### **RTICLE IN PRESS**

#### Chemosphere xxx (2016)  $1-9$  $1-9$



Contents lists available at ScienceDirect

### Chemosphere



journal homepage: <www.elsevier.com/locate/chemosphere>

## Scale-up of the electrokinetic fence technology for the removal of pesticides. Part I: Some notes about the transport of inorganic species

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- In EK soil remediation technology, size of the experimental setup matters.
- Same processes occurring, different observations made because of the dimensions.
- Increase in the intensity over the experiment due to the conductivity and pH.
- Depletion of ions from soil and concentration in the nearness of electrode wells.
- Rapid dynamic response: almost no relevant changes after a 15-day treatment.

#### **ARTICLE INFO**

Article history: Received 23 August 2016 Received in revised form 23 September 2016 Accepted 24 September 2016 Available online xxx

Handling Editor: Dr. E. Brillas

Keywords: Electrokinetic fences Soil remediation 2,4-D Oxyfluorfen Scale-up



### **ABSTRACT**

This work describes the application electrokinetic fence technology to a soil polluted with herbicides in a large prototype containing  $32 \text{ m}^3$  of soil. It compares performance in this large facility with results previously obtained in a pilot-scale mockup (175 L) and with results obtained in a lab-scale soil column (1 L), all of them operated under the same driving force: an electric field of 1.0 V cm<sup>-1</sup>. Within this wide context, this work focuses on the effect on inorganic species contained in soil and describes the main processes occurring in the prototype facility, as well as the differences observed respect to the lower scale plants. Thus, despite the same processes can be described in the three plants, important differences are observed in the evolution of the current intensity, moisture and conductivity. They can be related to the less important electroosmotic fluxes in the larger facilities and to the very different distances between electrodes, which lead to very different distribution of species and even to a very different evolution of the resulting current intensity. 2-D maps of the main species at different relevant moments of the test are discussed and important information is drawn from them. Ions depletion from soil appears as a very important problem which should be prevented if the effect of natural bioremediation and/or phytoremediation on the removal or organics aims to be accounted.

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<http://dx.doi.org/10.1016/j.chemosphere.2016.09.113> 0045-6535/© 2016 Elsevier Ltd. All rights reserved.



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### 1. Introduction

Nowadays, soil pollution is becoming one of the most important environmental problems for Humankind with important consequences in the availability and quality of water reservoirs for human supply. This issue should be faced from a multidisciplinary approach (civil, chemical and environmental engineers, biologist, geologist …), because of the complexity of the mechanisms involved in the reactivity and transport of pollutants, either those occurring naturally in the environment or those promoted by humans with the application of remediation technologies.

When electrochemical technologies are to be applied, this complexity becomes even worse, because of the interactions of those natural and anthropogenic processes with the processes promoted electrochemically. These electrochemically induced processes involve [\(Rodrigo et al., 2014](#page--1-0)):

- $-$  the important changes in the pH in the nearness of the electrodes (because of the electrolysis of water),
- transport processes of different species (pollutant or not) driven by the electric field applied (including electromigration, electrophoresis and electro-osmosis) and
- an increase in the temperature, caused by the ohmic resistance of soil.

All these processes interact among each other and with other chemical and physical processes (such as ion exchange, precipitation, volatilization, etc.) producing changes that, when properly engineered, contributes to the removal of pollutants from soil.

Many references can be found in the literature about all these processes ([Ribeiro et al., 2005; Reddy et al., 2009; Alcantara et al.,](#page--1-0) [2010, 2012; Gomez et al., 2010; Pazos et al., 2010; Reddy et al.,](#page--1-0) [2011; Ribeiro et al., 2011; Gomes et al., 2012; Cameselle and](#page--1-0) [Reddy, 2013; Vieira dos Santos et al., 2016](#page--1-0)). Most of them are carried out at the lab-scale because this is the level in which the process can be better characterized, with a higher accuracy in details, allowing even a good mathematical description of the system with lots of experimental results that allow researchers formulating the models and fitting their parameters. Then, processes need to be scaled-up, and here a problem arises. Financial support for doing large-scale studies is not easy to be obtained without the participation of companies, and in this case, they are interested in keeping the information to get a benefit and NDAs prevents a good diffusion of results. In addition, information taken from the fullscale restoration of polluted soil is very important, but it lacks the accuracy of the data obtained in research programs.

