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Use of Amendments to Restore Ecosystem Function to Metal Mining-Impacted Sites: Tools to Evaluate Efficacy

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Abstract As in situ use of amendments for restoration of metal-contaminated mining sites becomes increasingly accepted, the expected level of ecosystem function at these sites will increase. Use of appropriate tools to measure both the level and value of that function is critical to expand use of this approach. For these sites, amendment mixtures must reduce metal availability in situ and restore ecosystem function. Combinations of mixtures, typically consisting of a material with high metal binding capacity (cyclonic ashes, municipal biosolids, or other materials rich in Fe, Al, or Mn oxides), material to adjust soil pH (sugar beet lime, cement kiln dust, dolomitic limestone), and an organic residual to provide soil structure and nutrients (composts, animal manures, municipal biosolids) have been tested in multiple lab and field trials on metal-contaminated sites. This review focuses on field tests of this approach with the goal of providing methods to quantify reduction of hazard and restoration of functional systems. Methods to evaluate success of amendments including extractions to measure changes in metal availability, microbial function and diversity, phytoavailability of metals, and earthworm and small mammal assays are discussed. In most cases,

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² Crop Systems and Global Change Laboratory, Agricultural Research Service, USDA, Bldg. 007, BARC-West, Beltsville, MD 20705, USA measures of metal availability and ecosystem function are related. For example, surveys of small mammals on restored sites provide information on metal availability as well as suitability of restored habitat. Additional measures of ecosystem function including soil fertility, physical properties, and diversity of habitat are described. Finally, measures of the value of this approach for restoring ecosystems are detailed.

Keywords Metal mining wastes · Biosolids · Composts · Amendments · Ecosystem function · Bioavailability · Ecosystem services

Introduction

Mining and processing metal ores generates significant quantities of waste. The waste materials typically consist of a mixture of overburden, the mineral material overlying the ore deposit and tailings, and the residual material from mineral extraction. The level of contamination in these materials will depend on the efficiency of metal extraction. Aerial deposition of metals through smelter emissions and smelter slag are additional sources of contamination. These sites can impact significant area. For example, the Tri-State mining district in the USA includes portions of Kansas, Oklahoma, and Missouri [1]. Mining of Pb and Zn ores began in the mid-1800s with over 100 mines and 17 smelters in operation in Jasper County, MO during the peak of the mining operations. Metal-contaminated mine waste covers over 600,000 ha and the area includes multiple sites on the US EPA National Priorities List. Use of residual-based amendments to reduce metal availability in situ and restore ecosystem functions to these sites offers multiple benefits. However, it is necessary to identify appropriate combinations of amendments to reduce metal availability and reestablish ecosystem function. As part of this, determining

appropriate tools to measure efficacy of amendments is critical. Amendments must reduce metal availability to a range of receptors. In addition, the amended substrate must demonstrate the ability to support diverse ecosystem functions such as nutrient cycling, water retention, carbon retention, and providing habitat for diverse groups of species of flora and fauna. This review discusses historic use of amendments to restore disturbed sites where lack of nutrients and poor physical properties were the primary obstacles preventing establishment of a selfsustaining cover. Two approaches to restoration of disturbed sites that are also metal contaminated: use of metal-tolerant cultivars to reestablish a plant cover and use of in situ soil amendments to reduce contaminant availability will be summarized. A combined approach, where amendments are used to both limit contaminant availability and restore soil function, is outlined. We refer to this as an ecosystems approach to restoration of metal-contaminated mine wastes as it considers both the reduction in metal availability and establishing a functional ecosystem. This ecosystems approach is the primary focus of this review. In order for this approach to be adopted, it is critical to provide appropriate tools to measure efficacy of amendment combinations. Field studies of amendments to restore metalcontaminated sites and the tools used to evaluate both reductions in metal availability and level of ecosystem function will be discussed. A final section discusses attempts to value the ecosystems that have been reestablished on these sites. Quantifying the value of these systems is likely to be critical in gaining larger-scale use of this approach.

Historical Use of Amendments for Disturbed Soils

Use of soil amendments for restoration of disturbed soils is a well-established practice [2-4]. Here, we are referring to cases such as surface mining or deposition of mining-related wastes where the removal or replacement of the surface soil horizon has resulted in a surface material that is incapable of supporting a self-sustaining plant cover. Examples include coal mining, borrow pits, and sand and gravel mines. Amendments used at these sites were typically high organic matter residuals including municipal biosolids, composts, animal manures, and pulp sludges as well as inorganic residuals including coal combustion byproducts, sugar beet lime, dredged materials, water treatment residues, aluminumprocessing residuals, and foundry sands. There are many examples in the research of different organic amendments being used to restore a vegetative cover to disturbed soils. Initial research on this approach had focused on the efficacy of this practice, including testing of different rates and types of amendments for lands disturbed by coal mining with limited work on sand, gravel, and other types of mines [3, 5-7]. In each of these cases, the primary impediments to establishment of a plant cover on the disturbed surface were poor physical

properties and low nutrient status. Compared with the difficulty in remediating metal mining disturbed sites, these were comparatively simple infertile soils, but the prototype for problem solving of disturbed soils.

