



## Research article

# Identification and assessment of appropriate remediation management techniques for the recovery of soil-like material produced in landfill mining

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## ARTICLE INFO

Handling editor: Jason Michael Evans

## Keywords:

Landfill mining  
Soil-like material  
Resource recovery  
Phytoremediation  
Electrokinetic remediation

## ABSTRACT

Landfill mining has received major attention in recent years for the reclamation of waste disposal sites, including in developing countries such as India where significant efforts are being made to manage sites in this way. The bulk of the material obtained from landfill mining consists of fine-grained soil-like material (SLM) but its direct reuse in off-site applications is restricted due to the presence of harmful heavy metals, soluble salts and other pollutants. In this study, appropriate techniques for managing SLM to permit recovery and reuse are assessed. As a result, experimental investigation explores the efficacy of two remediation techniques considered appropriate for SLM management: electrokinetic remediation and phytoremediation. These were applied to SLM from a recently mined landfill and their ability to reduce heavy metal and other soluble salt burdens assessed. Electrokinetic remediation has shown considerable potential to mobilise and transport heavy metals and soluble salts through and from the SLM over an eight-week period. Phytoremediation experiments also demonstrated mobilisation and uptake of metals from the SLM over a similar duration although relatively low amounts were recovered as a result of the low biomass produced over this period. Both technologies have demonstrated potential for recovery of metals from SLM, as well as recovering the SLM itself as a potential resource.

## 1. Introduction

Landfilling of municipal solid waste (MSW) is a significant waste disposal route worldwide. Even where waste is diverted and recovered for beneficial use, such as through recycling and energy-from-waste schemes, landfill remains an important mechanism for management of large waste quantities and there is a significant legacy of existing and closed landfill sites. Multiple pressures drive the re-excavation of such sites, including resource recovery (e.g. materials of value, energy, fill materials and the land itself) and the management of various impacts on environmental and human receptors (Krook et al., 2012; Somani et al., 2018). To reduce accumulated legacy waste at dumps and reclaim the site for other purposes, so-called landfill mining can be a viable option (Somani et al., 2020). Whilst in many countries the approach is uncommon or is undergoing research, recently the National Green Tribunal (NGT), India, directed that mining of more than three thousand old MSW dumps in India should be undertaken to reduce the height of these, often unlined, 50–60 m high facilities (NGT, 2019).

Landfills of various types can have considerable resource potential

(Crane et al., 2017; Chandana et al., 2021), but this can be very difficult to extract economically – materials of value are distributed, diluted and heterogeneous whilst the land is often considered contaminated and unsuitable for construction. Soil-like material (SLM) is the predominant product of landfill mining. It accounts for 50–70% of the total mined waste (Mönkäre et al., 2016; Wanka et al., 2017; Hölzle, 2019; Somani et al., 2019; Singh and Chandel, 2020) and can contain significant quantities of potentially toxic elements and other problematic materials. Superficially it resembles a soil but due to its origins the nature of the grains will differ substantially with particles of plastics, wood, glass and other typical waste categories expected to be present. As a result, the interaction of chemical contaminants, and thus their fate and transport, may differ from behaviour in soil.

Concerns regarding heavy metal contaminants in SLM have been well reported in literature (Jain et al., 2005; Särkkä et al., 2018; Wolfberger et al., 2016; Hogland et al., 2018; López et al., 2018; Hölzle et al., 2022; Somani et al., 2023) and leaching can release unacceptable amounts of contamination (Prechthai et al., 2008; Masi et al., 2014; Somani et al., 2019). As a result, SLM is usually not the target resource

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from landfill mining but a byproduct; as such a significant component, however, opportunities for its beneficial reuse are urgently sought (Goli et al., 2022).

A major use of mined SLM has been as a cover material at the landfill site itself (Hogland et al., 2004; Jain et al., 2013; Bhatnagar et al., 2017). There are opportunities for the reuse of SLM in agricultural applications (Kurian et al., 2003; Prechthai et al., 2008; Masi et al., 2014; Rong et al., 2017; Datta et al., 2021) and geotechnical applications, i.e., as fill materials (Song et al., 2003; Oettle et al., 2010; Rawat and Mohanty, 2022). Liu et al. (2018b) reported the reuse of SLM as a microbial and bio-stimulation agent for the bioremediation of petroleum contaminated soil. Goli et al. (2022) presented the suitability of SLM as a buffering material. However, in such applications the contaminated nature of SLM, e.g., with contaminants such as harmful heavy metals and excess soluble salts, can be problematic and pre-treatment will be required (Somani et al., 2019). The authors are aware, from communication with landfill operators, of instances of the reuse of SLM in India in filling of low-lying areas and in agricultural applications without any pre-treatment, which may lead to environmental harm.

Much of the literature on SLM comprises studies highlighting its contamination potential, with little consideration thus far of the potential for treatment and recovery of SLM as a resource. As landfill excavation and mining is becoming significant in India and elsewhere, solutions for SLM treatment are required and the objective of this work is to identify and test appropriate techniques for its recovery and reuse. This study comprises two elements – firstly, an assessment of available technologies for decontamination or amelioration of SLM, based on existing contaminated land remediation and related technology, and secondly an experimental investigation into whether selected techniques considered appropriate for soil remediation are applicable to treat SLM.

## 2. Options appraisal of methods for reclamation of soil-like materials

Management of SLM contamination has previously been considered in a limited manner. Oettle et al. (2010) recommended blending SLM with locally available soil before reuse, although this is simply dilution rather than remediation. Washing of SLM (Wanka et al., 2017) was considered to be highly uneconomic, partly as a result of the challenge of dealing with contaminated fluid arisings. Mönkäre et al. (2016) have discussed the biological stabilisation of SLM, however, there was no mention about the removal of contaminants from SLM.

In an effort to identify feasible technologies for treatment and recovery of SLM, identification of objectives and constraints has been informed by the UK's Land Contamination Risk Management (LCRM) guidance (Environmental Agency, 2020) options appraisal framework. The purpose is to identify a shortlist of technically feasible technologies that satisfy requirements based on the likely form of the material. Step 1 of Stage 2 (Options Appraisal – Identification of feasible remediation options) comprises identification of management, technical and remedial objectives, consideration of regulatory controls and finally production of a shortlist of feasible remedial options. As it is not the purpose of this task to identify a site-specific single technology further use of this framework is not required.

The objectives of the options appraisal were identified, although unlike traditional site-based remediation, these are material-specific rather than site-specific. Ultimately, we wish to ensure that, after treatment, the material may be used in large volume fill and soil conditioner applications. Remediation technologies are required that:

- Are capable of treating large volumes of SLM with low-cost, 'appropriate' technologies. The product is unlikely to have significant inherent value as it will compete with low-cost virgin and other recycled materials. As a result, high-cost technology and/or process-

intensive techniques may not be appropriate (particularly in regions with less-developed remediation industries).

- Are capable of rapid, ex-situ treatment. This material will require treatment ex-situ, as it is a product of the sorting of excavated landfill contents, although this may take place in prepared beds and so be suitable for technologies considered to be applicable to porous media *in situ*. Rapidity is advantageous as it avoids long-term management requirements or a need for space. A 'rapid' process is one which may be considered as achievable on the scale of several months to a few years, rather than several years to decades.
- Treats the source of contamination, is capable of separating the prime contaminants (metals and inorganic salts) from the environmental matrix to a sufficient degree to permit reuse of the latter without further monitoring in the future. In the production of a material for reuse, the management of contaminant pathways or receptors is either not feasible or would require limitations to its use that would detrimentally affect its competitiveness in the market.

