Waste Management 158 (2023) 116–124

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Increasing recovery opportunities of metal(loid)s from municipal solid waste via landfill leachate recirculation

H. Lee [a], F. Coulon [a], D.J. Beriro [b], S.T. Wagland [a,*]

[a] School of Water, Energy and Environment, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK
[b] Digital Laboratories, British Geological Survey, Nottingham NG12 5GG, UK

ARTICLE INFO

Keywords:
Landfill
Metal recovery
Recirculation
Circular economy
Leachate

ABSTRACT

The recovery of 12 critical raw materials (CRM) from municipal solid wastes (MSW) via leachate recirculation was evaluated using a 4 L semi-pilot scale column percolation. The results showed that the recovery of the metal (loid)s was mainly influenced by order of importance: pH > organic content > type of metal(loid)s > age of the waste > redox potential. Among the CRM, Cd and Ni were the most mobile elements, while As and Cr were the least mobile. A comparison of leachate from the leachate recirculated columns before and after the initiation of recirculation indicates an increase in the concentrations of certain CRM and metalloids. The first recirculation cycle supported achieving 100 % recovery. CRM and metalloids in leachate can be recovered; however, the concentrations of CRM and metalloids are usually below 1 mg/L. In this regard, leachate recirculation may enhance the increasing concentration of CRM in landfill leachate. For example, after first recirculation cycle, Ni concentration increased from 0.05 mg/L to 0.11 mg/L. The results obtained from this study can develop further methodologies for the potential recovery of CRM and help foster further research into overcoming limitations for recovering CRM in landfill leachate.

1. Introduction

Minerals commodities are essential to our economy and lifestyle, and the global demand for metal(loid)s continues to rise. However, resource depletion and climate change are challenging problems that humankind faces and will become even more critical in the future with the global growth in economic activity (Watari et al., 2018). A very different system must be built by 2050 to reduce carbon emissions and avert catastrophic climate change drastically based on renewable energy sources (Moreau et al., 2019). Renewable energy technologies must be manufactured and maintained, which requires a flow and stock of mineral resources, particularly metals. Among them, Lithium (Li), nickel (Ni), cobalt (Co), Manganese (Mn), and aluminium (Al) are central to our transition to a low carbon economy, such as the production of electric cars, solar panels, wind turbines and other clean energy technologies (Cozzi and Gould, 2021). Thus, to provide the necessary resources to meet demand, the transition towards resource-efficiency is required (Lee et al., 2022a, 2022b).

There is indeed urgency in recovering minerals from discarded materials and urban solid wastes. In the early 2010 s, several studies have highlighted the potential of untapped resources of minerals that represent closed municipal solid waste (MSW) landfills and introduced the concept of enhanced landfill mining (ELFM) (Quaghebeur et al., 2013). ELFM aims to valorise and maximise the recovery and values of excavated wastes through Waste to Materials (WtM) and Waste to Energy (WtE) strategies (Quaghebeur et al., 2013). Among the resources recovered from ELFM, critical raw materials (CRM), rare earth elements (REE), and metal(loid)s are of strategic importance, and there is indeed a growing interest in maximising their extraction and recovery from landfill sites through WtM and WtE strategies (Kurniawan et al., 2021). Several studies have assessed the possibility of metal recovery from landfill sites through mining (Kaartinen et al., 2013; Wagner and Raymond, 2015; Parrodi et al., 2018; Lucas et al., 2019). However, there are still technical and economic challenges to mining minerals from landfills, which need to be overcome. The primary challenges are economic infeasibility, and metal remains buried for several decades, potentially subjected to corrosion and pollution before being excavated from landfill sites (Wagner and Raymond, 2015; Lucas et al., 2019; Lee et al., 2022a, 2022b). Another alternative which is seen as more suitable and sustainable approach, is landfill leachate recirculation (Lee et al., 2022a, 2022b; Guérin et al., 2004; White et al., 2011). Leachate recirculation can significantly influence metal behaviour and fate within the MSW

* Corresponding author.
E-mail address: s.t.wagland@cranfield.ac.uk (S.T. Wagland).

https://doi.org/10.1016/j.wasman.2023.01.011
Received 19 July 2022; Received in revised form 17 November 2022; Accepted 10 January 2023
Available online 17 January 2023

buried in the landfill sites. For example, studies have shown that the initial stage of leachate recirculation had low pH (5–6) due to the acidification stages, resulting in a relatively high concentration of metals (Bilgili et al., 2007; Qu et al., 2008). Leachate recirculation can also increase the chloride content as it is highly mobile and cannot be sorbed or transformed bio-chemically, which is an important controlling factor for metal release. Chloride affects the release of metals through the binding the metals on humic acids and through the adsorption of metals (Begeal, 2008; Damikouka and Katsiri, 2021).

However, the recovery of elements from landfill leachate is still not currently economically viable due to poor process efficiency and minerals that are often present at low concentrations. Further to this, our limited understanding of the chemistry and mechanistic processes occurring within the landfill waste mass and the leachate composition hinder our ability to enhance and optimise minerals recovery from MSW landfill sites.

Therefore, this research mainly addresses determining the effect of leachate recirculation on metal mobility and enhancing the metal recovery rate from landfill leachate. The results will provide information to understand metal behaviour and factors affecting metal mobility. Different ages of leachate samples were used for the experiment to investigate the influence of leachate age on metal(loid)s recovery.

2. Materials and methods

2.1. Sample collection and preparation

A total of about 120 kg of municipal solid waste (MSW) and 20 L of leachate samples were collected from two former MSW landfills located in the UK, an older site which had been closed for 20 years and a younger site which was still receiving waste. The older waste sample was collected in Gloucestershire, England, from a landfill operation between 1984 and 1991 without engineered controls; it covers an area of 2.3 ha and is up to 6 m deep. The Gloucestershire landfill received 60,000 m^3 of inert, industrial, commercial, and household waste. Older landfill leachate was collected from a closed site that was operated by Bedfordshire Country Council and is now under the responsibility of Bedford Borough Council. The site last accepted non-hazardous waste for disposal in October 1998. Leachate, used for the older waste from the Bedfordshire site, was extracted and treated by using the sequencing batch reactor (SBR) process. The younger waste sample was collected from an operational landfill located in Suffolk, England. The landfill receives minerals, mixed municipal waste, and other waste from the mechanical treatment of waste. Leachate for the younger waste was collected from the same site where the younger waste sample was collected. The samples were taken by a specialised private company hired by the landfill sites operator, placed in a cool box kept at 4 °C and transported back to the environmental analytical facility at Cranfield University. The samples were stored in a cold room at 4 °C until analysis. The waste samples were sorted manually into metal, paper, textile, wood, glass and other materials.

