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Assisted Natural Remediation of a Trace Element-Contaminated Acid Soil: An Eight-Year Field Study

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ABSTRACT

There are many remediation techniques for organic contaminated soils, but relatively few of them are applicable to trace element-contaminated soils. A field experiment was carried out to investigate assisted natural remediation (ANR) of an acid soil contaminated by As, Cd, Cu, Zn and Pb using one inorganic amendment, sugar beet lime (SL), and two organic amendments, biosolid compost (BC) and leonardeite (LE). The experiment was arranged in a completely randomized block design with four treatments in three replicates: 1) a non-amended control (NA); 2) SL amended at 30 Mg ha^{-1} year^{-1}; 3) BC amended at 30 Mg ha^{-1} year^{-1} and 4) LE amended at 20 Mg ha^{-1} year^{-1} plus SL amended at 10 Mg ha^{-1} year^{-1} (LESL). The amended plots received two doses of each amendment (DO2): one in October 2002 and another in October 2003. The plots were then divided in half into two subpolts and one subplot received another two doses of the same amendments (DO4) in October 2005 and October 2006. In 2011, the pH values of the amended soils were greater than that of the NA soil, with the SL-amended soil showing the highest pH. Total organic carbon (TOC) was also greater in the amended soil, and greater with DO4 than with DO2. Amendments reduced the concentrations of 0.01 mol L^{-1} $CaCl_2$-extractable Cd, Cu and Zn, especially in the SL-amended soil. Plant cover of colonizing vegetation was enhanced by amendments, but was independent of amendment doses. Changes in soil properties from 2003 to 2011 showed that the first amendment application of DO2 caused a high differentiation between all the amendment treatments and the NA treatment. After the second application of DO2, soil pH and TOC continued increasing slowly. Further two applications of amendments (DO4) caused differences only in the organic treatments. Plant cover increased over time in all the treatments including NA. It could be concluded that the slow and steady natural remediation of this soil could be enhanced by amendment application (ANR).

Key Words: amendment application, $CaCl_2$ extraction, colonizing vegetation, plant cover, soil contamination, soil property

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INTRODUCTION

There are many remediation techniques for organic contaminated soils, but relatively few of them are applicable to trace element-contaminated soils. This is due to the fact that trace elements can not be degraded and are relatively immobile; therefore, it is not possible to use many of the low cost options developed for organic contaminants to remove trace element contaminants (Adriano *et al.*, 2004; Vangronsveld *et al.*, 2009).

Sustainable strategies based on the utilization of plants and/or soil additives *in situ* have been used to remediate trace element-contaminated soils by reducing the bioavailability of residual contaminants (Adriano *et al.*, 2004; Kumpiene *et al.*, 2008). Trace elements can be retained in soil by sorption, precipitation and complexation reactions that take place naturally in soils and reduce the mobility and bioavailability of those elements. This process is called natural remediation (NR). The use of some wastes and byproducts as amendments can enhance this process, giving rise to the so-called assisted natural remediation (ANR) technique, which contrasts with most of the classic remediation techniques that drastically alter soil properties (Adriano *et al.*, 2004). The use of amendments in soil reclamation also fulfills two other objectives: i) to reduce waste disposal and revalue wastes by recycling organic matter and nutrients and ii) to restore soil quality (Lombi *et al.*, 2002a, b).

The remediation goals of classical remediation techniques recognized for most guidelines are typically the reduction of the total contaminant concentration, while for ANR the goal is the reduction of the bioavailability of contaminants (inactivation/stabilization). In order to gain wide acceptance, ANR must have a strong

