



In situ chemical stabilization of trace element-contaminated soil – Field demonstrations and barriers to transition from laboratory to the field – A review

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ABSTRACT

The chemical stabilization, or immobilization, of trace elements (metals and metalloids; TE) in contaminated soil has been studied for decades. A vast number of scientific publications are available on the method performance in laboratory settings, reporting that the application of various soil amendments to contaminated soil reduces TE mobility, bioavailability and toxicity. The most commonly used soil amendments include organic matter, iron oxides, phosphates, ashes, and lately biochar, alone or in combination with each other and/or lime. Most of the implemented field studies show a certain degree of improvement in soil and/or vegetation status following amendment. Regardless the positive performance of the technique in the laboratory, field validations and demonstrations remain scarce. The establishment of a field experiment often involves permits from authorities and agreements with site owners, both of which are considerably more time-consuming than laboratory tests. Due to conservative institutional structures, public authorities have been slow to adopt alternative remediation technologies, especially when the total TE concentration in soil remains the same and all of the associated risks are not yet convincingly described. For this reason, researchers should also focus on enhancing public knowledge of alternative remediation techniques so that future projects which aim to demonstrate the effectiveness of *in situ* immobilization techniques under natural conditions will be supported.

1. Introduction

Global concerns regarding environmental pollution and human health have prompted the active investigation of contaminated sites and suggestions for mitigation actions. The latter are often based on the presence of contaminants, rather than on a deeper understanding of their properties in soil. Some countries (e.g. Sweden) have set remediation goals and defined specific success indicators of this implementation. Intensive action has seen millions of Euros go towards site investigations and remediation (Skår, 2014; SEPA, 2018a). However, the number and extent of contaminated sites is so high that the set goals are too far from being met. Thus, new strategies for managing soil contamination are urgently needed.

Convenience and, to some degree, simplicity have driven soil remediation actions towards walk-away or ‘dig-and-dump’ solutions, *i.e.*

solving the problem of a contaminated site by removing trace elements (TE)-containing masses for subsequent disposal and replacing them with clean soil. While feasible for smaller sites, such solutions are hardly an option for agricultural land or large areas affected by *e.g.* mining and smelting activities. The removal and landfilling of TE-contaminated soil does not really serve the purpose either, because a landfill is, by definition, a contaminated site that will need to be managed in the future. Although several countries (*e.g.* UK, the Netherlands, France, Switzerland) have applied risk-based approaches in a local context – including a contaminant bioavailability concept – these types of approaches are not yet widely accepted. Furthermore, there is no general consensus on how to measure bioavailability and how many endpoints should be tested (Kumpiene *et al.*, 2017).

Contrary to organic compounds, TE cannot be degraded; hence TE-contaminated soil can be treated either by removing contaminants or

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contaminated soil fractions from bulk soil or by immobilizing TE within the soil matrix. Soil washing after excavation, followed by backfilling with the washed soil, has been practiced at highly contaminated sites (e.g. Banerjee et al., 1998). As is the case with the excavation and landfilling approach, soil washing is not an option for large areas. Hence, other measures, for example, rendering soil metals immobile and unavailable for biological uptake, might be more appropriate for managing environmental and human health risks in such cases.

Immobilization is a technique in which soil amendments are used to induce chemical changes in soil leading to reduced TE mobility and availability to biota. Immobilization or stabilization of trace elements in soil has been a subject of multiple studies and has received attention from both researchers and industry for its potential to replace the practice of excavation and landfilling. Hundreds of studies have provided evidence for how immobilization substantially improves soil properties and reduces trace element mobility, bioavailability and toxicity (e.g. reviewed by Beesley and Moreno-Jiménez, 2011; Bolan et al., 2014; Hettiarachchi and Pierzynski, 2004; Komarek et al., 2013; Kumpiene et al., 2008, etc.). However, the majority of these studies have been limited to the laboratory scale.

This review summarizes recent progress in the area of chemical stabilization, with a focus on upscaling laboratory approaches to field applications. The aim of the review is to understand the current potential of, and identify possible hindrances for, the applicability of TE immobilization in soil on a full scale based on the reported technological performance of field studies.

Literature on the type of amendments was narrowed down to the articles that described the most commonly used materials, namely, organic matter, biochar, Fe-based materials, ashes and phosphates. Any studies, in which amendments were added *in situ* for TE immobilization purposes, including aided phytostabilization and chemophytostabilization, were included. Studies, in which solidifying additives were used for stabilization/solidification were outside of the scope of this review. Peer-reviewed journal papers available through scientific databases Scopus and Web of Science were primarily used, but when needed, other types of publications available on-line, e.g. reports and theses published in English, were also included.

2. Soil amendments for the immobilization of trace elements - field applications

2.1. Phosphates

The use of phosphorus compounds to immobilize TE has been studied for decades. Most of the studies using P as a soil amendment have focused on the immobilization of Pb. Lead is able to form pyromorphite-type minerals, $Pb_5(PO_4)_3X$ where $X = F, Cl, Br, \text{ or } OH$, which are considered the most stable Pb compounds in the soil environment (Nriagu, 1974, 1984). When both added P and soil Pb dissolve, Pb is transformed from more soluble forms (mainly exchangeable Pb, but also present as carbonates, oxides and sulfates, or bound to Fe oxides and organic matter) to the least soluble mineral forms. These forms are stable under a wide range of natural pH and Eh conditions and do not dissolve after ingestion (Howard et al., 2013). The stability of the minerals further increases upon aging (Scheckel and Ryan, 2002, 2004; Lanphear et al., 2003). The types of synthetic and natural phosphorus compounds applied for Pb contaminated soils, immobilization mechanisms, treatment effectiveness, as well as analytical methods and techniques used to study the treatment effects have been comprehensively described in several reviews (e.g. Hodson and Valsami-Jones, 2000; Hettiarachchi and Pierzynski, 2004; Chrysochoou et al., 2007; Miretzky and Fernandez-Cirelli, 2008; Scheckel et al., 2013; Henry et al., 2015). The immobilization of other metals, such as Zn, Cu, and Cd, with P compounds may be attributed to co-precipitation and surface complexation mechanisms, result in compounds that are less stable than pyromorphite-type minerals (Cao et al., 2003a; Gucwa-Przepióra et al.,

2007).

Among all attempts to stabilize TE using soil amendments, immobilization of Pb as a pyromorphite might be the most successful case of the immobilization technique. A substantial number of studies - as reflected by the number of published reviews - have investigated this subject. Phosphate application to prevent Pb migration in soils was among the Best Management Practices (BMPs) suggested by the U.S Environmental Protection Agency for outdoor shooting ranges (USEPA, 2001). Nevertheless, only a few of these studies report results from field tests. A review by Chrysochoou et al. (2007) includes several field studies that were available in the peer-reviewed international literature by the time of publication. These comprise three sites (Jacksonville, USA; Jasper County Superfund Site, USA; and Joplin, USA), at which P amendments were used to immobilize TE in soil. Since then, additional results and updates of the previous field work have been published (i.e. Cao et al., 2003a, b; Scheckel et al., 2005; Tang et al., 2009), but there has not been much progress in establishing new field sites. Publications on four additional field sites were found, in Poland (Kucharski et al., 2005; Gucwa-Przepióra et al., 2007), England (Sneddon et al., 2008), China (Xiao et al., 2017) and USA (Brown et al., 2007a). In summary, the published information in peer-reviewed papers is available at least on seven field sites where contaminated soil was treated *in situ* using P amendments (Table 1). However, most of those studies were conducted on small-scale field plots (1–25 m²) and over a limited time period (max three years). Only one study reports results obtained 10 years after the establishment of the field plots (Tang et al., 2009). Additionally, two field studies have investigated the immobilization of dredged sediments: at Chopawamsic Creek, USA, using apatite (Scheckel et al., 2011) and in Lallaing, France, using synthetic hydroxylapatite (Bert et al., 2009).

2.1.1. Results of field studies

All of the field studies except one (Sneddon et al., 2008, Table 1) reported a significant decrease in soil extractable Pb concentrations upon P application. In the studies in which the observed decrease in Pb extractability was attributed to the formation of pyromorphite-type minerals (Jasper, Joplin and Jacksonville sites), the reduced uptake of Pb, decreased bioaccessible Pb (if measured) and decreased accumulation in plant shoots were also observed. In all of these studies, H_3PO_4 alone or in combination with other amendments served as the P source. Those studies, that attributed reduced TE extraction to changes in soil pH showed inconsistent results concerning changes in Pb uptake by plants (Kucharski et al., 2005; Gucwa-Przepióra et al., 2007; Xiao et al., 2017). The main P sources in these studies were $Ca(H_2PO_4)_2$ and a non-specified type of phosphate, along with a complementary addition of lime. The results of metal accumulation in roots and shoots (or rice grains) indicated that P amendment in these studies was more effective at reducing soil Cd than soil Pb concentrations.

