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LANDFILL LEACHATE TREATMENT BY MEMBRANE-BASED TECHNOLOGIES: COST-BENEFIT ANALYSIS, MEMBRANE CONCENTRATE MANAGEMENT, AND PERSPECTIVES

Ronei de Almeida

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Ronei de Almeida

Tese de Doutorado apresentada ao Programa de Pós-Graduação em Engenharia de Processos Químicos e Bioquímicos, Escola de Química, da Universidade Federal do Rio de Janeiro, UFRJ, como parte dos requisitos necessários à obtenção do título de Doutor em Ciências.

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Landfill Leachate Treatment by Membrane-based Technologies: Cost-benefit Analysis, Membrane Concentrate Management, and Perspectives.

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Poeminha do Contra

Todos esses que aí estão
Atravancando meu caminho
Eles passarão
Eu passarinho!

Mário Quintana

ABSTRACT

The current thesis explores the techno-economic aspects of nanofiltration (NF) and reverse osmosis (RO) processes treating landfill leachate. The management of membrane concentrate streams and related issues are analysed in this context. Experimental landfill cells were constructed to investigate the impacts of concentrate recirculation practice on the leachate quantity, quality, and treatment performance. Data from the 420-day monitoring experiment were analysed using a statistical tool. Membrane-based technologies stand out for their high cost-benefit. NF and RO systems removed organic matter, reported as chemical oxygen demand (COD), absorbance at 254 nm (UV_{254}), and ammonia nitrogen at removal efficiencies higher than 90%.The leachate treatment operating costs ranged from 0.132 to 3.35 USD per m^3 of treated leachate. In contrast, the management of the concentrated leachate generated by membrane filtration processes is not considered when the expenses for NF/RO implementation are estimated. Concentrate streams are commonly re-introduced into the landfill without additional cost for landfill managers. However, the consequences of this practice are not well-known, and the literature's findings show contrasting conclusions. Our assessment using simulated landfill cells indicated that membrane concentrate infiltration increases the organic content of methanogenic leachates. The pollution parameters' median values were higher in leachates drained from cells that operated with concentrate recirculation (i.e., 6729 vs 1367 mg L^{-1} [COD], 33.39 vs 17.39 cm^{-1} [UV_{254}]; p-value < 0.05). The recalcitrant organics' accumulation impacted the RO treatment efficiency by increasing organic fouling onto the membrane surface. In that direction, greener solutions for the leachate membrane concentrate management are recommended to guarantee the long-term sustainability of the leachate treatment chain. In this scenario, the use of leachate concentrate residue in the pyrolysis process of agroindustrial biomass was investigated to produce add-value products. This resource recovery study showed promising results, which could foster more sustainable strategies to close the landfill leachate treatment loop.

Keywords: municipal solid waste, pollution control, resource recovery, reverse osmosis, waste-to-resources

RESUMO

A presente tese explora os aspectos tecnológicos e econômicos dos processos de nanofiltração (NF) e osmose reversa (OI) para tratamento de lixiviados de aterros sanitários. O gerenciamento do concentrado de membrana e questões relacionadas também são analisados. Células experimentais de aterro foram construídas para investigar os impactos da recirculação do concentrado na quantidade, qualidade e desempenho do tratamento do lixiviado. O monitoramento das células foi realizado por 420 dias. Os processos de NF e OI foram capazes de remover >90% da demanda química de oxigênio (DQO), absorbância em 254 nm (UV_{254}) e nitrogênio amoniacal. Os custos operacionais do tratamento do lixiviado variaram de 0,132 a 3,35 USD por m^3 de lixiviado tratado. Em contrapartida, o gerenciamento do concentrado gerado no processo de filtração não é considerado quando as despesas do tratamento são estimadas. Em relação ao concentrado do processo de separação por membrana, esse rejeito é geralmente recirculado para as células de disposição de resíduos do aterro, sem custos adicionais para os gestores do aterro sanitário. No entanto, as consequências dessa prática não são completamente elucidadas e as informações disponíveis na literatura apresentam conclusões divergentes. O monitoramento das células experimentais de aterro indicou que a infiltração do concentrado aumentou o conteúdo orgânico do lixiviado gerado na fase metanogênica de decomposição dos resíduos. Os valores médios dos parâmetros de poluição foram maiores nos lixiviados drenados de células que operaram com recirculação do concentrado (6729 *vs.* 1367 mg L^{-1} [DQO], 33,39 *vs.* 17,39 cm^{-1} [UV_{254}]; p-valor < 0,05). O acúmulo no lixiviado de substâncias orgânicas recalcitrantes afetou o desempenho de tratamento por OI ao intensificar o *fouling* orgânico das membranas. Nesse sentido, estratégias baseadas na recuperação de recursos do concentrado de membrana foram recomendadas para garantir a sustentabilidade da cadeia de tratamento no longo prazo. A utilização do resíduo gerado após a evaporação do concentrado foi investigada no processo de co-pirólise com biomassa agroindustrial. Este estudo mostrou resultados promissores, os quais podem fomentar estratégias mais sustentáveis e contribuir no fechamento do ciclo da cadeia de tratamento de lixiviados de aterros.

Palavras-chaves: controle da poluição, osmose inversa, recuperação de recursos, resíduo sólido urbano, valorização de resíduos

FIGURES

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Chapter 1.   Introduction

This chapter highlights the thesis's motivation by introducing background information and the study's aims. The document structure and a statement of novelty and contribution are presented. The publications from this work are listed at the end of this chapter.

1.1 BACKGROUND

Landfill leachate (LFL) contains a wide range of pollutants of varying concentrations, from high values for organic matter, nutrients, and inorganics to low values for emerging contaminants (e.g., persistent organic pollutants, pharmaceuticals, personal care products, and plastic additives) (BUSCH et al., 2010; CLARKE et al., 2015; RAMAKRISHNAN et al., 2015). Hence, its treatment remains a major socio-environmental and economic issue in the municipal solid waste management chain. Treatment of LFLs must meet the wastewater disposal requirements established by regulatory authorities. Moreover, considering that the leachate physicochemical composition undergoes spatial and temporal variations, successful and efficient treatment must be ensured in both the active and post-closure landfill periods (FAN et al., 2006; STEGMANN, 2018).

The conventional technologies for leachate treatment are biological and physicochemical processes or their combination in integrated or sequential schemes (ABBAS et al., 2009; KURNIAWAN et al., 2010). Conventional biological systems alone cannot significantly treat methanogenic leachates, which contain contaminants resistant to biodegradation (TORRETTA et al., 2016). Additionally, high ammonia concentrations cannot be removed successfully by biological treatment, such as activated sludge, aerated lagoons, sequence batch reactor, or trickling filters (MOJIRI et al., 2021). Therefore, the option of advanced technologies should be considered. In this regard, membrane-based techniques (e.g., nanofiltration [NF] and reverse osmosis [RO]) have been demonstrated to be indispensable for leachate treatment due to their cost-effectiveness (CHEN et al., 2021).

NF and RO are high-pressure-driven membrane filtration processes. Membrane systems generate two output streams, which are named permeate and concentrate. The permeate is the treated water, and the concentrate, also known as retentate, is the residual stream (BAKER, 2012). A body of scientific studies recognizes NF and RO's importance for wastewater treatment, and several works have shown NF/RO treating landfill leachate in pilot and full-scale applications. On the other hand, economic data in the literature are scarce. Furthermore, companies that run leachate

treatment plants do not provide the actual process costs. Therefore, more information on this matter would help landfill managers and stakeholders to make smarter decisions.

Another significant aspect of membrane-based technologies is the management of concentrate streams. Due to its convenience and simplicity, the recirculation of membrane concentrate onto the landfill body is a commonly adopted management strategy in a leachate membrane-based treatment chain (ISWA, 2019). However, the negative experience of concentrate recirculation in some sites started to grow the debate about its long-term sustainability. Literature findings are contrasting in this matter, and research is still needed to elucidate the impacts of the concentrate infiltration practice on the leachate treatment chain.

The present thesis explores the technological and economic aspects of NF and RO treating landfill leachate. Besides, the impacts of concentrate recirculation, using experimental landfill cells, on the quantity, quality, and leachate treatment performance are investigated.

1.2 OBJECTIVES

1.2.1 Main aim

The main aim of this thesis is to evaluate the impacts of the recirculation process of concentrate streams, using experimental landfill cells, on the leachate quantity, quality, and treatment performance.

1.2.2 Goals

i.   Analyse the cost-benefit of NF process and the economic aspects of a RO system treating landfill leachate;

ii. Revise the state-of-art about management practices for NF/RO concentrate streams from landfill leachate treatment plants;

iii. Assess the influence of membrane concentrate infiltration practice, using simulated landfill cells, on the leachate quantity and its pollution parameters;

iv. Evaluate, comparatively, the performance of a bench-scale RO system treating the leachate collected from the experimental landfill cells;

v.   Investigate the co-pyrolytic conversion of leachate concentrate, using a lab-pyrolyser, as an innovative way to recycle minerals and manage membrane concentrates.

1.3 THESIS STRUCTURE

The overall structure of the study takes the form of eight chapters, including this introduction. Chapter two presents a brief literature overview on the main aspects of landfill leachate management and membrane-based technologies (i.e., nanofiltration and reverse osmosis). The third and fourth chapters are concerned with techno-economic aspects of NF in a lab-scale analysis and cost estimation of a full-scale RO facility for leachate treatment, respectively. The fifth chapter presents a state-of-art review about concentrate leachate management options, highlighting perspectives for future studies. Chapter six analysis the results of a 420-day monitoring experiment to assess the impacts of the membrane concentrate infiltration practice. Chapter seven addresses an innovative strategy to handle membrane concentrate streams. This section aims to provide insights into alternative approaches for membrane concentrate management within a circular bioeconomy context. Finally, major conclusions of this thesis are drawn in chapter eight.

