

Evaluation of cyclonic ash, commercial Na-silicates, lime and phosphoric acid for metal immobilisation purposes in contaminated soils in Flanders (Belgium)

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Metal immobilising capacities of Na-silicates are weak, while the active mechanism of cyclonic ash is the same as lime.

Abstract

In order to reduce the health risks associated with historically enriched metal smelting sites in Flanders (Belgium), the capacities of a non-beringite cyclonic ash and commercial Na-silicates to fix metals and create conditions to restore vegetation cover were evaluated and compared to lime and H₃PO₄. All tested amendments reduced Ca(NO₃)₂-extractable soil metal concentrations and reduced metal uptake in *Agrostis capillaris* seedlings. Sodium released by Na-silicates was possibly toxic to bean plants while an isotopic dilution technique revealed that metals were only weakly sorbed by silicates (i.e. reversible sorption). Cyclonic ash appeared more efficient than lime in both reducing oxidative stress in beans and Zn, Cu and Pb uptake in grasses. The metal fixing mechanism for both amendments appeared similar (i.e. irreversible fixation at constant pH), in contrast to H₃PO₄ where at least part of the immobilised Cd was irreversibly fixed across a range of pH.

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1. Introduction

Since the second half of the 19th century several industrial zinc producers have been active in the Northeast part of Flanders (Belgium), enriching numerous terrains with metals. In

this region (also called the Noorderkempen), an area of approximately 280 km² has been enriched with Cd (>1 mg Cd kg⁻¹ in surface soil sieved <2 mm). Studies have reported increased Cd concentrations in vegetables and livestock in the region (Ruttens et al., unpublished data). Staessen et al. (1999) reported decreased bone density, an increased risk of fractures among postmenopausal women, and kidney malfunctioning in people living in the region.

One of the main contamination and exposure routes in the region is wind dispersion of polluted soil particles coming from heavily contaminated soils lacking vegetation as a result of metal phytotoxicity (Bleeker et al., 2003; Kucharski et al., 2005). In 1990, 3 ha of a heavily impacted former smelter site (Maatheide) in Lommel (Flanders) were treated with beringite,

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a modified aluminosilicate that originated from the fluidized bed burning of coalmine slag (Vangronsveld et al., 1995). Following treatment, metal availability in soils substantially decreased resulting in the development of a healthy vegetation, while soil organisms (e.g. bacteria, nematodes) in the treated plots rose to normal levels (Bouwman et al., 2001; Vangronsveld et al., 1996, 2000). Based on changes in soil pH, and the results of selective and sequential extractions, a 3-step sorption mechanism was postulated (Mench et al., 1998; Vangronsveld et al., 1999). The first mechanism concerns metal adsorption onto highly accessible sites on the surface of the modified clay and on binding sites of the original soil components freed due to a 'liming effect' [beringite contained $\text{Ca}(\text{OH})_2$ and $\text{Mg}(\text{OH})_2$]. Secondly, coprecipitation of metals with Fe and Mn oxides was found (beringite contained Fe and Mn) while a third postulated mechanism was crystal growth and metal diffusion into the mineral structure, based on EXAFS data that found Zn-silicates in beringite treated plots (Hargé, 1997; Manceau et al., 2000; Mench et al., 1998).

The encouraging results achieved with beringite both in the lab and in the field convinced the Public Waste Agency of the Flemish Region [Openbare Afvalstoffenmaatschappij voor het Vlaams Gewest (OVAM)] that in situ metal immobilisation using cheap industrial by products could be a realistic, cost-effective strategy to reduce environmental and health risks posed by contaminated soils in the Noorderkempen. Beringite production however ceased in 1995 when the fluidised bed in Beringen (Flanders) closed down. Since then, numerous soil amendments have been screened for their metal immobilising capacities, including cyclonic ashes from other fluidised beds. In general, cyclonic ashes substantially reduced metal phytoavailability and phytotoxicity in contaminated soils, but acid extractions [e.g. Physiologically Based Extraction Test (PBET)] and isotopic-dilution data indicated that metals are released again upon acidification (e.g. Boisson et al., 1999; Geebelen et al., 2002a, 2003; Lombi et al., 2002a,b, 2003; Mench et al., 2003). These experiments indicated that the origin and composition of the burnt material and the burning conditions were 2 crucial factors determining the metal immobilizing potential of the ashes. A detailed screening of the coal refuse burning facilities throughout Europe resulted in the selection of cyclonic ashes from a Spanish coal burning plant. The burnt parent material as well as the burning conditions showed great similarity with beringite, resulting in comparable composition of the ashes (Table 1). These ashes were selected for further evaluation and are further referred to as cyclonic ash.