2.2. Experimental setup

Two sets of semi-pilot scale Perspex columns were set up to simulate leachate production from MSW of different ages and compositions (Table 1). The waste mixture was manually mixed thoroughly to ensure homogeneity before they were loaded into the columns. Leachate samples were used for the experiments. For younger waste samples (<5 years), a leachate sample was used, which was collected from the same site as the waste sample. For older waste samples (>10 years), an old landfill leachate sample was used which was collected from a closed site in Bedfordshire. Dimensions of each column were 21 cm diameter and 1.02 m height, with a working volume of 4 L (Fig. 1). One of the columns was filled with waste < 1-year-old (S.S) and the second with waste > 10 years old (S.G). In order to make comparison possible among different compositions from the samples, the samples were mixed well together manually. The outlet port at the bottom was used for recirculation leachate using a peristaltic pump. Gravel was filled at the bottom of the column to a height of 0.05 m to serve as a drainage layer and prevent clogging of the leachate outlets. Waste was compacted by pushing it and filled into the column to a height of 0.25 m. Cover layers consisting of 0.1 m sand were placed on top of the waste samples to decrease the amounts of air penetrating the column. Leachate was collected and stored in the leachate collection tank, which is a 5L plastic water container. Leachate was continuously recirculated for 10 cycles using the peristaltic pump with flow rates adjusted to 800 mL/day.

Table 1
Composition of the MSW samples collected from the landfill sites and the initial concentration of metal(loid)s in Gloucestershire (S.G) and Suffolk (S.S).

	S.S	S.G		S.S	S.G
Years	<1	>10	Metals (g)	2212	224
Waste samples (g)	59,470	62,230	Glass/ceramic (g)	3663	10
Paper/cardboard (g)	4692	1410	Textile (g)	3911	520
Plastic (g)	7094	4623	Soil/fine fraction (g)	35,944	54,437
Wood (g)	1112	842	Others	762	164
Li (mg/L)	0.84	0.11	Cu (mg/L)	4.97	0.01
Al (mg/L)	3.97	0.37	Zn (mg/L)	1.62	0.38
Cr (mg/L)	0.36	0.27	As (mg/L)	0.31	0.00
Mn (mg/L)	0.20	0.02	Cd (mg/L)	0.00	0.00
Co (mg/L)	0.11	0.02	Pb (mg/L)	0.07	0.00
Ni (mg/L)	0.59	0.05	Hg (mg/L)	0.00	0.00

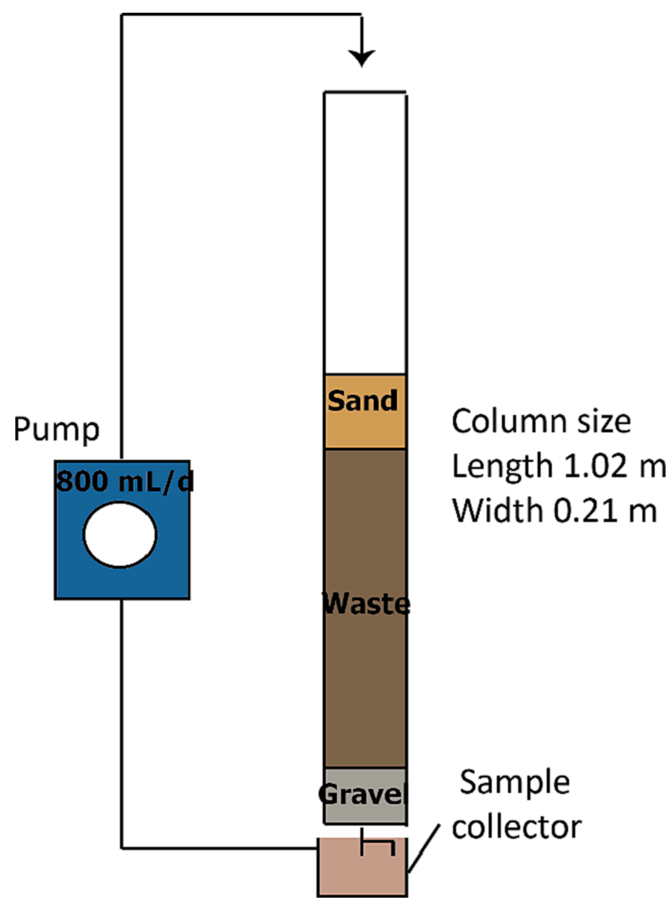

Fig. 1. Schematic diagram of the column percolation test.

2.3. Sampling and analytical methods

Leachate samples (250 mL) were collected after each cycle from the leachate outlet port. In order to keep the volume equilibrium of leachate before recirculation, the same volume of tap water (250 mL) was added to the leachate after sampling. The leachate was collected in polyethene bottles and stored in a cold room at 4 °C for a maximum of three weeks prior to physico-chemical and metal analysis.

2.3.1. Physico-chemical parameters

Physico-chemical analysis of each leachate sample from the landfills as well as from the experiment was conducted according to the Standard Method of Water and Wastewater (APHA, 2005); parameters included: pH, chemical oxygen demand (COD), redox potential, total organic carbon (TOC), volatile fatty acid (VFA), and conductivity (Table 2). All analyses were made in duplicate. The quality of measured values and analytical methods were verified by standards and blank samples. For VFA's analysis, the samples were centrifuged at 2500 rpm for 10 min and the supernatant was filtered using a 0.2 μm syringe filter (Sartorius AG, Geottingen, Germany), then the sample was stored at – 20 °C until analysis. The VFA content was determined by High performance liquid chromatography (HPLC) equipped with a Waters 996 photodiode array detector (USA). The mobile phase was 0.001 M sulphuric acid in HPLC grade water with a flow rate of 0.8 mL/min. The concentration of acetic acid, propionic acid, *n*-butyric acid, isobutyric acid, *n*-valeric acid and isovaleric acid, whose sum was reported as total VFA concentration. All analytical estimations were carried out in triplicate.

2.3.2. Metal(loid)s analysis

The metal(loid)s content was analysed by inductively coupled plasma mass spectrometry (ICP-MS) (Elan 9000 Perkin-Elmer SCIEX) after microwave-assisted acid digestion following the extraction procedure used in previous studies (Gutiérrez-Gutiérrez et al., 2015; Cipullo et al., 2018; Wagland et al., 2019). Briefly, Li, Al, Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Pb, and Hg were determined by pre-digesting 30 mL leachate with 1.5 mL concentrated trace metal grade nitric acid for leachate samples. For solid samples, 0.5 g of samples were mixed with 6 mL of HCl and 2 mL nitric acid in Teflon tubes and left overnight. The vessels for leachate samples were placed in the Mars Xpress microwave (CEM system, EPA 3015–8). Then, the vessels were cooled, and the solution was filtered through Whatman No. 2 paper and made up to 50 mL of deionised water. A blank digest was carried out in the same way. The microwave is an efficient and fast digestion technique method. Calibration standards were spiked with several certified standard solutions. The concentration ranges were 0.05, 0.1, 0.15 and 0.2 mg/L for the elements. Initially, the total quant method was used, followed by the quantitative method for selected metals, and the concentration of the metals was blank corrected. The quality of measured values and analytical methods were verified by standards and blank samples. Replicates analyse of samples showed a precision of typically $<$4 %.