The thesis structure based on its organisation and goals is illustrated in Figure 1-1.
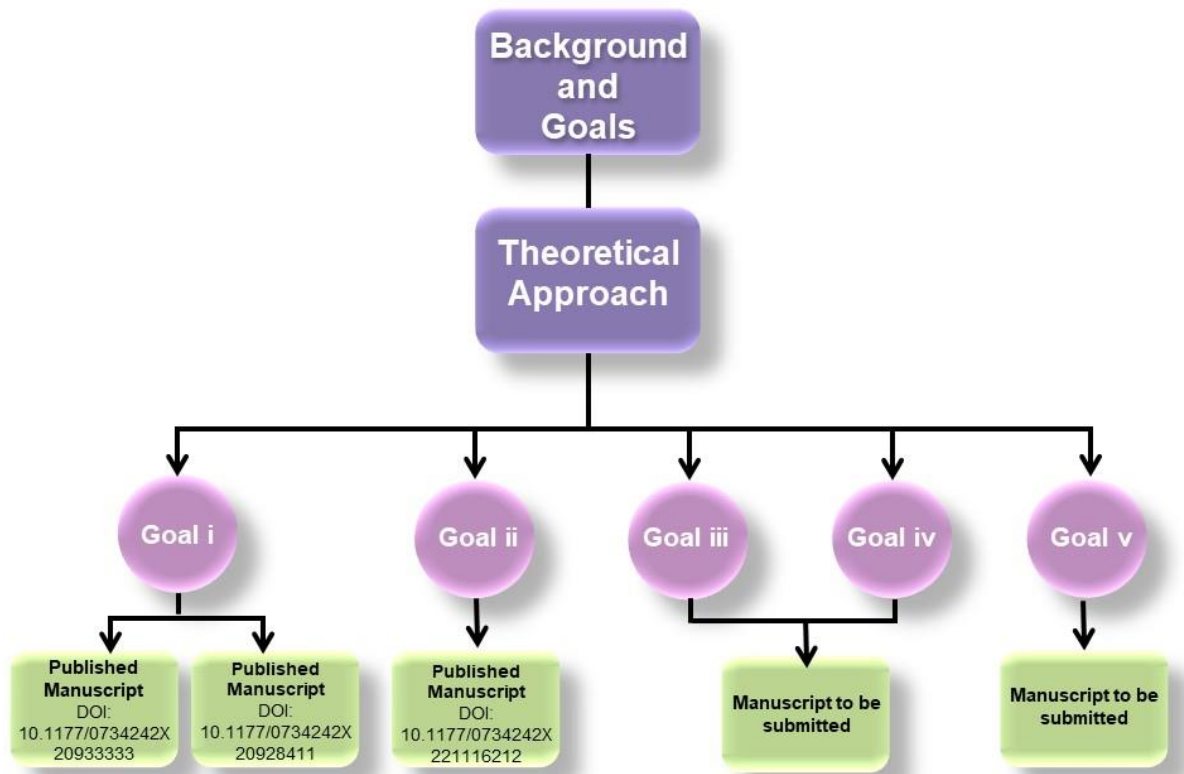
Figure 1-1 Thesis structure.

1.4 STATEMENT OF NOVELTY AND CONTRIBUTION

Membrane-based technologies have been used in several landfill facilities to treat wastewaters efficiently, but concentrate management is by far a critical issue for the leachate treatment chain. The impacts of the main existing management solution, i.e., membrane concentrate infiltration into the landfill, are not comprehensively understood, and the literature's findings show contrasting conclusions.

Firstly, this thesis adds to the current knowledge by investigating the consequences of membrane concentrate recirculation practice for leachate quantity, quality, and treatment performance. Second, to the best of the author's knowledge, a comprehensive literature review about leachate concentrate management focusing on membrane concentrate valorisation is presented for the first time. Finally, an innovative management solution based on the pyrolysis of the membrane concentrate residue is proposed and evaluated in a bench-scale experiment, which provides insights into alternative approaches for managing concentrate streams within a circular bioeconomy scenario.

1.5 PUBLICATIONS

Article papers

[1] **Appendix I.** De Almeida et al. A review on membrane concentrate management from landfill leachate treatment plants: The relevance of resource recovery to close the leachate treatment loop. ***Waste Manag. Res. J. a Sustain*. Circ. Econ**, v. -, 1–22, 2022.

[2] **Appendix II.** De Almeida et al. Nanofiltration applied to the landfill leachate treatment and preliminary cost estimation. ***Waste Manag. Res. J. a Sustain. Circ. Econ***, 38(10), p. 0734242X2093333, 2020.

[3] **Appendix III.** De almeida et al. Cost estimation of landfill leachate treatment by reverse osmosis in a Brazilian landfill. ***Waste Manag. Res. J. a Sustain. Circ. Econ***, 38(10), 0734242X2092841, 2020.

Related works

[4] **Appendix IV.** Technical and economic aspects of a sequential MF + NF + zeolite system treating landfill leachate. ***J. Environ. Sci. Heal. Part A***, 57 (8), 675-684, 2022.

[5] **Appendix V.** Almeida, R. et al. Lessons and challenges for the recycling sector of Brazil from the pandemic outbreak of COVID-19**. *Waste Disposal & Sustainable Energy***, 3,145-154, 2021.

[6] **Appendix VI.** Campos, J. C., De Almeida et al. **Monitoramento dos parâmetros físico-químicos e microbiológicos dos lixiviados de células experimentais de resíduos domiciliares com infiltração do concentrado de osmose inversa**. In: 31º Congresso da Abes, 1–11, 2021.

[7] **Appendix VII.** De Almeida et al. Techno-economic evaluation of landfill leachate treatment by hybrid lime application and nanofiltration process. ***Detritus***, 10, 170-181, 2020.

[8] Almeida, R.; De; Campos, J. C. Análise Tecnoeconômica Do Tratamento de Lixiviado de Aterro Sanitário. **Revista Ineana** 2020, 8(1), 6–27. (Appendix VIII)

[9] **Appendix IX.** Evaluation of coagulation–flocculation and nanofiltration processes in landfill leachate treatment. *J. Environ. Sci. Heal. Part A*, 54 (11), 1091-1098, 2019.

The related thesis works received the following awards: **Appendix X.** INEA Award 2019 (in Portuguese, 1º Prêmio INEA de Meio Ambiente). and **Appendix XI**. Menção Honrosa em Sessão de Apresentação do Centro de Tecnologia [CT] na XLII Jornada Giulio Massarani de Iniciação Científica, Tecnológica, Artística e Cultural [JICTAC 2020 – Edição Especial] [In Portuguese].

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Chapter 2.   Theoretical Approach

This chapter provides a brief background about landfill leachate management and membrane-based technologies (i.e., nanofiltration and reverse osmosis). It presents a literature overview concerning nanofiltration and reverse osmosis applications for leachate treatment. Fundamental principles discussed herein act as a groundwork for experiments and discussion of subsequent chapters.

2.1   LANDFILL LEACHATE MANAGEMENT

The composition of landfill leachate (LFL) varies depending on the gravimetric composition of landfilled waste, landfill age and mode of its operation, landfill status (i.e., active or closed), site conditions (e.g., climate, site geometry, soil properties, and geohydrology), among others (EL-FADEL et al., 2002; FARQUHAR, 1989). The typical LFL contains a significant concentration of biodegradable and non-biodegradable organics, inorganic compounds, and xenobiotic organic compounds (KULIKOWSKA & KLIMIUK, 2008; VACCARI et al., 2019). Recent studies identified other pollutants such as pharmaceuticals, plasticizers, and microplastics in untreated and treated leachate samples from both active and closed landfills (HE et al., 2019; SU et al., 2019). Thus, a considerable amount of literature about this topic can be expected in the forthcoming years. Nevertheless, major concerns of the landfill leachate are ammonia nitrogen (NH_3-N), salts (e.g., chloride, sulphate, carbonate, and bicarbonate), and organic matter – reported as five-day biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), and total organic carbon (TOC) (EHRIG & ROBINSON, 2010; ISKANDER et al., 2018).

According to published studies (COSTA et al., 2019; KURNIAWAN et al., 2010, 2006), landfill age plays an important role in leachate characteristics and; therefore, landfill leachate can be classified into three categories on an age basis: young, intermediate, and mature (Table 2-1). Overall, biodegradable organic matter (evaluated by BOD_5) reduces over time, and leachate organic matter stabilizes (KJELDSEN et al., 2002; LUO et al., 2020). In other words, the BOD_5/COD ratio (biodegradability index) decreases with the landfill age increases. The biodegradability index can even be more diminutive in tropical regions compared to others of temperate climate. The warmer conditions tend to boost microbial activity, which accelerates organic matter stabilization; thus, a high concentration of non-biodegradable compounds such as humic substances can also be a concern in the short term for landfills located in tropical regions (LEBRON et al., 2021). Besides, as earlier stated, the composition of landfilled residues can affect LFL characteristics; for instance, in regions where waste separation, pretreatment, and recycling of organic fraction are

effective, inorganic leachate parameters such as total dissolved solids (TDS), conductivity, and chloride may be more relevant (DE ALMEIDA & CAMPOS, 2020).

Several methods for landfill leachate treatment have been in use, such as co-treatment with sewage on wastewater treatment plants (BRENNAN et al., 2017; DERELI et al., 2021), leachate recirculation into the landfill body (BAE et al., 2019), constructed wetlands (BAKHSHOODEH et al., 2020), physicochemical processes ─ coagulation-flocculation (C/F), chemical precipitation, chemical oxidation, air stripping, carbon adsorption, and AOPs (DENG & ENGLEHARDT, 2006; FERNANDES et al., 2015; FERRAZ et al., 2013; FOO & HAMEED, 2009; LINS et al., 2015), and biological processes ─ aerated lagoons, sequencing batch reactor process, activated sludge process (ASP), membrane bioreactor (MBR), biofilms in rotating biological contactors, and trickling filters (AHMED & LAN, 2012; EL-GOHARY & KAMEL, 2016; ROBINSON, 2019).

Conventional treatments of LFLs are generally classified into three major groups: (1) biological processes (aerobic or anaerobic); (2) physicochemical processes; and (3) a combination of biological and physicochemical processes (Luo et al., 2020). Biological treatment is often used to remove biodegradable organics and total nitrogen due to its reliability, simplicity, and high cost-effectiveness (EHRIG et al., 2018). On the other hand, physical and chemical processes can be effective as a pretreatment for leachate's biological degradation since it helps to reduce the content of non-biodegradable substances, which can compromise the efficiency of the biological treatment (MOJIRI et al., 2021). Figure 2-1 depicts the performance of different leachate treatment processes according to the landfill age, i.e., young, intermediate, and mature.

Table 2-1 Typical landfill leachate physicochemical composition according to the landfill age in temperate and tropical regions.

Parameters	LFL age and leachate composition in temperate regions (yr)			LFL age and leachate composition in tropical regions (yr)		
	0—5 (young)	5—15 (intermediate)	>15 (mature)	0.5—2 (young)	1.7—2.1 (intermediate)	7.2—14.4 (mature)
pH	3—6	6—7.5	7.5—9.0	7.8—8.5	6.2—8.3	7.3—8.4
BOD_5 (mg L^{-1})	10,000—25,000	500—4000	<500	275—453	1—7068	1—12,766
COD (mg L^{-1})	15,000—40,000	1000—20,000	<100	1230—6027	164—17,440	576—21,137
BOD_5/COD	0.5—1	0.1—0.5	<0.1	—	0.006—0.3	0.002—0.3
Biodegradability	medium—high	medium	low	—	low	low
NH_3-N (mg L^{-1})	1500—4250	50—700	<30	526—1787	21.1—1120	133—2808
TDS (mg L^{-1})	10,000—25,000	2000—10,000	<100	—	70—5885	310—3480
Conductivity (mS cm^{-1})	15—41.5	6—14	—	8.90—10.87	0.677—14.59	3.92—25.63
Chloride (mg L^{-1})	1000—3000	100—2000	<100	2499—4204	—	—
Sulphate (mg L^{-1})	500—2000	50—1000	<50	—	—	—

BOD_5: 5-day biochemical oxygen demand. COD: chemical oxygen demand. NH_3-N: ammonia nitrogen. TDS: total dissolved solid.

