



The influence of humic acid on metal(loid)s leaching in landfill leachate for enhancing landfill mining

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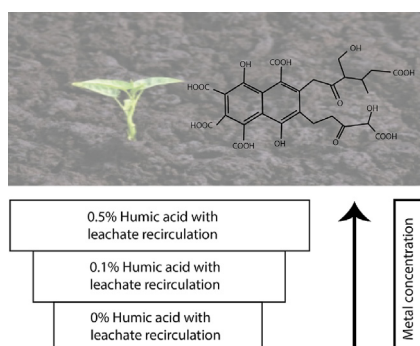
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HIGHLIGHTS

- Metal(loid)s have a strong affinity to humic acids; shows high efficacy on the recovery rate.
- High concentration of Pb, As, Al and Co outline recovery opportunities.
- The addition of humic acid inhibited the mobility of Li, Mn and Hg.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to investigate the effect of different concentrations of humic acid on the recovery rate of metal(loid)s in landfill leachate. The study focused on the release of 12 selected metal(loid)s, including critical raw materials (CRM) in landfills that were less than five years old and those that were more than ten years old. The experimental setup involved using different concentrations of humic acid (*w/v*) (0%, 0.1%, and 0.5%) at pH 4 and 6. The results of the study showed that humic acid was effective in releasing Al, Cr, Co, Ni, Cu, Zn, As, Cd, and Pb. On the other hand, an increase in humic acid concentration led to a decrease in the release of Li, Mn, and Hg. The immobilization of Li, Mn, and Hg was due to the coordination and adsorption of humic acid. The presence of humic acid accelerated the release of metal(loid)s by carboxylic acidity compared to the recovery rate of metal(loid)s in landfill leachate without humic acid. However, a higher concentration of humic acid did not always result in a stronger recovery rate. The recovery rate of metal(loid)s was related to the solubility and concentration of humic acid. These findings can inform the development of more efficient and environmentally-friendly methods of recovering metal(loid)s using humic acid as a leaching agent.

1. Introduction

Resource depletion poses a significant global challenge in the context of climate change. The current global energy mix predominantly relies on finite fossil fuels that, when burned, emit harmful greenhouse gases into

the environment (Månberger and Stenqvist, 2018). To limit the atmospheric concentration of carbon dioxide to 450 ppm by 2050 and mitigate the impacts of climate change, renewable energy sources must be developed and implemented (OECD, 2016; Hodgkinson and Salder-Smith, 2018). However, the production of renewable energy technologies requires a significant flow and stock of mineral resources, particularly metals, which are extractable but finite (Krishnan et al., 2021; Lee et al., 2022a, 2022b). Landfills offer a potential source of these metals through the process of

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landfill mining, which involves extracting minerals from waste materials that have been buried (Vollprecht et al., 2021). While this process has the potential to reduce landfill size and recover valuable materials, it is also associated with operational challenges, high energy requirements, and costs (Krook et al., 2012; Lee et al., 2022a, 2022b). An alternative approach is to focus on leachate treatment, which can remediate waste and recover valuable metals as secondary resources while generating electricity from organic compounds in the leachate (Iskander et al., 2016; Kurniawan et al., 2021; Lee et al., 2022a, 2022b). This approach offers several advantages, including maintaining the quality of metals and avoiding the decline in ore deposit concentration.

However, recovering metals from landfill leachate, which are present in very low concentrations, is a challenging task (Iskander et al., 2016; Lee et al., 2022a, 2022b). According to Kjeldsen et al. (2002), leachate metals may account for <0.02 % of the total metals present. To increase the concentration of metals in landfill leachate, solutions are needed. Previous work investigated the factors that affect metal mobility and enhance the metal recovery rate from landfill leachate (Lee et al., 2023). This showed that leachate recirculation, pH and organic matter are the critical factors that increase landfill leachate's recovery rate. The first cycle of leachate recirculation increases the metal recovery rate by up to 201 %; however, the recovery rate will be increased as the concentration of metal(loid)s increases, which will support the establishment of cost-effective metal extraction processes from landfill leachate.

Several approaches have been studied previously to leach processes using acid leaching, and chelation has been used, especially for treating contaminated soils. Ethylene diamine tetraacetic acid (EDTA) has been widely investigated as a ligand for its strong chelating ability for different heavy metals (Norvell, 1991; Sun et al., 2001). EDTA is a synthetic chelator that is a versatile driving agent that can form four or six bonds with metal ions (Shinta et al., 2021). However, EDTA has relatively high costs of approximately \$50/kg N2EDTA, and is not easily biodegradable and may remain absorbed by soil particles (Wasay et al., 1998; Barona et al., 2001; Palden et al., 2020). Also, the persistence of EDTA in the environment may create additional and unforeseen problems, such as contaminated groundwater (Shinta et al., 2021). Therefore, research into new biodegradable selective chelating agents is needed. Metal behavior is influenced by various reactions such as complexation, ion exchange, sorption and desorption, precipitation, and dissolution reactions (Bang and Hesterberg, 2004). Organic ligands, particularly humic substances, are known to impact metal mobility through complexation reactions by increasing or decreasing its sorption on mineral surfaces (Gungor and Bekbölet, 2010). Humic substances are a natural mixture of organic compounds and the primary constituent of organic matter in nature, which can be divided into three fractions based on their solubility behavior: humic acids, fulvic acids, and humins (Tobiašová et al., 2018).