In reporting recovery rates of metals from the leaching test, the following method of calculation is commonly used:

$$Recoveryrate = \frac{Lc}{Ic} \times 100$$

Where Lc, leaching concentration of metal after recirculation; IC, initial metal concentration.

Table 2
Physico-chemical parameters and methods of analysis.

Parameters	Instrument model	Instrument make
pH	Electronic pH meter	Jenway 3540
Conductivity	Conductivity meter	Jenway 3540
TOC	Shimadzu TOC-V	Shimadzu
COD	COD Cell	Spectroquant
Redox potential	Redox potential meter using micro-electrodes	Jenway Redox electrode
VFA	High-performance liquid chromatography	Waters-Alliance 2696

2.4. Statistical analyses

IBM Statistical Package for the Social Sciences (SPSS) 26 was used for the statistical analysis. Multivariate is a crucial method for investigating the relationship between variables and samples and can be performed to figure great value out on metals. A multivariable study was conducted considering the correlation matrices. In these matrices, each individual square contains the ordinal coefficient of Spearman which varies between 1 and −1 and the significance level of estimated correlation for each pair of variables (Bisquerra, 1989). Significance level was set at 0.05.

3. Results and discussion

3.1. pH, conductivity and oxidation–reduction potential (ORP)

Variations in pH, conductivity and ORP of the leachate samples collected at each of the ten recirculation cycles are given in Fig. 2. Leachates are generally characterised by pH values between 4.5 and 9 (Koumalas et al., 2019). The initial pH was lower (7.05) in young landfill leachate (S.S) compared to old landfill leachate (8.44) (S.G). The pH value was decreased for both samples during the first cycle, which reflects the acidogenesis phase. Afterwards, the pH value increased until six cycles in young landfill leachate and four cycles in old landfill leachate, which might be due to the aerobic degradation of the organic acid by the residual oxygen in the landfills and was attributed to the reduction of the partially ionised free VFA content (Bhalla et al., 2013). Values of pH remained relatively steady at around 7.7 and 8.7 from cycle 7 until the end of the experiment.

During monitoring, the recorded electrical conductivity (EC) in the leachate varied between 11.65 and 13.52 mS/cm for young landfill leachate (S.S) and 5.56 and 7.73 mS/cm for old landfill leachate (S.G). High values of EC indicate the presence of dissolved inorganic materials in samples. The conductivity values of leachate in both samples showed a decreasing trend by the recirculation cycle and showed young landfill leachate has a higher EC value. This finding is in agreement with the findings of Mousavi et al. (2021), suggesting the average EC showed that the electrical conductivity decreased with the landfill age.

The decrease in conductivity in this experiment is assumed to be due to the washout of some easily mobilised ions, metals, chloride and sulphate, also combined with other factors, such as the conversion of sulphate to sulphide under increasingly reducing conditions and the subsequent precipitation of sulphide as metal-sulphide, which would tend to withdraw significant ionic strength from the solution.. Metal variation in this study also showed a decreased trend. When solid waste stabilised, mobilizable ions were washed out continuously by leachate recirculation, and surplus oxygen gradually created oxidised conditions, which resulted in the partial dissolution of the precipitates in leachate. Consequently, it leads to an increase in conductivity after the decrease (Sekman et al. 2011).

The redox potential within a landfill determines the mechanism of waste degradation (Bilgili et al., 2007). The initial cycle of recirculation resulted in low ORP values. The ORP values decreased to 48.8 mV in young landfill leachate (S.S) and 136.9 mV in old landfill leachate (S.G) at the first cycle. After the first cycle, the ORP values increased to 55.6 mV in fresh waste and 222.4 mV in old landfill leachate, indicating the prevalence of anaerobic environmental conditions. ORP in old landfill leachate reached above 200 mV after ten cycles. The measurements are consistent with Top et al. (2019) results, which indicated a redox

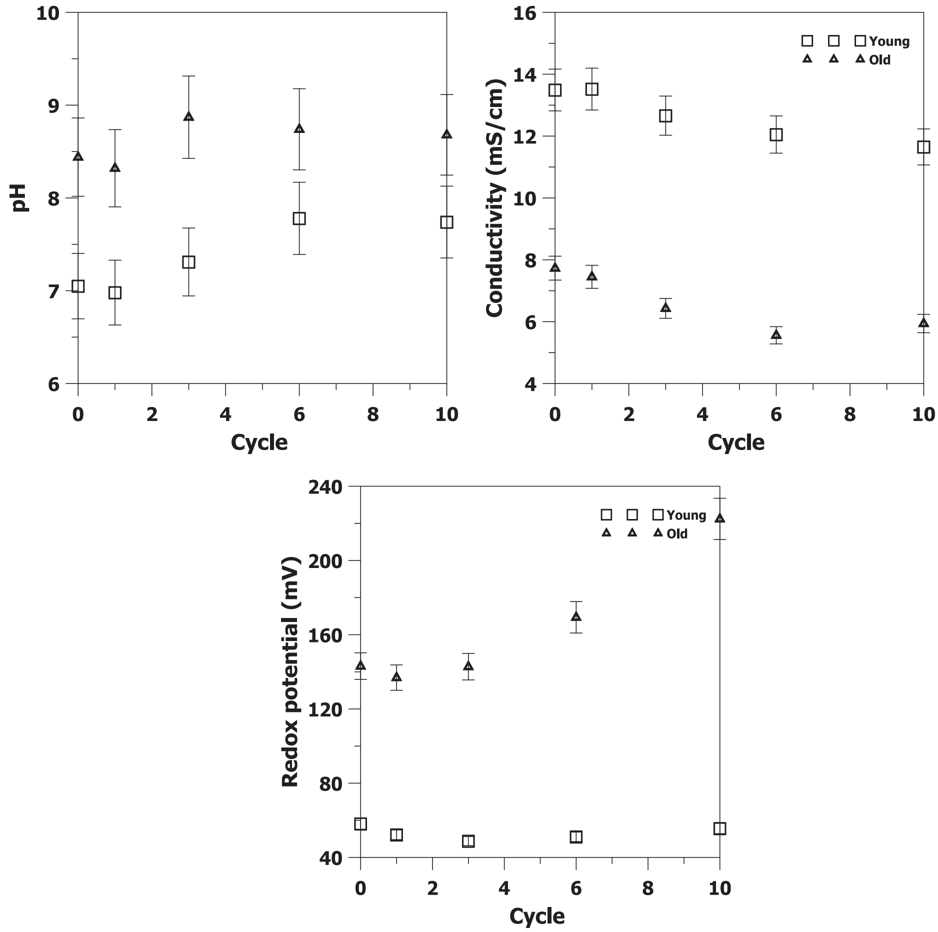

Fig. 2. Variations of pH, conductivity and ORP in the leachate from young (S.S) and old (S.G) landfill leachate.