Source: Adapted from Costa et al. (2019) and Lebron et al. (2021).
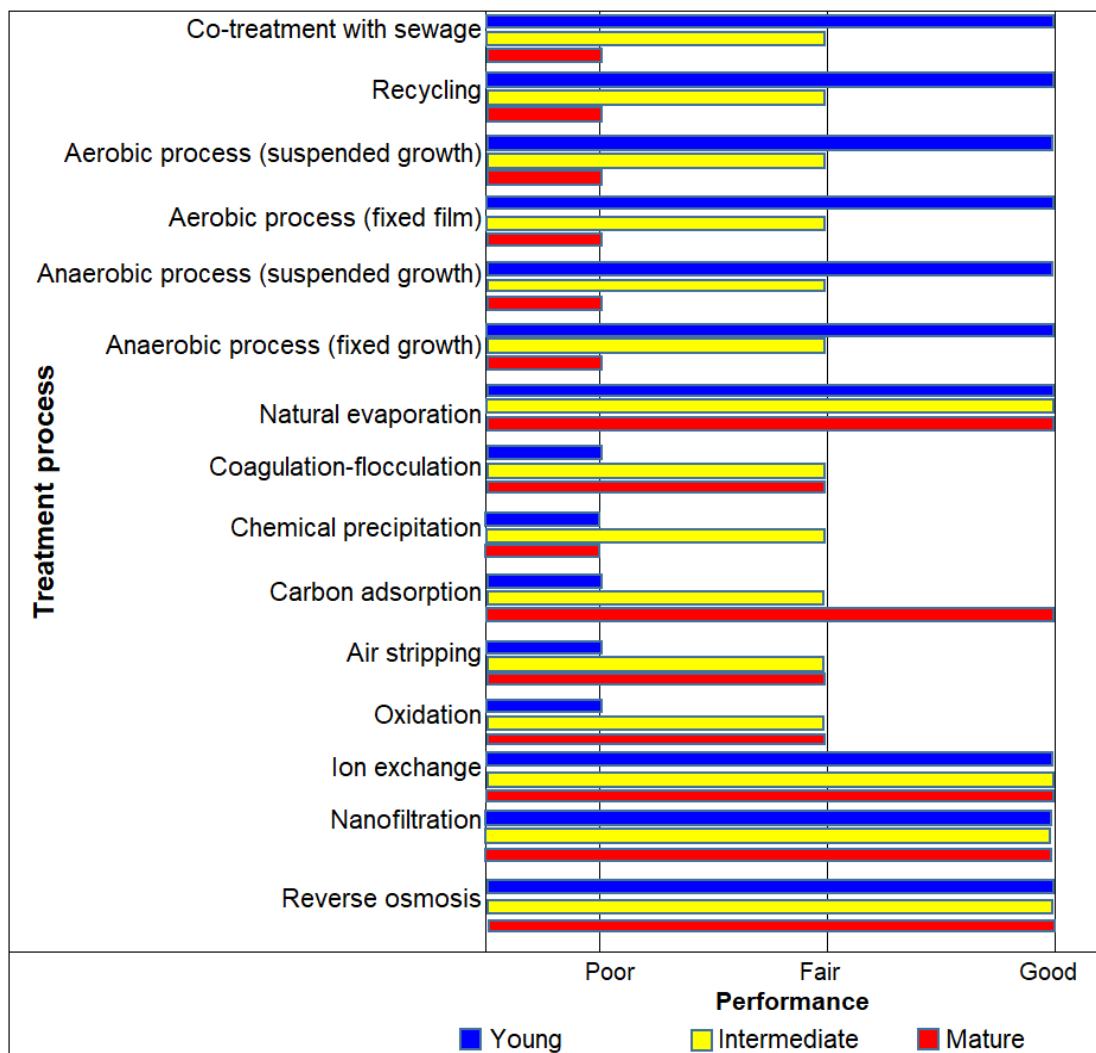
Figure 2-1 Performance of different leachate treatment processes according to the landfill age.
Source: Based on Luo et al. (2020).

As above-mentioned, several technologies are available for LFL treatment, and each of them has its merits and limitations. The selection of the best treatment route depends mainly on the LFL composition and economic feasibility. As leachate composition undergoes spatial and seasonal variation, treatment strategies cannot be stereotyped. Every scenario is unique, and treatment approaches should vary accordingly (MUKHERJEE et al., 2015).

From a techno-economic point of view, NF and RO seem to be the most promising and efficient methods among the existing technologies. NF and RO, either

as a foremost or polishing step in a leachate treatment chain, have shown to be an essential manner to meet the most restrictive standards for water discharge or reuse (CHEN et al., 2021). NF and RO processes can: 1) provide high-quality treated leachate (CHEN et al., 2021); 2) reduce the environmental footprint and size of the landfill leachate treatment plant (LLTP) (modular design/installation) (JAMALY et al., 2014; PETER-VARBANETS et al., 2009); 3) be automated and easily scaled (PETER-VARBANETS et al., 2009)**,** and; 4) be easily moved from site to site (KUMANO & FUJIWARA, 2008). Since the late 1980s, the use of RO has become a proven technology in use for LFL treatment. In 2018, there were over 300 leachate treatment RO plants installed worldwide (BALKEMA et al., 2018). For this reason and considering the scope of the present thesis, the following section focuses on NF/RO processes applied to the treatment of LFLs.

2.2 NANOFILTRATION AND REVERSE OSMOSIS

2.2.1 Fundamental concepts

Nanofiltration (NF) and reverse osmosis (RO) are high-pressure-driven membrane filtration processes. NF/RO systems generate two output streams during their operation: named permeate and concentrate. The permeate is the treated water, and the concentrate, also known as retentate, is the residual stream (Figure 2-2) (BAKER, 2012).
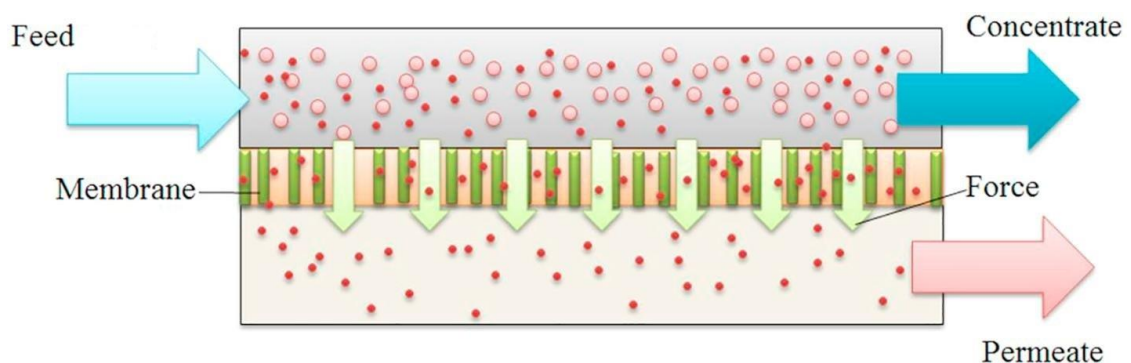
Figure 2-2 Schematic diagram of the membrane filtration process.

Source: Adapted from Ye et al. (2018).

There essential parameters determine the performance of membrane-based processes: solvent permeability or flux through the membrane, rejection of solutes, and yield or recovery of the membrane-based unit. The permeate flux (J [L m^{-2} h^{-1}]) is defined as the permeate flowrate per membrane's permeation area (Equation 2.1), while the membrane permeability (Lp [L m^{-2} h^{-1} bar^{-1}]) is the permeate flux per unit of applied pressure (Equation 2.2).

$$J = \frac{Q_p}{A_m} \tag{2.1}$$

$$L_p = \frac{J}{PTM} \tag{2.2}$$

Where: Qp (L h^{-1}) is permeate flow rate, A$_m$ represents the membrane's permeation area (m^2), and PTM is the transmembrane pressure (bars).

The rejection of solute "i" is defined by Equation 2.2.

$$Rejection\ (\%) = \left(1 - \frac{c_{p,i}}{c_{f,i}}\right) \times 100 \tag{2.3}$$

Where c$_{p,i}$ is the permeate concentration and c$_{f,i}$ is the feed concentration of solute "i"; Rejection is a dimensionless parameter, and its value varies between 100% (complete rejection of the solute) and 0% (solute and solvent pass freely through the membrane) (BAKER, 2012; URAGAMI, 2017).

The recovery of membrane-based system (R [%]) is the ratio of the permeate stream (Q$_p$) to the feed stream flowrate (Q$_f$) (Equation 2.4). It should be noted that the recovery parameter is linked to the design of an industrial application rather than a membrane characteristic (URAGAMI, 2019).

$$R\ (\%) = \frac{Q_p}{Q_f} \times 100 \tag{2.4}$$

The RO process uses a physical mechanism in which the operating pressure must be kept higher than the solution osmotic pressure ($\Delta P > \Delta \pi$) (Figure 2-3). The transport mechanism across the RO membrane follows the dissolution/diffusion model, where both solvent and solute dissolve in the dense surface layer of the membrane and diffuse separately due to the chemical potential gradient of each species. In wastewater treatment systems, RO membranes are primarily used to remove low molecular mass solutes such as salts and heavy metals (BAKER, 2012; WILF, 2014).
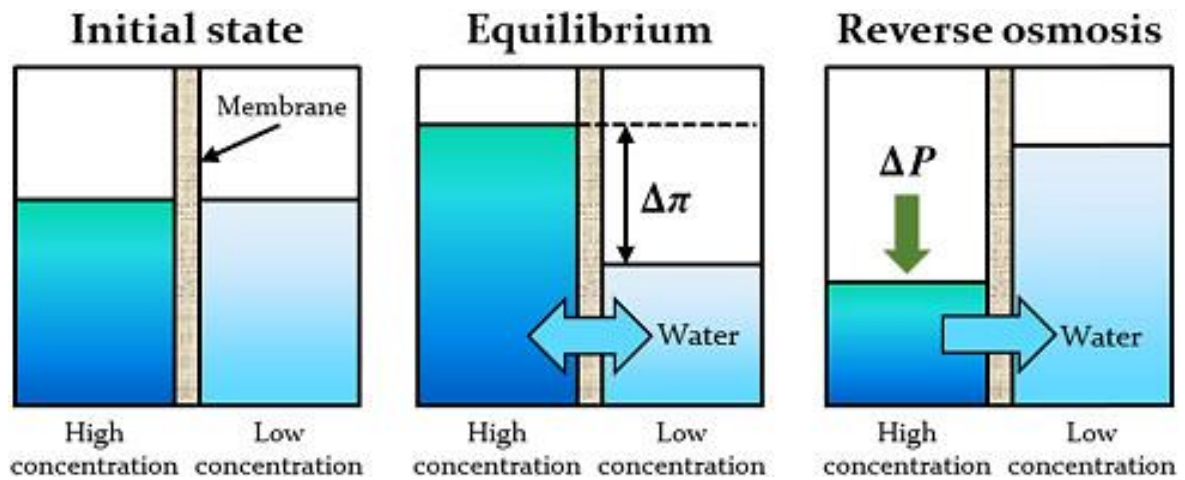
Figure 2-3 Reverse osmosis mechanism principles.

Source: Adapted from Kim et al. (2021.)

NF membranes have pore sizes of 0.5–2 nm, corresponding to a molecular weight cut-off (MWCO) of 100–500 Da. NF is also described as a process that removes particles and dissolved compounds smaller than 2 nm (MOHAMMAD et al., 2015). The principle of NF is very similar to that of RO but there are a just slight difference between RO and NF processes. RO membrane rejects monovalent ion as Na^+ and Cl^- ions and easily permeates water. However, NF membranes do not almost reject monovalent ion, reject multivalent ions such as Ca^{+2} and Mg^{+2} and permeate water. Therefore, a higher driving force in the pressure-driven separation process is required for RO systems but not for the NF process (ABDEL-FATAH, 2018; BAKER, 2012; URAGAMI, 2017). NF and RO systems can separate organic and inorganic compounds from the influent, producing water with low levels of dissolved solids (YANG et al., 2020).