There was also work done to verify the safety of the practice from the perspective of the amendments used for restoration rather than any hazards posed by the substrate. As much of the work was begun with municipal biosolids prior to the 1993 regulations regarding metal concentrations [8], there was also research done on the availability of metals and nutrients added to the substrate with the biosolids to plants and groundwater [3, 9–13]. Studies have typically shown minimal to no negative impact to groundwater or plants from metals added with biosolids. Spikes in nitrogen in groundwater have been observed following amendment addition. They are typically observed during the first year after amendment addition with concentrations decreasing over subsequent measurement intervals [13–15]. The addition of high carbon materials with biosolids or composts has also been shown to limit nutrient movement [2, 14]. In addition to demonstrating the safety of this practice, these studies have generally shown that use of organic amendments has resulted in accelerated soil formation [16]. More rapid recovery has also been shown with use of residuals in comparison to topsoiling or chemical fertilization [16, 17]. In addition, this approach has often resulted in high levels of ecosystem function on the treated soils. For example, restoration with amendments has allowed for a return of the disturbed lands to production agriculture with yields similar to those observed in undisturbed farmland [17-19]. This approach has become sufficiently accepted in parts of the USA that there is recommended guidance and/or regulations for types and application rates of amendments to disturbed lands [4, 8, 20, 21].

Metal-Contaminated Sites

Phytostabilization

Metal contamination can occur on agricultural soils, native soils, or soils in urban areas. In these cases, the ability of the soil to support a plant cover is not compromised. For surface residuals from metal mining-impacted areas, establishment of a self-sustaining plant cover requires reducing metal availability and restoring soil function. For these sites, the primary concern is a reduction in the phytoavailability and bioavailability of the contaminants and associated hazards to the ecosystem. In addition, typically, mining wastes are akin to subsoils with poor physical properties and with very low fertility. Phytostabilization is a term used to denote many approaches to using soil amendments and plants to alleviate the contaminant risk of contaminated soils including soils contaminated by metals. Initial efforts on these types of sites often focused on establishing a vegetative cover to stabilize the contaminated materials and so reduce hazards from wind and water erosion [22-26]. In many of the initial studies, work concentrated on identification of metal-tolerant plant species that are able to grow in a high metal substrate [27-30].

Reducing Metal Availability

A second or related approach has focused on reducing metal availability in situ using soil amendments so that plant growth on contaminated sites is not limited to tolerant species. A number of soil chemical mechanisms have been used. For example, phosphorus addition to Pb-contaminated sites has been used to reduce the bioavailability associated with Pb in situ though the precipitation of pyromorphite [31, 32]. There are also examples of high surface area oxides used to absorb Pb, Zn, and Cd. For example, Vagronsveld et al. [24] noted the ability of cyclonic ashes (berengite) to reduce Zn, Cd, and Pb availability to plants and allow for a persistent vegetative cover for at least 12 years after amendment addition in a gold mine spoil in Portugal. This observed reduction in availability may be the result of oxide adsorption and potentially the formation of metal-layered double hydroxides. Spectroscopy has shown that in contaminated soils, Zn can precipitate as a component of double hydroxide mineral [33, 34]. Addition of high Fe biosolid composts to Pb and As co-contaminated sites resulted in increased association of both elements with Fe oxides [35]. Increasing soil pH will also reduce the availability of most metals although in calcareous systems, increased metal solubility may be observed as a result of displacement off of adsorption sites by soluble Ca. These results suggest that using amendments to restore ecosystems to metal mining wastes will require an amendment or adjustment to reduce metal solubility.

Ecological Approaches

Most recently, phytostabilization has been combined with the use of amendments to establish a self-sustaining plant cover on metal-contaminated wastes by simultaneously reducing metal availability and restoring a functional soil to these sites. This has been referred to as the "ecological approach" (23 citing Jeffrey et al. 1975). In order for this approach to be successful, mechanisms for decreasing contaminant availability have to consider a range of potential pathways and receptors. Amendments also have to improve soil physical properties and fertility. In the same way that amendments restored productivity to disturbed soils, amendments have the potential to restore function to disturbed and contaminated soils, resulting in active systems both below and aboveground. With the return of plants and wildlife, it is critical to assure that the restored site does not become an attractive nuisance