increase in leachate during aerobic degradation of MSW. However, the result is inconsistent with James et al. (2004) and Ntwampe et al. (2008), who reported that ORP decreased with increasing pH as old landfill leachate has a higher pH value.

ORP has been considered as a parameter in controlling the fate of pollutants in the environments, particularly affecting the pollutant's reactions on the soil–water interface (Koumalas et al., 2019). However, metal mobility is driven by various mechanisms that include precipitation, sorption, and ORP. Therefore, ORP may be a key indicator of metal solubility, which is supported by the metal analysis in this study. The result showed that the ORP values tend to increase by the recirculation cycle, similar to results reported in the previous study (Bilgili et al., 2007). High redox potential (aerobic conditions) causes accelerated degradation of waste (Shearer, 2001).

3.2. *Chemical oxygen demand (COD), total organic carbon (TOC) and volatile fatty acids (VFAs)*

The chemical oxygen demand (COD), total organic carbon (TOC) and volatile fatty acids (VFAs) concentrations of the leachate collected during its recirculation are given in Fig. 3. COD indicates the oxidizability or reactivity of the organics, whereas TOC quantifies the amount of carbon in the organics (Montesantos et al., 2022). By comparing these two aspects, insight can be gained into the overall oxidation state of the organics. The leachate COD concentration showed similar behaviour to TOC concentration. COD increased by 1.3 % to 3.6 %, and TOC increased by 1.4 % to 4.5 % at the first cycle compared to the start value. It may be due to the hydrolysis of organic from the solid waste into the leachate was rapidly released in the initial stage. Leachate recirculation may be significant in improving the rate of hydrolysis and acidogenesis and redistributing nutrients (Nag et al., 2018). After increasing at the first cycle, the concentrations of both parameters then decreased and stabilised, which is similar to results reported in previous studies (Cossu et al., 2003; Nag et al., 2018; and Luo et al., 2019). The results from the previous studies and the results of the study show that leachate recirculation enhances rapid degradation and reaches stabilisation more quickly.

The decrease in COD and TOC concentration from leachate resulted from the dilution and washout mechanisms, the fact that microorganisms gradually adapted to the column conditions, and the dissolution rate of organics was slower than the rate of microbial degradation. The present study results showed that leachate recirculation has a positive effect on the rate of solid waste degradation in landfills and accelerates the biological stabilisation of organic fraction of wastes, and decreases the concentration of the pollutants (Bilgili et al., 2007). The concentration of COD and TOC in young landfill leachate was higher than in old landfill leachate. This indicated that the total amount of organic material in the landfill was decreasing with increasing age.

As shown in Fig. 4, the TOC, COD and VFAs concentration changes in the experimental were similar. VFAs increased by 2.3 % to 3.2 % at the first cycle compared to the start value. As a result of the high degradation rate in aerobic landfills, the concentrations of VFAs decreased since the first cycle, which is similar to results reported in previous studies (Sponza and Ağdağ, 2004; Bilgili et al., 2012). Young landfill leachate is well known to be characterised by high VFAs, COD and TOC (Renou et al., 2008; Bhalla et al., 2013). The results showed that VFAs of young landfill (S.S) leachate was about 78 % higher than old landfill leachate (S.G). Furthermore, VFAs of the leachate in both landfill leachate

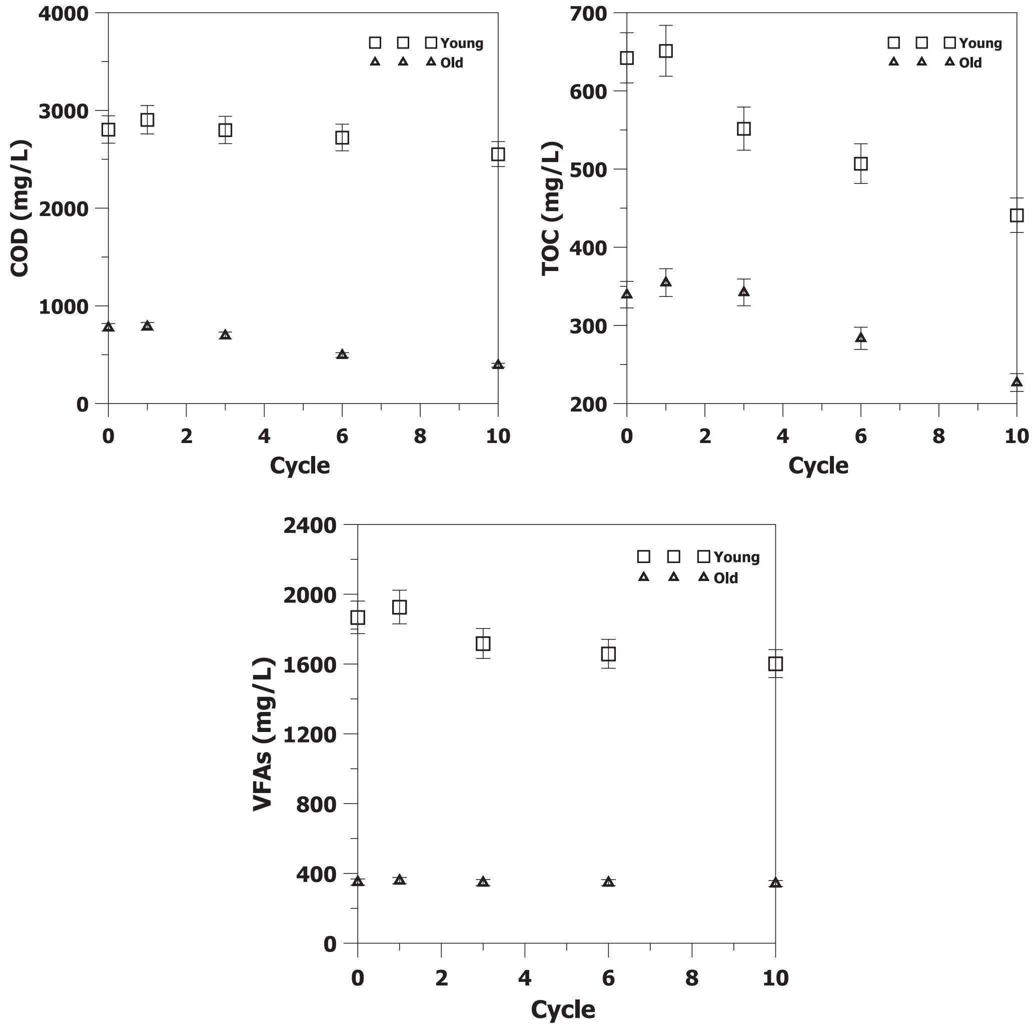

Fig. 3. Variations of COD, TOC and VFAs in the leachate from young (S.G) and old (S.G) landfill leachate.