Options have then been screened via a simple subjective matrix analysis to identify the most feasible options, based on their achievement of the objectives identified above. Initially, technologies considered appropriate for heavy metals or non-metals in the LCRM Options Appraisal Matrix have been assessed (Table 1). Of these, only flushing/leaching was found to be feasible for potentially low-cost ex-situ source treatment with the potential for SLM recovery, and even this has detrimental aspects regarding production of large volumes of contaminated liquids requiring treatment and/or disposal (Sharma and Reddy, 2004). However, the other appropriate techniques exist with which there is less experience in the UK but which have been applied internationally and have considerable research backing (Liu et al., 2018a) and are assessed in a similar manner in Table 1. Of these, bioremediation (considered here to be biological stabilisation when applied to metals and inorganics) is not considered a feasible option but electrokinetic treatment and phytoremediation are. Although electrokinetic treatment does produce contaminated fluids, they are likely to be considerably lower in volume than would be the case with flushing/leaching.

As a result of the above analysis treatability studies for a particular SLM have been carried out on electrokinetic treatment and phytoremediation (the leachability of metals and inorganic salts from SLM has previously been considered (Somani et al., 2019; Datta et al., 2021)). Electrokinetics comprises the application of a low power, typically direct current electric field to contaminated soils to transport contaminants principally by electromigration, movement of charged chemical species under a field, and electrically induced water flow known as electroosmosis (see Peppicelli et al. (2018) for further description). Phytoremediation employs the actions of plants (e.g. water uptake, associated microbial activity) to ameliorate pollution with metals being extracted from or immobilised in soil (see Ali et al. (2013) for further details). Both technologies have been applied successfully to wastes or related materials (including organic-rich matrices). For example, the geochemical speciation of metals in industrial waste was electrokinetically redistributed from less available to more labile fractions (Peppicelli et al., 2018) whilst Peng et al. (2011) combined electrokinetic remediation with bioleaching for the dissociation of metals from sewage sludge. Both technologies have previously been suggested as appropriate for recovery of resource from geological waste deposits (Sapsford et al., 2017).

## 3. Experimental materials and methods

Two technologies feasible for the remediation of SLM have been selected to explore the amenability of the material for decontamination and recovery of resource: electrokinetic (EK) and phytoremediation. Laboratory studies have been carried out to explore their potential to permit the offsite reuse of SLM contaminated with heavy metals and soluble salts.

**Table 1**  
Screening of metal/inorganic contaminant remediation options for application to SLM.

		Low cost/tech?	Rapid	Source treatment	SLM recovery?
Typical technologies listed in LCRM (EA, 2020)	Cover systems	Yes	Yes	No	No
	Hydraulic barriers	Yes	Yes	No	No
	In-ground barriers	Yes	Yes	No	No
	Excavation and disposal	Yes	Yes	No	No
	Natural attenuation	Yes	No	Yes	Yes
	Soil flushing/leaching	Yes	Possibly	Yes	Yes
	Chemical oxidation	Possibly	Yes	No	No
	Permeable reactive barriers	No	No	Yes	Possibly
	Soil washing	No	Possibly	Yes	No
	Hydraulic binders	Yes	Yes	No	No
	Vitrification	No	Yes	No	No
	Incineration	Yes	Yes	No	No
	Thermal desorption	No	Yes	Yes	Possibly
	Additional technologies (Liu et al. (2018a))	Electrokinetic extraction	Possibly	Possibly	Yes
Phytoremediation		Yes	Possibly	Yes	Yes
Bioremediation (stabilisation)		Yes	Possibly	No	No

### 3.1. Sample collection and site details

The SLM investigated in the present study was collected from an old MSW dumpsite located at Bhalswa, Delhi, India. Waste dumping commenced in 1990 and the estimated total quantity of waste accumulated at the site is around 8 million tonnes. The site is spread over 22 ha and the height of waste has reached 65 m above ground level. The site is still active and receives 2500–3000 tonnes waste per day.

Aged MSW samples were excavated using a backhoe excavator from trial pits (1m × 1m in plan) from 4 locations at Bhalswa dumpsite. Samples were taken from a depth of 3–5 m (approx.) from locations where waste was lying for more than 20 years. The excavated MSW was found to be 20–25 years old on the basis of dates mentioned on recovered food packets and newspaper clippings. After excavation, initial moisture content was measured and then samples were dried at the site for a period of about 8–10 days until the moisture content was reduced to 8–10%. MSW was then screened through sieves of size 80, 50, 20, and 4.75 mm. Finally, four samples of SLM (<4.75 mm) were transported to the laboratory for further characterization. The homogenization of samples was achieved through the coning and quartering method. A further description of dumpsite and sample collection is available in our previous publications (Somani et al., 2018; Somani et al., 2020; Datta et al., 2021a).

For comparing SLM with background soil, samples of local soil were collected beyond the periphery of the dumpsites (up to a distance of 0.5–5 km away) from a depth of 0.5–1.0 m from the ground level.

### 3.2. Material characterization

#### 3.2.1. Compositional analysis and particle size distribution of aged MSW

In the present study, total aged MSW refers to the unsegregated municipal solid waste collected from the trial pits. Composition analysis of MSW was carried out as per the procedure outlined by Zekkos et al. (2010). The collected samples varied between 120 and 140 kg, in agreement with recommendations in procedures for measuring the composition of unprocessed municipal solid waste (MSW) as per ASTM D5231-92 (2016) (91–136 kg), and were washed with tap water to separate finer particles associated with coarser fractions such as plastics, stones etc. After washing, the samples were air dried under sunlight for 4–5 days. Characterization of air dried MSW samples was performed by first sieving through a 20 mm sieve, separating the sample into <20 mm (finer) fraction and >20 mm (coarser) fraction. The coarser fraction was manually sorted into different constituents, such as gravel, textile, plastic, paper, wood and wood like, glass and miscellaneous items. Particle size distribution was carried out on further air-dried MSW samples using sieve sizes of 80 mm, 50 mm, 20 mm, and 4.75 mm in accordance with Indian standard of practice (IS, 1985 (Part IV)-1985).

### 3.3. Treatment techniques

#### 3.3.1. Electrokinetic experiments

Electrokinetic (EK) experiments were carried out in acrylic containers (Peppicelli et al., 2018; Fig. S2) with the central chamber (200 mm length, 100 mm breadth and depth) separated from electrode chambers by 10 mm thick acrylic plate perforated with 4 mm diameter holes and lined with coarse-grained filter paper. SLM (1.2 kg) was placed in the central chamber and compressed to a depth of 85–90 mm. The electrode chambers were then filled with deionized water to a final depth slightly below that of the SLM, thus preventing a layer of water overlying the SLM which would lead to a 'short circuit' with current avoiding the waste. The water percolated slowly into the central chamber to saturate the SLM. After saturation, compressed graphite plate electrodes (100 mm high, 90 mm wide and 10 mm thick), were installed in the electrode chambers.