[36]. In this approach, biological processes including nitrogen transformations, nutrient cycling, and decomposition are reestablished on the waste materials, transforming them into complex soil ecosystems. More recently advocates of this approach have suggested it as low cost, more sustainable and desirable alternatives to conventional remedial methods for metal-contaminated sites. Volchko et al. [37, 38] stress the importance of incorporating ecosystem function and the provision of ecosystem services into the consideration of remedial alternatives. Cundy et al. [39] describe gentle remediation options (GRO) as "techniques for contaminated sites that result in no gross reduction in soil functionality (or a net gain)." Munns et al. [40] note that the US EPA established a set of generic ecological assessment endpoints (GEAE) to the risk assessment process in 2003. These include receptors that are of high ecological relevance that are also susceptible to contaminants. This approach has recently been expanded to include ecosystem services with the focus on services that are beneficial to people. This addition is intended to complement the existing GEAE approach. It provides a way to directly link ecological risk to human well-being and, in the process, makes it easier to communicate the importance of this type of remedial approach. With this approach, it is required to define the direct benefit of the ecosystem service to people in order for it to be considered (Fig. 1).

Use of Ecosystem Endpoints

There are an increasing number of examples of research including greenhouse and field trials as well as full-scale use of amendments to both limit contaminant availability in situ and restore ecosystem functions. This approach has focused on combinations of amendments to achieve these goals. A number of different amendment combinations have been tested. A list of field trials including the nature and concentrations of the contaminants and amendments used is shown in Table 1. Evaluations of success of the amendment combinations have also expanded to include measures of ecosystem function as well as broader assessments of the bioavailability of metals in the treated materials. In each case, at least one combination of the amendments tested has demonstrated the efficacy of amendments to both reduce contaminant phytoavailability and bioavailability and to reestablish some level of ecosystem function to the sites.

In order for in situ amendment addition to provide longterm stabilization, it is critical to document that the sites are able to support functional ecosystems. For example, in a review of revegetation of mine tailings in arid environments, Mendez and Maier [26] set out soil, microbial, and plant criteria to evaluate success (Table 2). Ecosystems are complex with interrelated functions that are not fully understood or quantified. Understanding and quantifying the success of amendments to restore self-sustaining plant cover and Fig. 1 In situ restoration of alluvial tailings in Leadville, CO was used as a remedial alternative. Measures of ecosystem function were used to evaluate the efficacy of this approach [41]. The reestablishment of trout fishing on the river would qualify as an ecosystem service [40]



Re-establishment of an active trout fishing area constitutes an ecosystem service

function to these sites has to occur on multiple levels. On the first level, it is essential to document a reduction in the availability of the contaminants [55]. As the contaminants can potentially impact a range of receptors, this is not always clearcut. On the next level, the remedies have to restore function to soils. This typically involves a consideration of the soils' ability to supply and recycle nutrients to plants as well as the physical properties of the substrate. Finally, it is increasingly important to document restoration of ecosystem functions to these sites. Here, definitions of functions and criteria to quantify function can vary based on site location, characteristics, and desired end use. Examples of functions include microbial assays, net primary productivity, and nutrient cycling [56]. In many cases, these different levels of measures can overlap. For example, measures of microbial diversity and activity can be considered to reflect ecosystem function but have also been used as a measure of reduction of contaminant bioavailability [41, 42, 49, 51, 57-59]. Examples of different measures used for each category are discussed below.

Bioavailability Measures

Bioavailability is the availability of elements to animals which ingest crops or contaminated soils, or soils/dusts which are inhaled by animals. Different measures have been used based on the scale of the study and targeted receptors. As contaminants are left in place, these measures cannot focus on total concentrations. In certain cases, tests to measure toxicity of waste materials such as the US EPA toxic characteristic leaching procedure test have been used to assess changes in availability of contaminants [41, 60]. However, these extracts

have not typically been developed to predict in situ toxicity and are not designed for a focus on ecosystem functions. For these sites and goals, it seems more appropriate to consider impacts of the contaminants to the range of potential receptors. The bioavailable fraction of the total concentration has to be the focus [55]. What portion of the total metal constitutes the bioavailable fraction can vary based on the targeted receptor. Chemical extracts of soils have traditionally been used to predict nutrient availability in soils. These are often aggressive as they attempt to mimic the behavior of plants that alter soil chemistry in order to increase nutrient solubility [61]. In contrast, dilute salt extracts of soils are frequently used to predict phytoavailable fractions of total metals in contaminated soils. Here, the extracts are designed to mimic soil solution. Table 3 lists different versions of these extracts used to measure bioavailable metals in the field trials on mining contaminated sites. Common extracts include deionized water [52-54] and Ca(NO₃)₂ [42–44]. Kumpiene et al. [62] used soils from different sites across Europe that had been treated using different GROs including plant-based phytoextraction, phytostabilization, and in situ stabilization to assess different chemical extracts to predict ecotoxicological responses. The best correlations were found with NH₄NO₃ and NaNO₃ extractable trace elements and the ecotoxicological responses. Nitrate solutions avoid chloride complexation of Cd and some other elements by the extractants. Ecotoxicological responses were measured using plant growth and enzyme assays, earthworm preference tests, and nematode growth and reproduction tests. However, clear correlations were not found across extractions and assays for one of the soils where amendments had previously been shown to effectively reestablish vegetation with low metal uptake [52].