showed a variation corresponding to the variation of the pH. VFAs are short-chain organic acids; they contain a carboxyl group (-C(=O) OH) attached to the alkyl (R-) group (R-COOH). These organic acids may play an essential role in the mobilisation of metals through either the formation of soluble ligand: metal complexes or a decrease of pH. They have high intrinsic proton affinity; for instance, the oxygen atoms in the anions of the carboxyl group have low ionisation energies (Panina, 2002). In this context, the carboxyl group of VFAs are capable of monodentate coordination with divalent metal cations (1).

$$\text{R-COO}^{-} + \text{Metal}^{2+} \leftrightarrow \text{R-COO: Metal}^{+} \quad (1)$$

As a result, it may increase the release of metals from landfill leachate through the formation of metal: carboxylate complexes (Molaey et al., 2021). Accordingly, metal(loid)s had higher concentrations when VFAs increased in this study.

3.3. Metal(loid)s

Table 1 shows the initial concentration of metal(loid)s. The metal (loid) concentrations in the leachate collected during its recirculation over ten cycles are given in Fig. 4. These data demonstrate a decreasing trend in concentrations for both landfills, which is consistent with previous studies (Benson et al., 2007; Long et al., 2010; Yao et al., 2014; Yao et al., 2017).

Owing to leachate recirculation, metals in leachate were brought back to the landfill. The leachate migration in landfills may result in metals to be immobilised in landfills through absorption, complexation, and precipitation (Long et al., 2010). However, the metal(loid)s release in leachate was high at the first recirculation cycle. Increasing the moisture content of waste by leachate recirculation may improve the leaching process and biochemical reactions. Also, it was noted that the overall concentration of metal(loid)s was higher in young landfill leachate (S.S) compared to old landfill leachate (S.G). It is related to pH and the age of landfills; the pH varies according to the age of the landfill. As landfills age, the pH of leachates changes from values corresponding to acidic to alkaline solutions. Therefore, there is a higher concentration of metals in young landfill (S.S) is more acid than in old landfill (S.G), and therefore, there is a higher concentration of metals (Lee et al., 2022a, 2022b).

Table 3 shows the recovery rates for the metal(loid)s from the initial leachate sample recirculated ten times. Table 3 highlights that metal (loid)s recovery rate was over 100 % at the first cycle except for Al, Co, As, Pb and Hg in young landfill leachate and Al, Cr, Zn, and Hg in old landfill leachate. During the initial recirculation cycles, the pH of leachate was low as it is in the acidogenesis stage, so the metals had high solubility and dissolved into leachate. Also, there was metal dissolution into the liquid from the solid MSW sample due to the degradation of organic matter of the solid MSW sample and the leachate recirculation at the initial stage of the experiment. Therefore, high recovery rates were observed. After the first recirculation cycle, pH was increased to neutral as it reached the methanogenesis stage, and the metal could precipitate and be trapped in the samples. On average, the metal(loid)s recovery