This illustrates the general consensus that for reliable Pb immobilization to occur through the formation of pyromorphite-type minerals, low pH (usually achieved by the addition of phosphoric acid) is needed to induce the dissolution of soil Pb and make it available for immobilization reactions with dissolved P. If pH is not low enough, the immobilization most likely occurs through sorption reactions along with the formation of Pb sulfate (anglesite) and Pb carbonate (cerussite) (Zhang et al., 1998; Zhang and Ryan, 1998). These, as illustrated by the field studies, do not reliably decrease Pb availability in soil. However, the required soil acidification introduces the risk of downward migration of other metals present in the contaminated soil.

Among other most often mentioned drawbacks of using P amendments for Pb immobilization is that this approach requires large quantities of P, which, in addition to being a valuable nutrient with decreasing global availability, can lead to P leaching and contribute to eutrophication. Biogenic P sources, such as bone meal, have been suggested as a possible solution (Hodson et al., 2000, 2001; Sneddon et al., 2006). Research in laboratory settings employing bone meal has

Table 1
Field experiments of *in situ* chemical stabilization of trace element contaminated soil documented in scientific literature.

Amendment	Site, Country	Means of application	Plot size per treatment	Experiment duration	Targeted trace elements	Used plants	References
Phosphate							
TSP ^a	Joplin, USA	Manually applied	8 m ²	32 months	Pb	Kentucky Tall Fescue (<i>Festuca elatior</i>)	Scheckel et al. (2005); Scheckel and Ryan (2004); Ryan et al. (2004)
H ₃ PO ₄	Jacksonville, USA	Spraying H ₃ PO ₄ and manually mixing in PR	4 m ²	480 days	Pb, Cu, Zn, Cd	St. Augustine grass (<i>Stenotaphrum secundatum</i>)	Cao et al. (2002), 2003a, 2003b; Chen et al. (2003); Melamed et al. (2003)
H ₃ PO ₄ + PR	Jasper County, USA	Rototilling; Surface application; pressure injection	8 m ²	10 years	Pb	Kentucky tall fescue (<i>Festuca elatior</i>)	Yang and Mosby, 2006; Tang et al. (2009)
Calcium phosphate (Ca(H ₂ PO ₄) ₂ + lime + lignin phosphate + lime	Waryński, Upper Silesia, Poland	Manually mixed in	16 m ²	3 years	Cd, Zn and Pb	Tufted hairgrass (<i>Deschampsia cespitosa</i>)	Kucharski et al. (2005)
	Guangdong, China	Ploughed in	12 m ²	1 year	Cd, Pb	Indica Rice (<i>Oryza sativa</i> var. Indica)	Gucwa-Przepióra et al. (2007)
Diammonium phosphate (+ compost; biosolids or iron)	Tar Creek, USA	Mixed in with a hand-operated rototiller	1 m ²	18 months	Pb, Zn, Cd	Bermuda grass (<i>Cynodon dactylon</i>)	Brown et al. (2007a)
Sterilized bone meal	England	Excavated, mixed and backfilled		2 years	Zn, Pb, Cd	-	Sneddon et al. (2008)
Biochar							
Harwood	Merseyside, UK	Manually mixed in	0.15 m ²	8 months	As, Cd, Cu, Pb	-	Beesley & Dickinson (2011)
Wheat straw	Yixing, China	Surface application and plough	20 m ²	3 years	Cd, Pb	Rice (<i>Oryza sativa</i>)	Bian et al. (2014)
Wheat straw	Yixing, China	Ploughing and ranking	20 m ²	2 years	Cd	Wheat (<i>Triticum aestivum</i>)	Cui et al. (2012)
Wheat straw	Yixing, China	Ploughed in	20 m ²	5 years	Cd, Pb	Rice (<i>Oryza sativa</i>)	Cui et al. (2016)
Willow	Bezek Farm, Poland	Rototilling and sowing	18.5 m ²	18 months	Cd, Co, Cr, Cu, Ni, Pb, Zn	Wheat (<i>Triticum aestivum</i>) <i>Lepidium sativum</i>	Kończak and Oleszczuk (2018)
Wood-derived	Abergwyngregyn, UK	Spreading and harrowing	18 m ² (later subdivided to 9 m ²)	3 years	As, Cu, Cd, Ni, Zn	Field Bean (<i>Vicia faba</i>) Barley (<i>Hordeum vulgare</i>)	Lucchini et al. (2014)
Lantana camara	Dhambad, India	Mixing in with a spade	15 m ²	-	Ca, Co, Cu, Ni, Pb, Zn	Maize (<i>Zea mays</i>)	Masto et al. (2013)
Holm oak chips	Arganda del Rey, Spain	Rototilling	5 m ²	9 months	As, Cd, Cu, Ni, Pb, Zn	Barley (<i>Hordeum vulgare</i>)	Moreno-Jimenez et al. (2016)
Harwood	Castleford, UK	Excavated and mixed in	1 m ²	3 years	Ni, Zn	Ryegrass	Shen et al. (2016)
Miscanthus	Großbeeren, Germany	Manually mixed in	4 m ² - 6 m ²	2 years	Cu, Cd, Pb, Zn	Orchard grass (<i>Dactylis glomerata</i>)	Wagner and Kaupenjohann (2015)
Sewage sludge	Hunan, China	Ploughing and mixing	1000 m ²	4 months	Cd	Rice (<i>Oryza sativa</i>)	Zhang et al. (2016)
Rice straw	Wuhan, China	Ploughing and mixing	15 m ²	4 months	Cd	Lettuce (<i>Lactuca sativa</i>)	Zhang et al. (2017)
Ashes							
Berlingite (+ steel shots + organic matter)	Jales, Portugal	Mixed in manually	1080 m ²	4 years	As, Cd, Cu, Pb, Zn	<i>Agrostis castellana</i> <i>Holcus lanatus</i> <i>Cytisus striatus</i>	Bleeker et al. (2002)
Coal fly ash	Jinju, Gyeongnam, South Korea	Ploughed in	100 m ²	4 months	As, Cd, Cr, Cu, Mn, Ni, Pb, Zn	<i>Betula alba</i> Rice	Lee et al., 2006
Berlingite + compost	Lommel, Belgium	Mixed in	25 m ² 3 ha	15 months-5 years	Cd, Pb, Zn	<i>Festuca rubra</i> <i>Agrostis capillaris</i> <i>Lolium perenne</i>	Vangronsveld et al. (1995a), b, 1996

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Table 1 (continued)

Amendment	Site, Country	Means of application	Plot size per treatment	Experiment duration	Targeted trace elements	Used plants	References
Coal fly ash	Nord-Pas de Calais région, France	Ploughed in	3000 m ²	8–12 years	Cd, Pb, Zn, Cu	<i>Robinia pseudoacacia</i> <i>Alnus glutinosa</i> <i>Acer pseudoplatanus</i> <i>Salix alba</i>	Pourrut et al. (2011); Lopareva-Pohu et al. (2011); Demuyneck et al. (2014)
Berlingite	Louis Fargue (Coulhins), France	Mixed in	18 m ²	1 year	Cd, Ni	<i>Zea mays</i> L.	Boisson et al. (1998); Mench et al. (2006)
Coal Fly ash	Tongxi, Qingyuan City and Shanghai	Mixed in by tilling	10 m ²	2 rice cultivation season	Cd, Pb, Zn	<i>Lactuca sativa</i> Rice	He et al. (2016)
Coal fly ash + organic complex fertiliser	Dabaoshan mine, Shaoguan City, China Hunan Province, China	Ploughed in	18 m ²	6 months	As, Cd, Pb	Giant reed	Liu et al. (2017)
Iron							
ZVI ¹ as iron grit, FeSO ₄ (+lime)	Cornwall, Northhampton, St. Helens, UK	Ploughed in	2 m ²	~1 year	As	Various vegetable crops	Warren et al. (2003)
ZVI as steel shots	Louis Fargue (Coulhins), France	Mixed in	18 m ²	1 year 7 years	Cd, Ni	<i>Zea mays</i> L. <i>Lactuca sativa</i>	Boisson et al. (1998); Mench et al. (2006)
ZVI as steel shots + organic matter	Biogeco, France	Mixed in	18 m ²	6 years	Cu	<i>Cytisus striatus</i> <i>Populus nigra</i> <i>Rumex acetosella</i> <i>Agrostis capillaris</i> <i>Agrostis gigantea</i> and other grasses	Kumpiene et al. (2014); Touceda-Gonzalez et al. (2017b)
ZVI as steel shots (+ cyclonic ash)	Jales, Portugal	Mixed in	1080 m ²	4 years	As	<i>Holcus lanatus</i> <i>Cytisus striatus</i> <i>Betula alba</i> L.	Bleeker et al. (2002)
Red mud (+ gravel sludge)	Arnoldstein, Austria	Ploughed in	4 m ²	114 days, 5 years	Cd, Pb, Zn	Barley	Friesl et al. (2006); Friesl-Hagl et al. (2009); Kumpiene et al. (2014); Touceda-Gonzalez et al. (2017b)
Red mud ZVI as iron grit (+ compost)	Avonmouth, UK St. Médard d'Eyrans, France	Ploughed in	60 m ² 18 m ²	25 months 2 years 6 years	Cd, Cu, Pb, Zn Cu	<i>Festuca rubra</i> <i>Agrostis</i> spp. <i>Dactylis</i> spp. <i>Holcus</i> spp. <i>Populus</i> spp. <i>Salix</i> spp.	Gray et al. (2006) Kumpiene et al. (2011) Tiberg et al. (2016)
ZVI as iron grit	La Combe du Sault, France (Difpolmine)	Rototilling	18 m ²	18 months	As	Grasses and dicots	Jacquemin (2005); Guérin et al. (2008)
Fe oxides from water treatment residues	Collstrop, Hillerød, Denmark	Mixed in rotary bucket down to 1 m	100 m ²	14 months	As, Cr, Cu	n.a.	Nielsen et al. (2016)
Fe oxides from water treatment residues	Tar Creek, USA	Mixed in with a hand-operated rototiller	1 m ²	18 months	Pb, Zn, Cd	Bermuda grass (<i>Cynodon dactylon</i>)	Brown et al. (2007a)
CFH ^d + limestone; ZVI as iron grit FeCl ₃ /FeSO ₄ + lime	Steinsjøen, Norway Hawaii, USA	Mixed using a tractor blender Mixed in manually	4.5 m ² 0.5 m ²	4 years 612 days	Pb, Sb As	n.a.	Okkenhaug et al. (2016) Cutler et al., 2014
Organic matter							
Orgro ^o , sewage sludge compost	Baltimore, USA	Rototilled	10–50 m ²	1 year	Pb	Grass	Farfel et al. (2005)
Commercial compost (+ steel shots + berlingite)	Jales, Portugal	Mixed in manually	1080 m ²	4 years	As, Cd, Cu, Pb, Zn	<i>Agrostis castellana</i> <i>Holcus lanatus</i> <i>Cytisus striatus</i> <i>Betula alba</i>	Bleeker et al. (2002)