Table 1	Field studies that have tested the use of amendments to reduce metal bioavailability and reestablish ecosystem function on mining	impacted
soils		

Study	Time	Туре	Location	Site type	Contaminants (mg kg ⁻¹)	Amendment Rates at t ha^{-1}
Baker et al. [42]	2 years post amendment	Small-scale replicated plots	Kansas, USA	Mine waste materials—primarily overburden	Pb (2300–3400) Zn (6200–6800)	Composted manure (45 and 269) \pm CaCO ₃ and bentonite
Brown et al. [43]	2 years post amendment	Small-scale replicated plots	Idaho, USA	Pb, Zn mine waste	Cd 13–82 Pb 1100–8700 Zn 2500–19,100	Municipal biosolids, biosolid compost, wood ash, pulp and paper sludge, log yard debris in various combinations
Brown et al. [41]	2 years post amendment	Large scale (>1 ha)	Colorado, USA	Alluvial pyritic Pb, Zn mine tailings	Cd 9.5–27 Pb 1390–3170 Zn 1400–2520	Municipal biosolids (224) and lime (224)
Brown et al. [44]	6 and 18 months post amendment	Small scale replicated plots	Oklahoma, USA	Pb, Zn tailings and overburden	Cd 29 Pb 4000 Zn 6830	Di-ammonium phosphate, biosolids, compost, high Fe residuals, added singly and in combination
Trlica and Brown [45]	5 years post amendment	Small scale replicated plots	Colorado, USA	Alluvial pyritic Pb, Zn mine tailings	Cd 11.2 Pb 2060-4120 Zn 3370-3910	Lime kiln dust 224, municipal biosolids and woody material to achieve 25 and 50 g kg organic matter and C/N ratios from 8–50:1
Brown et al. [46]	Up to 7 years post amendment	Small scale replicated plots	Colorado, USA	Alluvial pyritic Pb, Zn mine tailings	Cd 75 Pb 2600 Zn 6700	Different types of lime (224 CaCO ₃ equivalent) + biosolids (224)
Brown et al. [1]	Up to 13 years	Large scale (>1 ha)	Missouri, USA	Pb/Zn tailings and overburden	Cd 18–57 Pb 1100–4900 Zn 3200–10.000	Biosolids 110–336 + lime 24–48, mushroom compost 224, poultry manure 224
Cordova et al. [47]	5–9 months post amendment	Small scale replicated plots	Chile	Smelter contaminated	Cu 194–607 Pb 39–76 Zn 110–149	Lime (6.7), lime (6.7) + compost (133) each \pm planting
Friesl-Hanl et al. [48]	5 years post amendment addition	Small scale replicated plots	Austria	Agricultural soils contaminated by smelter deposition	As 32 Cd 5.6 Pb 913 Zn 545	Red mud 1 % w/w, gravel sludge + red mud 2.5 + 0.5 % w/w, limestone 0.5 % w/w,
Galende et al. [49]	6 months post amendment addition	Small scale replicated plots	Spain	Mine waste materials-primarily overburden	Pb 28,380–46,635 Zn 25,774–71,139	Cow slurry, poultry manure compost, pulp sludge mixed with poultry manure
Madejón et al. [50]	Up to 2 years post amendment	Small-scale replicated plots	Spain	Tailings dam spill onto agricultural soil	As 211 Cd 4.4 Cu 119 Pb 471 Zn 381	Municipal biosolids compost (2×30) , a high humic acid low grade coal (2×25) + sugar been lime (2×10) and sugar beet lime (2×30)
Pepper et al [51]	Up to 10 years	Large scale (>1 ha)	Arizona, USA	Copper tailings	Cu 570–2500 Mo 45–195	Municipal biosolids at 248, 270 and 371
Stuczynski et al. [52]	1–5 years post amendment addition	Large scale (>1 ha)	Poland	Smelter wastes	Cd 540 Pb 7900 Zn 30,900 Cd 2310 Pb 23,800 Zn 75 100	Municipal biosolids 300 + lime as mixed oxide and carbonate at 31, Carbonate cap of 30 cm depth with 300 biosolids applied above the cap
Ulriksen et al. [53]	7 months post amendment addition	Small-scale replicated plots	Chile	Aerial deposition of smelter waste	As 32 Cu 291 Pb 28 Zn 145	Lime (6.7), lime (6.7) + compost (133) each \pm planting
Vangronsveld et al. [54]	5 years post amendment	Large scale (>1 ha)	Belgium	Smelter waste	Zn 2000–16,000	Berengite (high Al residue from coal combustion) (120) + MSW compost (100)