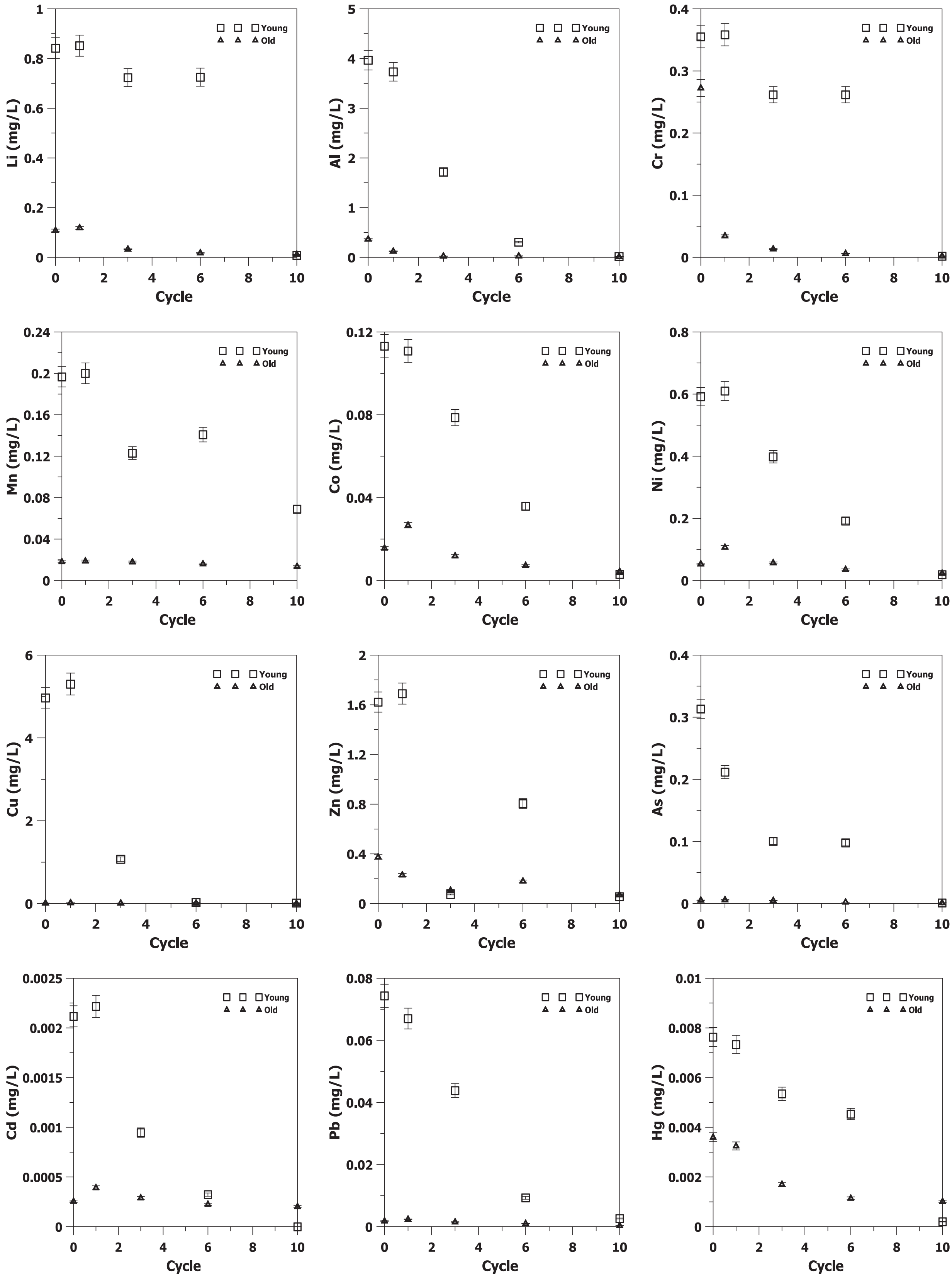

Fig. 4. Variations of metal(loid)s in the leachate from young (S.S) and old (S.G) landfill leachate.

Table 3
Recovery rates of metal(loid)s from young (S.S) and old (S.G) landfill leachate according to recirculation cycle.

Cycle time	Li (%)		Al (%)		Cr (%)		Mn (%)		Co (%)		Ni (%)	
	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G
1	101	109	94	33	101	13	102	104	98	171	103	201
3	86	30	43	6	74	5	63	99	70	76	67	107
6	86	16	8	7	74	2	72	89	32	46	32	67
10	1	11	0.5	3	0.5	1	35	75	3	28	3	43
Average	69	41	36	12	62	5	68	92	50	80	52	105
Variance	1561	1581	1373	147	1392	21	566	124	1315	3036	1406	3636
Cycle time	Cu (%)		Zn (%)		As (%)		Cd (%)		Pb (%)		Hg (%)	
	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G	S.S	S.G
1	107	166	104	61	68	110	105	154	90	130	96	90
3	22	98	5	28	32	89	45	114	59	83	70	47
6	0.6	87	50	48	31	43	15	88	13	56	59	32
10	0.3	40	3	19	0.4	24	0	79	4	18	3	28
Average	32	98	40	39	33	67	41	109	41	72	57	49
Variance	1920	2034	1702	276	566	1193	1604	831	1237	1665	1164	608

rate was 14.59 % higher at the first recirculation cycle in old landfill leachate.

Zn showed similarity similar tendency to Mn and Li was similar with Cr in young landfill leachate. Co exhibited the same behaviour as Ni and Cd in old landfill leachate. Zn and Mn showed another increase at the third recirculation cycle in the old landfill leachate, and Zn increased again at the third recirculation cycle in the young landfill leachate. This may be caused by the exposure of the metal to various processes such as absorption–desorption, precipitation-dissolution and complexation-dissolution, which means the metals in leachate may be the express proportion after saturation of balances in waste (Long et al., 2010). Another possible reason is the degradation of waste, which release a part of the metals bound with the organic matter in the waste.

Al, Cu and Zn have relatively higher concentrations than other metals, which can be a great opportunity. Cu showed an excellent recovery rate at the first recirculation cycle. However, the recovery rate by recirculation of Al and Zn were not effective. Co, Ni, Cd and Pb in old landfill leachate showed a high recovery rate at the first recirculation cycle. The results demonstrate that the amount of metals recovered in the leachate may be potentially increased by recirculation.

There are several factors affecting the mobility of metal(loids), Table 4 shows the correlations between metals and other variables.

According to the Spearman's correlation coefficient values, all metal (loid)s concentration is statistically correlated with the low pH value. Therefore, pH is the most critical factor for controlling metal mobility, and metals have minimum solubility at a pH range between 7.5 and 9. Redox potential has been considered one of the critical factors controlling the mobility of metals. However, previous research that studied redox potential influence on metal mobility has been limited to a few selected metals, such as As and Cr (Sommer and Lindsay, 1974; Chuan et al., 1996; Cao et al., 2001). It was pointed out that As and Cr mobility is directly correlated with the redox potential, bringing about the dominant species change. The results showed that the mobility of Cr is directly correlated with redox potential, which is consistent with the previous studies; however, As was not correlated with redox potential in this research. Negative correlations of Li, Al, Cr, Mn, Co, and Ni were observed with redox potential. However, Al, Cu, Zn, As, Cd, Pb and Hg did not show redox chemistry, which indicates that redox potential indirectly affects their release. All metal(loid)s show a strong positive correlation with COD, VFAs and conductivity. TOC and VFAs showed a similarity tendency with COD. However, Cu, Zn, As, and Cd did not correlate with TOC unlike COD and VFAs. It indicates that metals release was strongly affected by pH, COD and VFAs.

4. Conclusion

This work presents and verifies an effective method to increase metal concentration by recirculating to enhance metal recovery from landfill leachate with an understanding of the factors that impact metal solubility. Leachate recirculation increases the metal recovery rate up to 201 %. Among the 12 selected metal(loid)s, Li, Cd, Mn, Cu, and Ni can be obtained over 100 % leaching after the first leachate recirculation cycle regardless of the age of the waste. Therefore, leachate recirculation gives an economically feasible option to accelerate secondary resource utilisation. Good correlations between metal concentration, pH, organic matter, and redox potential give ground for a better understanding of the

Table 4
Spearman's correlations coefficients for metals and physico-chemical parameters for [S.S/ S.G] landfill leachate recirculation samples taken after ten cycles.

	Li (%)	Al (%)	Cr (%)	Mn (%)	Co (%)	Ni (%)
COD	0.83**	0.67*	0.68*	0.91**	0.73*	0.74*
pH	−0.88**	−0.84**	−0.77**	−0.94**	−0.88**	−0.88**
Conductivity	0.86**	0.74*	0.75*	0.93**	0.79**	0.80**
Redox	−0.75*	−0.57	−0.64*	−0.82**	−0.65*	−0.66*
TOC	0.90**	0.80**	0.75*	0.97**	0.84**	0.86**
VFAs	0.87**	0.78**	0.72*	0.96**	0.82**	0.83**
Age	−0.79**	−0.62	−0.61	−0.88**	−0.67*	−0.69*
	Cu (%)	Zn (%)	As (%)	Cd (%)	Pb (%)	Hg (%)
COD	0.61	0.59	0.72	0.61	0.72*	0.66*
pH	−0.78**	−0.72*	−0.84**	−0.79**	−0.87**	−0.80**
Conductivity	0.68*	0.66*	0.77*	0.68*	0.78**	0.73*
Redox	−0.51	−0.52	−0.62	−0.52	−0.63	−0.61
TOC	0.75*	0.72*	0.84**	0.75*	0.84**	0.77**
VFAs	0.74*	0.70*	0.82**	0.73*	0.82**	0.73*
Age	−0.56	−0.54	−0.68*	−0.55	−0.68*	−0.58