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Table 1 (continued)

Amendment	Site, Country	Means of application	Plot size per treatment	Experiment duration	Targeted trace elements	Used plants	References
Wastewater biosolid compost, leonardite and olive leaves compost + lime	El Vicario (Aznaicóllar dam accident area), Spain	Mixed with rotary tiller	56 m ²	14 years	As, Cd, Cu, Pb, Zn	<i>Lamarckia aurea</i> <i>Chrysanthemum coronarium</i> <i>Raphanus raphanistrum</i> <i>Poa annua</i> <i>Bromus rubens</i>	Burgos et al. (2008); Madejón et al. (2006), 2009, 2010, 2018a, b; Montiel-Rozas et al. (2016a), b; Pérez-de-Mora et al., 2011; Soler-Rovira et al. (2010); Xiong et al. (2015)
Cow manure + lime, solid olive mill waste + lime	Aznalcóllar dam accident area, Spain	Ploughed in	32 m ²	3 years	Cd, Cu, Pb, Zn	<i>Brassica juncea</i>	Clemente et al. (2005), 2006
Organic matter + ZVI as steel shots	Biogeco, France	Mixed in	18 m ²	6 years	Cu	<i>Cytisus striatus</i> <i>Populus nigra</i> <i>Rumex acetosella</i> <i>Agrostis capillaris</i> <i>Agrostis gigantea</i> and other grasses	Kumpiene et al. (2014); Touceda-Gonzalez et al. (2017b)
Greenwaste compost	St Helens, UK	Capping	5 m wide strips	2 years	As, Zn, Pb, Cu	Lettuce and sunflower (<i>ex situ</i>)	Clemente et al. (2010)
Compost from solid olive mill waste, Pig slurry	Sierra Minera, Spain	Mixed in	2 m ² 3 m ²	2.5 years	As, Cd, Cu, Pb, Zn	<i>Atriplex halimus</i> <i>Bituminaria bituminosa</i>	Clemente et al. (2012); Pardo et al. (2014a), b
Sugarcane bagasse compost (SC)	Taiwan	Mixed in	2400 m ²	18 months	Cu, Zn, Cr, Ni	Rice	Huang et al. (2018)
Biosolid compost from wastewater plant	Spain	Rototilled	56 m ²	4 years	As, Cd, Cu, Pb and Zn.		Madejón et al. (2009)
Compost + biochar	Arnoldstein, Carinthia, Austria	Mixed in with rotary hoe	27 m ²	2 years	Cd, Pb, Cu and Zn	<i>Miscanthus × giganteus</i>	Kareer et al. (2018); Touceda-Gonzalez et al. (2017b)
Municipal solid wastes + bark chippings compost	Mina Touro, Spain	Mechanical tillage	25 m ²	3 years	Cu	<i>Salix caprea</i> <i>Mauerbach</i> <i>Salix viminalis</i>	Touceda-Gonzalez et al. (2017a); Quintela-Sabaris et al., 2017
Compost (+ lime)	Gironde, France	Amendments were mixed 25 cm depth	3 m ²	2 years	Cu	<i>Populus nigra</i> Mixed stand of <i>Populus nigra</i> <i>Salix viminalis</i>	Šimek et al. (2018)
Municipal waste compost (+ beringite)	Lommel, Belgium	Mixed in	25 m ² 3 ha	15 months-5 years	Cd, Zn	<i>Amorpha fruticosa</i> <i>Agrostis capillaris</i> <i>Festuca rubra</i>	Vangronsveld et al. (1995a), b; 1996
Biosolids + lime	Piekary Slaskie, Poland	Ploughed with a chisel plow	3300 m ² and 4000 m ²	5 and 20 years	Zn, Cd, Pb	<i>Lolium perenne</i> <i>Festuca rubra</i> <i>Poa pratensis</i> <i>Festuca arundinacea</i> <i>Festuca ovina</i> <i>Agrostis alba</i> <i>Lolium perenne</i> <i>Agrostis canina</i> <i>Agrostis capillaris</i>	Stuczynski et al. (2007); Siebielec et al. (2018); Kumpiene et al. (2014); Touceda-Gonzalez et al. (2017b)
Manure compost	Weining district, China	Not specified	1600 m ²	4 years	Pb, Zn, Cd	<i>Dactylis polygama</i> <i>caespitosa</i> <i>Puccinellia distans</i> <i>Festuca heterophylla</i> <i>Arundo donax</i> <i>Braussonetia papyrifera</i> <i>Robinia pseudoacacia</i> <i>Cryptomeria fortunei</i>	Luo et al. (2018)

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Table 1 (continued)

Amendment	Site, Country	Means of application	Plot size per treatment	Experiment duration	Targeted trace elements	Used plants	References
Biosolid, chipped woody debris + lime	Leadville, USA	Mixed in with a hand-operated rototiller; Surface applied with rear through spreader and tilled in with a ripper	8 m ² 72000-123400 m ²	5 years; 2 years	Cd, Pb, Zn	<i>Lolium multiflorum</i> Seed mixture of various grasses	Brown et al. (2007b), 2009 Brown et al. (2005)
Biosolid + wood ash	Bunker Hill, USA	Mixed in manually	4 m ²	2 years	Pb, Zn, Cd	Western wheat grass (<i>Pascopyrum smithii</i>) Vetch (<i>Vicia sativa</i> L.) Bermuda grass (<i>Cynodon dactylon</i>)	Brown et al. (2003)
Municipal biosolids + lime, Biosolids compost	Tar Creek, USA	Mixed in with a hand-operated rototiller	1 m ²	18 months	Pb, Zn, Cd		Brown et al. (2007a)

n.a. – not applied.

Co-amendments in parenthesis indicate treatments with the main amendment alone and additionally combined with the indicated co-amendment.

^a TSP- triple superphosphate.^b PR – phosphoric rock.^c ZVI – zerovalent iron.^d CFH - ferric oxyhydroxide powder.

shown similar immobilization effects as treatment with inorganic P at far lower P/Pb ratios. Nevertheless, the following field study (Sneddon et al., 2008) did not find any positive impact of bone meal addition on Pb immobilization over a period of two years, but did showed significant Cd and Zn solubilization. This mobilization was attributed to the organic component of bone meal (Sneddon et al., 2008). Cao et al. (2003a) provided a similar explanation (i.e. dissolved organic carbon) for the increased Pb and Cu leachability observed in laboratory tests following P addition. Organic matter, especially low molecular weight organic acids (e.g. citric acid), can inhibit pyromorphite formation in soil (Zuo et al., 2016). Hence, organic matter-rich soils stabilized with P amendments will yield worse metal immobilization results than sandy soils. Certain organic components of bone meal (e.g. various proteins), along with their degradation products (e.g. ammonia and nitrate), may be toxic to soil organisms and were implicated as the reason for earthworm toxicity following bone meal amendment (Atuah and Hodson, 2011). Although other P compounds without organic components, e.g. potassium hydrogen phosphate, triple superphosphate, and rock phosphate, were shown to have varying effects on *Eisenia fetida* earthworms, and it was suggested that earthworm toxicity depends on a combination of factors such as increased electrical conductivity, pH and P concentration in soil (Atuah and Hodson, 2011; Maenpaa et al., 2002; Ownby et al., 2005).

The increased mobilization of oxyanions is another common side effect of P addition to soil. Phosphates are structural analogues of arsenates and can replace soil As, especially at high P concentrations and high soil pH (Carabante et al., 2010). Similarly, selenium, vanadium, molybdenum, antimony and tungsten ions can be replaced by phosphate ions, which can lead to the increased leaching of these elements (Eich-Greatorex et al., 2010; Larsson et al., 2017; Mikkonen and Tummavuori, 1994).

