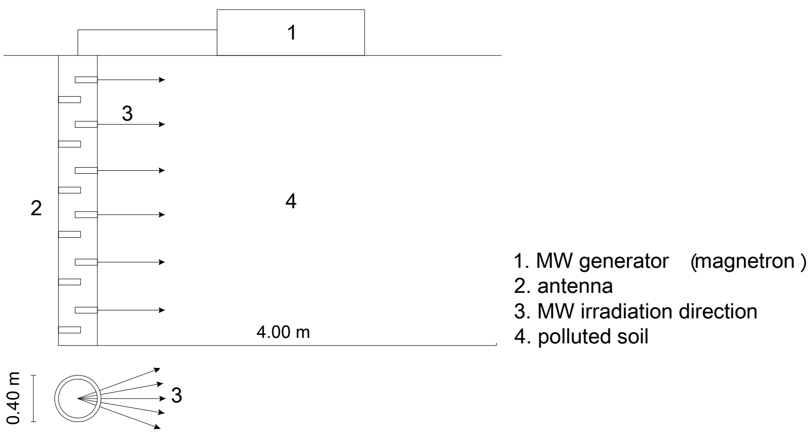

Figure 1: Schematic of the in situ MW treatment apparatus adopted for modelling

Table 1: Parameters used for the modelling

Parameter	*Value*	*Parameter*	*Value*
Soil density, ρ (kg m^{-3})	2,000	Incident electric field, E_0 (V m^{-1})	1,000
Soil heat capacity, C_p (kJ kg^{-1} $°C^{-1}$)	800	Initial soil temperature (°C)	20
Soil thermal conductivity, k (W m^{-1} $°C^{-1}$)	0.8	Temporal step for calculation (s)	0.2
Initial soil moisture (%).	14	Diameter of the antenna (m)	0.4
MW wavelength, λ_0 (m)	0.125		

Mathematical model results allow to predict variation of T, Dp, E, ε' and ε" with time (t) and distance (d).

4. Results and discussion

The variation of T, D_p, E, *ε'* and *ε"* with time (t) and distance (d) obtained by the simulation is reported in Figure 2 and 3.

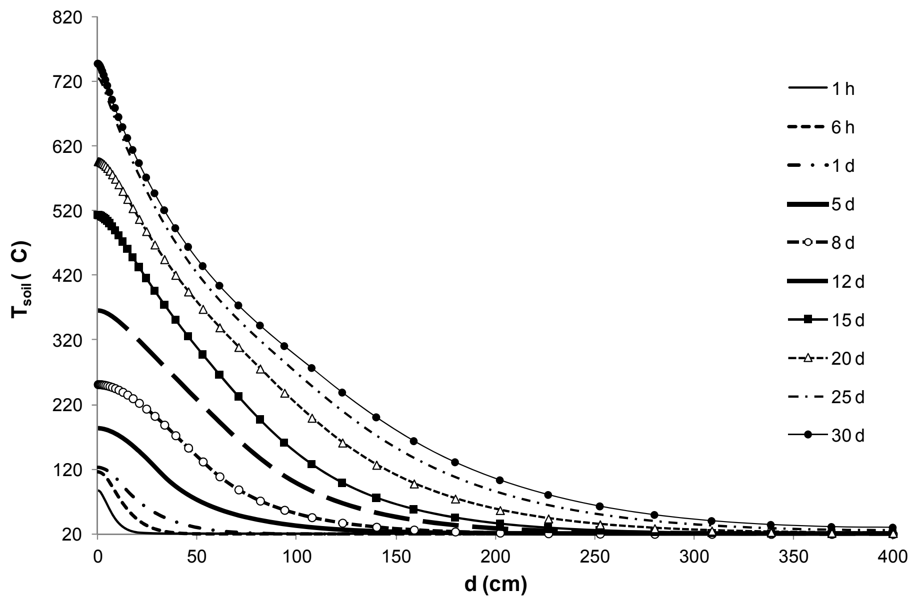

Figure 2: Variation of soil temperature with time (t) and distance from the antenna (d)

Results show a decrease of *T* when *d* increases due to the spatial decreasing of the electric field (*E*) penetrating the soil medium. Overall, the heat absorbed by soil rose resulting in an increasing of *T* with the remediation time.

All curves are characterized by a particular shape due to the overlapping of both MW absorbing phenomena and conduction heat transfer processes. In addition, it is shown that a *T* higher than 100 °C is achievable for a distance up to about 2.5 m and for the maximum remediation time simulated.

This behaviour is due to the progressive changing of dielectric properties (*ε'* and *ε*) of the soil that results in an increase of the deep penetration (D_p) value and, consequently, in a progressive penetration of the microwave (as electric field, *E*) into the soil (Figure 3).

Therefore, even if after a selected remediation time a soil layer is not more able to absorb MW heat due to its dielectric properties, the progressive spatial variation of *E* results in a constant increase of *T* for increasing *d* value. This highlights the possibility to heat polluted soil also for high distance from the MW source.

Obtained results are in agreement with the limited studies carried out (Kawala and Atamanczuk, 1998) where a pilot-scale MW apparatus was used to simulate a three days-soil treatment.