** . Correlation is significant at the 0.01 level.
* . Correlation is significant at the 0.05 level.

chemical processes and indicate that organic matter's significant effects on metal mobility. To reach sufficient acidic process conditions to increase metal mobility, additional acid would be necessary. Therefore, further work is required to explore the influence of organic matter on metal recovery with leachate recirculation to maximise the metal recovery rate.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

The authors would like to thank Richard Andrews, Jane Hubble, Nigel Janes, Rodrigo Leitao and Nuannat Simmons, from the Environmental Science and Technology Soil Laboratories at Cranfield University, for their assistance in the laboratory work.

References

Begeal, C., 2008. The effect of chloride ion on heavy metal partitioning and transport in an urban watershed: patroon creek, albany, ny. Geol. Theses Dissertation. 9. https://scholarsarchive.library.albany.edu/cas_daes_geology_etd/9.
Benson, C.H., Barlaz, M.A., Lane, D.T., Rawe, J.M., 2007. Practice review of five bioreactor/recirculation landfills. Waste Manag. 27, 13–29. https://doi.org/10.1016/J.WASMAN.2006.04.005.
Bhalla, M., Saini, M., Jha, M., 2013. Effect of age and seasonal variations on leachate characteristics of municipal solid waste landfill. Int. J. Res. Eng. Technol. 2 (2013), 223–232.
Bilgili, M.S., Demir, A., Özkaya, B., 2007. Influence of leachate recirculation on aerobic and anaerobic decomposition of solid wastes. J. Hazard. Mater. 143, 177–183. https://doi.org/10.1016/J.JHAZMAT.2006.09.012.
Bilgili, M.S., Demir, A., Varank, G., 2012. Effect of leachate recirculation and aeration on volatile fatty acid concentrations in aerobic and anaerobic landfill leachate. Waste Manag. Res. 30, 161–170. https://doi.org/10.1177/0734242X11417983.
Bisquerra, R., 1989. Introducción conceptual al análisis multivariable. Promoc Public Univ, SA Barcelona.
Cao, X., Chen, Y., Wang, X., Deng, X., 2001. Effects of redox potential and pH value on the release of rare earth elements from soil. Chemosphere 44, 655–661. https://doi.org/10.1016/S0045-6535(00)00492-6.
Chuan, M.C., Shu, G.Y., Liu, J.C., 1996. Solubility of heavy metals in a contaminated soil: effects of redox potential and pH. Water, Air Soil Pollut. 90, 543–556.
Cipullo, S., Prpich, G., Campo, P., Coulon, F., 2018. Assessing bioavailability of complex chemical mixtures in contaminated soils: Progress made and research needs. Sci. Total Environ. 615, 708–723. https://doi.org/10.1016/J.SCITOTENV.2017.09.321.
Cossu, R., Raga, R., Rossetti, D., 2003. The PAF model: an integrated approach for landfill sustainability. Waste Manag. 23, 37–44. https://doi.org/10.1016/S0956-053X(02)00147-2.
Cozzi, L., Gould, T., 2021. World Energy Outlook 2021. IEA Publ. 1–386.
Damikouka, I., Katsiri, A., 2021. Natural attenuation in marine sediments: investigation of the effect of chloride concentration on the mobility of metals. Recent Dev. Innov. Strateg. Environ. Sci. Eur. https://doi.org/10.1007/s11356-020-09852-4/Published.
Guérin, R., Munoz, M.L., Aran, C., Laperrelle, C., Hidra, M., Drouart, E., Grellier, S., 2004. Leachate recirculation: moisture content assessment by means of a geophysical technique. Waste Manag. 24, 785–794. https://doi.org/10.1016/J.WASMAN.2004.03.010.
Gutiérrez-Gutiérrez, S.C., Coulon, F., Jiang, Y., Wagland, S., 2015. Rare earth elements and critical metal content of extracted landfilled material and potential recovery opportunities. Waste Manag. 42, 128–136. https://doi.org/10.1016/J.WASMAN.2015.04.024.
Hernández Parrodi, J.C., Höllen, D., Pomberger, R., 2018. Characterization of fine fractions from landfill mining: A review of previous investigations. Detritus 2, 46–62. https://doi.org/10.31025/2611-4135/2018.13663.
James, C.N., Copeland, R.C., Lytle, R.A., 2004. Relationships between oxidation-reduction potential, oxidant, and pH in drinking water. WQTC Conference. American Waterworks Association, San Antonio, TX, USA.
Kaartinen, T., Sormunen, K., Rintala, J., 2013. Case study on sampling, processing and characterization of landfilled municipal solid waste in the view of landfill mining. J. Clean. Prod. 55, 56–66. https://doi.org/10.1016/J.JCLEPRO.2013.02.036.
Koumalas, A., Barampouti, E.M., Dounavis, A., Mai, S., 2019. Leachates from landfill sites in Thessaloniki, Greece: Effect of aging. Environ. Res. Eng. Manag. 75, 30–39. https://doi.org/10.5755/j01.erem.75.4.23073.
Kurniawan, T.A., Singh, D., Xue, W., Avtar, R., Othman, M.H.D., Hwang, G.H., Setiadi, T., Albadarin, A.B., Shirazian, S., 2021. Resource recovery toward sustainability through nutrient removal from landfill leachate. J. Environ. Manage. 287, 112265 https://doi.org/10.1016/J.JENVMAN.2021.112265.
Lee, H., Coulon, F., Beriro, D.J., Wagland, S.T., 2022a. Recovering metal(loids) and rare earth elements from closed landfill sites without excavation: Leachate recirculation opportunities and challenges. Chemosphere 292, 133418. https://doi.org/10.1016/J.CHEMOSPHERE.2021.133418.
Lee, H., Coulon, F., Wagland, S.T., 2022b. Influence of pH, depth and humic acid on metal and metalloids recovery from municipal solid waste landfills. Sci. Total Environ. 806, 150332 https://doi.org/10.1016/J.SCITOTENV.2021.150332.
Long, Y.Y., Shen, D.S., Wang, H.T., Lu, W.J., 2010. Migration behavior of Cu and Zn in landfill with different operation modes. J. Hazard. Mater. 179, 883–890. https://doi.org/10.1016/J.JHAZMAT.2010.03.087.