Three test conditions were maintained: Treatment Ep – EK system with pH control, Treatment E – EK system without pH control, and Treatment C – control system (no EK or pH control). In EK experiments, power was supplied by a bench-top power supply (max 30 V) and current was monitored by a digital ammeter. A constant current of the order of 20 mA was maintained over the experimental period of 60 days. Cathodic pH increases rapidly under EK conditions (see Acar and Alshawabkeh, 1993) and can be responsible for immobilisation of metals near the cathode. As a result, cathodic pH was controlled in treatment Ep by adding 2 ml of 5M hydrochloric acid at hourly intervals via a peristaltic pump. This volume was determined by initially carrying out a stoichiometric analysis of the generation of hydroxyl ions at a current of 20 mA followed by trial-and-error fine adjustments to maintain a consistent pH in the cathode chamber.

Prior to treatment, triplicate samples were obtained from homogenised SLM for analysis. After the experiment, SLM was collected from each treatment, from three equally spaced locations denoted as 'anode' (1.5 cm from the anode end of the SLM), 'central' and 'cathode' (1.5 cm from the cathode end). Subsamples of each were taken for (i) total heavy metal analysis (chromium, copper, lead, nickel, zinc) and (ii) leaching behaviour (see section 3.4.1) of heavy metals and leachable soluble salts (sodium, potassium, calcium, magnesium, sulphates, chlorides, nitrates, phosphates). Heavy metals and soluble salts were determined in liquid samples taken from electrode chambers on a weekly basis. Samples were analysed using methods described in section 3.4.

#### 3.3.2. Phytoremediation

Pot experiments with ryegrass (*Lolium perenne* L.) were conducted in order to assess the potential for metal uptake from SLM as well as changes in heavy metal and ion leachability. Ryegrass has been widely used for the removal of heavy metals from soil, sludge, and water

through phytoremediation (e.g. Guo et al., 2014; Zhu et al., 2015; Zhang M. et al., 2019; Li et al., 2020), and was selected here as a tool to demonstrate the feasibility of phytoremediation. In total, 30 pots were used in the experiment, each containing 80 g SLM amended with 48 ml quarter-strength Hoagland's solution (Sigma Aldrich, UK). Fifteen ryegrass seeds were sown in each of 15 pots (P1–P15), with the remaining 15 pots (C1–C15) used as controls (no ryegrass seed). All pots were watered with 24 ml quarter-strength Hoagland's solution (O) every week. Plastic trays (petri dishes) were placed under each pot and any leachate arising after watering was collected for the analysis of heavy metals.

Five replicate pots from each treatment were destructively sampled after 4 weeks (P1–P5 and C1–C5), 6 weeks (P6–P10 and C6–C10) and 8 weeks (P11–P15 and C11–C15). Subsamples of SLM taken both at the start of the experiment and from all pots after destructive sampling were analysed for total heavy metals and leaching characteristics. All plant biomass was dried and digested for metal analysis.

### 3.4. Analytical methods

#### 3.4.1. Leaching characteristics of SLM

Single batch leaching tests (Swedish standard method SS-EN, 20037-2, 2003, as used by Kaartinen et al. (2013) and Wanka et al. (2017)) were performed on SLM before and after remedial treatment. A liquid to solid ratio of 10 L/kg (10 g SLM, 100 ml deionized water) was mixed in a mechanical rotary shaker for 24 h at 100 rpm. After shaking, it was allowed to settle, centrifuged at 4000 rpm (relative centrifugal force of 5304) for 15 min and then filtered through 2.5 µm filter paper (Whatman no. 42).

#### 3.4.2. Solid samples extraction for heavy metals analysis

For heavy metal analysis, SLM was ground to <0.075 mm in a pestle and mortar. Around 0.2–0.5 g of sample was mixed with 8 ml of aqua regia (mixture of nitric and hydrochloric acids in a molar ratio of 1:3) before undergoing microwave digestion (180 °C for 20 min with a ramp rate of 10 °C per minute) following USEPA 3050B (1996). After cooling to room temperature, the digested mixture was filtered through Whatman No. 42 filter paper into a volumetric flask (50 mL) and filled up to the required volume using deionized water.

For the acid digestion of plant biomass, since the amount of biomass was low it was mixed with pure sand and subjected to acid digestion.

#### 3.4.3. Sample analysis

The samples obtained from liquid sampling, leaching tests and acid digestion of solid samples were analysed for heavy metals (chromium [Cr], copper [Cu], nickel [Ni], lead [Pb], zinc [Zn]) and other cations (sodium [Na], potassium [K], calcium [Ca], magnesium [Mg]) via inductively coupled plasma - optical emission spectrometry (ICP-OES - PerkinElmer Optima 2100 DV). Anions (sulphates [SO<sub>4</sub>], chlorides [Cl], nitrates [NO<sub>3</sub>], phosphates [P<sub>2</sub>O<sub>5</sub>]) in leaching tests were determined using a Hach spectrophotometer using standard reagents following the manufacturer's instructions.

### 3.5. Statistical analysis

One-way ANOVA was carried out using MS-Excel and the statistical significance was attributed at  $P < 0.05$ . Heavy metal profiles were also drawn using MS-Excel software.

## 4. Results and discussion

### 4.1. MSW characterization

The excavated waste from the Bhalswa dumpsite mainly comprised of fines (<20 mm; 72% by weight) as shown in Fig. S1 (Supplementary Information). Construction and demolition waste (CDW), mainly

consisting of stones, brickbats, and concrete fragments comprised 15%. Combustible materials (plastic, wood, and textiles) were found to total <10%. The percentage of recyclable fractions (glass, metals etc.) was very low due to the very efficient traditional (informal) system of collection of these items by waste collectors called 'Kabadiwalas' prevalent for several decades. The composition observed is in accordance with previous findings reported in the literature (Kurian et al., 2003; Hogland et al., 2004; Rong et al., 2017).

The particle size distribution of the excavated MSW is presented in Table S1 (Supplementary Information). The proportion of SLM (sand + clay + silt, of size less than 4.75 mm) depended on the testing method, with washing via wet sieving causing a significant increase in finer fractions (from 45–48% to 65–68%). Proportions following wet sieving are higher in comparison to those reported by previous investigators due to the detachment of fine particles adhered to coarser particles.

#### 4.1.1. Characterization of SLM

The characterization of the SLM used in the present study is shown in Table 2. High electrical conductivity, organic content, and concentrations of heavy metals, sulphates and chlorides are noted, and are considerably higher than in background soils from the study area (data not shown). The concentration of heavy metals and soluble salts higher than the prescribed regulatory thresholds (SWM Rules, 2016) as well as with the background soils prevents the direct use of SLM in offsite applications (e.g., earthfills and compost).

#### 4.2. Electrokinetic remediation

Profiles of heavy metal concentrations in the SLM over time for all three treatments, compared to initial concentrations, are presented in Fig. 1, whilst the variation of metal concentration with time in the electrode chambers is presented in Fig. 2. Chemical symbols are used in figures and tables for the sake of brevity. In both electrokinetic treatments, chromium, copper, zinc and lead migrated through the SLM towards the anode gradually over time, with slightly greater transport for the first three of these with pH control (Ep). One zinc concentration (Ep experiment, near the anode, 24 days) was unexpectedly high and is considered to be an outlier; it is not considered further as later data were in line with expectations. These outcomes concur with the significantly elevated concentrations of all metals observed in the anode chambers of both electrokinetically-treated specimens, with little to none observed in the cathode chambers. Only nickel differed, with apparent transport towards both anode and cathode. However, as nickel was detected only in the anode chamber and not the cathode chamber, this suggests that transport is indeed towards the anode here, but that nickel in SLM near the cathode was less mobile. Only small metal concentrations were observed in the control specimen electrode chambers indicating that non-electrokinetic transport is low. Initial chromium, copper and lead concentrations in SLM were elevated above regulatory limits (Fig. 1) whilst after EK treatment concentrations in the majority of the specimens (apart from the anode end) were significantly reduced, at (chromium) or below (copper, lead) the limits. Both nickel and zinc initial concentrations were close to their respective limits. After EK treatment, nickel had been redistributed but concentrations were still close to the limit at the electrode ends, whilst zinc had been removed in most of the specimen such that concentrations were well below the limit.

