

# Progress in assisted natural remediation of an arsenic contaminated agricultural soil

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*Restoration occurred in a contaminated soil six years after the combined application of iron grit and beringite.*

## Abstract

A contaminated soil was collected in a field adjacent to a derelict As<sup>(III)</sup> smelter in Reppel (Bocholt, Belgium). A single soil treatment (% by soil weight) based on either iron grit (SS, 1%), beringite (B, 5%), or iron grit (1%) + beringite (5%) (BSS) was applied. Untreated and treated Reppel soils and a control soil were placed in lysimeters inside a greenhouse and cropped annually. The efficiency of soil treatments in decreasing As and metals in exposure sources and restoring soil functions was assessed 6 years after the treatments commenced. Decreases in extractable Cd, Mn, Zn and As occurred in the BSS soil. Only BSS treatment reduced both As and metal concentrations in leachates. BSS treatment produced best growth of lettuce and cabbage, the highest shoot and pod yields for dwarf bean, the lowest As, Cd and Zn concentrations in plant tissues, and partly restored Rhizobium nodulation on bean roots. The epigeic earthworm (*Dendrobaena octaedra*) could only survive in the BSS soil. Depurated living worms from the BSS soil had Cd concentration similar to those in control worms, but higher As, Ca, Fe, and Zn concentrations. Based on physiologically based extraction test (PBET), As bioaccessibility was reduced from 12% (untreated soil) to 7.4% (BSS) and 3% (SS), but only the SS treatment decreased the bioaccessibility of Cd (−30%) and Pb (−35%). The range of chemical and biological indicators suggested that BSS amendment was the most effective treatment for restoration of normal soil functions 6 years after initial treatment of the Reppel soil.

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## 1. Introduction

Natural remediation of trace element-contaminated soils can be promoted by in situ inactivation of contaminants through the use of soil amendments (Mench et al., 2000a, 2003a; Brown et al., 2005). Generally, formation of insoluble trace element chemical species reduces leaching through the soil profile and the labile pool available for biological action (Boisson et al., 1999; Geebelen et al., 2003; Ruttens et al., submitted for publication). Plants further cover the soil surface, preventing erosion,

reducing water percolation, and increasing biodiversity (Mench et al., 2003a,b; Brown et al., 2005). Iron, Al and Mn oxides commonly occur in soils and react with metals and As. The OH-OH distance in Fe, Mn, and Al oxides matches well with the coordination polyhedra of many trace metals. Such hydroxyl groups form an ideal template for bridging trace metals (Charlet and Manceau, 1993; Hargé, 1997). Reactions with metals can be promoted when these (hydr)oxides are combined with alkaline materials (Mench et al., 2000a). The efficacy of iron grit (SS, 1% by air-dried soil weight), beringite (B, 5%), and iron grit (1%) + beringite (5%) (BSS) to immobilize metals and As was investigated, following a 3-week reaction time, using a contaminated soil series including one soil from Reppel, Belgium (Boisson et al., 1999). This village in the Limburg province is