Humic and fulvic acids have a similar structure but differ in molecular weight and functional group contents (Evangelou, 1998; Gungor and Bekbölet, 2010). Humic acid, which has a higher molecular weight and contains fewer oxygen-containing functional groups than fulvic acid, is generally present in higher concentrations in soil than fulvic acid (Spark et al., 1997). Humic substances (humic and fulvic acids) can bind metals to their hydroxyl and carboxyl groups (Leung and Kimaro, 1997; Bozkurt et al., 2000; Klavins et al., 2006; Gutiérrez-Gutiérrez et al., 2015). Humic acid is known to complex with heavy metal ions, owing to its main functional groups, including carboxylic acids, alcohols, phenols, carbonyls, phosphates, sulphate, amides, and sulphides, which can interact with metal species in solution (Senesi, 1992). Moreover, humic acid is organic and benefits plant growth through nutrient uptake and hormonal effects (Ampong et al., 2022).

To increase the concentration of metal(loid)s in landfill leachate with less environmental disturbance, it is essential to understand the solubility characteristics of humic acid and how it influences metal release. Previous research has focused on enhancing decomposition and accelerating stabilization by landfill leachate recirculation. These studies demonstrated that not all metals are bound to the humic acid and could have retained heavy

metals in the landfill by adsorption or surface complexation (Šan and Onay, 2001; Xiaoli et al., 2007; Qu et al., 2008; Long et al., 2010; Yao et al., 2014; Luo et al., 2019; Yao et al., 2019). However, leachate recirculation has the opportunity to increase the mobility of metals, and the appropriate amount of humic acid may assist in increasing metal mobility; thus, combining those methods can enhance the metal's recovery rate. Therefore, this study investigates the impact of changing the humic acid concentration on metal recovery from landfill leachate by adding humic acid and conducting recirculation operations on 12 metal(loid)s.

2. Materials and methods

2.1. Sample collection and preparation

Municipal solid waste (MSW) was collected from two former MSW landfills located in the UK. 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 5/6 m deep. The Gloucestershire landfill received 60,000 m³ of inert, industrial, commercial, and household waste. The younger waste samples were 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. 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 for the purpose of this research into metal, paper, textile, wood, glass and other materials, and the composition of the samples in this experiment by wet weight is in Table 1.

2.2. Experimental set-up

Twelve sets of semi-pilot scale Perspex columns were set up to simulate leachate production from MSW of different ages and compositions (Table 1). Dimensions of each column were 21 cm diameter and 1.02 m height, with a working volume of 4 L (Fig. 1) (Lee et al., 2023). Six of the columns were filled with waste <1-year-old (S.S) and the other six with waste >10 years old (S.G). 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. The waste weighing 15 kg was compressed by pushing and then deposited into the column, filling it 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 5 L plastic waster container. Leachate was continuously recirculated for 5 cycles using the peristaltic pump with flow rates adjusted to 800 mL/day. A slow flow rate for the peristaltic pump with leachate, such as <100 mL/day, may occur clogging. Although a high recirculation flow rate can accelerate the stabilization of landfills, excessive recirculated leachate will impose a heavy working load on the leachate collection system, which is not secure and cost-effective (Feng et al., 2018). Therefore, the flow rate was set as 800 mL/day.

To measure the effect of humic acid on metal(loid)s mobility, 0 %, 0.1 % and 0.5 % of humic acid were set at pH 4 and 6. The pH was adjusted using 1.8 M nitric acid solution or 1 M sodium hydroxide solution. Humic acid was added to tap water at concentration of 0.1 g and 0.5 g per 100 mL of tap water; 4 g of humic acid was added to 0.1 %, and 20 g of humic acid was added to 0.5 % with 4 L of tap water.

2.3. Sampling and analytical methods

Leachate samples (250 mL) were collected after each cycle from the leachate outlet port. To keep the volume equilibrium of leachate before

Table 1

Composition of the MSW samples collected from the landfill sites; sample no.1 is from Gloucestershire (S.G) and sample no.2 is from Suffolk (S.S).

Samples no.	Years	Waste sample (g)	Paper/cardboard (g)	Plastic (g)	Metals (g)	Glass/ceramic (g)	Textile (g)	Wood (g)	Soil/fine fraction (g)	Other (g)
1	>10	62,230	1410	4623	224	10	520	842	54,437	164
2	<1	59,470	4692	7094	2212	3663	3911	1112	35,944	762

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 experiment was conducted, parameters included: pH, chemical oxygen demand (COD), redox potential, total organic carbon (TOC), volatile fatty acid (VFA), and conductivity (further details in supplementary data Table 1). 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:

$$\text{Recovery rate} = \frac{L_c}{I_c} \times 100$$

where L_c , leaching concentration of metal after recirculation; I_c , initial metal concentration.