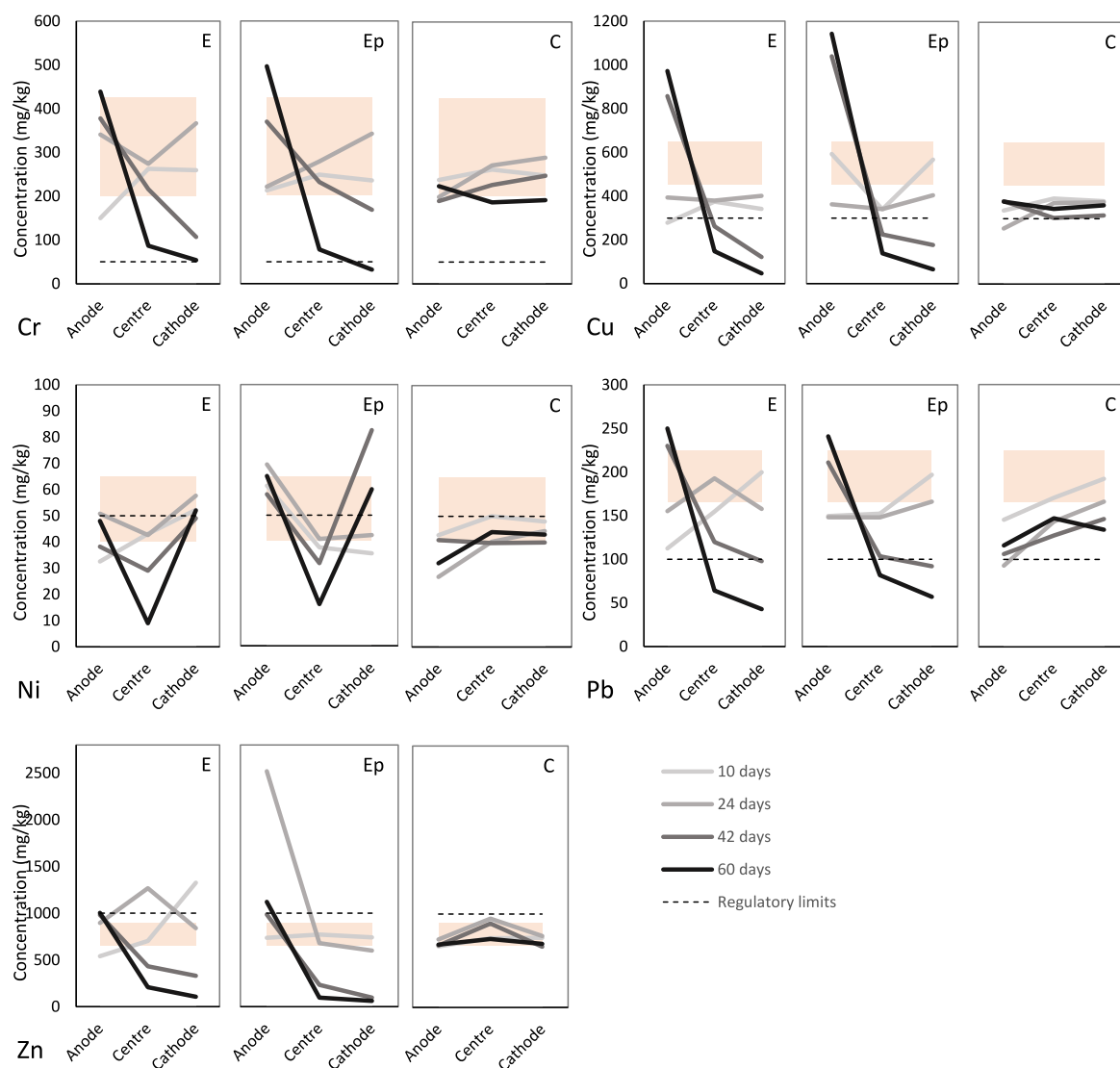
Transport to the anode of positively charged ions is unexpected under electromigration and suggests that metals are present in the form of a negatively charged complex with other materials. Given the high organic matter content of the SLM (15–18%), we suggest that metals are associated with organic matter in SLM, and that transport is largely governed by the mobility of negatively charged metal-organic matter complexes. Metal movement towards the anode has previously been observed in electrokinetic treatment of organic-rich media and attributed to complexation with natural components of organic matter such as humic and fulvic acids (Park et al., 2010). Such complexing agents have

**Table 2**

Characteristics of SLM used in the present study (- represents values not given in the Solid Waste Management (SWM) rules of India (MoEF&CC, 2016)).

Property	SLM	Background soil	Regulatory limits (SWM Rules, 2016)
Particle size distribution	Clay (<0.002 mm)	6–9%	–
	Silt (0.002–0.075 mm)	35–40%	–
	Sand (0.075–4.75 mm)	50–55%	–
pH	7.9–8.4	7.2–7.4	6.5–7.5
Electrical conductivity (dSm <sup>-1</sup> )	5.5 <sup>a</sup> –6.8 <sup>a</sup>	0.3–0.32	4
Organic content (%)	15–18	1–1.2	–
Total soluble solids (mg/kg)	22,000–25,000	500–700	–
Sulphates (mg/kg)	7500–9500	300–350	–
Chlorides (mg/kg)	3500–4500	200–250	–
Zinc (mg/kg)	650–900	44–67	1000
Copper (mg/kg)	440 <sup>a</sup> –650 <sup>a</sup>	14–14.5	300
Lead (mg/kg)	165 <sup>a</sup> –225 <sup>a</sup>	5–6.5	100
Nickel (mg/kg)	40–65 <sup>a</sup>	20–21	50
Chromium (mg/kg)	200 <sup>a</sup> –425 <sup>a</sup>	17–25	50
Total organic carbon (%)	6.8–8.4	0.67–0.69	>12
Total Nitrogen (%)	1.09–1.40	0.05–0.07	>0.8
Total Phosphorous (%)	0.68–0.74	0.03–0.04	>0.4
Total Potassium (%)	0.34–0.66	0.04–0.05	>0.4

<sup>a</sup> values exceed the maximum permissible limit.



**Fig. 1.** Profiles of heavy metal concentration in SLM across specimens over time (shaded band – initial range of concentrations; dashed line represents regulatory limits as given in Table 2; E – EK treatment, Ep – EK treatment with pH control, C – control treatment).