Table 1
Composition of beringite and cyclonic ashes from Spanish origin, on oxide basis (in %) measured using X-Ray fluorescence

	Beringite	CA Spain
SiO_2	51.0	38.5
Al_2O_3	31.0	24.6
CaO	4.1	7.6
Fe_2O_3	6.6	4.3
K_2O	3.5	2.6
MgO	1.6	1.2

Manceau et al. (2000) observed the formation of Zn silicates in beringite treated soil. This generated the idea of using commercial silicates (water glass) for metal immobilisation purposes. Na-silicates have replaced phosphates in laundry and dishwasher detergents where they bind divalent cations and maintain a high pH. Other applications include the cleaning of metal surfaces, bleaching of pulp, de-inking of recycled paper, and water and wastewater treatment (CEES, Centre Européen des Silicates). Commercial Na-silicates are produced when sand is heated under increasing pressure and in the presence of Na_2O resulting in the formation of single or short strains of SiO_4 tetraheders (Fig. 1a). According to the European Silicate Producers (CEES, Centre Européen des Silicates), reaction of Na-silicates with naturally dissolved polyvalent metals results in insoluble silicates or amorphous silica (Fig. 1b). Washing powders contain dispersants that prevent the cation-silicate complex from precipitating.

This paper examines the metal immobilising efficacy and the effect on plant growth of cyclonic ashes and Na-silicates.

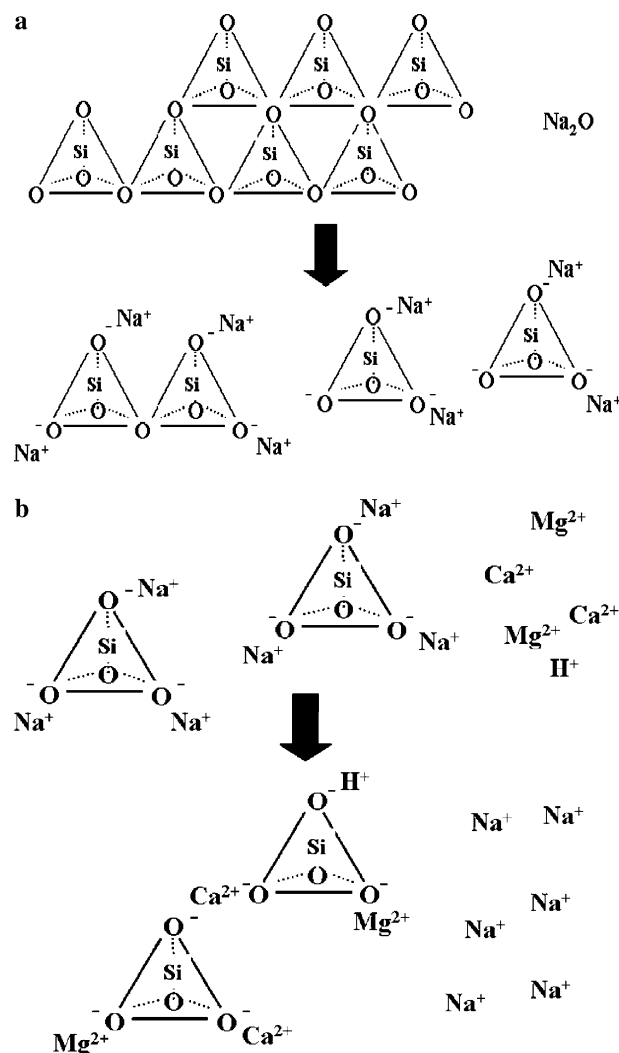


Fig. 1. (a) Commercial Na-silicates are produced by heating sand under increasing pressure in the presence of Na_2O . (b) Reaction of Na-silicates with dissolved polyvalent cations (Ca, Mg,...) results in the formation of insoluble silicates or amorphous silica (CEES, Centre Européen des Silicates).

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