The applicability of P amendment to Pb immobilization in large-scale remediation projects is still debatable. In their review, Chrysochoou et al. (2007) covered the criticism of this method as a sustainable technique for the remediation of Pb-contaminated shooting ranges. The main arguments against P amendment were basically the same as other authors have highlighted, namely P leaching and eutrophication, leaching of oxyanions, and limited reliable evidence of treatment performance. Also, the achieved efficiency - which spanned transforming 11–55% of Pb to the least soluble forms - was not considered high enough to justify the other shortcomings. Nevertheless, this should not hinder further research, especially demonstrations that cover larger spatial and time scales, and focus on overcoming the identified limitations of this technique.

2.2. Biochar

Scientific literature on the potential of biochar (BC) to immobilize TE in soil first appeared in 2008 and the amount of published research is rapidly increasing since. Several review papers have been recently published describing BC properties, potential benefits and limitations of BC application to soil (Chen et al., 2018; El-Naggar et al., 2019; Kavitha et al., 2018; Shaaban et al., 2018). Compared to phosphates, which were first described as a possible means of immobilizing soil Pb in 1974 (Nriagu, 1974), studies of BC have been extended to field studies considerably faster. There are currently 10 peer-reviewed papers available that describe BC application for metal immobilization in field (Table 1).

Biochar is a sorbent material for the removal of inorganic and organic pollutants that is a less expensive and more efficient alternative to active carbon. Biochar is generated by the anaerobic thermal degradation of biomass and organic waste materials; it is rich in carbon, porous and contains oxygenated functional groups (Lehmann and Joseph, 2009). Biochar composition and properties depend on the feedstock material and pyrolysis conditions; thus, a wide variety in BC types can be found in the literature. However, it is common that researchers wrongly identify other types of pyrogenic carbonaceous

material (e.g. pyrolyzed chars with low carbon contents) as BC. Although numerous studies have been conducted on the sorption properties of different types of BCs in the removal of TE (Cd, Cr, Cu, Ni, Pb, Zn, etc.) from aqueous and soil systems (Ahmad et al., 2014; Beesley and Moreno-Jimenez, 2011; Oustrière et al., 2016; Xu et al., 2017), knowledge about their application as a soil amendment in polluted sites, factors controlling TE stabilization using BC amendments and the medium-to long-term behavior of BC under natural conditions still remains limited.

The application of BC as soil amendment has been tested at sites affected by anthropogenic activities, e.g. mining, smelting, farming. The immobilization of TE by BC depends on a number of factors, e.g. feedstock material, BC physico-chemical characteristics, BC application rate, and has been extensively studied in the laboratory using batch leaching and pot experiments (Beesley et al., 2010; Bopp et al., 2016; Houben et al., 2013; Kelly et al., 2014; Meier et al., 2017; Trakal et al., 2018; Uchimiya et al., 2010). The results show that BC has high sorption capacity for trace metals, with materials characterized by relatively high O/C molar ratios performing best due to the presence of oxygen functional groups that have a high affinity for metals. In addition, metal retention by BC usually increases as BC surface area, porosity, cation exchange capacity (CEC) and ash content increases. Finally, higher BC application rates tend to increase the immobilization of metals in soil (Chen et al., 2018), although certain exceptions - which depend on the origin of BC - have been reported (e.g. Oustrière et al., 2016). An increase in BC addition rates implies the need for an increased production of BC. This could be considered as a major limitation, since the large-scale application of BC amendment to agricultural and soil remediation purposes would necessitate a more sustainable BC production system.

2.2.1. Biochar as a medium-to long-term amendment in field studies

Most studies that have analyzed the effectiveness of BC amendments were usually conducted under laboratory conditions with short equilibration times, ranging from days to weeks. However, long-term equilibration time is a key aspect in assessments of the efficiency of remediation efforts at contaminated field sites. In particular, detailed information is needed about the immobilization mechanisms and possible changes over time (Komarek et al., 2013; Kumpiene et al., 2008). The effectiveness of BC amendments depends on the characteristics of the polluted site, the experimental plot size, application time range, and environmental factors such as humidity and temperature. The effectiveness of BC amendment is usually assessed by investigating the presence of pollutants in leachates and soil pore water (SPW), metal uptake in plant tissues and potential increases in plant biomass and soil microbial communities.

Field studies conducted over the short-to medium-term, i.e. < 1 year, show contradicting results for different TE. In most cases, BC amendment leads to a decrease in the concentration of extractable or soluble forms of cationic metals (Masto et al., 2013; Zhang et al., 2017; Zheng et al., 2015), but for some elements, especially oxyanionic metalloids (e.g. As), the application of BC yields a minimal effect, or even increases extractable concentrations (Beesley and Dickinson, 2011; Moreno-Jiménez et al., 2016; Zheng et al., 2015). This is expected since most functional groups of the BC surface have a negative charge, which favors the electrostatic repulsion of oxyanions. Also, the presence of dissolved organic matter promotes the solubility of As through competition for binding sites. Various field studies conducted over a period of 2–3-years found a significant decrease in the soluble and bioavailable forms of Cd, Ni, Pb and Zn after biochar amendment to contaminated soils (Bian et al., 2014; Cui et al., 2012; Lucchini et al., 2014; Shen et al., 2016; Wagner and Kaupenjohann, 2015). This reduction was mainly attributed to a liming effect and chemical stabilization through sorption on the BC surface and/or pores. The increase in soil pH following BC amendment is known to reduce the solubility of metals, which favors the precipitation of mineral phases (Ahmad et al., 2014; Xu et al., 2017; Wagner et al., 2015). However, this change may also

increase the dissolved organic carbon (DOC) concentration in a soil system (Wagner and Kaupenjohann, 2015) leading to the increased extractable concentration of some elements, e.g. Cu and Pb (Oustrière et al., 2016).

The ability of BC to immobilize trace metals remains rather constant over time and can even increase as the BC ages (Bian et al., 2014; Cui et al., 2012; Wagner and Kaupenjohann, 2015). Cui et al. (2016) observed the transformation of Cd and Pb from the exchangeable and bioavailable metal forms into more stable forms over a five-year study of wheat BC stabilized soil. Similarly, no change or even a decrease in metal concentrations in soil pore water (SPW) were observed during pot experiments in which material ageing was monitored for 2–3 years (Egene et al., 2018; Oustrière et al., 2017; Van Poucke et al., 2018). Biochar is highly recalcitrant and resilient to degradation; thus, it may be possible to sustain the positive effects of its application for longer periods of time when compared with other organic amendments, such as compost or peat, which can easier mineralize through physical or biological processes (Egene et al., 2018).

Any assessment of the effectiveness of BC application should also include an evaluation of its effects on vegetation and plants, i.e. through analyses of crop yield, changes in physiological characteristics, and metal accumulation in plant tissues, as well as ecotoxicological tests. Most of the field studies included in this review found that BC amendment resulted in a significant decrease in trace metal (especially Cd, Pb and Zn) uptake by plants (Bian et al., 2014; Cui et al., 2012; Moreno-Jimenez et al., 2016; Wagner and Kaupenjohann, 2015; Zhang et al., 2016; Zheng et al., 2015), as well as an increase in the production of shoot and root biomass (Bian et al., 2014; Kończak and Oleszczuk, 2018; Masto et al., 2013; Wagner and Kaupenjohann, 2015; Zhang et al., 2016; Zhang et al., 2017). Increased plant biomass can usually be explained by BC-induced TE immobilization, but the higher nutrient (P, K or Ca) content in BC amended soils can also stimulate plant growth. For example, the application of 10% BC increased available and total K by ~50% and ~20%, respectively, over an 18-month period (Kończak and Oleszczuk, 2018). Also, the liming effect of BC may decrease Al toxicity, increase CEC and the bioavailable P concentration in the amended soils (Major et al., 2010; Masto et al., 2013; Wagner and Kaupenjohann, 2015).

Although increased BC application rates favor the immobilization of most trace metals and improve the nutrient supply of the soil, elevated BC amounts may also cause unwanted detrimental effects, such as a deficiency of phytoavailable nutrients, reduction in CEC, and/or inclusion of phytotoxic compounds, e.g. some TE and PAHs, which may eventually inhibit biomass growth (Lucchini et al., 2014; Quilliam et al., 2012, 2013). In addition, changes in pH, DOC, soluble P or other geochemical conditions might even increase the mobility of TE (Beesley et al., 2010; Beesley and Dickinson, 2011; Moreno-Jiménez et al., 2016; Zheng et al., 2015). Hence, depending on the BC characteristics and the contaminants present in the soil, BC amendment may reduce the available concentrations of some elements and, at the same time, increase the concentrations of others. Nevertheless, the balanced application of BC amendment in field studies has shown promising results in terms of TE immobilization and plant growth, although additional work must be conducted before BC is widely applied on a commercial scale. Most of the studies were conducted in small-sized plots with (1–20 m²), with only one study covering an area of over 1000 m² (Zhang et al., 2016). The spatial variability of soil contamination in a field is one of the important factors that determines the contaminant immobilization success. The variability of results in field trials tends to decrease as plot size increases (Petersen, 1994). Future studies should also cover longer time periods. The longest field study lasted only five years (Cui et al., 2016), and the influence of changes in environmental factors and agricultural practices were not sufficiently considered. These factors may promote the leaching of contaminants and influence the effectiveness of BC amendments. Also, most of the BC field studies used high BC application rates, 10–50 t/ha, presumably to ensure

positive TE immobilization results, but this approach could prove costly and unsustainable on the commercial scale.