Lucas, H.I., López, C.G., Parrodi, J.C.H., Vollprecht, D., Raulf, K., Pomberger, R., Pretz, T., Friedrich, B., 2019. Quality assessment of nonferrous metals recovered by means of landfill mining: A case study in Belgium. Detritus 8, 79–90. https://doi.org/10.31025/2611-4135/2019.13879.
Luo, Z., Chen, W., Wen, P., Jiang, G., Li, Q., 2019. Impact of leachate recirculation frequency on the conversion of carbon and nitrogen in a semi-aerobic bioreactor landfill. Environ. Sci. Pollut. Res. 26, 13354–13365. https://doi.org/10.1007/s11356-019-04817-8.
Molaey, R., Yesil, H., Calli, B., Tugtas, A.E., 2021. Influence of volatile fatty acids in anaerobic bioleaching of potentially toxic metals. J. Environ. Manage. 285, 112118 https://doi.org/10.1016/J.JENVMAN.2021.112118.
Montesantos, N., Fini, M.N., Muff, J., Maschietti, M., 2022. Proof of concept of hydrothermal oxidation for treatment of triazine-based spent and unspent H2S scavengers from offshore oil and gas production. Chem. Eng. J. 427, 131020 https://doi.org/10.1016/J.CEJ.2021.131020.
Moreau, V., Dos Reis, P.C., Vuille, F., 2019. Enough metals? Resource constraints to supply a fully renewable energy system. Resources 8. https://doi.org/10.3390/resources8010029.
Mousavi, M.S., Feng, Y., Mccann, J., Eun, J., Marques, C., 2021. In Situ Characterization of Municipal Solid Waste Using Membrane Interface Probe (MIP) and Hydraulic Profiling Tool (HPT) in an Active and Closed Landfill Characterization of Municipal Solid Waste Using Membrane Interface Probe (MIP) and Hydraulic Profiling Tool (HPT) in an Active and Closed. https://doi.org/10.3390/infrastructures6030033.
Nag, M., Shimaoka, T., Komiya, T., 2018. Influence of operations on leachate characteristics in the Aerobic-Anaerobic Landfill Method. Waste Manag. 78, 698–707. https://doi.org/10.1016/J.WASMAN.2018.06.044.
Ntwampe, S.K.O., Sheldon, M.S., Volschenk, H., 2008. Oxygen mass transfer for an immobilised biofilm of phanerochaete chrysosporium in a membrane gradostat reactor. Brazilian J. Chem. Eng. 25, 649–664. https://doi.org/10.1590/S0104-66322008000400003.
Panina, N.S., Belyaey, A.N., Simanova, S.A., 2002. Carboxylic acids and their anions. Acid and ligan properties. Russ. J. Gen. Chem., 72, pp. 91-94, https://doi.org/10.1023/A:1015353530785Popenda, A., 2014. Effect of redox potential on heavy metals and As behavior in dredged sediments. Desalination and Water Treatment, 52:19-21, 3918-3927. https://doi.org/ 10.1080/19443994.2014.887449.
Qu, X., He, P.J., Shao, L.M., Lee, D.J., 2008. Heavy metals mobility in full-scale bioreactor landfill: Initial stage. Chemosphere 70, 769–777. https://doi.org/10.1016/J.CHEMOSPHERE.2007.07.013.
Quaghebeur, M., Laenen, B., Geysen, D., Nielsen, P., Pontikes, Y., Van Gerven, T., Spooren, J., 2013. Characterization of landfilled materials: screening of the enhanced landfill mining potential. J. Clean. Prod. 55, 72–83. https://doi.org/10.1016/J.JCLEPRO.2012.06.012.
Renou, S., Givaudan, J.G., Poulain, S., Dirassouyan, F., Moulin, P., 2008. Landfill leachate treatment: Review and opportunity. J. Hazard. Mater. 150, 468–493. https://doi.org/10.1016/J.JHAZMAT.2007.09.077.
Sekman, E., Top, S., Varank, G., Bilgili, M.S., 2011. Pilot-scale investigation of aeration rate effect on leachate characteristics in landfills. Fresenius Environ. Bull. 20, 1841–1852.
Shearer, B., 2001. Enhanced biodegradation in landfills. Master of Science Thesis. Virginia Polytechnic Institute and State University, Virginia, USA.
Sommer, L.E., Lindsay, W.L., 1974. Effect of pH and redox on predicted heavy metal-chelate equilibria in soils. Soil Sci. Soc. Am. J. 43, 39.
Sponza, D.T., Ağdağ, O.N., 2004. Impact of leachate recirculation and recirculation volume on stabilization of municipal solid wastes in simulated anaerobic bioreactors. Process Biochem. 39, 2157–2165. https://doi.org/10.1016/J.PROCBIO.2003.11.012.
Top, S., Akkaya, G.K., Demir, A., Yıldız, S., Balahorli, V., Bilgili, M.S., 2019. Investigation of Leachate Characteristics in Field-Scale Landfill Test Cells. Int. J. Environ. Res. 13, 829–842. https://doi.org/10.1007/s41742-019-00217-5.
Wagland, S.T., Coulon, F., Canopoli, L., 2019. Developing the case for enhanced landfill mining in the UK. Detritus 5, 105–110. https://doi.org/10.31025/2611-4135/2019.13772.
Wagner, T.P., Raymond, T., 2015. Landfill mining: Case study of a successful metals recovery project. Waste Manag. 45, 448–457. https://doi.org/10.1016/J.WASMAN.2015.06.034.
Watari, T., McLellan, B.C., Ogata, S., Tezuka, T., 2018. Analysis of potential for critical metal resource constraints in the international energy agency's long-term low-carbon energy scenarios. Minerals 8. https://doi.org/10.3390/min8040156.

White, J.K., Beaven, R.P., Powrie, W., Knox, K., 2011. Leachate recirculation in a landfill: Some insights obtained from the development of a simple 1-D model. Waste Manag. 31, 1210–1221. https://doi.org/10.1016/J.WASMAN.2010.10.022.
Yao, J., Kong, Q., Li, W., Huayue, Z., Shen, D., 2014. Effect of leachate recirculation on the migration of copper and zinc in municipal solid waste and municipal solid waste incineration bottom ash co-disposed landfill. J. Mater. Cycles Waste Mang. https://doi.org/10.1007/s10163-013-0217-7.
Yao, J., Qiu, Z., Kong, Q., Chen, L., Zhu, H., Long, Y., Shen, D., 2017. Migration of Cu, Zn and Cr through municipal solid waste incinerator bottom ash layer in the simulated landfill. Ecol. Eng. 102, 577–582. https://doi.org/10.1016/J.ECOLENG.2017.02.063.