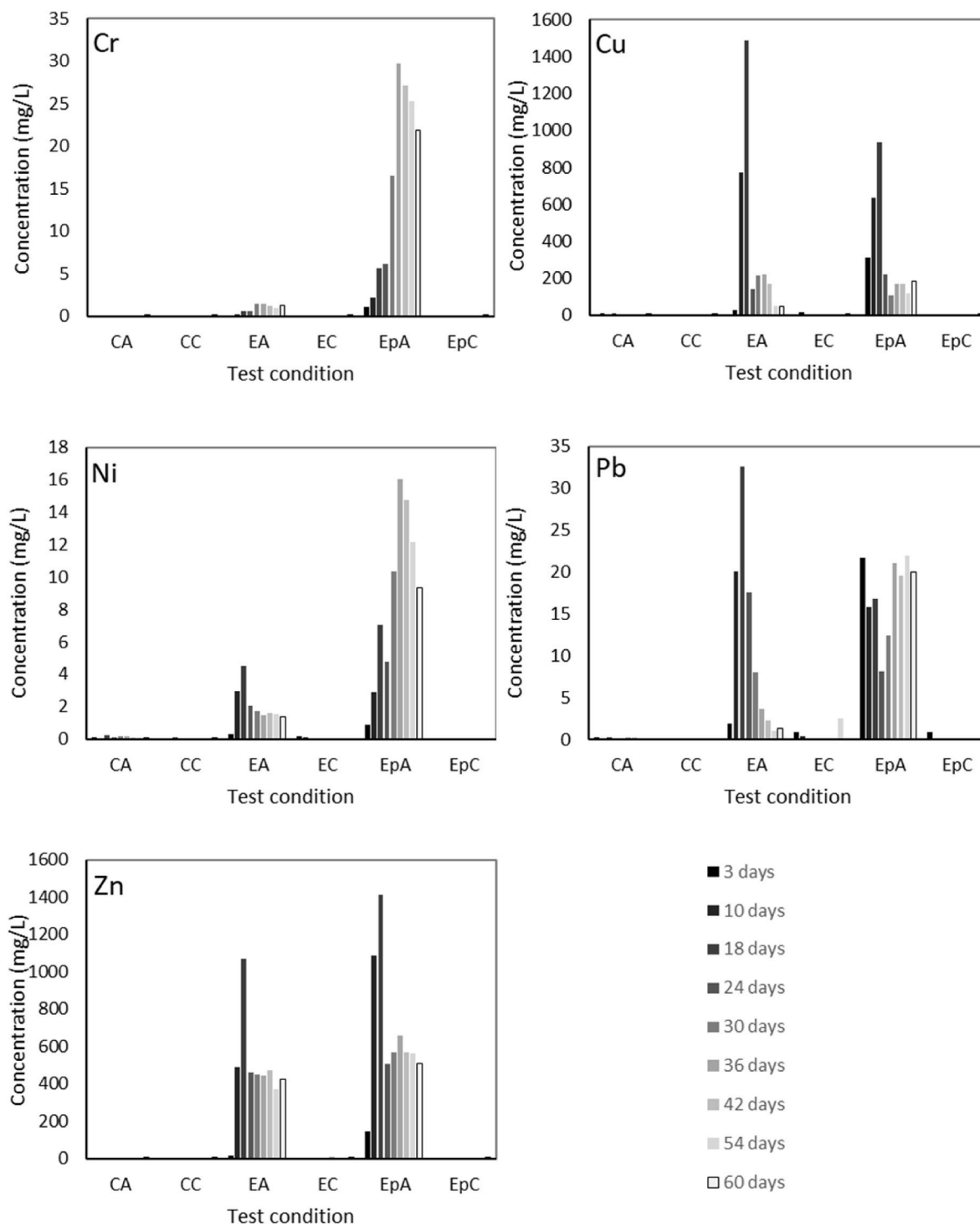


Fig. 2. Variation of heavy metals in electrode chambers over time (CA/CC – control anode/cathode; EA, EC – electrokinetic anode/cathode; EpA, EpC – electrokinetic with pH control anode/cathode).

also been used as a means of mobilising metals (Bahemmat et al., 2016).

Initially, the neutral to slightly alkaline pH suggests that the net charge of DOM molecules will likely be negative, so we hypothesise that heavy metal ions associate with DOM in the SLM and then transport towards the anode, as observed. Metal concentrations at the anode chamber reached a maximum in the first 3–5 weeks and then mostly decreased. This may be attributed to three possible mechanisms associated with the low pH (~pH 1 from 3 days), oxidising environment at the anode under EK which would permit the metal ions to begin moving back towards the cathode: (i) disassociation of metals from organic

matter; (ii) breakdown of DOM molecules; (iii) charge on DOM molecules switching from negative to positive due to the pH being below the likely point of zero charge for organic matter (Bakatula et al., 2018).

The transport to the anode of two metals in particular, chromium and nickel, and to an extent lead, was found to be of greater magnitude in the Ep experiment with pH control. This may be attributed to the extent of association of metals such as nickel with DOM (Ashworth and Alloway, 2004), as with pH control (cathode pH restricted to between 8.5 and 11.5 in Ep experiment compared to ~14 in E experiment) increased mobility of DOM towards the anode will be permitted (Curtin et al.,

2016). Copper and zinc had similar behaviour under the two treatments which may be related to the much larger quantities of these metals present.

The concentrations of cations (sodium, potassium, calcium, magnesium) and anions (sulphate, chloride) collecting in the electrode chambers are shown in Fig. 3. In the control experiment, concentrations of all ions were similar at both ends of the specimen at all time points, as expected. However, cation concentrations decreased steadily with time whilst anion concentrations were broadly stable which may be linked to high cation exchange and buffering capacities in SLM as reported by Liu et al. (2018b) and Goli et al. (2022). In the E and Ep experiments, cation

behaviour diverged with monovalent alkali metals (sodium, potassium) observed to move away from the anode and increase initially in the cathode chamber, which is expected behaviour under electromigration, although cathode chamber concentrations decreased with time in the latter half of the experiment (similar to that observed in the control experiment). These ions are less likely to associate strongly with DOM and so provide further evidence for the role of organic matter in the unexpected behaviour seen with divalent cations. Divalent cations (calcium, magnesium) demonstrated similar behaviour to that of the heavy metals with movement to the anode. Anions (sulphate, chloride) demonstrated expected electromigratory behaviour in moving towards