Bioaccessibility is measured by a chemical method which has been shown to extract an amount of a contaminant which is well correlated with the bioavailability of the contaminant measured by an animal feeding trial. Soil extracts can also be designed to predict bioaccessibility to other receptors. An example is the physiologically based extraction test (PBET) procedure [32]. The PBET procedure was initially developed to predict the portion of total Pb in soils that would become soluble and is absorbed in the human gastrointestinal system, but versions of this extract have also been developed to predict metal toxicity to waterfowl [32, 63, 64] and terrestrial birds [65]. It is important to note that much better correlation of bioaccessibility with

 Table 2
 Criteria for evaluating successful in situ stabilization of mine wastes including tailings and overburden

Soil	Microbial	Plant
Improved aggregation	Increased population of heterotrophic bacteria and fungi	Shoot metal concentrations acceptable for wildlife
Reduction of erosion and runoff	Decrease in autotrophic iron and sulfur-oxidizing bacteria	Biomass and percent cover comparable to undisturbed neighboring sites
Reduced metal bioavailability and mobility		Self propagation
		Colonized by native species Plant cover and productivity for >10 years

Adapted from Mendez and Maier [26]

mammalian bioavailability was demonstrated for pH 2.5 than the EPA set pH 1.5 for this extraction [32].

Plant Response/Metal Uptake

Plant metal uptake is one of the most accepted measures of metal availability. The majority of field trials included measures of plant metal concentrations as a means to demonstrate efficacy of the amendments in alleviating phytotoxicity (Table 3). Plant metal concentrations can reflect the potential for phytotoxicity but may also indicate potential for revegetation to pose hazards to wildlife that use the site for grazing [36, 52, 66]. Stuczynski et al. [52] specifically grew hay on the restored site for use in a feeding trial to determine whether the plants would pose a risk to wildlife or livestock grazing on the site. This study illustrates errors in risk prediction based on feeding Cd salt-amended diets to estimate dietary Cd risks. Crops grown on contaminated sites will contain high levels of Zn if Cd is increased because in Zn-Pb mine wastes, Zn is commonly 100-200 times higher than Cd. In Stuczynski et al. [52], comparable levels of Cd were fed in control forage plus Cd salt as in the forage grown on the remediated site, but tissue Cd was markedly lower for the crop on remediated soil than for the control forage plus Cd salt. Feeding Cd salts strongly overestimates risk from crop Cd for nearly all contaminated sites. These results supported the findings of no increase in kidney or liver for the forages grown on the remediated smelter wastes. Similar results were shown for Cd in Swiss chard grown on soils amended with biosolids; crop Cd was increased significantly but kidney and liver Cd were not increased [67]. Other measures of metal availability or metal-induced stress to plants have been used. Vangronsveld et al.

[54] measured enzymes in plants as indicators of metal stress. This approach was recently tested as part of a method identification exercise for GROs [62]. Across a number of sites, the authors found that plant stress enzyme activity increased with increasing extractability of trace elements. Growth assays have also been used with soils collected from field trials [41, 62]. The advantages of greenhouse assays are that a single species can be used across multiple sites and that unique field conditions will not prevent growth. However, greenhouse trials can sometimes fail to predict field factors that can limit response. For example, in a field trial in Leadville, CO, low rainfall, and associated elevated electrical conductivity prevented plant establishment over multiple field seasons, something that was not anticipated in the greenhouse trial that preceded the field work [46]. Greater salinity tolerance of plants seeded, or irrigation after amendment of extreme acidity may be required at some sites [26].

Microbial Function

Increasingly, measures of microbial function are being used to evaluate reductions in metal availability and ecosystem function in amended sites. Microbes directly inhabit the contaminated substrate and are responsible for many of the soil reactions that allow for a self-sustaining vegetative cover. Measuring microbial diversity and function is increasingly common in both lab and field studies. On a basic level, studies have measured respiration as an indication of an active microbial community [41, 49, 52, 57]. Total microbial biomass has also been measured [41, 42, 68]. Functionality measures include the ratio of $NH_4/$ NO₃, an indicator of the microbial ability to oxidize ammonia [1, 41] and nitrification [42]. Studies have also included quantification of microbial enzymes [42, 49, 52, 58, 68]. There are also examples of tests of microbial diversity being used to assess the complexity of the restored systems in comparison to uncontaminated soils [51, 59, 69]. Pepper et al. [51] quantified the diversity in the microbial community in biosolid-amended Cu mine tailings in comparison to a control site and to untreated tailings. The group used DNA extraction followed by PCR amplification to characterize the richness and diversity of the microbial community on the amended tailings. They found that the community in the treated tailings, 10 years posttreatment, was similar in microbial number, activity, and diversity to native soils. As genetic tools to analyze microbial populations in soil become increasingly common, it is likely that this type of analysis will be used more frequently to assess ecosystem function in restored sites [51, 70].