2.3. Ashes

The application of ash as a soil ameliorant after woodland burning to stimulate vegetation growth has been practiced by primitive hunters and plant-gathering peoples as early as since interglacial times (Steensberg, 1957). By the early Neolithic Period, woodland clearance for arable farming had started and ploughing wood ash into the soil had already become a common practice (Huang et al., 2006). The use of wood ash has continued since, but is nowadays mainly used for the recycling of nutrients in forest soil (Bramryd and Fransman, 1995; Pitman, 2006).

Ever since contemporary societies began to incinerate various solid fuels – peat, coal and currently municipal solid waste (MSW) – for energy recovery, ash has become an excess material that needs to be managed. Two types of solid waste are generated during solid fuel burning, namely, fly ash (FA), which comprises finely-sized particles and additives collected in incineration air pollution control (APC) systems and bottom ash (BA), which represents the non-combustible fraction remaining on the hearth of a furnace (Chandler et al., 1997). Bottom ash is much more heterogeneous than fly ash and seldom considered for soil amelioration purposes. Fly ash is a fine homogenous material mainly composed of spherical glassy particles, silica, alumina, iron and calcium oxides as well as a number of other elements, e.g. S, Cl, Cr, Cd, Zn, As, Se, Br, Pb, etc. The chemical and mineralogical composition of FA varies greatly depending on the type and composition of the source material as well as the type of incinerator (Brännvall and Kumpiene, 2016). Fly ash is a highly reactive material and, due to ageing and weathering, some key properties (e.g. pH, buffering capacity and element solubility) change over time (Brännvall, 2010). The pH of FA from woody fuel is highly alkaline, whereas coal FA can be acidic or highly alkaline depending on the coal composition. Since TE composition of ashes can differ substantially, this would also affect TE mobility and phytoavailability in contaminated soil (Labidi et al., 2017).

Most studies that have investigated the use of ash amendment for TE immobilization purpose have focused on coal fly ash and have been limited to laboratory or small-scale greenhouse experiments. Most of the research has shown that fly ash can reduce the mobility, extractability, ecotoxicity and phytoavailability of TE (mainly cationic elements) in contaminated soils (e.g. Boisson et al., 1999; Jala and Goyal, 2006; Kumpiene et al., 2007, 2009a; Mench et al., 2006; Ohecova et al., 2014). In contrast, the effect of ash application on anionic elements (e.g. As) is highly variable (e.g. Beesley et al., 2018; Mollon et al., 2016) and may depend on final soil pH, the amount ash applied as well as the Ca content of the ash. There are currently only a few published large-scale field studies available that have evaluated how fly ash application influences TE immobilization in contaminated soil (Table 1).

2.3.1. Results of field studies

Field studies of ash as a soil amendment are performed for two purposes: to improve soil nutrient status and facilitate biomass growth (mainly in forestry); or to reduce phytotoxicity of contaminated soil and promote revegetation of barren soil surfaces. Field studies that include the latter objective i.e. contaminant immobilization, have been conducted in France, Belgium, Portugal, China, India and South Korea (Table 1). The size of the plots used in these studies varies from 10 m² to 3 ha. The results mainly show a substantial decrease in TE (Pb, Cd, Zn, Cr, Se, and Cu) extractability and phytoavailability to various plants following the application of ash to contaminated paddy, acidic, and smelter-affected soils (Dermatas and Meng, 2003; He et al., 2016; Hseu et al., 2010; Liu et al., 2017; Vangronsveld et al., 1995a, b). In contrast, the studies report increased phytoavailability of Zn, Cu, Ni, Pb, and Cd when fly ash was applied to clean soils in order to evaluate FA effects on

soil and plant productivity (Kalra et al., 1998; Tripathi et al., 2009; Tsadilas et al., 2014).

The highly complex composition of ash means that the mechanisms through which FA immobilizes TE are also complex. Ashes are composed mainly of aluminosilicates, but high pH (ca 12) and calcium oxide concentration of FA can cause a liming effect, which may be one of the main mechanisms through which FA immobilizes TE. The pH neutralization induced reactions in soil (e.g. ion exchange and sorption) might be reversible upon soil acidification (Nachtegaal et al., 2005). Nevertheless, the main advantage of ash over lime is its high buffering capacity towards acidification. Slow aging of FA results in weathering of aluminosilicates and increase in non-crystalline clay content, which may contribute to long-term TE fixation (Zevenbergen et al., 1999). In addition, ash contains substantial amounts of major nutrients such as P and K and, along with a long-lasting pH neutralizing capacity (Bolan et al., 2014), has been shown to facilitate healthy plant growth for more than a decade after application at heavily contaminated sites in Belgium (Vangronsveld et al., 2009). In addition, coal fly ash amendment showed a positive impact on calcium chloride-extractable Cd and Zn concentrations in smelter-contaminated soils during decade-long field experiments (Demuyne et al., 2014; Labidi et al., 2017; Lopareva et al., 2011).

In addition to the various benefits of fly ash amendment, e.g. improvements in soil texture, nutrient status, water holding and pH buffering capacities, there are several drawbacks such as the high salinity of ash and the addition of several potentially toxic TE (boron and molybdenum in particular) that are discussed (Pandey and Singh, 2010). Furthermore, an excess of reactive ash constituents (Ca, Al, and Fe) can bind nutrients leading to their imbalance in treated soil (Schumann and Sumner, 1999; Doran and Martens, 1972). This was the reason suggested for previous findings that ash amended soil (80 Mg ha⁻¹) produced only half of the maize biomass that a fertilized (20N–20P₂O₅–20K₂O) control plot produced (Schumann and Sumner, 1999). Although the use of ash for soil amelioration is an ancient practice, it has not been extended to the remediation of contaminated soils, despite good results concerning reduced TE availability in soil. The positive impacts of balanced ash application might outweigh the negative impacts. Field studies that include long time periods, i.e. over a decade, should increase confidence in this method, and it is remarkable that ash amendment has not received more attention in the context of soil remediation.

2.4. Fe-based amendments

Iron-based amendments have been intensively studied in the context of contaminated soils for their potential to act as analogues of naturally ubiquitous soil Fe phases, which are a crucial component of the soil sorption complex. Increasing the sorption capacity of soils can be achieved by either directly adding various Fe oxides or indirectly adding their precursors, i.e., zero-valent iron (ZVI) in the form of iron grit, iron sulfates, and iron (red) mud, among others (Komarek et al., 2013; Kumpiene et al., 2008).

Arsenic and other oxyanions are the most common contaminants targeted by iron-based amendments. The pH of contaminated soil is usually below the isoelectric point of iron oxides (ca. pH < 8). At such pH values, Fe oxides are predominantly positively-charged, which makes them highly effective in sorbing anionic contaminants (e.g. As, Sb). Laboratory studies have shown the As immobilization efficiency of Fe oxides to be as high as 99%. Due to their amphoteric nature, Fe oxides can simultaneously sorb cationic elements, e.g. Cd, Pb, Zn, Cu as well, with similar or slightly lower efficiencies. The use of Fe-based materials for soil remediation has been comprehensively reviewed by Cundy et al. (2008) and Komarek et al. (2013).

2.4.1. Results of field studies

A wide range of iron-based materials and compounds have been

tested for their application to soil remediation attempts. Their overall efficiency can be increased by using nano-sized analogues or composites. While these novel materials exhibit promising adsorption properties, their production costs still remain high.

Similarly to the other discussed soil amendments, a vast body of scientific literature is available on Fe-induced TE immobilization in soil under laboratory conditions. Research describing how these amendments perform under field conditions remains scarce. Field-scale experiments, found to be documented in scientific papers, have been performed at eleven sites with plot sizes ranging from 1 m² to ~1000 m² (Table 1). The twelfth, and probably largest full-scale application of Fe amendments for As immobilization, was implemented in 2006 on a 10 ha area at a former gold extraction and processing site in La Comb du Sault, France (Difpolmine; Jacquemin, 2005). Unfortunately, the follow up of the project could not be found in peer-reviewed scientific literature. One identified study described the responses of wild small mammals at this site to As after remediation (Drouhot et al., 2014). One additional study evaluated the ecosystem at this site 18 months after aided phytostabilization was performed (Guérin et al., 2008).

In addition to those 12 cases, one field study was found on immobilization of dredged sediments in Bremen, Germany, using Fe-bearing materials (Müller and Pluquet, 1998).