The detection limit test was conducted, including all steps of the sample analysis. The blank test was repeated, and the standard deviation of parallel determinations was calculated. To establish the method detection limits (MDLs) for each metal(loid)s, the standard deviation of the average derived from the blank samples was multiplied by four. The resulting MDLs for the selected metal(loid)s ranged from 1.0 to 8.4 ng/L. The data below the detection limit was presented as < MDL in Table 2.

2.4. Humic acid analysis

A stock solution of HA was prepared by dissolving 1 g of HA in 1 L of deionised water for investigating the impact of the presence of HA in leachate on the leachability of metals. HA content was determined by ultraviolet light absorbance at 254 nm. The values were converted using a calibration curve. For that purpose, serial dilutions of the concentrated HA stock solution were prepared: 1, 2, 5, 7, 10, 15, 20, 50, 100 mg. UV_{254} was measured

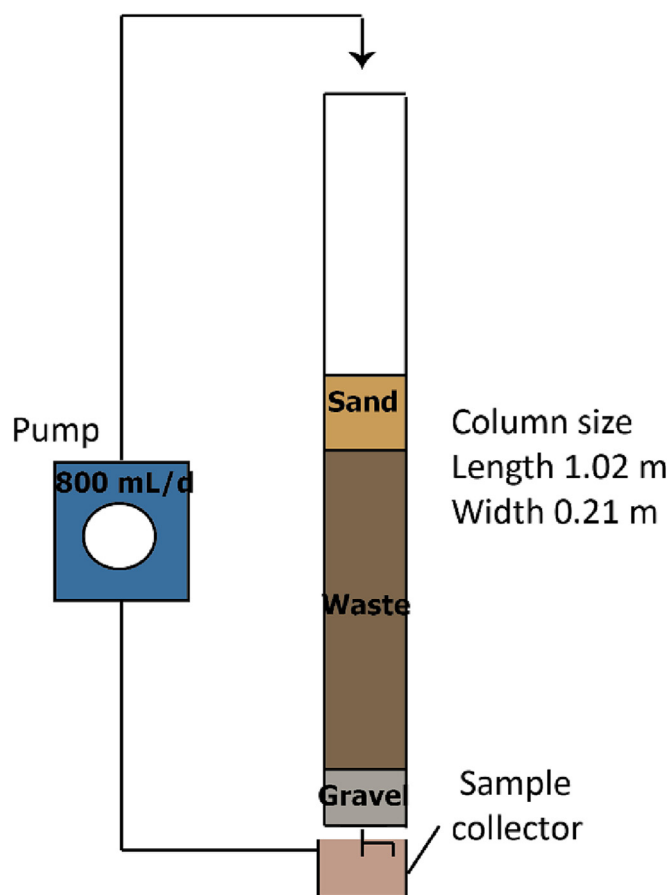


Fig. 1. Schematic diagram of the column percolation test (Lee et al., 2023).

Table 2

The initial concentration of selected metal(loid)s in the solutions at pH 4 and 6 that were circulated in the waste matrix of Gloucestershire (S.G) and Suffolk (S.S).

(mg/L)	S.S		S.G		(mg/L)	S.S		S.G	
	pH 4	pH 6	pH 4	pH 6		pH 4	pH 6	pH 4	pH 6
Li	0.06	0.12	<MDL	<MDL	Cu	0.19	0.19	0.05	0.05
Al	0.62	0.06	0.02	0.04	Zn	0.12	0.19	0.06	0.12
Cr	0.01	<MDL	<MDL	0.02	As	0.01	<MDL	<MDL	<MDL
Mn	0.10	0.45	0.10	0.03	Cd	<MDL	<MDL	<MDL	<MDL
Co	0.01	0.01	<MDL	<MDL	Pb	0.01	<MDL	<MDL	<MDL
Ni	0.04	0.04	<MDL	0.03	Hg	0.33	0.09	0.01	<MDL

through a 1 cm quartz cell. Samples for UV₂₅₄ were filtered through 0.45 μm membrane filter. The test was conducted in duplicate.

The E₄/E₆ ratio (absorbance ratio at 465 nm and 665 nm) has been widely used to study the humic acid fraction. The ratio is inversely proportional to molecular weight, acidity and degree of humification. The value of E₂₈₀ is used as an indicator for the aromaticity of the sample's structure because electron transition occurs in this UV region for phenolic arenes, benzoic acids, aniline derivatives, polyenes, and polycyclic aromatic hydrocarbons with two or more rings (Uyguner and Bekbolet, 2005). E₄/E₆ ratio and E₂₈₀ were measured through a 1 cm quartz cell. Samples for E₄/E₆ ratio and E₂₈₀ were filtered through a 0.45 μm membrane filter. The test was conducted in duplicate.