 Table 3
 Measures to assess reduction in bioavailability as a result of amendment addition and ecosystem function in amended systems

Study	Soil extracts	Microbial assays	Soil biota	Plant measures	Mammal
Baker et al. [42]	Ca(NO ₃) ₂ extractable metals, DGT metals	Total microbial biomass , phospholipid fatty acid (PLFA) followed by fatty acid methyl esters (FAME), nitrification using aerobic incubation, enzyme assays including B-glucosidase, acid phosphatase, alkaline phosphatase, and arylsulfatase			
Brown et al. [43]	Ca(NO ₃) ₂ extractable metals			Plant Cd, Pb, Zn concentrations, biomass	
Brown et al. [41]	Water soluble, weak acid extractable, exchangeable, and TCLP metals	Microbial biomass C and N, CO ₂ -C respiration and NH <i>L</i> (NO ₂ ratio	Earthworm survival and metal uptake	Plant Cd, Pb, Zn concentrations	Small mammal number and whole body Cd, Pb, and Zn concentrations
Brown et al., [44]	NH ₄ NO ₃ extractable metals, extractable Fe and Al			Plant Cd Zn concentrations	PBET
Trlica and Brown [45]				Plant Cd and Zn concentrations	
Brown et al. [46]	Ca(NO3)2 extractable metals			Plant Cd and Zn concentrations	
Brown et al. [1]	Ca(NO ₃) ₂ extractable metals	NH ₄ /NO ₃ ratio	Earthworm survival and metal uptake	Plant Cd and Zn concentrations	Small mammal number and kidney Cd, Pb, and Zn concentrations
Cordova et al. [47]	0.1 KNO ₃ extractable Cu, Cu ²⁺ ion activity				
Friesl-Hanl et al. [48]	1 M NH ₄ NO ₃ extractable metals, soil pore water Zn			Agronomic plant metal concentrations in grain and straw	
Galende et al. [49]	CaCl ₂ extractable Pb, Zn, root elongation assay	Basal respiration, substrate-induced respiration, enzyme activities including b-glucosidase, arylsulfatase, acid phosphatase, urease, community level profiles using Biolog EcoPlates			
Madejón et al. [50]	EDTA extractable, CaCl ₂ extractable				
Pepper et al. [51]		Heterotrophic plate counts, bacterial community DNA extraction with PCR amplification, cloning and sequencing of 16S r RNA used for richness estimates and diversity index, nitrification, sulfur oxidation, and dehydrogenase			
Stuczynski et al. [52]	De-Ionized water extractable Cd, Pb, and Zn	Glucose induced respiration, total respiration, enzyme assays including alkaline phosphatase, acid phosphatase, urease, aryleulfatase, and dehydrogenase		Grass yield and metal concentration	Feeding study with calves fed with hay grown on the treated materials
Ulriksen et al. [53]	De-ionized water and KNO ₃ -extractable Cu			Plant tissue Cu, As, Zn and Pb concentrations	
Vangronsveld et al. [54]	Water extractable metals			Plant growth and morphological parameters, plant enzyme assays	

Soil Biota

On the next level, population counts and diversity of soil biota have been quantified on treated contaminated sites soils [41,

71]. Bouwman and Vangronsveld [71] measured nematode populations at a Zn smelter 12 years after a metalimmobilizing agent (cyclonic ash), and a municipal waste compost had been added to the site and found that 27 species of nematodes were present in the amended soils. However, a number of species that were commonly found on neighboring uncontaminated sites were not found on the restored soil, suggesting that only partial function had been restored. A shift in nematodes might have resulted in the raised pH of the remediated site (required to prevent Zn phytotoxicity) compared to nearly uncontaminated very acidic soils, so comparisons must consider the multiple changes in soils when remediation is required. This type of assay is only possible for field trials. Earthworm assays, based on survival and or survival and metal accumulation, can be used for field studies as well as laboratory incubated materials [1, 41, 62]. Brown et al. [1, 41] studied earthworm survival and metal uptake on treated tailings and found that amendments reduced toxicity and resulted in increased biomass. Results from earthworm assays can also be used to estimate the potential for contaminant transfer through the wildlife food chains [36, 41].

Mammals

One of the concerns with in situ amendments is that restoring vegetative cover on sites will result in creating an "attractive nuisance." Animals will return to the sites and be exposed to contaminants, effectively increasing the hazard posed by the contaminants. Brown et al. [1, 41] trapped small mammals and measured whole body and kidney concentrations of Cd, Pb, and Zn. Kidney pathology was also examined to determine if exposure to contaminants had resulted in damage to this organ. Results showed that amendments had restored sites sufficiently to attract high populations of small mammals and that damage to organs was only observed in a small minority of the animals that were captured. This type of study is expensive and is only possible on large-scale field trials. Vangronsveld [54] reported evidence of rabbits returning to the restored site but did not assess impacts to small mammals.