In six of the field studies, ZVI grit (steel shots) served as the source of metallic Fe. These products are micro- to mesosized (0.1–2.5 mm) steel abrasives that are predominantly composed of metallic Fe, but include some impurities of Cr, Cu, Ni, Pb, Zn and Mn. Iron oxide precursors (Fe chloride and/or Fe sulfate) were applied at two sites, while four studies used industrial waste materials containing Fe oxides (red mud, water treatment residues). These industrial waste materials contained less iron (24–32%) and more of other compounds (Si, Mn, Ca, Al, Cr, OC, etc.) than the other Fe compounds.

Both laboratory and field studies have shown that using ferrous sulfate as the source of Fe oxides decreases soil pH. This might not negatively affect soil As, as a lower pH contributes to more positive charges on Fe oxides. However, soil acidification negatively impacts on the leaching and plant uptake of cationic metals (Warren et al., 2003). Thus, lime must be added to counteract soil acidification.

The use of red mud showed varying effects on both metal immobilization (ranging from slight immobilization to an 80–99% reduction in ammonium-nitrate extractable Cd, Pb, Zn and Ni) and on metal uptake by plants (including a slight decrease, no effect or increased Cd uptake) (Friesl et al., 2006; Gray et al., 2006). The observed decreased metal mobility was suggested to be a result of increased soil pH (Friesl et al., 2006). It is likely that pH-induced changes in the soil underlie the variability in results. Similar outcomes have been noted in phosphate treatments when the immobilization of metals was a result of pH changes in the soil (and likely the induced ion exchange reactions), rather than chemisorption or precipitation of the desired mineral species. Gray et al. (2006) compared red mud with lime and reported no difference between the treatments in terms of the extractable Cd, Pb, Ni and Cu 25 months post-treatment. At this time point, both treatments also showed similar plant biomass values. Although red mud contains substantial amounts of iron oxides (24%, Gray et al., 2006), it could be classified as an alkaline soil amendment (pH > 10). Hence, the liming effect may be the predominant mechanism through which red mud immobilizes TE, and the effectiveness of this mechanism diminishes rather quickly over time. The effects of Fe oxides are expected to be more long-lasting. For example, Cu was still strongly bound to Fe (hydr)oxides in ZVI-amended soil six years after treatment (Tiberg et al., 2016). In general, higher initial concentrations of contaminants in soil will translate to better immobilization efficiencies even if the final TE concentration in solution (absolute values) might be similar. For this reason, the element immobilization at a former wood treatment site reaching as high as 95% was not enough to meet Danish groundwater quality criteria (Nielsen et al., 2016). Although field tests usually show

significantly less efficient TE immobilization (or no effect at all) relative to laboratory tests (e.g. Bleeker et al., 2002), several studies have reported values that were close to the theoretical maxima obtained under laboratory conditions. High Sb (up to 89–93%) and Pb (up to 94–97%) immobilization efficiencies, measured as water extractable elements and which were in line with - or even outperformed - results from laboratory column tests, were achieved in a small-arms shooting range in Norway using ferric oxyhydroxide powder (CFH) and ZVI (Okkenhaug et al., 2016). Similar efficiencies were achieved for Cu, Cr and As at an abandoned wood impregnation site treated with iron water treatment residues (Nielsen et al., 2016). In the latter study, soil down to 1 m depth was roughly mixed with a rotary screening bucket mounted onto an excavator, while in the former study a 1 m³ tractor blender was employed. Soil mixing is a crucial step in evenly distributing soil amendments in the soil and may be the main reason that these two studies were able to achieve such high immobilization efficiencies. Spreading amendment on soil surface followed by rototilling was a common practice in the other sites. This practice may not have provided sufficient homogeneity in soil-amendment mixture and could thus explain the lower efficiencies reported in those studies. Unfortunately, different ways of incorporating amendments is seldom studied.

Iron-induced immobilization of elements at field sites does not always lead to the expected recovery of the ecosystem (Guérin et al., 2008). The results of the La Comb du Sault site follow-up study showed that the vegetation in the phytostabilised areas was still poorly developed 18 months after the soil treatment, while the phytostabilised and control areas did not differ in the bacterial community structure, arbuscular mycorrhizal response and the presence of earthworms (Guérin et al., 2008). Furthermore, Fe amendments do not necessarily reduce element phytoavailability (Boisson et al., 1998; Warren et al., 2003). Elements bound to poorly crystalline Fe oxy (hydro)oxides, which are the first minerals that form in ZVI-amended soil, can be quantified by dissolution in oxalate. This method - a step in some types of sequential extraction procedures - has been used to demonstrate the extent to which elements are immobilized in Fe amended soils (Kumpiene et al., 2011, 2012). Oxalates are ubiquitous in plants and can be exuded by roots, especially in response to toxicity induced by certain soil elements, e.g. Al (Ma and Miyasaka, 1998). Released oxalate, along with other organic molecules exuded from roots, might induce the desorption of Fe-bound elements (Gonzaga et al., 2009; Quartacci et al., 2009). As a result, Fe-immobilized elements might still be available for plant uptake despite decreased concentrations in the soil pore water and exchangeable fractions. For example, decreased As concentration bound to poorly crystalline Fe oxy (hydro)oxides and substantial accumulation of As in plant litter were observed in a long-term field lysimeter experiment (Kumpiene et al., 2012). Moreover, impurities of Fe amendments, e.g. Ni and Cr, might increase the total or leachable concentrations of these elements, but this is not always linked to their uptake by plants (Boisson et al., 1998; Gray et al., 2006).

The main concerns regarding the use of Fe oxides for TE immobilization in soil are their redox sensitivity and the metastability of the most reactive iron oxyhydroxide phases. The corrosion of metallic Fe added to soils produces ferrihydrite, a metastable poorly crystalline oxyhydroxide that reacts with dissolved TE and reduces their mobility. Iron oxyhydroxides will slowly transform into more crystalline Fe oxides (e.g. goethite, hematite) that have lower specific surface areas and reactivities. This might cause the release of sorbed TE and increase their leaching over time. In addition, ions that compete with those immobilized by Fe oxides can replace sorbed TE over time. This is particularly relevant in As contaminated sites at which P fertilizers are applied to facilitate the revegetation of barren soils and mine tailings. Although As has a higher affinity for Fe oxides than P, the addition of large amounts of P, especially at higher pH, has been shown to replace up to 50% of bound As (Carabante et al., 2010). On the other hand, various sorbates (e.g. silicates, phosphates, and organic compounds) can

influence Fe crystallization by passivating Fe oxide surfaces (Schwertmann and Cornell, 2000). This might retard or even block ferrihydrite transformation in soils. Previous studies have shown that sorbed TE (e.g. As) can also inhibit the further crystallization and dissolution of Fe-(hydr)oxide minerals (Baleeiro et al., 2018; Paige et al., 1997; Melitas et al., 2002), by this maintaining TE immobilization efficiency over prolonged periods of time. Although preliminary studies indicate that the latter process might prevail in Fe-amended soil, spectroscopic confirmation from long-term field studies is needed.

Due to the redox sensitivity of Fe oxides, Fe amendments should be applied to well-aerated soils above the groundwater table, as prolonged water-saturated conditions can lead to drop in redox potential and subsequent reductive dissolution of Fe oxides and bound TE (Kumpiene et al., 2009b, 2013). In a field study which included water-logged conditions at a depth of 0.5–1 m, Nielsen et al. (2016) reported only a partial redox decrease, which did not affect Fe reduction or the release of bound contaminants over a 14-month experiment. A potentially more alarming consequence of redox changes that needs to be monitored during Fe amendment is the reduction of arsenate (As^{5+}) to arsenite (As^{3+}) (Kumpiene et al., 2009b; Nielsen et al., 2016), which is a more mobile and toxic As species in soil.

2.5. Organic matter

Numerous sources of organic matter, e.g. manure, sewage sludge, sawdust, bark chips, woodchips etc., are used as soil amendments, directly, or after pretreatment through, e.g. composting (Gadepalle et al., 2007). Composting is a process during which organic wastes are degraded, mainly through microbiological processes. As a result of this degradation, compost normally shows high contents of plant nutrients and humus substances (Medina et al., 2015), which is one of the reasons of using compost as soil amendment. Since the material sources for composting are so diverse, a wide variety of properties, including presence of potentially contaminating substances (e.g. pharmaceuticals, TE in compost from sewage sludge), exist among different compost materials (Davis and Wilson, 2005; Gadepalle et al., 2007; Lu et al., 2012). This, in turn, can negatively affect soil quality (Rate et al., 2004; Fang et al., 2018), as well as public acceptance of these amendments. The use of biosolids (treated sewage sludge) as a soil ameliorant, for example, has raised public concerns in many countries. Despite of numerous benefits of biosolids application to land (Lu et al., 2012), this material is considered to be one of the main sources of metal contamination in those soils (Mahar et al., 2015).

Excessive concentrations of certain TE (e.g. Cu, Ni, Zn) in contaminated soil usually decrease plant growth and biological soil activity, eventually leading to a loss of soil organic matter (Viventsova et al., 2005). The use of compost as soil ameliorant is therefore expected to counteract these processes by improving soil properties, facilitating plant growth and microbial activity and promoting the establishment of mesofauna and animal communities (Abbott et al., 2001; Burkert et al., 2000; Cassman et al., 1996; Gadepalle et al., 2007; Lal, 2003; Maurya and Shah et al., 2003; Singh, 1995; Shafi et al., 2007; Williamson and Johnson, 1981). Moreover, compost addition may create suitable conditions to immobilize metals through the formation of metal complexes with organic constituents.