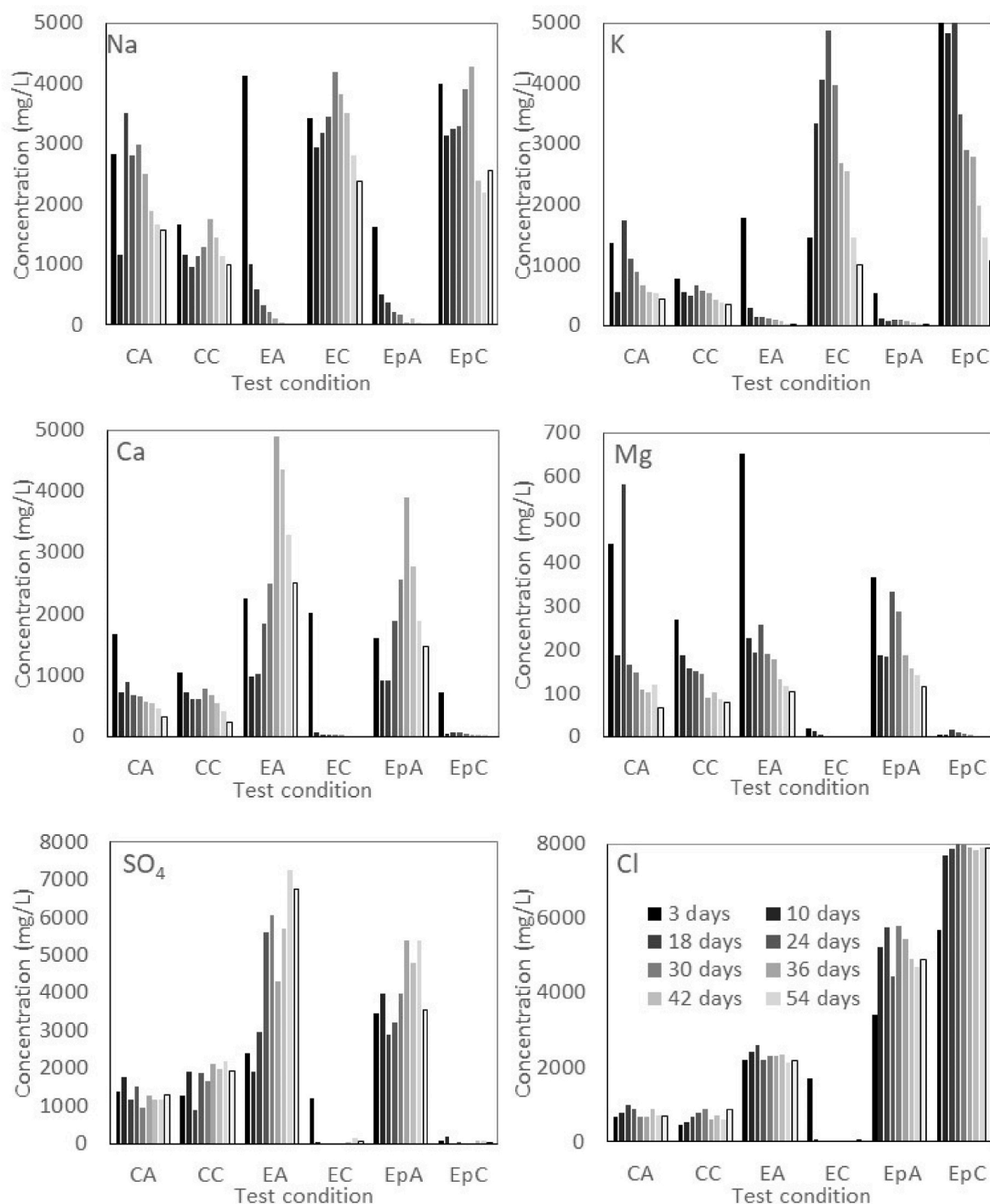


Fig. 3. Ion concentrations in electrode chamber fluids (CA/CC – control anode/cathode; EA, EC – electrokinetic anode/cathode; EpA, EpC – electrokinetic pH control anode/cathode).

the anode chamber. The high chloride concentration in the cathode chamber (Ep treatment) resulted from continuous addition of hydrochloric acid for pH control.

#### 4.2.1. Effects of electrokinetics on leachability

Leachable heavy metals demonstrate a similar behaviour to total metal contents with the majority of leachable heavy metals present in the anode region in E and Ep experiments, with none found in the cathode region (Fig. 4). Electrokinetic transport is the major reason for this; higher pH near the cathode may contribute in immobilising some metal in this region (E setup) but similar outcomes were found in the Ep setup with cathode pH control.

Leachable soluble solids and individual leachable cations and anions determined in all three test conditions are shown in Fig. 4. Anions (sulphate, chloride, nitrate, phosphate) all showed increasing leached amounts towards the anode in both E and Ep setups whilst cation behaviour differed based on their charge with monovalent ion leaching increasing as expected towards the cathode and divalent ions towards the anode, matching the total element concentrations given above and the trends observed in electrolyte fluids. Enhanced leaching of sodium, potassium and magnesium in Ep compared to the E setup may have been caused by the addition of hydrochloric acid to the cathode in the former, leading to ion exchange on SLM grain surfaces.

### 4.3. Phytoremediation

Pot experiments were conducted for 8 weeks to assess the ability of ryegrass to grow in SLM and extract or amend the behaviour of contaminants and associated ions.

#### 4.3.1. Fate of heavy metals

Distribution of the various heavy metals between the SLM, plant root biomass, plant shoot biomass and leachate solutions are presented in Fig. 5 for both planted and control pots. Overall metal recovery from all pots, both planted and control decreased with time for all metals. Plant uptake was a very small proportion of the total for each metal, which may be explained partly by the small levels of biomass present after 8 weeks (average/standard deviation mass per pot of roots was  $21.7 \pm 5.3$  mg and of shoots was  $3.4 \pm 0.9$  mg). Concentrations in the biomass (Table S2) were comparable to or higher than concentrations obtained in phytoremediation of contaminated soils and organic-rich wastes with ryegrass (Zhang et al., 2019; Paradelo et al., 2020) and other species (e. g., willow - Page et al. (2014)). With greater biomass, seedling density it might reasonably be expected that the mass uptake would be increased. In all cases metal concentrations in planted SLM were less than in unplanted controls (Fig. S3), whilst root and shoot concentrations tended to increase with time (albeit with considerable variability and at low levels (Table S2)). Heavy metal concentrations in planted SLM after 8 weeks were found to be approaching or below regulatory thresholds for copper, nickel and zinc, whilst those for lead and chromium were above these thresholds (Fig. S3). It should also be noted that unplanted controls also exhibited nickel and zinc concentrations below the thresholds on average, although the variability with nickel meant that satisfaction of these limits was not fully achieved.

Heavy metals determined in leachate samples collected from every pot after weekly watering are presented in detail in Fig. S4. Leaching of chromium, copper and nickel from control pots was consistently lower than that from planted pots. Enhanced contaminant mobility may arise through the action of plants, particularly production of root exudates by ryegrass – these typically comprise a range of organic acids and

