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Research article

Electrokinetic remediation of soils contaminated by potentially toxic metals: Dedicated analytical tools for assessing the contamination baseline in a complex scenario

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ABSTRACT

In order to assess the capabilities of a remediation technology, and to judge of its efficacy, it is necessary to evaluate the initial average contamination level of the soil, an operation that can be difficult because of the inhomogeneity of the contamination itself. The goal is even more challenging when different contaminants are present, greatly differing both in terms of nature and of concentration. By referring to an industrial site contaminated mainly by As, Cd, Cu, Pb, Sb, Tl and Zn, we present a new approach for the necessary processing of sampling data, in order to establish the pre-intervention baseline: an estimate of the average contamination has been obtained through a suitable integration of the volume underlying the distribution curve of each contaminating species. This information, otherwise not accessible by means of sampling of discrete points, is useful in evaluating the effectiveness of the remediation technology under investigation, and can also be considered for other reclamation approaches as well. Since “chemometrically acceptable” results are typically achieved by increasing the number of samples (with related analytical investments), the proposed approach can help keep low these ancillary costs, while providing results that are more reliable.

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1. Introduction

Potentially toxic metals, deriving from industrial activities or from mining sites, are quite common contaminants, especially in industrial sites, and the resulting soil contamination is one of the most critical forms of environmental pollution, due to the serious effects that these contaminants exert on human health and ecosystems (Duruibe et al., 2007). In addition to their own toxicity, once dispersed in the soil, these metals and their compounds may remain bioavailable for long periods or migrate towards aquifers, thus spreading the contamination to other environmental matrices. Various methods are known for the reclamation of soils and groundwaters contaminated by potentially toxic metals (Hashim

et al., 2011), yet the technologies currently used are extremely invasive, resulting in a low level of environmental and social sustainability (US EPA, 2006a). Even though the soil washing still remains the common choice, the ElectroKinetic Remediation Technology (EKRT) probably represents the most promising solution, being an *in situ* process potentially able to allow results similar to *on-site* and/or *off-site* interventions, but with substantially higher levels of sustainability and acceptability (Vocciante et al., 2016).

As a remediation approach, the application of electric fields of suitable intensity through a given soil matrix leads to the mobilization of the target contaminants and their migration toward suitable electrode wells, which represent also the way for allowing the extraction of the pollutants, through the periodic collection of the process water (Acar et al., 1995). The mobilization of the contaminants, which is the main problem of this remediation technique, requires the addition of proper compounds that react with the compounds of concern (Ottosen, 2014). To improve the efficiency of the remediation approach, an effective strategy consists in the preliminary saturation of the soil with a suitable electrolyte

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solution (Vocciante et al., 2017), in order to facilitate the mobilization of the target species (under the action of the electric field) by means of complexation phenomena.

In previous works, we dealt with the set up (Rosestolato et al., 2015) and then the environmental assessment (Vocciante et al., 2016) of an innovative EKRT configuration for the remediation of a multi-metal-contaminated real site. We discussed the enhancements introduced, highlighting and validating their peculiarities, which result in a greater effectiveness as well as a reduction of the environmental footprint of the intervention at the same time.

As anticipated in the above-cited papers, the modified EKRT approach may prove effective in dealing a highly complex contamination by potentially toxic metals, irrespective of the nature of the species and of their concentration. In addition, we showed how an advanced water management system, preferably coupled with a reliable wastewater recovery system, is fundamental to avoid the waste of water resources, thus allowing to keep down the costs as well as the ecological footprint related with the implementation of this technology and to maximize its benefits (Vocciante et al., 2017).

To further increase the eco-sustainability, the additional coupling with alternative sources of electricity like solar (Vocciante et al., 2016) or wind (Souza et al., 2016) is under examination.

In this contribution, we present an original approach for the necessary processing of the sampling data, in order to establish the pre-intervention baseline. The US Environmental Protection Agency has provided a general non-mandatory guidance on the development of Data Quality Objectives (DQOs) related with operations of environmental data collection (US EPA, 2006b; 2006c). It is generally assumed that the quality of data pertaining to a contaminated site is primarily determined by the nature of analytical methods used to collect the information. However, the major source of uncertainty in environmental data sets is due to sampling variability, as a direct consequence of the heterogeneity of environmental matrices; actually, the uncertainty related with the sampling procedure can be up to 90% or more (Crumbling et al., 2001). Calculation programs exist that allow determining the number of samples necessary for estimating a given average concentration with the desired accuracy; however, they rely upon simple statistical equations (Gilbert et al., 2001), which involve big approximations, like a random sampling and the assumption that the matrix to be sampled is homogeneous.