Previous studies showed that 0.1 to 2 M of oxalic acid increased metal release, and 0.5 % humic acid leached out Cd, Pb, Ni, Cu and Zn, twice that of 0 % humic acid (Das et al., 2021; Usharani and Vasudevan, 2016). Therefore, the effect of humic acid concentration on the release of selected metal(loid)s was studied at 0.1 % and 0.5 % at pH 4 and 6 from young and old landfill leachate. The leachate was collected from the bottom of the columns after circulating 4 L of tap water through the waste matrix. Subsequently, humic acid was introduced to investigate its impact on metal(loid)s leachability. According to previous studies (Bhalla et al., 2013; Moody and Townsend, 2017; Lee et al., 2022a, 2022b), the pH value of authentic landfill leachate is generally <6.5 for leachate <5 years old and >7.5 for leachate over 10 years old. However, our previous research (Lee et al., 2022a, 2022b) and findings from the lower pH range indicated a higher release of metal(loid)s. Considering the leaching efficiency of humic acid, which was comparatively higher at pH 4 and 6, the pH values selected for this study were 4 and 6.

2.5. ATR-FTIR spectra

Attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) was performed at room temperature in the wavenumber range between 400 and 4000 cm⁻¹ with 16 scans at a spectra resolution of 4 cm⁻¹ to detect any interaction between the metals and humic acid. FTIR spectroscopy shows the intensity of the peak absorbance of the function groups, which can identify the chemical bond in metals. The freeze-dried leachate samples were used for the device Bruker Platinum ATR.

2.6. Statistical analyses

IBM Statistical Package for the Social Sciences (SPSS) 26 was used for statistical analysis. Multivariate analysis of variance is a critical 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 matrixes. 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. Solubility of humic acid

It is well known that the solubility of humic acid in leachate increases with increasing pH (Fan et al., 2006; He et al., 2006; Qu et al., 2008;

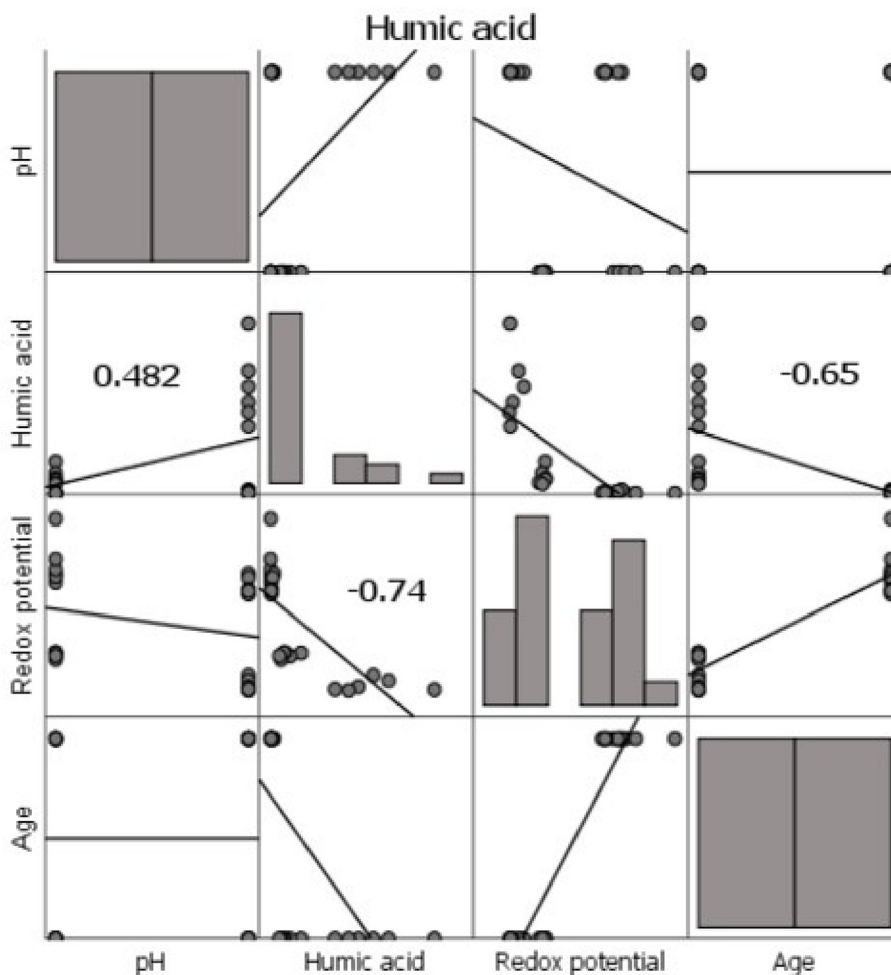


Fig. 2. The leached concentration values of humic acid in landfills, according to pH, redox potential, and age.

Table 3

Recovery rates of metal(loid)s from young (S/S) landfill leachate at pH 4 and different HA concentrations (0.1 and 0.5 %).