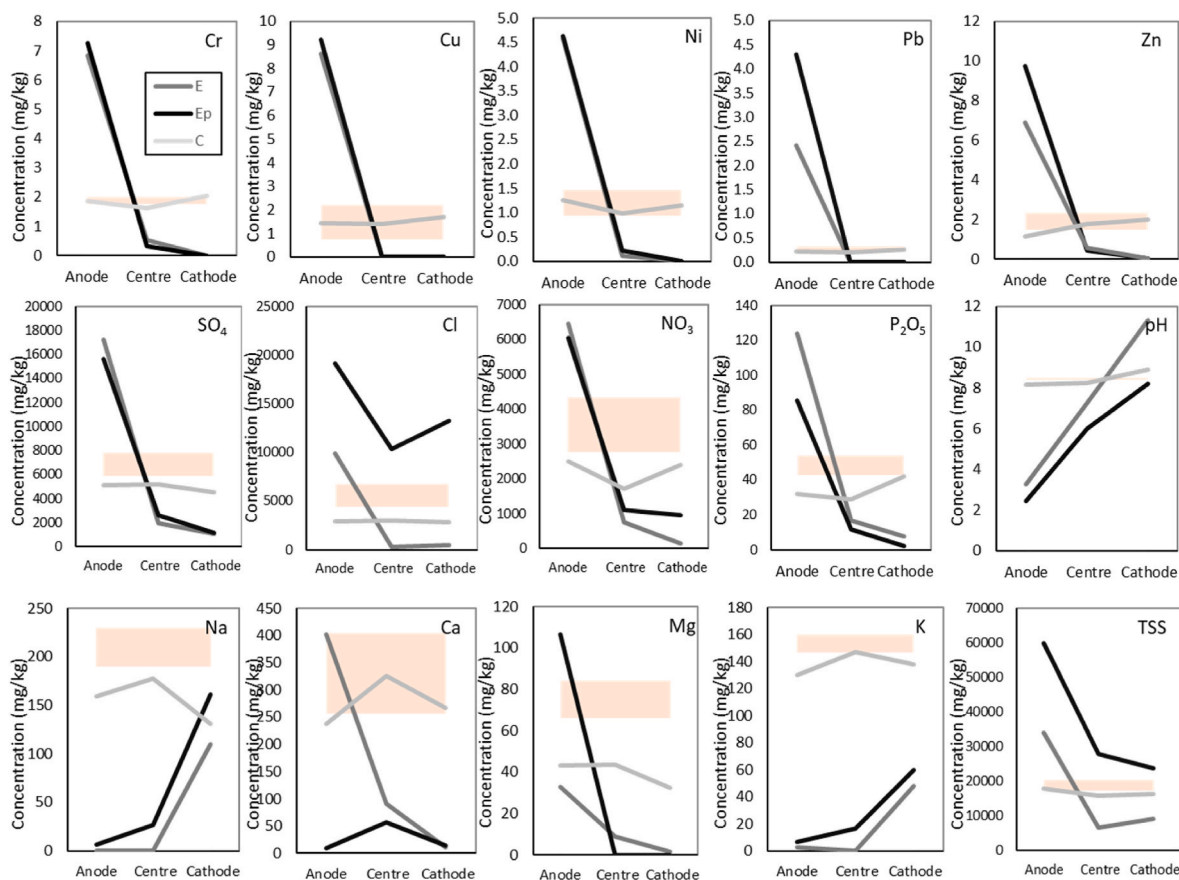


Fig. 4. Leachable heavy metal and other ion concentrations from SLM following the various treatments in EK experimentation (shaded band – initial range of concentrations; E – EK treatment, Ep – EK treatment with pH control, C – control treatment).



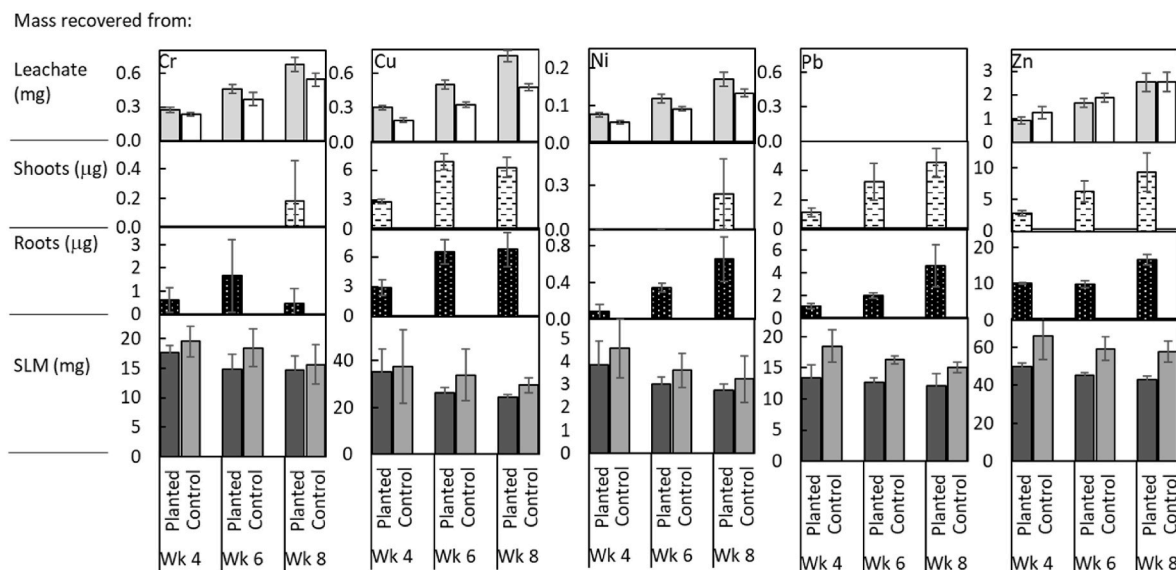


Fig. 5. Comparison of heavy metal mass recovery from SLM, root biomass, shoot biomass and leachate in control and planted pots over the phytoremediation experiment. Error bars represent  $\pm 1$  standard deviation.

chelating species which can help mobilise metals (Paynel et al., 2001). Increased leachate concentrations may be problematic and may more than offset any metal recovery in biomass. With zinc there was less distinction between planted and unplanted pots which may have been due to greater zinc concentration (i.e. an initially large available fraction

could dwarf additional zinc made available through root exudate leaching). Lead in leachate was always below the detection limit in both unplanted or planted pots, which may be indicative of its low availability in this material and possibly the susceptibility of lead to phytostabilisation (Engendorf et al., 2020).

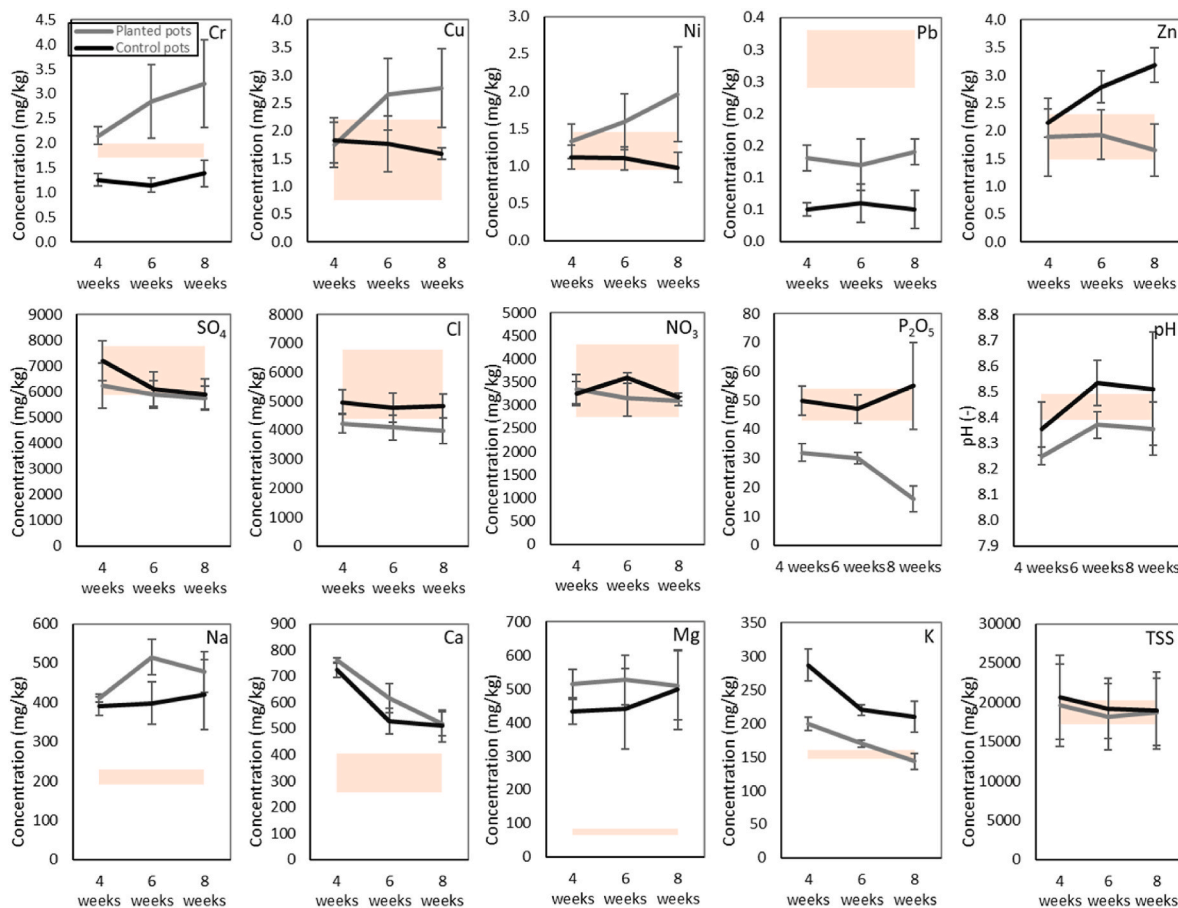


Fig. 6. Leachable metal and other ion concentrations from SLM (average and standard deviation; units – mg/kg; shaded band – initial range of concentrations; for heavy metals, significant differences between planted and control pots are given by \* ( $p < 0.05$ ), \*\* ( $p < 0.01$ ) and \*\*\* ( $p < 0.001$ )).