The standard operating mode of full-scale NF/RO systems is cross-flow with high internal flow rate concentrate recirculation. Typical NF/RO systems modules include tubular, spiral wound, hollow fibre, and disc tube (YANG et al., 2020). The higher cost-benefit of the spiral wound module makes it the most used configuration. Spiral wound modules are available in standardized' design, in a range of standard diameters (2.5 ,4 and 8 inches) to fit pressure vessels that can hold several modules in series connected by O-rings. The module uses flat sheet membranes wrapped around a central tube. The membranes are glued along three edges over a permeate spacer to form 'leaves'. The permeate spacer supports the membrane and conducts the permeate to the central permeate tube (Figure 2-4) (MAYNARD & WHAPHAM, 2020; URAGAMI, 2017).
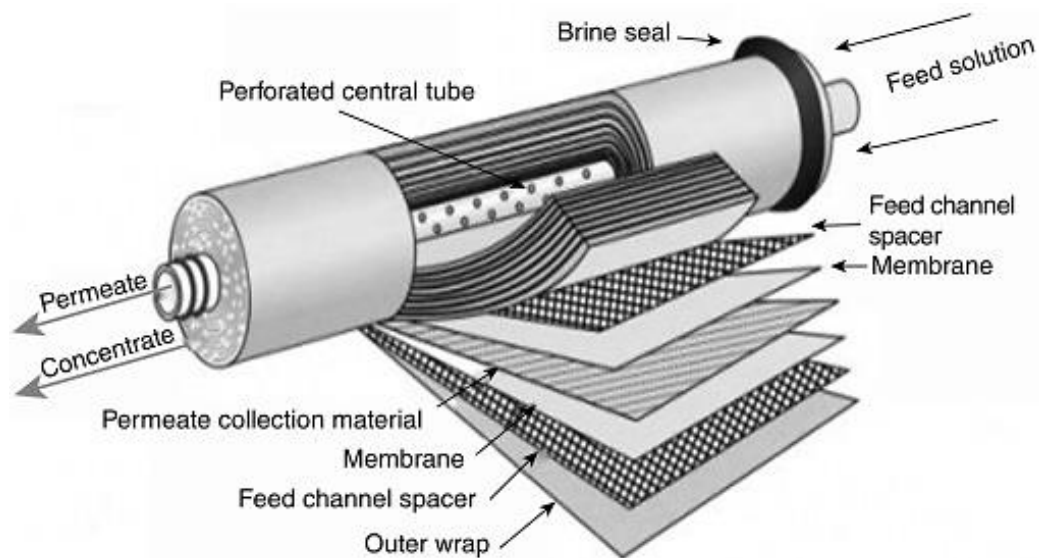
Figure 2-4 Spiral wound module design.
Source: Maynard & Whapham (2020)

Due to their modular configuration, NF/RO modules are available in containers from various suppliers and adapted to the required conditions. NF/RO spiral wound systems are generally used in a stand-alone mode, and depending on the effluent requirements, several steps can be combined, where wastewater gets filtered in two or more steps before final discharge (DE ALMEIDA et al., 2020; ISWA, 2019).

2.2.2 NF/RO processes in the leachate treatment chain

NF and RO have been widely applied in full-scale LFL treatment projects (CHEN et al., 2021; DI MARIA et al., 2018; LEBRON et al., 2021). A body of scientific research recognizes their importance for LFL treatment (ANNA TAŁAŁAJ et al., 2021; DOLAR et al., 2016; MARIAM & NGHIEM, 2010; RAMASWAMI et al., 2018; SMOL & WŁODARCZYK-MAKUŁA, 2017), and several studies have operated NF/RO in pilot and full-scale application (Table 2-2), demonstrating the membrane technology maturity. For example, according to Argun et al. (2020), the NF process used as a final step of an LFL facility located in Turkey is essential to meet the local leachate disposal limits. The NF system is composed of 60 spiral wound modules and polyethersulfone membranes. It operates at pump pressure <18 bar, recovery rate of 85%, and permeate flux of 16 L m^{-2} h^{-1}. COD, NH_3-N, TDS, and colour removal efficiencies are, on average, 84%, 70%, 51%, and >99%, respectively. Several full-scale RO systems are also documented in other studies that looked at the RO process (CINGOLANI et al., 2018; RUKAPAN et al., 2012; THEEPHARAKSAPAN et al., 2011). In Poland, a 72 m^3 d^{-1} LLTP has performed the disc-tube RO system at an operating pressure of 65 bar, permeate flux of up to 50 L m^{-2} h^{-1} and recovery of 75%. Treatability results showed removal efficiencies of BOD_5, COD, and NH_3-N greater than 90% (TALALAJ, 2015).

Table 2-2 NF and RO in the pilot- and full-scale application for landfill leachate treatment.

Treatment scheme	Location	Scale	Module type	Membrane	Operational conditions		Treatability (% removal)		Reference
					Permeate flux (L m^{-2} h^{-1})	Recovery (%)	COD	NH$_3$-N	
NF	India	Pilot	Flat sheet membrane	NF-300	~53–266	~50–80	82.6–88.6	68–70	CHAUDHARI & MURTHY (2010)
Sedimentation + anoxic/aerobic tanks + UF + **NF**	Turkey	Full	Spiral wound	PVDF membrane	17	Data not available	99	>93	YANAN et al. (2012)
MBR + **NF**	Brazil	Pilot	Spiral wound	NF90-2540	5.8–6.9	60	80–96	85–95	AMARAL et al. (2015)
MBR + **NF**	Turkey	Full	Spiral wound	PES membrane	16	85	84	70	ARGUN et al. (2020)
MBR + **NF**	China	Full	Data not available	Data not available	Data not available	Data not available	>80	70	SHAO et al. (2021)
MBR + **RO**	Korea	Full	Spiral wound	SW-4040	Data not available	Data not available	97	96	Ahn et al. (2002)
ASP + flocculation/sedimentation + **two-stage RO**	German	Full	Novel thin open channel spiral wound	FT30	6.5–4.2	70	99.7	99.9	LI et al. (2009)
Two-stage RO	German	Full	Novel thin open channel spiral wound	FT30	6.5–8.1	70	99.5	98.9	LI et al. (2009)
Coagulation + sand filtration + MF + **RO**	Thailand	Full	Spiral wound	LFC3-LD	Data not available	50	87.5	>88	THEEPHARAKSAPAN et al. (2011)
Coagulation + sand filtration + **RO**	Thailand	Full	Spiral wound	LFC3-LD	Data not available	50	>98	Data not available	RUKAPAN et al. (2012)
Two-stage RO	Czech Republic	Pilot	Spiral wound	SW30-4040	42	94	97.3	94.0	ŠÍR et al. (2012)
Sand and cartridge filtration + **two-stage RO**	Romania	Full	Disc-tube	BIO-10058-v2	12–22	Data not available	94–96	>94	ȘCHIOPU et al. (2012)
Buffer tank + **RO**	Poland	Full	Disc-tube	CD9-RO	Up to 50	75	98.8	99.1	TALALAJ (2015)
UF+ three-stage **RO**	Italy	Full	Disc-tube	Gel GPT-BW 30	13–32.5	91–95	92.4–99.2	46.2–95.8	CINGOLANI et al. (2018)
Two-stage RO + Air stripping	China	Full	Disc-tube	Data not available	Data not available	>83%	>99	>99	WU & LI (2021)

ASP: activated sludge process. COD: chemical oxygen demand. MBR: membrane bioreactor. MF: microfiltration. NF: nanofiltration. NH$_3$-N: ammonia nitrogen. PES: polyethersulfone. PVDF: polyvinylidene fluoride. RO: reverse osmosis. UF: ultrafiltration.

2.3   MEMBRANE FOULING AND MANAGEMENT OF CONCENTRATE STREAMS

Two issues are identified as the main shortcomings of implementing membrane filtration processes: 1) membrane fouling, which decreases the permeate flux and water quality, and 2) concentrate stream management. Membrane fouling requires extensive pretreatment or chemical cleaning of the membrane, resulting in a short lifespan of membranes. That is still the bottleneck problem in membrane promotion and application, as fouling can also increase operational costs. Before NF/RO filtration, pretreatment steps are taken to remove suspended solids and colloids and, consequently, prevent fouling and biofilm growth on the membrane surface (JAMALY et al., 2014).

Membrane fouling can be represented by a resistances-in-serie model (Equation 2.5). Resistances in serie represent the theoretical resistance to permeate flux due to a build-up of a layer of dirt or other fouling substances onto the membrane surface (COLLADO et al., 2020).

$$J = \frac{TMP}{\mu R_f} = \frac{TPM}{\mu(R_M + R_{rev.} + R_{irrev.} + R_{irrecov})} \qquad (2.5)$$

Where TMP is the transmembrane pressure (kg m−1 s-2), μ is the dynamic viscosity (kg m s−1), R_f is the total fouling resistance, R_M is the intrinsic membrane resistance and $R_{rev.}$, $R_{irrev.}$ and $R_{irrecov.}$ are the reversible, irreversible and irrecoverable fouling resistances, respectively (all resistances in m^{-1}). In a practical way, reversible fouling is removed by physical cleaning, irreversible fouling is eliminated by chemical cleaning, and irrecoverable fouling refers to those foulants that cannot be removed by any cleaning step (COLLADO et al., 2020; URUGAMI, 2017).

Finally, a challenge must be dealt with is managing membrane concentrate streams. The development of technologies and process breakthroughs in the water desalination field have been helping to tackle the concentrate management issue in other areas. There is a pool of patented technologies to improve the overall feed water recovery, aiming to reduce concentrate volume and achieve minimal liquid discharge (MLD) or zero liquid discharge (ZLD) (JOO & TANSEL, 2015; SUBRAMANI &

JACANGELO, 2014). However, their high costs linked mainly to energy requirements limit the implementation of full-scale ZLD systems. Furthermore, it is important to note that MLD/ZLD systems are associated with unintended environmental impacts as a result of their high energy demand and carbon footprint. Other major issues of MLD/ZLD systems include fouling, scaling, and expensive metallic materials (VOUTCHKOV & KAISER, 2020). Therefore, future research to addresses these drawbacks are needed. The membrane concentrate management from leachate treatment plants is comprehensively reviewed in Chapter 5 of this thesis.

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Chapter 3.   Nanofiltration Applied to Landfill Leachate Treatment and Preliminary Cost Estimation

The cost-benefit of the nanofiltration process treating landfill leachate is analysed in this chapter. A limited number of studies in the existing literature examine nanofiltration performance based on a cost-benefit perspective. The current chapter provides an overlook of techno-economic aspects of the nanofiltration technology for landfill leachate treatment. Nanofiltration was performed using a bench-scale filtration module with commercial polymeric membranes. At the end of each filtration, a cleaning protocol was applied to recover the initial membrane permeability. The concentration of recalcitrant compounds, expressed as humic substances, was reduced from 821 ± 86 to 84 ± 8 mg L^{-1} in the permeate, and chemical oxygen demand complied with the wastewater discharge standards imposed by local legislation. The capital cost for a full-scale installation was estimated at MUS$ 0.772, and the specific total cost, treated leachate per volume unit, was estimated at US$ 8.26 m^{-3}.