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theoretical background, be demonstrated in the laboratory, and be successful in field experiments. The first two have been largely demonstrated, but results from field-scale validations are relatively scarce. The main objective of field experiments is the evaluation of sustainability (durability) that is crucial for the acceptance of the inactivation/stabilization strategies, since it is difficult to make long-term stability predictions based on short-term laboratory tests (Vangronsveld *et al.*, 2000; Kumpiene *et al.*, 2008). This is especially important when using organic amendments because of the possible changes in the solubility and stability of trace element-organic matter complexes. The effectiveness of trace element stabilization in contaminated soils is usually assessed over several weeks or months, and only a few treatments have been evaluated after several years. Therefore, more information from long time-scale field experiments is needed (Vangronsveld *et al.*, 2000, 2009; Pérez de Mora *et al.*, 2007). Such studies should consider the effect of ANR on: i) soil physicochemical properties, with particular emphasis on trace element concentrations (total and bioavailable) and ii) the development of a vegetation cover and the uptake and accumulation of trace elements by plants (Adriano *et al.*, 2004; Madejón *et al.*, 2006b; Vangronsveld *et al.*, 2009). Moreover, it is necessary to verify the adequacy and the effect of different types of amendments, appropriate application doses, and the persistence of amendment effects over time (Pérez de Mora *et al.*, 2007; Madejón *et al.*, 2010). The cost of ANR is much lower than those of the other remediation techniques. The present study used ANR in a soil moderately contaminated by trace elements. The main objective was to evaluate the effect of the applications of three amendments 8 years after the first application and the rate of change of some chemical properties during the whole experimental period, in attempting to: i) help in future remediation of similar contaminated areas and ii) study the differences between NR and ANR over time.

MATERIAL AND METHODS

Study site

An experimental field at El Vicario (37°26′21″ N, 6°12′59″ W) that was affected by the toxic mine spill in Aznalcóllar, southwest Spain (Grimalt *et al.*, 1999) was selected as the study site. The field is located on the right bank of the Guadiamar River, 10 km downstream from the Aznalcóllar mine. The only remediation work carried out at this site was the initial removal of the sludge together with a layer of underlying topsoil. The soil is a loam (21.1% clay, 29.1% silt and 49.8% sand) classified as Typic Xerofluvent (Soil Survey Staff, 2010). Characteristics of this soil at the 0–15 cm depth before the remediation are shown in Table I. The climate is semi-arid Mediterranean, characterized by a complex pattern of spatial and seasonal variability with wide and unpredictable rainfall fluctuations from year to year (Martínez-Casasnovas *et al.*, 2002). Average annual temperature is 19 °C (minimum: 9 °C in January; maximum: 27 °C in July) and average annual rainfall is 484 mm (Madejón *et al.*, 2010).

TABLE I

Selected properties of the soil studied and the amendments used

Property	Soil	Amendment[a]		
		SL	BC	LE
pH	3.86±1.32[b]	9.04±0.08	6.93±0.03	6.08±0.07
Moisture (g kg^{-1})	–	250	450	280
TOC[c] (g kg^{-1})[d]	9±2	67±16	195±12	289±4
Kjeldhal N (g kg^{-1})[d]	9.0±1.0	9.8±0.4	13.1±0.6	11.7±0.2
P (g kg^{-1})[d]	4.2±0.8	5.1±0.6	12.4±0.2	0.4±0.0
K (g kg^{-1})[d]	23.0±4.0	5.3±0.5	9.3±0.2	39.7±0.8
As (mg kg^{-1})[d]	211±103	2±0	6±2	35±4
Cd (mg kg^{-1})[d]	4.44±1.16	0.43±0.15	0.73±0.40	0.83±0.11
Cu (mg kg^{-1})[d]	119±27	51±8	121±6	28±2
Mn (mg kg^{-1})[d]	645±25	297±10	257±25	66±1
Pb (mg kg^{-1})[d]	471±216	39±7	137±26	22±2
Zn (mg kg^{-1})[d]	381±136	138±31	258±18	65±1

[a] SL = sugar beet lime; BC= biosolid compost; LE = leonardite.
[b] Mean±standard deviation ($n = 48$ for the soil; $n = 3$ for the amendments).
[c] Total organic carbon.
[d] On dry weight basis.