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the site of a former As<sup>(III)</sup> refinery, which operated from 1910 to 1965. There were no regulations in place concerning the treatment or disposal of the wastes and dust. Consequently up to 8 ha of this site were highly contaminated by the storage of Zn and As products, i.e. up to 34 700 mg As, 1670 mg Zn, 131 mg Cd per kg soil dry weight (DW) creating a phytotoxic soil (Boisson-Gruppen, 1999). Agricultural fields, adjacent to this derelict As smelter also contained increased arsenic contents and were slightly phytotoxic. Due to the large surface to be remediated, conventional remedial options are too costly. Therefore, alternative technologies such as in situ inactivation for reducing trace element exposure from the soil are needed, especially to reduce the major threat posed to this agricultural ecosystem. In short-term pot experiments, the most effective amendments for reducing 0.1 M calcium nitrate-extractable Cd, Cu, Ni, Pb, and Zn in the Reppel soil were B and BSS (Boisson et al., 1999). The lowest water-extractable As fraction was found in the Reppel soil amended with either B, SS or BSS. The phytotoxicity of the untreated (Unt) and amended soils was assessed using a plant biotest and two plant species (dwarf bean, maize). According to biometric and biochemical plant parameters the effectiveness of soil amendments in remediating the Reppel soil was: BSS > SS > B. Subsequently, a small-scale field experiment was established in 1997 using large mesocosms (Mench et al., 2000a). The “soil-soil solution-plant roots” exposure pathway was initially investigated. Arsenate is usually the predominant As species in aerobic soils, whereas arsenite dominates under anaerobic conditions (Smith et al., 1999). In soil pore water collected by moisture sampler, As<sup>(V)</sup> was mainly present in the (1%) SS-treated soil before and after this amendment corroded in the contaminated soil (Boisson-Gruppen, 1999). Concentration of As<sup>(III)</sup> represented only 0.17% of total As in solution, and was 1.6 µg L<sup>-1</sup> in the Unt soil solution. Monomethyl-arsenic and dimethyl-arsenic were identified in the soil pore water but their concentrations were not relevant compared to As<sup>(V)</sup>. Although the relative As<sup>(III)</sup> concentration increased to 1.5% in the SS-soil solution, absolute As<sup>(III)</sup> concentrations decreased (1.35 µg L<sup>-1</sup>). The oxidation of As<sup>(III)</sup> by newly formed Mn oxides may be an explanation for this but if it occurred, it is not a significant in terms of As exposure and

risk assessment. Metal redistribution between solid phases in year 4 after soil treatment was studied using sequential extraction. Compared to Unt soil, the percentage of Zn associated with amorphous minerals and other compounds increased in the B-treated soil whereas Zn associated with organic compounds decreased (Pannetier, 2000).

When opting for the in situ immobilisation strategy with contaminants remaining in place, important questions need to be answered. These include: What is the sustainability of the soil treatment? Is maintenance necessary? What is the effectiveness regarding the various exposure pathways? What are the costs? Such questions can only be investigated in long-term studies. This implies that regular biomonitoring and assays are needed to investigate the restoration of ecosystem functions using a range of ecological receptors and relevant pathways. One option is to combine soil extracts to mimic the plant behavior, bacterial biosensors, direct toxicity tests and animal feeding trials, and in vitro extracts to predict the human exposure through direct soil ingestion (Brown et al., 2005).

This work reports on the range of evaluation tools used, the soluble and exchangeable fractions of trace elements in the soils, the behavior and the composition of terrestrial bioindicators exposed to the contaminated Reppel soils, 6 years after in situ soil treatment using either iron grit, beringite, or the combination of these additives. The bioindicators are three plant species (lettuce, cabbage, and dwarf bean), soil microorganisms (*Rhizobium* spp.), and an epigeic earthworm. In addition, Scheifler et al. (2005) reported the responses of snails. Lettuce is a well-known Cd/Zn accumulator (Mills and Jones, 1991), and dwarf bean is a metal-sensitive plant species largely used in plant testing (Van Assche et al., 1988). The innovative aspects of the research are: i) the assessment of different exposure pathways in untreated and remediated soils from a long-term small-scale field experiment in year 6 after soil treatment and ii) the investigation of differentially-sensitive terrestrial targets.

## 2. Materials and methods

The soil (3000 kg) was collected from an agricultural field annually cropped with maize adjacent to the derelict As smelter at Reppel (Bocholt,

Table 1  
Composition of the Reppel and Control soils and the soil additives

	pH <sub>water</sub>	CEC <sup>2</sup>	C	N	P <sub>2</sub> O <sub>5</sub> <sup>1</sup>	Ca	Mg	K	Fe
			g kg <sup>-1</sup>						
Control soil	5.6	5.9	19.6	0.78	31				1.09
Reppel soil	6.6	3.86	17.6	1.14	0.21				
Beringite	11					24.6	8.9	1.9	32.8
Iron grit	8.5		10						970
	As	Cd	Cr	Cu	Mn	Ni	Pb	Zn	
			mg kg <sup>-1</sup>						
Control soil	1.1	0.07		3.5		2.7	8.2	10.3	
Reppel soil	169	0.60		26.9		9.9	35.6	70.1	
Beringite		9	950	120	1100	120	203	630	
Iron grit	70	0.03	3500	1010	7710	739	20	104	

<sup>1</sup>Olsen et al. (1954), <sup>2</sup>cmol kg<sup>-1</sup>.

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