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3.1 INTRODUCTION

Landfill leachate treatment is expensive, multi-processed, and different from each other due to the complex composition, high loading, and seasonal variation (LIU et al., 2015). Several technologies and treatment schemes are available for their treatment (DE ALMEIDA et al., 2019; DI MARIA et al., 2018; RENOU et al., 2008; TALALAJ et al., 2019): (1) co-treatment of sewage and leachate on the site or transfer to the central wastewater treatment plant; (2) recirculation into the bioreactor landfill body; (3) physicochemical treatment (coagulation-flocculation, precipitation, chemical oxidation, activated carbon adsorption); (4) biological processes (anaerobic treatment, aerated lagoons, activated sludge plants, membrane bioreactor (MBR), biofilms in rotating biologic contractors, and trickling filters); and (5) membrane processes (main nanofiltration and reverse osmosis). Despite this, to provide a treated effluent that can be discharged into natural water streams or reused, one single treatment technology is not available and a combination of treatment technologies is essential.

The selection of the best treatment scheme depends on the characteristics of the leachate, extent of treatment required, and economic parameters. Reported studies have shown that although biological processes are widely applied for leachate treatment (FERNANDES et al., 2017), this is not sufficient in reaching the existing discharge standards due to the recalcitrance, i.e., the presence of non-biodegradable and/or low biodegradable compounds (CAMPOS et al., 2013; TALALAJ et al., 2019), characteristic of the mature landfill leachate.

The refractory organic is mainly humic substances (HS). According to Lima et al. (2017), HS can increase the solubility of hydrophobic compounds and solubilize metals such as Ni, Cd, and Zn by complexation, modifying the bioavailability and biotoxicity of landfill pollutants. The presence of the refractory organic matter in the leachate illustrates the complexity of this type of wastewater, limiting the efficiency of biological processes. Thus, for the removal of these compounds, advanced treatments such as membrane filtration are required.

Leachate treatment by membrane process has received much attention over the last two decades. Typical membrane technologies include reverse osmosis (RO),

nanofiltration (NF), and ultrafiltration (UF) (BAKER, 2012). Separation efficiency depends on the type and molecular size of leachate contaminants, the membranes used, and the operational pressure (ZIREHPOUR & RAHIMPOUR, 2016).

Yao (2013) points out that NF technology is capable of achieving several objectives regarding leachate treatment, e.g., control of organic, inorganic, and microbial contaminants. Comparatively, the advantage of using an NF membrane is the request for lower operating pressures, and has higher fluxes than RO membranes, better retention than a UF membrane, high rejection of polyvalent ions and organic matter with a molecular weight of 200-1000 Da, and has relatively low operating and maintenance costs (YAO, 2013). Moreover, due to its unique properties as compared to UF and RO membranes, the NF membrane has a significant advantage that is the capability to remove recalcitrant organic matter from landfill leachate (CHAUDHARI & MURTHY, 2010).

Fernandes et al. (2017) stressed that most of the conventional biological and physicochemical technologies implemented at the landfill facilities are not capable to treat leachates efficiently and are currently being replaced by/or complemented with membrane technologies. Also, Brazilian researchers have highlighted the need to use efficient and economically viable technologies for the treatment of leachate in Brazilian landfills (COSTA et al., 2019; DE ALMEIDA et al., 2019). However, to the best of our knowledge, the most advanced leachate treatment technologies are not feasible in most of the municipalities due to, for instance, the high cost of implementation and maintenance, and the volumes of leachate to be treated. Consequently, municipalities are required to implement treatment processes that are incompatible with the characteristics of the leachate, resulting in treated leachate on disagreement with established disposal legislation.

Previous studies focused on the investigation of the technical feasibility of NF. However, there are a limited number of works that examined leachate treatment costs. Also, companies that run leachate treatment plants do not provide this data. Within this context, to cover this knowledge gap, the purpose of this work was to assess the

efficiency of the NF process and estimate the costs involved in a full-scale NF plant used as a final treatment for landfill leachate.

3.2 MATERIALS AND METHODS

3.2.1 Sample characterization

This study has used the leachate pre-treated by physicochemical process from the Seropédica landfill, located in Rio de Janeiro State, Brazil. The samples used were obtained from a previous bench-scale investigation (DE ALMEIDA et al., 2020).

The Seropédica landfill activities started in April 2012 and permitted Gramacho dumpsite closure, which had been the largest dumpsite in operation in Latin American from 1976 to 2012. Seropédica landfill receives over 10,000 tons of waste per day. It operates with the reception, treatment, and final disposal of solid waste from non-hazardous industry activities and municipal solid waste (MSW). The physical composition of MSW shows that, as expected for low- and middle-income countries, organic matter is the main fraction (> 50%). The landfill has an on-site facility with leachate treatment flow rate up to 1,000 m^3 d^{-1}. The plant comprises air stripping and biological processes (aerobic-anoxic conditions) (ALMEIDA, 2018).

The biochemical oxygen demand (BOD_5) was evaluated by the respirometric method (method 5210-B) from the standard methods (APHA/ AWWA/ WEF, 2012). Chemical oxygen demand (COD) (method 5220-D) and true color (method 2120-C) also was determined according to standard procedures (APHA/ AWWA/ WEF, 2012) using a DR2800™ portable spectrophotometer and reactor Hach (DRB200). Before true color determination, samples were filtered through 0.45 μm cellulose membrane filters.

Ultraviolet absorbance at 254 nm (UV_{254}) was measured using a Shimadzu UVmini-1240. The conductivity was determined using a conductivity meter MS Tecnopon mCA 150. Turbidity analysis was performed with a turbidity meter Poli Control-Ap2000 and pH was measured using a pH meter Quimis (Q400AS). Chloride (Cl^-) concentration was determined by ion chromatography, using a Metrohm system

with a Metrosep A Supp 5 - 150/4.0 conductivity detector equipped with an anion column. The mobile phase was an aqueous solution of sodium carbonate and sodium bicarbonate, a flow rate of 0.7 mL. min^{-1}, with 40ºC column temperature. Ammonia nitrogen (N-NH$_3$) was measured using an Orion Star^{TM} Thermo pH/ISE Portable Multiparameter Meter following the American standard methods (method 4500-E) (APHA, 2012). Humic substances (HS) were determined by the modified spectrophotometric method. The procedure used is as described by Sheng et al. (2007).

3.2.2 Nanofiltration process

3.2.2.1 Experimental setup

NF was performed using a bench-scale filtration module. A schematic of the experimental setup used in the present study is shown in Figure 3-1.

The experimental system consisted of a feed tank, a membrane module, two pressure gauge, a recirculation pump (B-01), flowmeters (FI-01 and FI-02), flow control valves in the feed (V-1), permeate (V-3, V-4) and concentrate streams (V-2). The system has a capacity of 5 L, an effective circular membrane area of 77.7 cm^2, the material of construction of 316 stainless steel cells (PAM Selective Membranes Inc.).

The leachate was tested with two polymeric commercial membranes (SR100 and NP030). SR100 membrane was supplied by Koch Membrane Systems and NP030 was supplied by Lenntech. According to manufactures, these membranes are polyamide thin-film composite and polyethersulfone thin-film composite, respectively. These polymers due to their chemical resistance and mechanical stability are the dominant NF membrane materials used at an industrial level for wastewater treatment (Baker, 2012). Based on this and practical experiences, we selected them. The membranes were compacted for 2 h using distilled water at 2 bars. The characteristics of the membranes supplied by manufacturers are presented in Table 3-1.
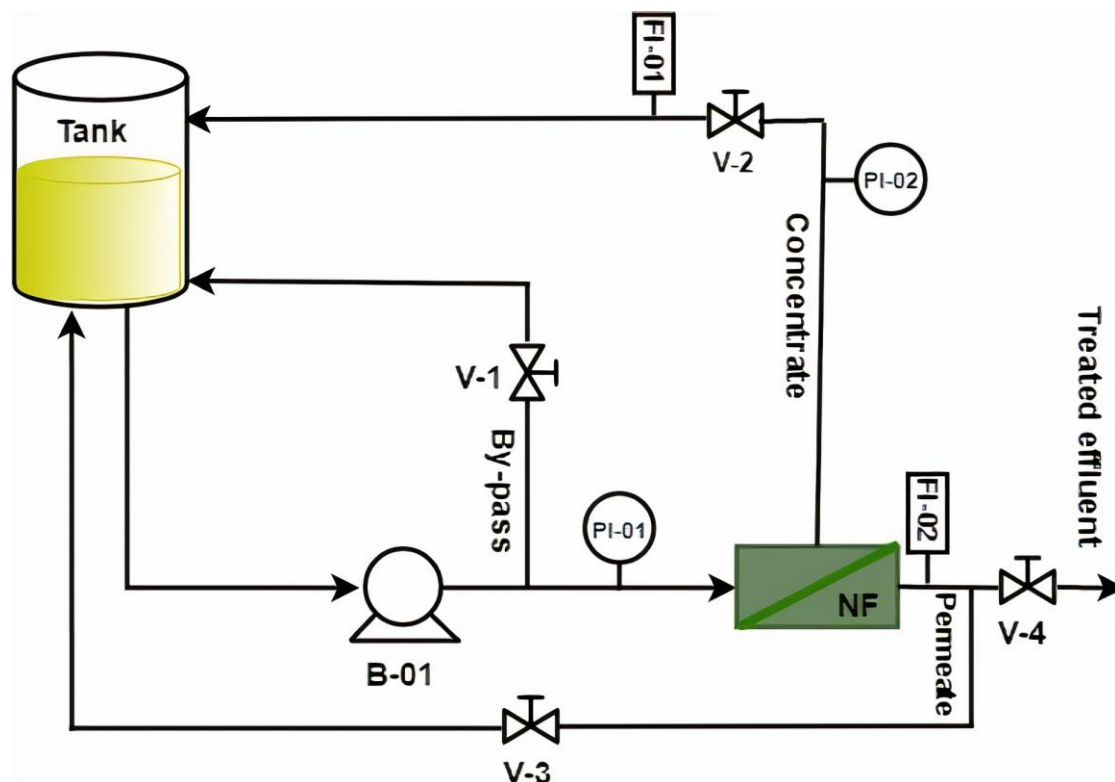
Figure 3-1 Schematic diagram of the nanofiltration experimental setup.

B-01: Pump (feed); PI-01:Pressure gauge (feed); PI-02: Pressure gauge (concentrate); V-01:Control valve (by-pass); V-02: Control valve (concentrate); V-03: Control valve (permeate); V-04: Control valve (permeate); FI-01: Flowmeter (concentrate); FI-02: Flowmeter (permeate); M: Nanofiltration membrane.

Table 3-1 Characteristics of polymeric membranes used in this study.

	Membrane SR100	Membrane NP030
Membrane chemistry	Polyamide	Polyethersulfone
Molecular weight cut-off (Da)	200	400
Hydraulic permeability ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)	1.2-2.5	1.5-3.5
Allowable pH (continuous operation)	4-10	0-14
Rejection percent	> 99[1]	80-95[2]

(1) Test Conditions: 5.000 mg L^{-1} $MgSO_4$ in deionized water at 6.6 bar applied pressure, 15% recovery, 25°C, pH 7.5.
(2) Test Conditions: 2000 mg L^{-1} Na_2SO_4, 40 bar, 20 °C, stirred cell 700 rpm.