Since at least the second hypothesis is hardly verified in real cases, we are presenting an alternative approach, by showing how a suitable integration of the underlying volume of the contamination distribution curve of a given contaminant can allow for a better estimate of the average contamination. This information, which is not accessible directly with a satisfying degree of confidence from a discrete sampling, can be useful in evaluating the effectiveness of the remediation technology.

2. Materials and methods

2.1. Site characteristics

The contaminated soil was collected from a disused industrial site located in the southern part of Italy, once subjected to various productive activities, including the extraction/production of zinc from sphalerite (the chief ore for this metal), which determined a complex contamination by potentially toxic metals (Vocciante et al., 2016). Table 1 summarizes the specifications of the site of reference.

From a lithostratigraphic point of view, the site is characterized by the presence of three main horizons (Table 2):

The data considered in this paper, and discussed in what follows,

were obtained by analysing the samples taken during the preparation of the test field for the EKRT application discussed in (Vocciante et al., 2016, 2017), and are a proof of how the stage of preparation of the test field itself may be usefully exploited to better characterize the soil before the intervention.

In fact, during the preparation of the test field, a number of soil samples has to be extracted, because of the creation of the different wells, which can be usefully exploited for the execution of further analytical investigations. 35 samples were obtained in correspondence with the electrode housings (21 anodes and 14 cathodes, distributed along 5 parallel rows of 7 electrodes). Additional 24 samples were obtained during the realization of the wells dedicated to monitoring and/or dewatering operations, which were arranged in 4 rows of 6, as schematized in Fig. 1.

In correspondence with each survey, soil samples were taken and subjected to analytical characterizations. Different aliquots were collected: from the superficial layer, from 0.0 to 0.5 m (which was then completely removed from the test field, being substantially represented by concrete paving), from the intermediate soil portion, comprised between 0.5 and 1 m, and then from the deeper portion, representative of the subsequent 2 m (from –1 to –3 m). In the following, only this latter portion will be discussed.

2.2. Analytical characterization

The analytical determinations, carried out by a certified private laboratory (LabAnalysis, on behalf of Syndial, ENI group), focused on both physical parameters, such as pH, redox potential, humidity, density (see Table 1), and the measurement of concentrations of the various chemical species (metals, chlorides, organic carbon, hydrocarbons). In some cases, the soil samples were also subjected to speciation (following the Tessier's protocol (Tessier et al., 1979) – additional data have been provided as Supplementary material), in order to identify the forms in which the various metal species are present (this has interest since it determines the actual availability of the specie, which may even be not easily removable). Table 3 reports the analytical values (in terms of total concentration, mg kg⁻¹ d.w.) obtained for the analysed metal species; values exceeding the limits laid down by the Italian Legislative Decree no. 152/06 (Annex 5 of Title V, Part IV, Table 1 B: land for commercial/industrial use) have been underlined.

Data in Table 3 show how the contamination by heavy metals is actually limited to seven species: As, Cd, Cu, Pb, Sb, Tl and Zn (in alphabetic order). The considered soil layer appears heavily contaminated, with exceedances also more than 10 times higher than the legal limits. In addition, the contamination is distributed in a very heterogeneous manner, as indicated by the values of the standard deviation %, which in some cases is even higher than 100%.

2.3. Determination of the contamination baseline

The data produced by an environmental monitoring are always the result of the analysis of a limited number of samples; although the analytical uncertainty is minimized by conventional laboratory analysis, sampling uncertainty is generally not addressed because of the high per-sample costs. As a result, a low-sampling density is often considered, although the numerical values obtained are subsequently used to estimate the contamination values of the entire site. Consequently, the obtained mean values are always a quite rough estimate of the true value and may deviate considerably from the real scenario of the existing contamination.

Moreover, there are numerous sources of uncertainty (errors) in the environmental data itself:

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