Humic acid (%)	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5
Cycle time	Li (%)		Al (%)		Cr (%)		Mn (%)		Co (%)		Ni (%)	
1	206	47	280	1442	143	271	98	81	120	196	175	143
2	171	49	122	913	87	214	91	95	100	157	146	117
3	153	57	94	625	72	165	86	105	88	141	128	100
4	132	58	81	302	59	98	75	88	76	112	108	78
5	117	60	79	362	50	106	70	96	66	115	94	86
Average	156	54	131	729	82	171	84	93	90	144	130	105
Variance	1230	35	7248	2*10 ⁵	1359	5345	130	84	454	1192	1002	682
Humic acid (%)	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5
Cycle time	Cu (%)		Zn (%)		As (%)		Cd (%)		Pb (%)		Hg (%)	
1	83	90	156	183	23	228	725	624	132	258	29	4
2	62	82	151	193	19	148	659	737	124	262	1	2
3	52	71	145	206	17	99	684	783	112	239	1	2
4	41	54	127	162	14	53	589	546	83	155	3	2
5	36	63	111	195	12	73	555	668	63	182	0	1
Average	55	72	138	188	17	120	643	672	103	219	7	2
Variance	356	209	345	273	20	273	4894	8669	833	2341	158	1

Zhang et al., 2018; Lee et al., 2022a, 2022b). The relationships between humic acid, pH, landfill age, and redox potential are shown Fig. 2. A significant positive correlation was observed between pH and humic acid ($p < 0.05$), which is consistent with the findings reported by Lee et al. (2022a, 2022b). When the pH is neutral to high, humic acids tend to be more negatively charged because of the ionization of COOH and phenolic OH groups. On the other hand, a strong negative correlation was observed between humic acid and redox potential ($p < 0.01$) as well as landfill age ($p < 0.01$). This suggests that the leaching of humic acid increases with a decrease in landfill age and redox potential. In young landfills, the concentrations of volatile fatty acids and humic acids are high mainly due to the anaerobic fermentation of organic compounds (Lee et al., 2022a, 2022b). In contrast, humic acids tend to be lower in older landfills (>10 years) (Han et al., 2009). A comparison of redox potential with the concentration of humic acids shows lower redox potential with higher concentrations of humic acid. It may be due to the consumption of organic carbon as landfill ages. Humic acids contain various redox-active functional groups, such as hydroxyl and phenol, thereby enabling them to participate in the redox reaction directly (Bai et al., 2020; Lacroix et al., 2021). pH and redox potential are the two critical parameters controlling the mobility of metals. Decreasing redox potential can be increased metal release in landfill environments (Cao et al., 2001). Therefore, it is crucial to understand the

relationship between each factor that affects metal release to increase the metal recovery rate in landfill leachate.

3.2. Effect of humic acid concentration on metal recovery

In both landfill sample types, most of the metal(loid)s releases were a consistent increase in the release of metal(loid)s with increasing concentrations of humic acid (further details in supplementary data Fig. 2, Fig. 3, Fig. 4, Fig. 5). The results indicate that the presence of humic acid significantly enhanced the release of most metals in both landfills. Tables 3 and 4 show the recovery rates of metal(loid)s when exposed to humic acid concentrations of 0.1, and 0.5 % at pH 4, after five times recirculation of the initial leachate sample (further details in supplementary data Table 3 and Table 4). While the initial recirculation of leachate may release metal (loid)s bound to hydrolysed organic matter, the release of metal(loid)s may subsequently decrease as degradation continues. This is due to reactions such as precipitation and transformation. Results show that for both landfills, an overall increasing trend was observed in the release of metal (loid)s with increasing humic acid concentration. In young landfill leachate, for a humic acid concentration of 0.1 %, metal(loid)s recovery rate exceeded 100 % in the first cycle except for Mn, Cu, As, and Hg at pH 4, and Mn, Cu, and Cd at pH 6. In the old landfill leachate, Al, Cr, Mn, Ni,

Table 4

Recovery rates of metal(loid)s from old (S/G) landfill leachate at pH 4 and different HA concentrations (0.1 and 0.5 %).

Humic acid (%)	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5
Cycle time	Li (%)		Al (%)		Cr (%)		Mn (%)		Co (%)		Ni (%)	
1	90	169	274	17,892	122	3044	139	156	75	436	151	1565
2	75	128	3200	9007	279	6003	127	180	114	285	395	2317
3	72	97	5517	10,390	391	418	109	129	145	268	655	365
4	62	45	5943	23,619	5516	8831	84	11	181	472	1917	2138
5	59	108	6025	38,024	207	2159	57	109	141	706	739	1542
Average	72	110	4192	19,786	1303	4091	103	117	131	434	771	1640
Variance	151	2060	6*10 ⁶	1*10 ⁸	5*10 ⁶	1*10 ⁷	1099	4276	1557	31,347	5*10 ⁵	4*10 ⁵
Humic acid (%)	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5	0.1	0.5
Cycle time	Cu (%)		Zn (%)		As (%)		Cd (%)		Pb (%)		Hg (%)	
1	17	80	69	73	371	5930	371	530	46	1892	31	12
2	50	50	76	99	422	2278	216	348	342	924	15	122
3	55	44	106	117	626	1661	210	278	278	763	14	38
4	60	76	150	30	639	1294	482	297	760	601	8	21
5	58	109	82	186	684	1540	257	499	210	646	11	19
Average	48	72	97	101	548	2541	307	390	329	966	16	42
Variance	316	674	1096	3340	20,013	3*10 ⁶	13,710	13,607	70,603	3*10 ⁵	78	2081