The interaction between organic matter and soil metals is complex. The functional groups of organic matter, such as alcohols, phenols, carbonyls and carboxyls, can dissociate their hydrogen ions and sequester metal cations through chelation, i.e. the formation of a ring structure (Bolan et al., 2003; Mahar et al., 2015; Park et al., 2011). If the added organic matter itself remains stable, chelated metals will remain immobilized in the soil. Organic acids that can form soluble complexes with metals, could, however increase mobility of these metals (Wiszniewska et al., 2016), although the phytoavailability of dissolved organic matter-metal complexes is lower than that of free ions in soil solution (Brown et al., 2003; Hernandez-Soriano and Jimenez-

Lopez, 2012; Li et al., 2000; Wang et al., 2014; Wiszniewska et al., 2016). If the organic moiety can get attached to metal (loid) directly through a covalent carbon-metal bond, the methylation of a metal (loid) would occur. Formation of methylated compounds of e.g. Hg, As, Sb, Se, can have an opposite effect and increase element bioavailability (e.g. Bradley et al., 2017). Most of the methylated compounds are formed biotically by microorganisms in a reducing environment (Mason, 2012). Hence, organic matter should be cautiously used as a soil amendment when metal (loids) that can form methylated species are present in the contaminated soil.

The stability of soil organic matter itself depends on many factors, including pH, redox potential, OM origin, ratio between low and high molecular weight organic acids, etc. Therefore, the impacts of various organic soil amendments on metal mobility and phytoavailability may differ considerably from case to case.

2.5.1. Results of field studies

Numerous indoor and outdoor pot studies have shown that a range of compost amendments decrease the phytoavailability of As (Cao and Ma, 2004), Cu, Fe, Zn, Mn and Pb (Castaldi et al., 2005; Udovic and McBride, 2012; Walker et al., 2004), although contradictory results have also been observed (Cao et al., 2003c). Similarly, field studies established in the USA (Brown et al., 2003, 2005, 2007a, b; Farfel et al., 2005), Taiwan (Huang et al., 2018), Spain, Poland, France and Austria (Clemente et al., 2006, 2010; 2012; Kareer et al., 2018; Kidd et al., 2015; Kumpiene et al., 2014; Madejón et al., 2018a; Quintela-Sabaris et al., 2017; Touceda-Gonzalez et al., 2017a, b) (Table 1) show highly variable results regarding TE mobility, bioavailability and plant growth after addition of compost for the treatment of various contaminated sites. In many of these field studies, the addition of organic soil amendments did not affect - or even increased - the mobility and phytoavailability of TE. The results for oxyanions, such as As, showed the highest variability. Most of the soil organic matter molecules are predominantly negatively charged and therefore cannot directly bind oxyanions. Mechanisms of As immobilization upon compost addition, which was observed in some studies (Quintela-Sabaris et al., 2017), are still unclear and may be the result of indirect effects, e.g. changes in soil sorption properties and formation of ternary complexes between organic matter, cations (e.g. Fe) and anions (Ritter et al., 2006). Although recent studies question the possibility for As to get incorporated into Fe-OM complexes (Sundman et al., 2015).

For the aforementioned reasons, the sole application of biodegradable organic compounds might not be the most suitable approach for immobilizing metals in contaminated soil. Combining biosolids with liming materials (i.e. lime-stabilized biosolids) has been shown to successfully restore vegetation cover over several decades and reduce the solubility of cationic TE (Cd, Pb, Zn) in areas degraded by mining and smelting activities in Piekary Slaskie, Poland (Stuckzynsky et al., 2007; Siebielec et al., 2018), Bunker Hill, USA (Brown et al., 2003), Leadville, USA (Brown et al., 2005, 2007a, 2009), Tar Creek, USA (Brown et al., 2007b) and Jasper County, USA (Brown et al., 2002a, 2002b). The purpose of the liming materials is to prevent pH decreases over the long-term. Thus, the likely mechanism of metal immobilization in such cases is lime-induced metal precipitation rather than sorption to organic matter, which was reported in a recent study from Taiwan (Huang et al., 2018). In cases when the soil pH did not change after compost addition, an increase in soil CEC was suggested as the reason for the reduced soluble fractions of Cu, Zn, Cr, Ni in contaminated paddy soil (Huang et al., 2018). Nevertheless, elevated uptake of TE by plants on the restored sites (e.g. Brown et al., 2009; Stuckzynsky et al., 2007) might become of concern and more information on mass balance of TE is needed to comprehensively describe the performance of such management practices.

2.6. Other amendments

Other soil amendments tested in field studies comprise lime, limestone, zeolites, gravel sludge, etc., which have been applied alone or in combination with the other reviewed materials (Mench et al., 2010). Lime, a traditional agricultural soil ameliorant, has been tested for metal immobilization as well. Lime decreases metal mobility due to the alteration of soil pH, but this effect is often short-lived. For this reason, annual lime application may be necessary to ensure metal stability in soil (Seoane and Leiros, 2001). Therefore, using lime as a sole mean for metal immobilization is uncommon. Lime is usually used as a co-amendment to counteract the acidifying effect of other amendments such as Fe sulfate or phosphoric acid.

Zeolites have been tested in several field studies in Prescot (Merseyside, UK) (Rebedea, 1997), Northrhine-Westfalia (Germany) (Müller and Marschner, 2005) for immobilization of metals. Although some promising results in terms of decreased water extractable metals (Cd, Cu, Zn) were observed in the amended soils (Rebedea, 1997), TE immobilization and plant uptake were not always coherent and varied depending on the tested soil, element and zeolite type.

Gravel sludge, a waste product of the gravel industry that contains illite, calcite and quartz, has been tested in field studies in Arnoldstein, Austria (Friesl et al., 2006), Dottikon, Rafz and Giornico, Switzerland (Krebs et al., 1999). These studies showed varying results both for TE extractability and plant uptake depending on soil, element and plant species.

Field studies using these and some other alternative amendments are uncommon, results are variable, and it is difficult to draw generic conclusions about their suitability for long-term TE immobilization at contaminated sites.

3. Discussion

Long-term field studies are crucial not only for evaluating the stabilizing efficiency of the amendments, but also for better understanding the behavior of the amendments under natural climatic conditions. The addition of an exogenous phase to a soil, *i.e.*, the stabilizing amendment and potentially new contaminants present as impurities, can alter the soil physicochemical equilibrium (Michalkova et al., 2016; Vitkova et al., 2017). Nevertheless, amendments are expected to alter the soil equilibrium so that the immobilization of TE can occur.

The field studies included in this review evaluated the amendment efficiency to immobilize TE directly by using plants grown onsite and/or through various extraction procedures that used *e.g.*, H₂O, 0.1 M Ca (NO₃)₂, 1 M NH₄NO₃, 1 M NH₄-acetate, sequential extractions, or bioaccessibility tests (PBET). Although single and sequential soil extractions can be useful for comparing different amendments, the direct sampling of the soil solution from field sites (Nielsen et al., 2016; Okkenhaug et al., 2016) is necessary for providing more precise information about both the changes in contaminant mobility and the efficiency of the amendment, as well as crucial for subsequent site monitoring. An intermediate step of outdoor lysimeters might also serve useful (*e.g.* Marchand et al., 2011). It is unfortunate that some of the reviewed field studies only used one type of indicators (either chemical or biological) to describe immobilization efficiency. Studies that follow the revegetation of barren contaminated sites do not always report how the studied amendments affect element leaching and extractability, while other studies do not include biological endpoints. This limits the broader understanding of the immobilization efficiency achieved in field.

Plants and the associated microorganisms (endophytes, mycorrhizae) are commonly considered as a part of immobilization strategies (Vangronsveld et al., 2009). If not purposely planted, vegetation will start colonizing as soon as the toxicity in soil decreases and it becomes a more suitable habitat. In addition to several benefits, notably, physical stabilization of surface soil, decreased erosion and water percolation,

and even immobilization of contaminants in the root zone, vegetation can also bring about undesired effects. These include the ability of plants to exude organic molecules and alter soil pH, which, as shown in some studies, might increase TE leaching and uptake by plants (*e.g.* Kumpiene et al., 2011). Even moderate concentrations of TE in shoots might lead to a substantial transfer of contaminants from soil to the biosphere when the plant biomass generated by the plant cover is not collected and/or the biomass provides insufficient dilution of the TE. Soil bacteria and mycorrhiza might also contribute to these processes (*e.g.* Ciadamidaro et al., 2017; Dharni et al., 2014; Phanthavongsa et al., 2017). As this undermines the primary objective of TE immobilization, field studies should always consider both chemical and biological endpoints, as well as include a mass balance, so that all of the relevant contaminant transfer pathways have been taken into account. Recent work has shown how coupling advanced spectroscopic techniques (XAS) with speciation modeling can be useful for predicting the long-term efficiency of amendments and the fate of the targeted contaminants (Tiberg et al., 2016). Such approaches should be more commonly applied in further studies evaluating the suitability of amendments for stabilizing TE.