This brief summary suggests that a range of measures can be used to assess reduction in contaminant availability at metal mining-impacted sites where amendments have been used to reduce metal availability and restore ecosystem function. While a range of soil extractions have been used and typically show reduced extractable metals with certain amendments, this alone is not sufficient to demonstrate a reduction in toxicity. Kumpiene et al. [62] found high correlations between reduced extractable metals and other ecosystem endpoints in a controlled study using soils from a range of sites. Brown et al. [57] also reported a relationship between extractable metals and plant response for a range of amendments tested on a single soil in a multilaboratory study. Relationships have also been seen with reduced extractable metals and increased microbial function and populations [42]. Mild extractions similar to soil solution (0.01 M Ca(NO₃)₂ or 1 M NH₄NO₃) are much better predictors of metal remediation for plants than are strong extractants (DTPA), and higher solution/soil ratios are required to avoid saturation of chelating extractants [27]. Measures of microbial populations, activity, and diversity are likely to become more common with increased availability of tools to measure these variables. Plant assays are also frequently used and appear to be effective for demonstrating reductions in bioavailability, particularly when native rather than metal-tolerant species are tested. Testing efficacy higher up on the food chain will likely be limited due to associated costs and the need for larger-scale field trials. However, this type of approach may be helpful for regulatory acceptance.

Soil fertility and Physical Properties

For amended sites to have full ecological function, it is critical that the treated site soils can function as living soils. This involves both the ability of the substrate to supply required plant nutrients over the long-term as well as having the necessary physical characteristics to support plant growth. There are also a range of tests and accepted protocols to measure soil fertility and soil physical properties (phytoavailable metals; nutrients; salinity; SAR; Ca/Mg ratio, etc.). Despite this, not all studies include these measures. For example, while Kumpiene et al. [62] tested a set of measures to assess the efficacy of different GROs, no measures related to soil function, fertility, or physical properties were included. Some of the field tests of amendments have included at least a portion of these measures (Table 4). Changes in soil physical properties would include changes in bulk density, water holding capacity, texture, and organic matter or total C. While soil carbon and organic matter are not technically physical properties, there is a direct relationship between soil organic matter content and soil quality [72]. Use of organic amendments and increased soil C has also been associated with improved soil physical properties including increased water holding capacity and reduced bulk density [73, 74]. Many of the field studies included measures of soil organic matter or soil carbon. A few studies have included measures of soil water holding capacity after amendment addition [1, 47, 53] (Fig. 2). Increased water holding capacity was observed with organic matter amendment addition for each of these studies. Newly established plant cover will be more resilient if plants are not subject to drought stress. It would appear that increased water holding capacity would increase the potential for long-term success. Brown et al. also measured bulk density and found reduced bulk density (0.82 g cm^3) on sites where high rates of biosolids had been applied in comparison to the untreated tailings (1.52 g cm^3) .

Most of the field studies included measures of soil pH (Table 4). Soil pH is a critical variable for metalcontaminated sites as pH is a major factor controlling the solubility and availability of metals to many receptors. Increasing pH will decrease the solubility of most metals to

Table 4 Measures of soil fertility, physical properties, and plant nutrient concentrations on field sites restored using a combination of amendments

Study	Soil nutrients	Soil physical properties	Plant nutrients
Baker et al. [42]	pH, NH ₄ -N, NO ₃ -N, P, K	Total C	
Brown et al. [44]	pH, EC	Total C	
Trlica and Brown [45]	C:N ratio		
Brown et al. [46]	pH, EC		
Brown et al. [1]	Mehlich III Cu, K, P, total N	Organic C, Bulk density, % H ₂ O at 1 bar, texture	
Cordova et al. [47]	EC, pH, N,P, K, Ca, Mg	OM%, Field capacity (% w/w)	Plant concentrations of nutrients: N, P, K, Ca, Mg, Zn, Mn, Fe
Friesl-Hanl et al. [48]	pH, EC, CEC	Texture, organic C	Plant concentrations of nutrients: Cu, Mn, Mg, and P
Galende et al. [49]	pH, CEC, available P, total N, exchangeable K, Ca, and Mg	Texture, organic matter	
Madejón et al. [50]	pH, total S	Organic C	Plant concentrations of macronutrients: N, K, P, Ca, S, Mg, Mn
Stuczynski et al. [52]	pH and EC		
Ulriksen et al. [53]	Available N, P, K, Ca and Mg, EC	% OM, soil water holding capacity	
Vangronsveld et al. [54]	CEC, pH	OM	

plants and also reduce extractable metal concentrations. In contrast, increasing pH can increase availability and solubility of metalloids (Se, Mo). Soil pH is also a factor in determining the availability of many plant nutrients and so measures of soil pH were considered here to be a measure of soil fertility. Electrical conductivity (EC) is another parameter that will impact access to nutrients and the sustainability of in situ amendments. Particularly in arid areas, amendments with high EC can limit plant response even if contaminant availability is reduced [46, 52]. Thus, irrigation may be required to remove



Fig. 2 Reported changes in soil water holding capacity following amendment addition from field trials. Brown et al. [1] measured percent $_{\rm H2O}$ at 0.1 bar, Cordova et al. [47] measured percent $_{\rm H2O}$ w/w, and Ulriksen et al. [53] measured percent water holding capacity. All observed changes were statistically significant

excessive salinity [26], or a capillary breaking layer installed below improved topsoil [52].