Table 5
Correlations coefficients for metals and recirculation cycle, pH and humic acid for [young/ old] landfill leachate recirculation samples.

	Li (%)	Al (%)	Cr (%)	Mn (%)	Co (%)	Ni (%)
Recirculation	-0.18	-0.40*	-0.29*	-0.19	-0.40**	-0.45**
Humic acid	-0.28*	0.55**	0.45**	-0.12	0.52**	0.46**
pH	-0.11	-0.51**	-0.37**	-0.11	-0.52**	-0.56**
Age	-0.56**	-0.18	-0.28	-0.23	-0.49**	-0.64**
	Cu (%)	Zn (%)	As (%)	Cd (%)	Pb (%)	Hg (%)
Recirculation	-0.48**	-0.36**	-0.41**	-0.37**	-0.39**	-0.29*
Humic acid	0.24*	0.38**	0.58**	0.49**	0.51**	-0.12
pH	-0.56**	-0.49**	-0.53**	-0.38**	-0.51**	-0.30**
Age	-0.64**	-0.62**	-0.21	-0.49**	-0.40**	-0.14

** Correlation is significant at the 0.01 level.

* Correlation is significant at the 0.05 level.

As, and Cd reached over 100 % recovery rate at pH 4, and Al, Mn, Co, Ni, As, Cd, and Hg at pH 6, for the same humic acid concentration. When the humic acid concentration was increased to 0.5 %, metal(loid)s recovery rate exceeded 100 % in the first cycle except for Li, Mn, Cu, and Hg at pH 4, and Li, Mn, and Hg at pH 6 in young landfill leachate. In old landfill leachate, Li, Al, Cr, Mn, Co, Ni, Cd, and Pb reached over 100 % recovery rate at pH 4, and Li, Al, Cr, Mn, Co, Ni, Cu, As, Cd, and Hg reached over 100 % recovery rate at pH 6. These findings align with those of Usharani and Vasudevan (2016), which suggest that humic acid can effectively leach out metals.

Li can be recovered using several different methods, such as co-precipitation and solvent extraction; however, it requires specific conditions such as temperature and pH (Meshram et al., 2014). When the

humic acid concentration was 0.1 %, the recovery rate of Li in young landfill leachate was 100 %, while in the case of old landfill leachate, a humic acid concentration of 0.5 % was required to achieve a similar recovery rate. This difference could be due to the fact that the sorption of Li onto humic acid is more significant than for other metals. The recovery rate of Li, Mn, Ni, Cd and Hg was relatively lower when humic acid was added 0.5 % in young landfill leachate. Humic acid has complex functional groups such as hydroxyl group (-COOH), which can provide electrons to coordinate with metals to form complex compounds or chelates. The coordination is related to the concentration of humic acid. Therefore, the combined effects of coordination and adsorption make the recovery rate of those metals lower when adding more humic acid.

Cationic metal interacts with the phenolic and carboxyl groups on the molecular chain of humic acid to form spherical compounds, which can effectively lower the concentration of available metals (Wu et al., 2017). The improvement of Li, Mn and Hg leaching rates is not apparent with increasing humic acid content. It may be because excessive humic acid can chelate with metal ions and reduce the concentration of Li, Mn and Hg in the leaching solution (Zhang et al., 2018). The recovery rate of metal(oids) using humic acid 0.1 % on average from young landfill leachate was 96.40 % and 1208.24 % at pH 4 and 6. The recovery rate of metal(oids) using humic acid 0.5 % on average from young landfill leachate was 144.03 % and 267.16 % at pH 4 and 6. The recovery rate of metal(oids) using humic acid 0.1 % on average from old landfill leachate was 1831.13 % and 1603.18 % at pH 4 and 6. The recovery rate of metal(oids) using humic acid of 0.5 % on average from old landfill leachate was 387.74 % and 216.10 % at pH 4 and 6. The recovery rate of some metals, such as Al was remarkably higher than other metals; the composition and distribution of the sample may cause it (Lee et al., 2022a, 2022b).