3.1. Institutional hindrances to the method application in field

Although hundreds of laboratory studies implemented over several decades have indicated that TE immobilization has significant potential for remediating contaminated soil, the technique has barely reached the large-scale demonstration phase. The results from field studies have not lead to the expected impact on the decision-maker level, partly because many relevant stakeholders still may not be acquainted with the method (Adriaensen et al., 2008; Cundy et al., 2013; Onwubuya et al., 2009). Despite the substantial body of scientific evidence on the effectiveness of TE immobilization, this technique is often met with skepticism, not only by authorities, but also by scholars themselves (*e.g.* Chrysochoou et al., 2007). Convincing evidence from large-scale field demonstrations is critical and necessary to swaying authorities and scholars alike. The establishment of field experiments implies permits from authorities as well as agreements with site owners, both of which are considerably more cumbersome and time-consuming than laboratory tests.

Previous research suggests that resistance towards new environmentally preferable technologies can often be explained by the functioning of the *institutional setting* (*e.g.*, Montalvo Corral, 2002). Such institutional settings, which include rules, policies and attitudes (Peters, 2012), are unfortunately often signified by path dependency (Pierson, 1996), *e.g.* officials involved in the decision-making of soil remediation strategies have preferences for conventional solutions ('dig-and-dump') even if an alternative *in situ* technique could offer a better outcome for the society at a lower cost. The failure to adopt alternative remediation techniques, such as *in situ* chemical immobilization of TE in soil, is therefore likely to be partly explained by issues related to cost competitiveness and the necessity of learning and knowledge building (Cundy et al., 2015, 2016; Chen and Li, 2018).

In many countries, the actor that caused the pollution is responsible for solving the problem, with the polluter pays principle commonly being applied. If the pollution occurred a long time ago, or if the party that caused the problem cannot be held accountable (*e.g.* out of business), the responsibility is transferred to the land owner (Andersson et al., 2008; SEPA, 2018b). In any case, the responsible party, *e.g.* land owner, will be eager to handle the problem of contaminated soil at the lowest cost possible. As evidence for applying stabilization methods to full-scale scenarios is scarce, the actual costs of *in situ* soil stabilization in comparison with the established landfilling solution are not well-known. The economic perspective on technical change maintains that gradual cost reductions in a new method will be a result of learning-by-doing activities associated with the dissemination of a new technology (Jaffe et al., 2002). Hence, the costs of large-scale immobilization

techniques can be expected to decrease once they are commonly used. However, as the currently used ‘dig-and-dump’ solution may cause negative external costs, *i.e.* if contaminated soil continues to leach TE at the landfill, or contaminants are spread during soil transportation (through dusting and wearing of vehicles), the full cost of the landfill alternative is not revealed or paid for. Instead, when traditional landfill is used as the management solution for contaminated soil, the external costs associated with soil remediation will be transferred to society, as the responsibility for a closed landfill will fall on the local authority. As a result, landfills can be considered to be over-utilized from a societal perspective. One way to adjust for this market failure would be to internalize the external costs (including landfill construction, maintenance, closure and long-term aftercare) into the cost associated with moving the contaminated soil to landfills, thus making the landfilling option more expensive.

Public authorities shape the formal institutional framework (*i.e.* policy and rules) and make decisions on the environmental permits for handling soil contamination. In most countries, policies and rules concerning the management of contaminated soil are mainly set at the national level, often by national environmental protection agencies. The ways in which polluted soil is handled are significantly affected by decisions at the regional and, in particular, municipal levels, as these local authorities often make land-use planning decisions. Considering that public authorities are responsible for making efficient decisions, it is important that they are well informed about the full costs and benefits of the different methods for handling contaminated soil. For this reason, providing reliable information on the costs as well as expected effects on humans and the environment associated with different remediation techniques is a task that can have far-reaching impacts and repercussions. Although cost-benefit analyses can be used to estimate how various remediation alternatives will profit society (*e.g.* Johansson, 1993), these methods are limited by the fact that the extent of the external costs is generally unknown and that some impacts are described with substantial uncertainties. Furthermore, it is questionable whether the benefit transfer model (*i.e.* estimating the economic value of remediation by using data from previous projects) can provide reliable estimates as the impacts of remediation techniques vary considerably between sites. Furthermore, performing cost-benefit analyses for every contaminated site would require extensive resource. Söderqvist et al. (2015), who used a polluted site close to Gothenburg, Sweden to assess how uncertainty influences the cost-benefit approach, concluded that cost-benefit analyses should be used in conjunction with other assessments.

It is, however, likely that the public authorities will prioritize a more established option to a less familiar solution - even if the new solution is superior from a societal cost perspective - due to the lack of knowledge and experience. Previous research suggests that organizational change often depends on public authorities’ abilities to learn about the available possibilities along with previous experiences of adopting new technologies (*e.g.* Haas, 1990; Hall, 1993). Precisely for this reason, knowledge transfer with the public authorities - especially at the local level - will be crucial to increasing the share of management alternatives other than landfilling (Cundy et al., 2015). Other researchers have proposed the use of different decision support tools (*e.g.* Onwubuya et al., 2009).

When Lemming et al. (2010) assessed the health and environmental effects associated with a PCE-contaminated site, they found the health costs to be negligible, while the environmental costs contributed to about 30–50% of the estimated remediation costs. Still, institutional frameworks may be conservative when possible environmental or human health risks are involved, and this is particularly noticeable when there is inadequate evidence about these effects. For example, if the public authorities *believe* that a new remediation technique involves a risk to society, *e.g.* that the problem with contaminated soil cannot be solved unless the soil is moved, they will not consider this alternative a sufficiently safe solution. Public authorities are often obliged to adhere

to the precautionary principle, *i.e.* they have a social responsibility to protect the public from exposure to harm when scientific knowledge covering the risks is incomplete. The attitudes of the general public may also hinder a shift towards applying alternative remediation techniques. For instance, beliefs among those who will use the land (*e.g.* for residential purposes) that the landfill is a safer alternative from a health perspective may well translate to the general public opposing decisions to use *in situ* immobilization technologies. For this reason, strategies for advocating *in situ* immobilization methods must include relevant and reliable information that is geared towards the general public. This will first enhance public knowledge building - which helps prevent ill-founded opposition - and then ensure societal support (*i.e.* legitimacy, *e.g.* Scharpf, 2017; Schmidt, 2010). In this respect, how information from public authorities and land owners regarding new treatment solutions is framed can also be expected to affect public values and attitudes towards change. Proponents of new remediation techniques need more information on the factors that affect stakeholder perceptions of these techniques, as previous research on failures to adopt alternative remediation approaches has identified stakeholder participation and acceptance among the general public to be crucial factors (Cundy et al., 2013; Onwubuya et al., 2009).

In sum, an inertia that may hinder authorities to embrace alternative treatment solutions and impede change of the management strategies for contaminated soil is that institutional frameworks are conservative in nature and can be expected to hold a certain (inherent) resistance to change. In this way, institutional structures and path dependency help explain why public authorities are slow to adopt alternatives. There are, however, possible ways forward. One way to advocate the adoption of *in situ* treatment could be to make the full costs of the current norm, *i.e.* the landfill solution, transparent, after which policies could internalize the complete external costs. It might, for example, be possible to impose a tax on moving contaminated soil. At the same time, researchers should focus on disseminating content that highlights the benefits and risks of alternative technologies with the end goal being the improvement of public knowledge of *in situ* immobilization options.

4. Conclusions

Despite the substantial body of literature that describes TE immobilization in contaminated soil performed on the laboratory scale, large-scale field validations/demonstrations remain scarce. Moreover, there is a considerable number of field tests that have not been described in peer-reviewed scientific literature (*e.g.* briefly described in conference papers, reports, brochures or governmental websites), and only provide fragmental or superficial information on the method implementation and performance. Field studies are not always aligned with the policies of peer-reviewed journals, *i.e.* publishing novel content rather than follow-up results. With such a scarce amount of well-documented scientific studies, it is not surprising that the market penetration of the technique is poor. This is not helped by path dependency and conservative institutional structures, with the public authorities being slow to adopt alternative remediation technologies, especially when the total TE concentration in soil remains the same and all the risks are not convincingly described.

Successful laboratory studies are often concluded with a statement that the method should be tested in the field. Institutional barriers (*i.e.* permissions from authorities and land owners) contribute to the slow transition from the laboratory to large-scale field demonstrations. Therefore, it is important to enhance knowledge-building of public authorities and society at large about the benefits of alternative remediation techniques, after which it may be easier to gain support for the large-scale demonstrations that will provide scientific evidence for the effectiveness of *in situ* immobilization at contaminated sites.

Most of the implemented field studies show a certain degree of improvement in the soil and/or vegetation status following soil

amendment. As soil toxicity decreases, plants and microorganisms will colonize the treated soil, which will induce dissolution/precipitation reactions and drive the geochemical soil conditions away from equilibrium. The net-effects of such processes on TE circulation can only be evaluated by monitoring the sites over extended time periods. This further motivates continuing existing field studies and establishing new sites at contaminated areas.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apgeochem.2018.12.003>.

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