More traditional measures of soil fertility include extractions to measure available soil nutrients as well as plant digests to assess nutrient concentrations in plant tissue. Several of the studies included one or both of these measures (Table 4). In mine waste-impacted materials, in addition to metal toxicities, nutrient deficiencies can be a limiting factor for plant growth. Use of organic amendments to restore soil function should include a consideration of the required macronutrients and micronutrients for plant growth. As the importance of and potential for establishing these sites as functional ecosystems is increasingly recognized, it is likely that traditional measures of soil fertility, quality, and productivity will be included in tools to evaluate amendment mixtures and remedial options. Long-term sites where disturbance rather than metal contamination was the primary factor limiting plant growth (e.g., coal mine sites) have shown that amendments are sufficient to reestablish productivity [19, 45]. It is likely that similar results will be seen with the appropriate use of amendments.

Ecosystem Services

Community Diversity

All of the field trials listed in Table 4 have shown the ability of amendments to limit contaminant availability, improve fertility, and restore a plant cover to treated materials. The restored systems appear able to support plant life that is typical for the particular region rather than only metal or salt-tolerant ecotypes [41, 50, 54, 75]. Córdova et al. [47] measured plant species diversity, percent cover, and biomass in Cu mining impacted materials amended with compost and limestone. Two revegetation techniques were tested in the trial: spontaneous and assisted revegetation. The researchers found that the native seed bank was sufficient to establish a native cover on the amended soils. Brown et al. [75] altered the C/N ratio of amendments through different ratios of municipal biosolids and woody material on alluvial Pb and Zn tailings in Leadville, CO. Plots were seeded with a mixture of native plants. Species diversity and percent cover were measured 1, 2, and 5 years after amendment addition. The treatment that had the lowest C/N ratio (8:1) had the highest biomass at year 5 as well as the lowest diversity. Results indicated that a C/N ratio greater than 20:1 resulted in increased species diversity. Vangronsveld et al. [54] observed multiple types of fungi on a high metal mine waste area amended with cyclonic ash and compost. The authors also noted evidence of animal browsing. Small mammal return to amended sites was also reported by Brown et al. [1, 41]. These results suggest that appropriately used amendments can restore ecosystem functions to the impacted areas.

Quantification of Value

To date, quantification of ecosystem services on restored sites has been limited [1, 76]. These benefits can include a range of ecosystem services such as nutrient and waste recycling and carbon sequestration. There is also a potential for social benefits such as increased open space and associated recreation opportunities. Finally, there are a range of economic benefits including agricultural production and biomass generation. Many of these benefits have been realized at disturbed noncontaminated sites restored with residuals. For example, high rates of carbon sequestration have been observed on sites reclaimed with organic residuals in comparison to sites restored with topsoil or fertilizer [19, 45, 77]. US EPA sampled contaminated sites restored with amendments to quantify soil carbon restoration [76]. At Leadville, CO, EPA quantified soil carbon storage at a number of sites where biosolids or compost and limestone equivalent had been applied to highly acidic (pH 3.5 due to sulfide oxidation) Pb-Zn alluvial tailings to reduce metal toxicity 10 years prior. Unamended tailings were used as a reference point. They found net carbon sequestration of 52–86 t C ha⁻¹, equivalent to 190–315 t CO₂ ha⁻¹ in the amended sites. Brown et al. [1] compared the ecosystem impacts of the use of amendments to restore Pb and Zn mine wastes to topsoiling. The cost of using harvested topsoil to cover contaminated materials was estimated by calculating the time required to build 15 cm of topsoil and the per hectare price paid to farmers to leave sensitive land fallow under the USDA Conservation Reserve Program. In comparison, the greenhouse gas costs and benefits of using municipal biosolids including avoided emissions of fugitive gasses, transport emissions, fertilizer credits, and soil carbon sequestration were calculated. The authors estimated that it would require 1875 years to replace 15 cm of topsoil resulting in a cost of US\$241,070 per hectare. In contrast, diverting biosolids from incineration to land application for the same amount of land would result in avoidance of 485 t of CO_2 and an additional credit of -156 t CO_2 .

Conclusion

Use of amendments to reduce metal availability and increase soil function appears to be transitioning from laboratory scale to field trial and full-scale use. A range of tools exists to measure success of these efforts and can be used to assess functioning of these systems on multiple levels. Full valuation of the benefits of this type of approach, however, is still being developed.

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Compliance with Ethical Standards

Conflict of Interest On behalf of all authors, the corresponding author states that there is no conflict of interest

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