The membrane system was operated with constant transmembrane pressure (6, 7, 8 and 9 bar) at 120 L h^{-1} cross-flow rate. The permeate flux (flow rate per unit membrane area, L m^{-2} h^{-1}) was monitored and the best operating pressure was

defined. Under ideal pressure conditions, the NF system was fed with 3 litres of pre-treated leachate.

The NF unit was conducted in the concentration mode of filtration, which means the permeate is discharged or collected while the concentrate is recycled to the feed tank. The volume reduction factor (VRF) of the NF was VRF = 2.5. The volume reduction factor is expressed by the following definition: $VRF = V_f /(V_f - V_p)$, where V_f and V_p are the initial volumes of the feed and permeate collected, respectively.

During the filtration, the valves V-2 and V-4 remained opened, valve V-3 was closed, and the feed flow rate and the pressure were controlled by the frequency inverter connected to the pump B-01 and through the valve V-1 (by-pass). At the end of the NF, samples of permeate were collected to be analyzed. The permeate was stored at 4°C.

3.2.2.2   Membrane regeneration

At the end of each experiment, the membranes were regenerated to recover its initial permeability. The system was water washed before the chemical cleaning procedure, which involved the use of an aqueous solution composed of 3 mol L^{-1} of sulfuric acid, followed by membrane soaking in a 1.5-2.5% aqueous solution of sodium dodecylbenzene sulfonate (SDBS). The sulfuric acid solution was recirculated for 2 hours with the permeate line closed. Thereafter, the membrane was submerged for 24 hours on SDBS solution. Once this process was ended, the system was flushed with distilled water to eliminate the cleaning solution traces. All the experiments were performed in triplicate.

3.2.3 Cost estimation

The cost estimation was performed based on the results obtained in the tests infiltration module and extrapolated to a full-scale NF plant with 1,000 $m^3 d^{-1}$ feed flow

rate, being represented, in this work, by the capital costs (CAPEX), by the operational costs (OPEX) and specific total cost (TC) (cost per unit of permeate, US$ m^{-3}).

For preliminary estimation of the expenses of NF-full scale, the following considerations were made: (i) the leachate treatment plant would operate 365 days a year and would be out of operation only during periods of routine maintenance, chemical cleaning and integrity testing (GUERRA & PELLEGRINO 2012); (ii) the NF process would operate with 60% recovery (R = 60%); (iii) the membrane modules used would be 40 m^2 in the working area (BAKER, 2012), and was considered the value of the square meter of the polymer membrane of US$ 40.00 (BAKER, 2012); (iv) The full-scale NF permeate flow (J = 12 L m^{-2} h^{-1}) was based on manufacturer data; (v) final disposal of the concentrate generated in the process was not considered since this waste is disposed of the landfill itself without additional costs for the leachate treatment plant.

3.2.3.1 CAPEX

CAPEX was determined by adding up the acquisition costs of the membrane modules and housing, pumps, valves, pipes, instrumentation that constitute a permeation unit, and start-up (SALEHI et al., 2014; SINGH & CHERYAN, 1998). Membrane and housings acquisition costs are approximately 25-35% of the total investment value (SALEHI et al., 2014). Most membrane modules are provided with their housing. The cost of membranes and housings was calculated from the square meter cost of membranes (GUERRA & PELLEGRINO, 2012) (Equation 3.1).

$$C_m = C_{mem/m^2} \times A_m \tag{3.1}$$

Where: C_{mem} (US$): cost of membranes; $C_{mem/m2}$ (US$ m^{-2}): cost related to 1 m^2 of membrane; A_m (m^2): total permeation area.

The membrane area required for the operation of the NF-plant depends on the design permeate flow rate, and permeate flux of polymeric membrane used (Equation 3.2). On the other hand, to obtain Equation 3.2, it is necessary to estimate the effective

permeate flow rate (Equation 3.3), system uptime ratio (Equation 3.4), and system downtime (Equation 3.5).

$$A_m = \frac{Q_e}{J} \qquad (3.2)$$

$$Q_e = \frac{Q_p}{R_{op}} \qquad (3.3)$$

$$R_{op} = \frac{(24 - t_{inop})}{24} \qquad (3.4)$$

$$t_{inop} = t_{mr} + t_{ti} + t_{lq} \qquad (3.5)$$

Where: Q_e (L h^{-1}): effective permeate flow rate; J (L m^{-2} h^{-1}): permeate flux; Q_p (L h^{-1}): projected permeate flow rate; R_{op}: system uptime ratio (h h^{-1}); t_{inop} (h day^{-1}): system downtime; t_{mr} (10 min day^{-1}): routine maintenance time; t_{ti} (20 min day^{-1}): time for membrane integrity testing; t_{lq} (h day^{-1}): time to perform chemical cleaning of the membranes (50 cleanings of 6 hours per year). The routine maintenance time and membrane integrity test are estimated in hours per day.

For a cost estimate regarding pumps, the total cost was estimated by multiplying the pump cost by the number of modules in the plant (Salehi et al., 2014). The peripheral price that includes costs with valves, pipes, and instrumentation was estimated at 20% of the membrane cost (Guerra and Pellegrino, 2012). The startup cost, which is the amount of capital required to start the operation, corresponding to 8% of fixed investment, was also considered as investment cost (Amaral et al., 2016).

3.2.3.2 OPEX

For the composition of OPEX, the costs of energy consumption for the operation of the NF system, investment depreciation, membrane exchange, maintenance, hand labor, and membrane cleaning were included (SINGH & CHERYAN, 1998). The energy required to operate the full-scale NF-unit was estimated at 25% of the

membrane costs (SINGH & CHERYAN, 1998). A percentage of 5% of the initial investment associated with preventive and corrective maintenance of the membrane was considered as maintenance cost. As an estimated cost of chemicals used to clean the membranes, a value of 2% of the initial investment. Investment depreciation is typically considered for a period from 7 to 14 years (SALEHI et al., 2014). The membrane costs are not included in this estimate and are considered an independent operating expense. The depreciation was calculated by subtracting the membrane cost from the capital cost, divided by the period considered in the calculation. Guerra & Pellegrino (2012) estimated the cost related to hand labour at approximately 2% of the cost of capital, an amount adopted in this work.

3.2.3.3 Total cost

The TC of treated leachate was obtained by Equation 3.6, which accounts for the OPEX normalized by the annual volume of permeate and the CAPEX normalized by volume of permeate added annually to the time, in years, of operation of the full-scale NF-plant, determined by Equation 3.7.

$$TC = \frac{OPEX}{V_t} \qquad (3.6)$$

$$R_{CAPEX} = \frac{CAPEX}{V_t \times n} \qquad (3.7)$$

Where, R_{CAPEX}: normalized capital cost per volume of treated effluent (US$ m^{-3}); n: the operating period of the leachate treatment plant considered in years; V_t: total volume of treated effluent (m^3).

3.3 RESULTS AND DISCUSSION

3.3.1 Effluent characterisation

Table 3-2 presents the characterization of the pre-treated leachate samples used for this work and the discharge limits established in Brazilian legislation.

Table 3-2 Parameters obtained in the characterization of the pre-treated used in this study.

Parameters	Number of samples	Min	Max	Average±SD	Brazilian legislation
pH	12	10.7	11.5	11.1±0.1	5-9
COD (mg L^{-1})	10	2116	2368	2258±230	250
HS (mg L^{-1})	6	782	866	821±86	-
UV$_{254}$ (cm^{-1})	10	12.98	15.38	14.25±1.36	-
NH$_3$-N (mg L^{-1})	8	10.9	18.5	14.8±1	20
True color (mg Pt-Co L^{-1})	8	1120	1430	1290±144	-
Cl$^-$ (mg L^{-1})	3	523	632	585±73	-
Condutivity (mS cm^{-1})	10	12.48	12.57	13±1	-
Turbidity (NTU)	9	36	45	39±4	-
BOD$_5$ (mg L^{-1})	3	698	773	736±70	-

SD: Standard Deviation.

The average ammonia nitrogen concentration found was 14.8±1 mg L^{-1}, which indicated that pre-treated leachate is following the standards of disposal of Brazilian law, Conama #430/2011. However, the samples show some typical properties of mature leachate because of the substantial concentration of organic matter (COD, 2258±230 mg L^{-1}). Likewise, BOD$_5$/COD (0.21) has indicated low biodegradability. Besides, the average concentration of HS was 821±86 mg L^{-1} and UV$_{254}$, which indicates the aromatic organic matter contents, varied from 13 to 15 cm^{-1}. Also, all the samples were characterized by a very high content in dissolved salts, notably chlorides (585±73 mg L^{-1}) and conductivity (13±1 mS cm^{-1}). These characteristics fits well with Costa et al. (2019) and also confirms our earlier results (DE ALMEIDA et al., 2020).

Alfaia et al. (2017) pointed out that about 51.4% of the municipal solid waste deposited in Brazilian landfills is composed of organic matter. This may be the reason for the high organic content of the leachate from this site. On the other hand, the treatability of landfill leachate depends on its composition and characteristics. Thus, leachate characteristics such as organic matter (COD, BOD$_5$/COD ratio, HS) are

determinants for the selection of suitable treatment.

In our view, the physicochemical and biological leachate treatment processes applied at this site are incompatible with the characteristics of the leachate, resulting in treated leachate on disagreement with Brazilian legislation. Therefore, a further treatment process is needed. In this respect, NF represents a sound technology to complement leachate treatment because it has the advantages of providing a high permeate flux at low operating pressure and maintaining a high organic matter rejection rate (CHAUDHARI & MURTHY, 2010; DE ALMEIDA et al., 2019).

3.3.2   Nanofiltration process

3.3.2.1   Flux and regeneration

The permeate flux was assessed throughout the process (400 min), returning the permeate stream to the feed tank. After 250 min of operation, the process was stopped, and the membrane regeneration protocol described above was applied, then the filtration process was returned for a further 150 min to assess the permeate flux recovery. The results are shown in Figure 3-2.