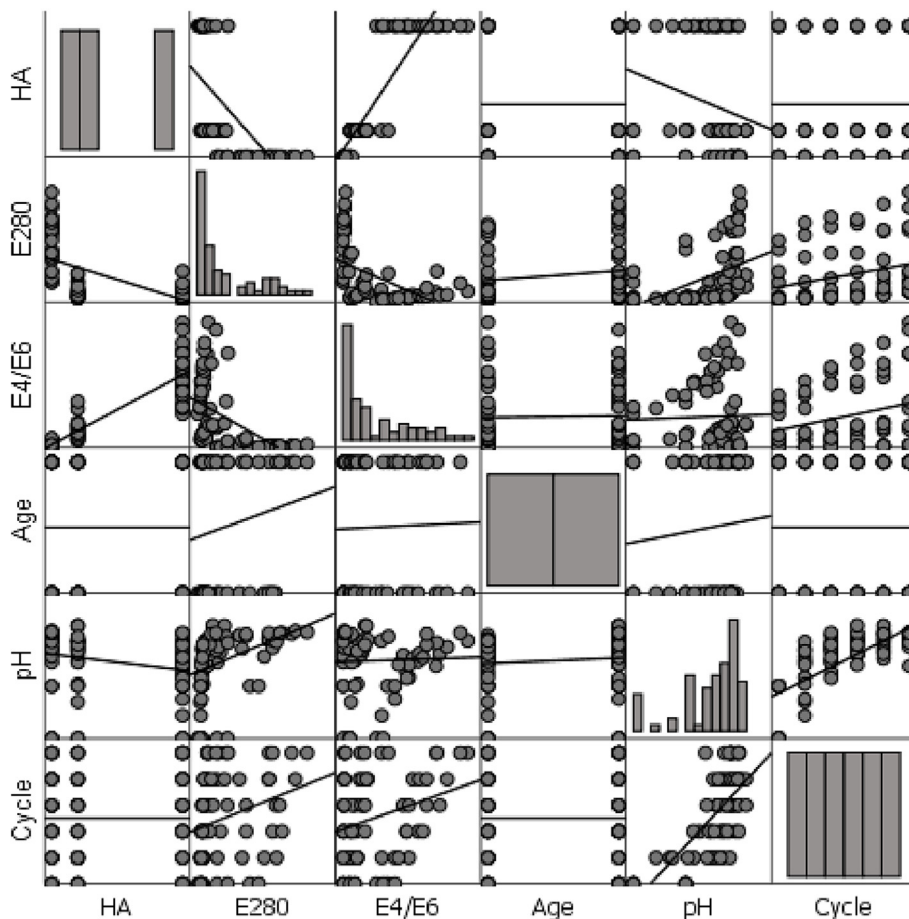


Fig. 3. Visualizing correlations regarding humic acid characteristics in landfill leachate.

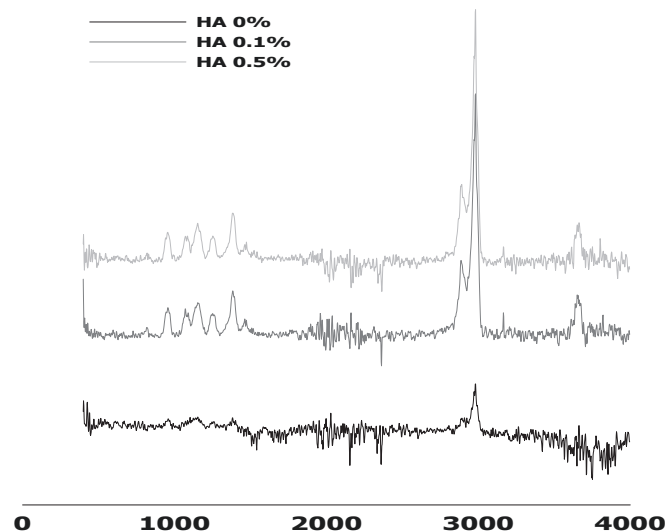


Fig. 4. FTIR spectra peak in landfill leachate which was assigned to a stretching frequency of H₂O molecule coordinated metal ions and methyl group (CH stretching), following the humic acid concentration (0, 0.1, 0.5 %).

For some metals, such as Al and Cr, recovery using humic acid from young landfill leachate may be more effective at pH 6. It may be related to humic acid solubility as they are insoluble in acidic conditions, where young landfill has lower pH value. pH plays an essential role in the stabilising process of humic acid toward metals. Most metals reached over 100 % recovery rate at 0.5 humic acid concentration at pH 6. However, metal release differs depending on the interaction between pH and humic acid and the existence of metal in different forms. These results indicate that some metals release may be increased at neutral pH than at acidic pH.

Several processes, such as proton-induced dissolution, oxidative dissolution caused by oxygen attack, ligand-induced dissolution caused by humic acid adsorption or surface complexation, and protection of the mineral surfaces from proton and oxygen attacks by adsorbed humic acid may enhance or inhibit the dissolution of metals in the presence of humic acid (Chou et al., 2018).