At this point, it is worth to say that scale-up in disciplines such as chemical or environmental engineering does not only mean "make things with a larger size". Unfortunately, this is a very common mistake, typically associated to researchers or professionals not directly related to these technological disciplines. In contrast, this concept also involves a deeper understanding of the processes, which have been previously characterized at lower scale with smaller devices (for which operation conditions can be more easily controlled and details about processes can be more easily elucidated) (López-Vizcaíno et al., 2016). Thus, the key in scale-up is the definition and understanding of the "controlling mechanisms" in full-scale, rather than the study of the fundamentals of a process, which for sure, in a lower scale and with more controlled conditions can be obtained with a higher accuracy giving very valuable data. However, rather often, these data cannot explain the real behavior of the system and here is where this important concept arises. In the case of soil remediation, there is a coexistence of three electricity-driven processes (electrokinetic, electrochemical and electric heating processes), which also coexists with other chemical processes (such as ion exchange reactions such as precipitations, etc.) and physical processes (hydraulic fluxes, evaporation, etc.). The prevalence of any of these mechanisms over the others can lead to a very different performance of the technology ([Alshawabkeh](#page--1-0) [et al., 1999; Yuan et al., 2006, 2007; Karagunduz et al., 2007;](#page--1-0) [Buchireddy et al., 2009; Li et al., 2016\)](#page--1-0) and it is worth to evaluate how the size of the experimental setup influences on these mechanisms if results carried out at small scale aims to be extrapolated for full-scale applications.

This work reports important information about the scale-up of electrokinetic fence (EKF) technology for the remediation of soil polluted with herbicides, by comparing results obtained in a prototype of 32  $m<sup>3</sup>$  with those obtained in a mockup of 175 L. These lower-scale results were obtained in different works previously published in the literature ([Risco et al., 2015, 2016e](#page--1-0)), within a wider-scope research program in which different electrodes placement were compared for the efficient removal of pesticides from soil [\(Risco et al., 2016a, 2016b, 2016c, 2016d\)](#page--1-0). At this point, a preliminary work about scale-up informs about the many inputs that should be accounted for proper scale-up study  $(López-Vizcaíno$ [et al., 2016](#page--1-0)). Within this general scope, Part I is focused on the description of the processes that affect to inorganic species contained or produced in soil during the application of the remediation technology.

### 2. Materials and methods

#### 2.1. Materials

The soil used in this work is provided from a region of Castilla la Mancha (Spain), with important agrarian activities. This soil has been used in others works carried out to our research group [\(Risco](#page--1-0) et al., 2015, 2016a, 2016b, 2016d, 2016e; López-Vizcaíno et al., [2016\)](#page--1-0). It is classified as low plasticity soil, according with ASTM D2487 ([International, 2006\)](#page--1-0) and ASTM D4318 ([International,](#page--1-0) [2010a](#page--1-0)) and as silty loam within the textural classification of USDA ([Staff, 1993](#page--1-0)) (clay = 4.9%, silt = 68.2% and sand = 26.9%). The mineralogical composition is described in detail elsewhere (López-[Vizcaíno et al., 2016](#page--1-0)).

Two different commercial pesticides have been used to simulate an accidental spill: ESTERON 60 supplied by Dow AgroSciences, and FLUOXIL 24 purchased from CHEMINOVA AGRO. The active component of ESTERON 60 is the 2,4-dichlorophenoxyacetic acid  $(2,4-D)$  with a composition of 60%  $(v/v)$ , in an emulsified solution with calcium dodecylbenzenesulfonate. FLUOXIL 24 is composed by  $24\%$  (v/v) of Oxyfluorfen, non-polar and hydrophobic herbicide, dissolved in aqueous solution by the emulsifier action of xylene (59%), cyclohexanone (13%) and calcium dodecylbenzenesulfonate  $(4\%)$ .

#### 2.2. Soil remediation prototype

The EK remediation test was carried out in an especial facility built in the Institute of Chemical and Environmental Technologies (ITQUIMA) of the UCLM (Ciudad Real, Spain), which consists of two electrokinetic soil remediation prototypes, with soil-treatment capacities of 16 and 32  $m^3$ , respectively, of which only the second reactor has been used in this work. Dimensions of this later prototype are 2 m of height and a square plant of 16 m<sup>2</sup> (4 m  $\times$  4 m). Specific parameters construction of reactor were described in literature previously ([L](#page--1-0)ó[pez-Vizcaíno et al., 2016](#page--1-0)). Fig. SM-1 shows a scheme of the plant and section of the cell.

The electrode configuration selected to be studied in this prototype corresponds to an electrokinetic fence (EKF), using a sequence of six alternating electrodes (three cathodes and three

Please cite this article in press as: López-Vizcaíno, R., et al., Scale-up of the electrokinetic fence technology for the removal of pesticides. Part I: Some notes about the transport of inorganic species, Chemosphere (2016), http://dx.doi.org/10.1016/j.chemosphere.2016.09.113

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