#### 4.3.2. Leaching characteristics of SLM following phytoremediation

The leaching characteristics of SLM following treatment were determined in order to assess the effect of phytoremediation on the availability of heavy metals and soluble salts still remaining in the SLM. Leachable heavy metals remaining in the SLM are shown in Fig. 6. Generally, unplanted controls exhibited decreasing or static available fractions of metals with time, as leaching removed the most available material. The presence of ryegrass had the effect of increasing leachability for all metals, providing further evidence for an enhancement of availability through mechanisms likely to include release of exudates (Luo et al., 2019).

Leachable total soluble solids (TSS), anions (sulphate, chloride, nitrate, phosphate) and cations (sodium, potassium, calcium, magnesium) determined in SLM collected after 4, 6, and 8 weeks are also shown in Fig. 6. Leachable anions and TSS were broadly similar between planted and control pots apart from phosphates, where planted plots had lower concentrations. Leachable cations were similar in both planted and control pots, apart from potassium where planted pots had lower concentrations. That both phosphates and potassium were lower in availability in planted pots may be as a result of their role as essential macronutrients in plant growth. Leachable cations were elevated relative to in untreated SLM, which may result from use of Hoagland's solution where many of these ions are present. In particular, phosphate, calcium and potassium levels are significant compared to those in untreated SLM (~20–50% per watering event).

#### 4.4. Overall discussion

Both remediation techniques tested have demonstrated potential for resource recovery, both in terms of the metals present in the SLM through EK transport to electrodes and plant uptake and storage in biomass, but also the recovery of processed SLM as a product of some value. Each has increased the mobility and availability of contamination and demonstrated its partial extraction although in the eight-week treatment neither was fully complete. Electrokinetic processing caused rapid separation of metals and SLM and much of the SLM was remediated to below regulatory thresholds. In this particular SLM we have hypothesized that metal mobility and extraction appeared to be linked with that of organic matter. Depending on the organic content this may impact the mobility of metals in SLM from different landfills and with different levels of organic matter, but the outcome here suggests that metals are highly mobile. The technique should be combined with metal extraction from the system through recovery of metals at the electrodes, for example. Phytoremediation was somewhat inconclusive because of poor mass balances though reduced SLM concentrations and evidence of biomass uptake in comparable quantities to previous phytoremediation studies indicate that SLM can similarly be treated. An increase in biomass is required, however, as SLM metal concentrations were not reduced to below regulatory thresholds in the time available.

Both technologies cause increased leachability of heavy metals, at least in certain regions of the SLM. This indicates that we have not reached a suitable endpoint in either case, and so there is a period where enhanced availability may cause SLM to pose an increased risk until sufficient metals and salts are removed from the system altogether. Leachate management will be a key part of these technologies where used.

Application of either technology will require a degree of management for limited quantities of arisings where contamination is concentrated, specifically water and SLM located near electrodes (electrokinetic treatment) and plant biomass (phytoremediation). This will necessarily be situation-specific, although numerous opportunities exist for waste treatment (e.g. sorption, precipitation/coagulation) whilst opportunities for resource recovery are now also becoming possible (Sapsford et al., 2017). Revisiting the options appraisal for remediation technologies (Table 1), the experiments demonstrate that both techniques may be sufficiently rapid to bring contamination levels

below regulatory limits within a period of several weeks, providing evidence that both technologies have the potential to be considered rapid, with the caveat that this will depend on site conditions. Phytoremediation, therefore, may be considered as a potential low cost, sufficiently rapid source treatment that permits recovery of SLM for reuse. Electrokinetic treatment is also appropriate from a technical point of view, with only the cost being uncertain. Energy consumption over the EK experiments is estimated to be less than 0.7 kW-hours per kg SLM. And if low cost energy is available this may be feasible.

## 5. Conclusions

In most of the previous studies the use of soil-like material has been suggested as landfill cover. However, due to the production of huge volumes of SLM, its bulk utilization in the embankment, structural fill material in reclamation projects, as a subgrade material and fertilizer should be explored further. The characterization of SLM from MSW dumpsites identified elevated concentrations of heavy metals and soluble salts which prevents the unrestricted use of SLM in many offsite applications. An options appraisal of methods for remediation and reclamation of SLM has been conducted based on UK guidelines. As a result, electrokinetic remediation and phytoremediation techniques have been explored for the remediation of SLM before re-use in offsite applications. Electrokinetic remediation caused significant movement of heavy metals and soluble salts towards the electrodes over a period of 60 days. An unexpected movement of chromium, copper, lead and zinc and other divalent cations (calcium, magnesium) was observed towards the anode and attributed to the high organic content in the SLM and association of metals with anionic dissolved organic matter. Despite the anomalous behaviour, heavy metals and other ions were highly mobile under electrokinetic processing suggesting that they are not strongly bound to SLM and that there is potential for significant removal *via* EK. Given the economic constraints in developing countries like India, a thorough analysis of the expenses associated with EK remediation is essential. Consequently, a more careful and precise examination of cost should be taken into account to arrive at a well-informed decision regarding the appropriateness of the technique for managing SLM and enabling its recovery and reuse. Phytoremediation caused a reduction in the concentration of total heavy metals in SLM over a period of 8 weeks and metal uptake into biomass was observed. The findings demonstrate that electrokinetic remediation and phytoremediation are capable of recovering heavy metals from SLM. This is encouraging for 'landfill mining' projects, which otherwise have a serious question mark in terms of post-mining utilization of SLM. Until recently, full-scale landfill mining activities occurred somewhat sporadically around the world, but the current large-scale implementation of landfill mining across India, and the potential for this to be replicated in other locations worldwide, demonstrates that there is now a need for mechanisms of appropriate treatment and reuse of SLM as the most significant product of such processes.

### Credit author statement

Mohit Somani: Conceptualisation, methodology, investigation, visualisation, writing – original draft, funding acquisition.; Michael Habbottle: Conceptualisation, methodology, writing – review and editing, visualisation, supervision.; Manoj Datta: Supervision.; G.V. Ramana: Supervision.; T.R. Sreekrishnan: Supervision.

### Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Mohit Somani reports financial support was provided by British Council India.

## Data availability

No data was used for the research described in the article.

## Acknowledgements

The first author acknowledges the support of the British Council for funding his research placement at Cardiff University through the Newton-Bhabha PhD Placement Scheme for Indian Scholars (project ID 540368875).

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2023.119300>.

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