By the analysis of the permeate fluxes obtained during the filtration process of the membranes SR100 and NP030 (250 min), it was verified that the permeated flux, in the four pressures evaluated, was higher in the NP030. This can be explained by comparing the molecular weight cut-off of membranes (Table 3-1). As a reference, Mariam & Nghiem (2010) performed the NF process for the treatment of pre-treated leachate collected from Whytes Gully landfill (Australia) using commercial membranes NF270 and SR2. The authors also reported similar results.
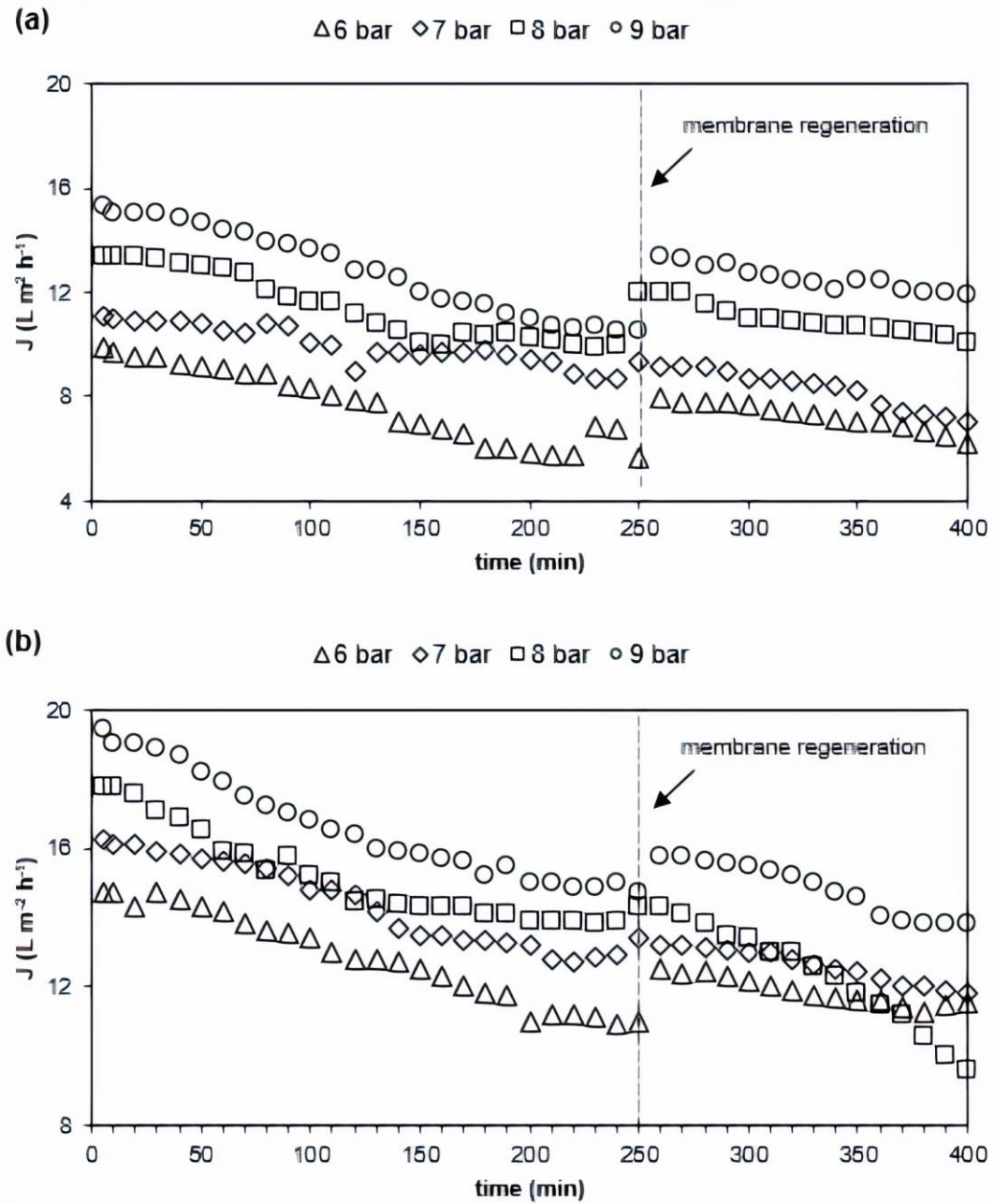
Figure 3-2 Monitoring of the permeate flux during the nanofiltration process with membranes SR100 (a) and NP030 (b), at 6, 7, 8, and 9 bar operational pressure.

At the pressure of 6 bar, in the cross-flow rate of 120 L. h^{-1} (250 min operation), the permeate flux of the SR100 membrane ranged from 9.9 to 5.7 L m^{-2} h^{-1}, whereas for the NP030 membrane, this value was 14.8 to 11.0 L m^{-2} h^{-1}. At higher pressure (9 bar), the range of values was 15.3 - 10.6 L m^{-2} h^{-1} (SR100) and 19.4-14.7 L m^{-2} h^{-1} (NP030). Nonetheless, the operating pressure of 8 bar was defined as the optimum operating condition because it had more stable permeate flow values during the

filtration (Figure 3-2) and due to the technical limitations of the bench module. At 8 bars of pressure, permeate fluxes of the SR100 and NP030 decreased from 13.9 L m^{-2} h^{-1} to 11.1 L m^{-2} h^{-1} after 250 min of operation and from 17.9 L m^{-2} h^{-1} to 15.1 L m^{-2} h^{-1}, respectively.

It is observed that in the nanofiltration, the permeate flux, at constant pressure, decreases continuously. Sir et al. (2012) point out that the continuous decrease of the permeate flux along the leachate treatment by membrane may be associated with the adsorption of humic and fulvic acids on the surface of the membrane, which can cause fouling and lead to shallow permeate flux, making the process unfeasible. Jiang et al. (2017) state that organic fouling by humic acids is mainly due to hydrophobic interactions between organic matters and membrane surface. From this point of view, due to the high content of recalcitrant substances, organic fouling plays a major role in membrane filtration of landfill leachate.

By the analysis of the permeate fluxes obtained during the nanofiltration process was observed that the NP030 membrane is more susceptible to fouling. As mentioned above, the leachate treatment was tested with two polymeric commercial SR100 and NP030 membranes. The former has a hydrophilic surface and the latter has a hydrophobic surface. Fouling in membrane systems is closely related to membrane material (ABUABDOU et al., 2020). According to Jiang et al. (2017), membranes with smooth and hydrophilic surfaces demonstrated less fouling tendency than those with rough and hydrophobic surfaces. Therefore, SR100 is expected to have the best anti-fouling performance.

As shown in Figure 3-2, the membrane cleaning protocol applied in this evaluation was able to recover, for both membranes, approximately 80% of the permeate flux. After regeneration of the membranes and 150 min of operation, the drop in permeate flux values was about 25%. Zirehpour & Rahimpour (2016) point out that NF and RO membranes have to be, once a week, cleaned with special cleaning agents, and in this case, their service lives could be estimated at 1.5 years. Since a variety of contaminants of the leachate, the membrane cleaning is a complex subject, indicating the characteristics of the contaminated sediment membrane, for the

selection of the most economical and effective cleaning agents and cleaning solution is essential (GUO et al., 2012).

3.3.2.2   Effluent treatment

The concentration of HS was considerably reduced, and their final value corresponded to a reduction of 90% and 87% of the level present in the pre-treated leachate, given the filtration process with membranes SR100 and NP030, respectively. Regarding COD, the concentration was reduced from 2258 ± 230 mg L^{-1} in the pre-treated effluent to 193 ± 20 mg L^{-1} (SR100) and 225 ± 20 mg L^{-1} (NP030) in the permeate. The COD values decreased to below 250 m L^{-1} established by local legislation as wastewater discharge standard.

The results of the physicochemical analysis are summarized in Table 3-3. NF was very effective at removing all pollutants. The reduction rate of UV_{254} was greater than 75%. BOD_5 was eliminated by more than 98%, while over 90% of true color was reduced. The turbidity was reduced from 39 ± 4 NTU to 0.40 ± 0.05 NTU in the permeate, which corresponds to 99% of removal.

Comparatively, with the NF270 and SR2 membranes, Mariam and Nghiem (2010) obtained a reduction percentage of organic matter (reported as total organic carbon, TOC) ranging from 89 to 93%. In their study, the decline in permeate flux ranged from 14 to 60%. Similarly, organic fouling was the key factor for the decline in permeate stream. In our previous research, we obtained 89% TOC removal and 30% permeate flux decline (DE ALMEIDA et al., 2019).

Table 3-3 Physicochemical parameters of pre-treated leachate and effluent after the application of the nanofiltration.

Parameters	Number of samples	Min	Max	Average±SD	Number of samples	Min	Max	Average±SD	Min	Max	Average±SD
	Pre-treated leachate				**Nanofiltration**						
						SR100			**NP030**		
pH	12	10.7	11.5	11.1±0.1	10	7.8	8.3	8.0±0.1	7.9	8.2	8.1±0.1
COD (mg L^{-1})	10	2116	2368	2258±230	10	174	205	193±20	220	278	225±20
HS (mg L^{-1})	6	782	866	821±86	5	75	98	84±8	95	121	109±10
UV$_{254}$ (cm^{-1})	10	12.98	15.38	14.25±1.36	8	2.46	3.44	2.88±0.25	2.55	3.07	2.87±0.50
NH$_3$-N (mg L^{-1})	8	10.9	18.5	14.8±1	5	7.9	10.2	8.9±1	5.6	12.5	9.4±1
True color (mg Pt-Co L^{-1})	8	1120	1340	1290±144	8	67	97	83±7	110	135	122±13
Cl$^-$ (mg L^{-1})	3	523	632	585±73	3	225	371	298±33	302	343	318±30
Conductivity (mS cm^{-1})	10	12.48	12.57	13±1	10	6.34	7.21	6±1	9.00	9.32	8±1
Turbidity (NTU)	10	36	45	39±4	10	0.25	0.56	0.40±0.05	0.50	1.10	0.77±0.05
BOD$_5$ (mg L^{-1})	3	698	773	736±70	3	5	9	7±1	7	12	11±1

SD: Standard Deviation.

In the current study, additional removals of Cl^- (>45%) and conductivity (>35%) were obtained. A similar result was achieved by De Almeida et al. (2019) with nanofiltration membrane SB90 (molecular weight cut-off range, 500 - 700 Da). Claudhari & Murthy (2010) observed 62 and 65% rejection of chloride from two leachate samples with NF. On the other hand, the maximum chloride rejection was 14% in a study performed by Trebouet et al. (2001). As also reported by some other studies (DE ALMEIDA et al., 2019; SAMHABER & NGUYEN, 2014), monovalent ions like Na^+ and Cl^-, which are present in significant amounts in landfill leachates, can permeate an NF membrane. Baker (2012) points out that chloride is a monovalent anion, and the rejection of this type of ion is not characteristic of the NF membrane. However, its replacement can occur due to the precipitation of Cl^- in the membrane or even by the transport of ions, to maintain the membrane electroneutrality. In complex aqueous matrices, such as leachate, the presence of a wide variety of ions also presents a wide range of complex interactions, making it difficult to predict the efficient removal of monovalent contaminants.

According to Claudhari & Murthy (2010), for Cl^-, rejection is lower at acid pH values because the protons can neutralize the negative sites on the membrane surface, then chloride ions can pass easily through the membrane. Kim et al. (2006) evaluated the effect of pH on the rejection of inorganic salts using NF membrane and the rejection of Cl^- increased with an increase in the feed pH. In the present study, possible the pH of the effluent (11.1±0.1) contributed to the rejection values obtained. In a more recent study, Amaral et al. (2015) obtained chloride and conductivity rejection above 80% in leachate treatment by NF, at feed pH 8.2±0.2.

It is moreover noteworthy that the presence of HAs in leachate was associated to the permeate flux decline and increase in the concentration of inorganic salts in the permeate during membrane filtration performed by Sir et al. (2012). The authors reported that the average rejection of inorganic salts decreased by 20% when 50 mg HA L^{-1} was added to the leachate. As mentioned above, changes in waste composition can have effects on leachate characteristics. In developed countries, as a result of the separation, recycling, and pretreatment of organic waste fractions, inorganic leachate parameters are more important, e.g., heavy metals, sulfide, chlorides, and salts. In this

case, the replacement of the NF membranes by the reverse osmosis membranes is recommended.