In order to achieve a metal immobilisation, a soil temperature higher than about 500 °C is needed (Hsieh et al., 2007), therefore the MW in situ application for a 30 days-remediation application of radionuclides polluted sandy soil is limited to a maximum treatable distance of about 50 cm.

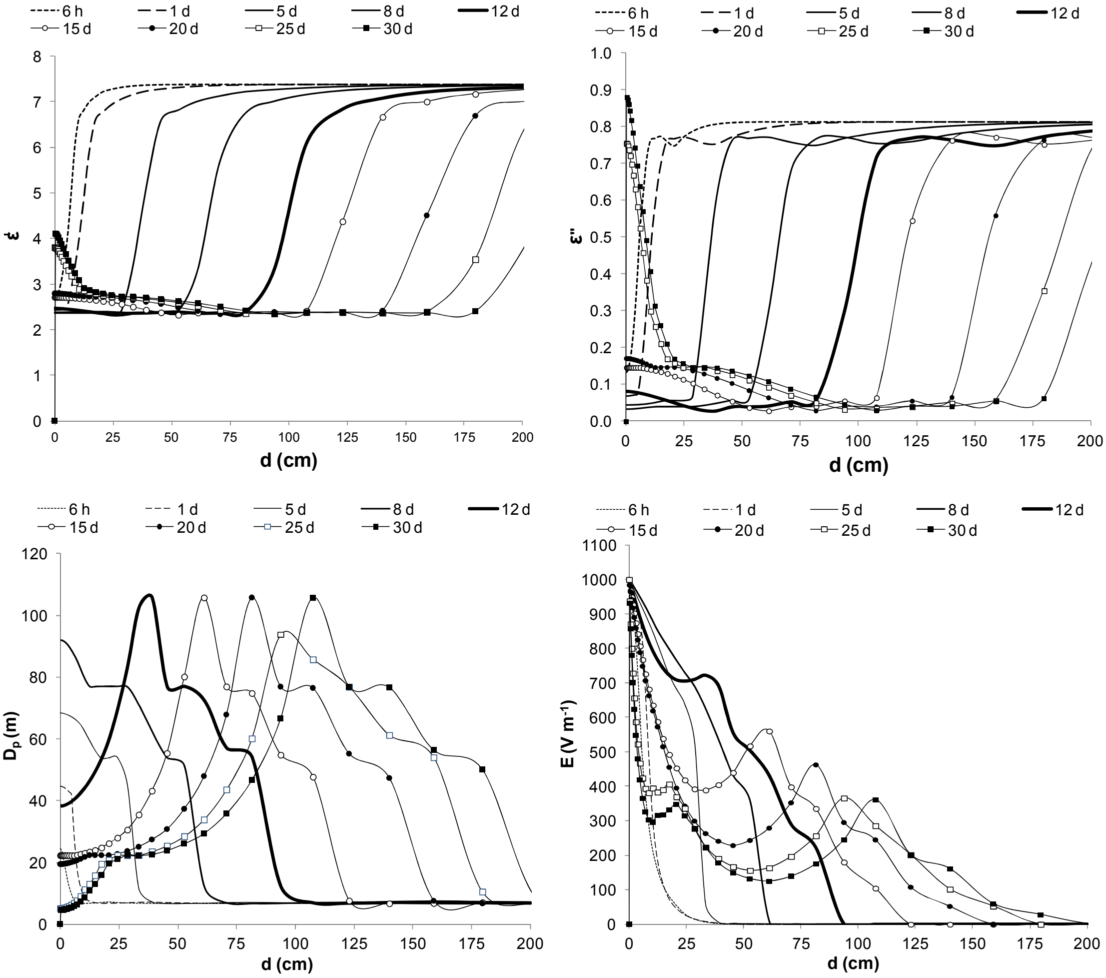

Figure 3: Variation of parameters D_p, E, ε' and ε'' with time (t) and distance from the antenna (d)

5. Conclusions

In this paper we used a mathematical model to assess the treatability of radionuclides polluted soils by microwave heating.

The increasing of soil temperature due to MW irradiation heavily influences the changing of relevant process parameter such as *ε′* and *ε″* with treatment time and distance during all the irradiation period resulting in a progressive increase of the ability of the electric field to penetrate the soil.

The progressive penetration of the MW into the medium generates, after a treatment of 30 days, a soil temperature higher than the initial value for a maximum distance from the MW source of about 4 m making the investigated treatment a suitable remediation technique.

In situ MW remediation of radionuclides polluted sandy soil is applicable for a limited distance of about 50 cm from the antenna if a remediation time of 30 d is considered. Powers higher than 4 kW or remediation time longer than 30 d should be used in order to successfully treat radionuclides polluted soils. Dielectric materials, able to significantly increase the soil temperature or specific chemical materials able to improve the soil stabilisation processes, could be adopted to make cost-effective the MW treatment.

The mathematical approach presented in the work is able to describe MW remediation processes, thus it can be considered a useful planning tool.

Complete research activity will include modeling using different operating conditions and types of soil.

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