The correlation between humic acid, metals, recirculation cycle, and pH are shown in Table 5. There was a strong positive relationship between humic acid and Al, Cr, Zn, As, Cd, Pb ($p < 0.01$) and Cu ($p < 0.05$). However, Li shows a strong negative relationship with humic acid ($p < 0.05$). Additionally, there was a strong positive relationship between Li and Mn, Cu, Zn, Cd, Pb, Hg ($p < 0.01$) and Co, Ni ($p < 0.05$). Metal(loid)s leaching tended to decrease with increasing recirculation, pH value and landfill age. This study showed that an increase in humic acid concentration led to the release of most metals, increasing their availability and recovery rate in landfill leachate.

3.3. Chemical characteristics and FTIR spectra of humic acid

The adsorption coefficient at 280 nm (E₂₈₀) and the ratio of absorption coefficients at 465 and 665 nm (E₄/E₆) were measured in this study (Fig. 3). There was a strong relationship between humic acid and E₂₈₀ and E₄/E₆ ($p < 0.01$). The E₄/E₆ value increased while the concentration of humic acid increased, whilst E₂₈₀ decreased. The ratio is generally known to be inversely proportional to molecular weight and proportional to acidity. However, E₄/E₆ showed no clear trend with landfilling age and pH. The ratio is an index of humification with larger values indicative of greater humification. It implies the presence of aliphatic compounds with increasing humic acid (Uyguner and Bekbolet, 2005). The value of E₂₈₀ indicates the total aromaticity of the samples (Kang et al., 2002). In the study, increasing landfill age shows the highest E₂₈₀, which may be that younger landfill leachate was at the initial stage of humification.

Different functional groups can be identified from characteristic peaks by FTIR peaks. Fig. 4 shows the FTIR spectra of leachate samples that give information about specific molecular structures and various functional groups, as well as studies of interactions between humic acid and metals (Khan and Schnitzer, 1972). The leachate's spectral features indicated that the leachate's aromatic character was in the order of HA 0.5 % > HA 0.1 % > HA 0 %. This result is in agreement with humic acid characteristics.

All spectra in different concentrations of HA were similar. Adsorption bands observed at the wavelengths of 3751 cm⁻¹ (H₂O stretching), 3533 cm⁻¹ (N—H stretching), 3355 cm⁻¹ (O—H stretching), 2981 cm⁻¹ (C—H stretching), 2359 cm⁻¹ (C—O stretching), 2029 cm⁻¹ (Aromatics), 1682 cm⁻¹ (C=O stretching), 1623 cm⁻¹ (C—O stretching), 1107 cm⁻¹ (C—O stretching) and 668 cm⁻¹ (C—S stretching). The characteristics of functional groups were similar; however, the band's strength differed depending on the humic acid concentration. Increasing humic concentration leads to relatively stronger absorption bands at 1682 cm⁻¹, while having a lower absorption band at 1623 cm⁻¹. The observed intense peaks indicate the existence of carboxylic. The presence of groups near the COOH group of a carboxylic acid affects the acidity, which increases metal mobility.

Leachate with humic acid has stronger aliphatic bands, carboxylic groups, and aromatics. However, the strengths of the bands, which give differences in the relative distribution of each functional group, have some differences. The main metals binding sites of humic substances include the carboxylic acid site (Sawalha et al., 2007; Qu et al., 2008). Therefore, the high content of humic acid increases metal recovery as the acidity of the carboxylic acid is strong.

4. Conclusion

This study investigated the effect of humic acid on metal recovery in landfill leachate. The results showed that humic acid had a significant impact on increasing metal recovery in landfill leachate. Among the tested metals, Cd, Al, Zn, and Li had the highest recovery rates with 0.1 % humic acid concentration in young landfill leachate at pH 4, while Hg, Pb, Al, and As had the highest recovery rates at pH 6. For 0.5 % humic acid concentration in young landfill leachate, As, Al, Cr, and Pb had the highest recovery rates at pH 4, while Hg, As, and Al had the highest recovery rates at pH 6. In old landfill leachate, Al, Pb, and As had the highest recovery rates with 0.1 % humic acid concentration at pH 4, while Al, Cr, and As had the highest recovery rates at pH 6. For 0.5 % humic acid concentration in old landfill leachate, Al, Cr, and Pb had the highest recovery rates at pH 4, while As, Al, and Cr had the highest recovery rates at pH 6. Using humic acid for metal recovery in landfill leachate is a sustainable method that can help reduce costs and mitigate environmental impacts. These findings provide insight into the potential for organometallic contribution to develop innovative and cost-effective technologies for the recovery of metals from landfill leachate while considering the potential environmental and economic impacts.

CRedit authorship contribution statement

H. Lee: Conceptualization, Investigation, Writing – original draft. **F. Coulon:** Supervision, Conceptualization, Writing – review & editing. **S.T. Wagland:** Conceptualization, Supervision, Funding acquisition, Writing – review & editing.

Data availability

Data supporting this study are openly available from Cranfield University Online Research Data [CORD] at <https://doi.org/10.17862/cranfield.rd.22306042>.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.165250>.

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