3.3.3 Cost estimation

Regarding cost estimates for the process, the information obtained from the experimental units was used to assess leachate treatment costs. The CAPEX for a full-scale plant was estimated at MUS$ 0.772, and OPEX was found to be equal to US$ 3.35 m^{-3}. The treatment cost of the pre-treated effluent by NF is equivalent to US$ 8.26 m^{-3}. According to an estimate made by Samhaber & Nguyen (2014), the total cost of the industrial NF treatment process ranges from US$ 1 m^{-3} to US$ 6 m^{-3}. However, few studies mention the use of NF to treat landfill leachate (AMARAL et al., 2016; CHAUDHARI & MURTHY, 2010; DE ALMEIDA et al., 2019) and none of these texts have examined in detail the landfill leachate management costs. The costs for treatment of leachate vary from simpler processes such as co-treatment with sewage (about 18 - 27 € m^{-3}) to more sophisticated technologies such as reverse osmosis (15 - 40 € m^{-3}) (DE ALMEIDA et al., 2019).

A sensibility analysis was performed to evaluate the impact of increased recovery on effluent treatment costs. For a recovery rate of 70%, there was a 5% decrease in OPEX per m^3 of permeate. While in an 80% recovery scenario, these costs were reduced by about 17%. The increase in the recovery rate enables a decrease in operating cost per cubic meter of treated leachate. However, this increase in permeate recovery would result in an increase in polarization concentration at the membrane surface, resulting in severe risk of fouling and lower efficiency of removal of effluent pollution parameters. Figure 3-3 shows the relative composition, in percentage, of CAPEX and OPEX.
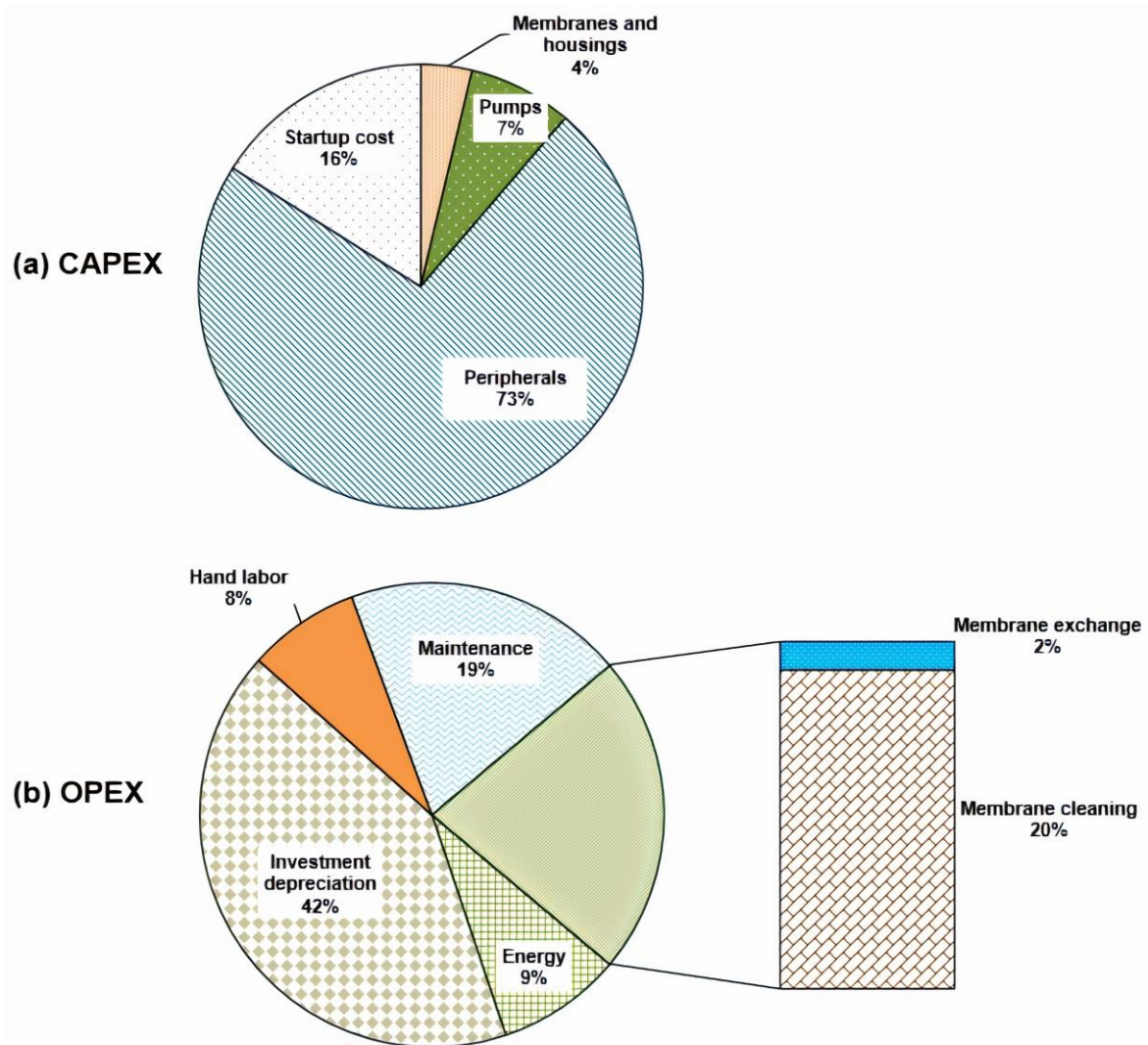
Figure 3-3 Relative composition, in percentage, of (a) CAPEX and (b) OPEX.

CAPEX has highlighted the expenses with the installed system, which account for the acquisition of peripherals. On the other hand, peripherals cost was related to the m^2 of membrane used in the NF process. Also, membrane costs are proportional to plant size, which point out to an economy of scale.

Regarding the OPEX, the percentage value related to the investment depreciation and membrane (exchange and cleaning) stands out, 42% and 22%, respectively. Operating expenses are incurred over the life of the project and include a variable component that can be managed continuously. Thus, it is evident that good operational practices throughout the useful life of the leachate treatment plant, such as periodic cleaning of membranes, may reduce operating costs and make the NF more economically attractive.

The costs for managing the concentrate were not considered since it can be disposed of in the landfill without additional costs for the NF-plant. Opinions on the environmental and economic consequences of recirculation membrane concentrate practice are not unanimous, and specific studies present in the scientific literature are scarce. According to Talalaj (2015), this practice accelerated waste decomposition. On the other hand, Calabrò et al. (2018) reported that the frequency of specific parameters, e.g. ammoniacal nitrogen, Cl^- and SO_4^{-2}, was found to be higher in the leachate recovered after concentrate recirculation.

3.4 CONCLUSIONS

This study dealt with the application of NF on a bench-scale, followed by a cost estimative of the applied process in a full-scale NF plant. The conclusions from this work are:

i. Due to the complexity of landfill leachate, to provide treated leachate that can be discharged into natural water streams or reused is not achievable using a single technique but a combination of different processes is required. NF can be used as the downstream stage of an integrated treatment in which the upstream treatment is covered by a physicochemical process.

ii. The concentration of recalcitrant substances, expressed as HS, was reduced from 821 ± 86 mg L^{-1} in the pre-treated leachate to 84 ± 8 mg L^{-1} in the permeate, which corresponds to 90% rejection. The COD values decreased to below 250 m L^{-1} established by local legislation as disposal standard.

iii. The CAPEX for a full-scale plant was estimated at MUS$ 0.772, and OPEX was found to be equal to US$ 3.35 m^{-3}. The total cost to treat the effluent by NF was estimated at US$ 8.26 m^{-3}.

iv. Membrane fouling seems to be an inevitable problem of NF technology. In this respect, future researches should focus on novel membrane materials (e.g., nanomaterials, polymer/carbon nanotubes, nanocomposite, graphene), and membrane surface modification.

v. Nanofiltration concentrate produced during the leachate treatment is a weakness of the membrane process and proper management is a challenging task. In addition, opinions about concentrate recirculation on landfill cells are conflicting and vary greatly between published studies. Therefore, further researches are needed and should focus on technologies for concentrate volume minimization or system configurations for reuse, treatment, and discharge.

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Chapter 4.   Cost Estimation of Landfill Leachate Treatment by Reverse Osmosis in a Brazilian Landfill

This chapter explored the economic aspects of a full-scale reverse osmosis facility. The capital cost for this full-scale plant was estimated at MUS$ 1.413, and operating costs ranged from US$ 0.132 to US$ 0.265 m3 per year. The cost of leachate treatment has been estimated at US$ 8.58 m-3, considering the operation of the treatment facility for 20 years after landfill closure. The presented study intends to provide a comprehensive analysis, highlighting the economic benefits of membrane-based technologies for landfill leachate treatment.

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4.1 INTRODUCTION

Landfill leachate is a highly contaminated wastewater generated by the decomposition of substances contained in solid waste, which has large amounts of contaminants measured in terms of Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD_5), ammonia, and inorganic salts (KJELDSEN et al. 2002). Leachate treatment process is expensive and different from each other due to the complex composition, also, its composition is variable from different landfills (TALALAJ et al. 2019). For this reason, the treatment methods have not been standardized so far. In this context, landfill leachate management is one of the major environmental and economic issues faced by landfill owners.

Reverse osmosis (RO) seems to be one of the most promising and efficient methods among existent processes for landfill leachate treatment used as the downstream stage of integrated treatment or integral treatment (YAO, 2013). Several studies have been using RO in the landfill leachate treatment (CINGOLANI et al. 2018; DI MARIA et. 2018; DOLAR et al. 2016; SCHIOPU et al. 2012; TALALAJ et al. 2019). In the early 2010s, Šír et al. (2012) reported that RO-plants were installed in more than one hundred landfill sites in northern Europe, North America, and the Far East. Besides, membrane filtration processes have been used at landfills in Brazil, for example, at the landfills of Seropédica, São Gonçalo, Gramacho and Campos in Rio de Janeiro State, Rio Claro and Osasco in São Paulo State (COSTA et al. 2019; DE ALMEIDA et al. 2019; DE ALMEIDA et al., 2020).

The published literature reports studies that evaluated the technical feasibility of RO in the treatment of leachate. In general, the process performance in terms of removal of pollution parameters, e. g., COD, ammonia nitrogen, and chloride, were investigated (KOŠUTIĆ et al. 2014; KUUSIK et al. 2014; TALALAJ 2015; ŠÍR et al. 2012; SMOL et al. 2015). This is due to the ability to retain contaminants dissolved in leachate with high efficiency. Cingolani et al. (2018) reported that RO can remove pollutants from leachate with an efficiency up to >99%. For this reason, it is preferred over other technological options. However, to the best of our knowledge, very few authors have examined in detail the landfill leachate management costs. Moreover,

companies that run leachate treatment plants do not provide the actual process costs. Within this context, the main objective of this work was to estimate the cost of landfill leachate treatment from a full-scale RO plant located in Rio de Janeiro State, Brazil.

4.2 MATERIALS AND METHODS

4.2.1 Case study

The sanitary landfill of this study is located in Rio de Janeiro State, Brazil. It operates with the reception, treatment and final disposal of solid waste from non-hazardous industry activities, health services, different types of construction, and municipal solid waste. The inter-municipal sanitary landfill activities started in January 2012, with an initial construction investment of MUS$ 2.45. The capacity to receive solid waste is about 2,500 tons per day, and the estimated useful life is 15 years. Leachate treatment is estimated to continue to operate 20 years after the landfill closure. Even closure of sanitary landfills will not instantaneously eliminate the problem related to leachate because even capped, it will produce this wastewater stream (ALFAIA et al., 2017).

The leachate treatment plant has been operating since 2014, has a treatment capacity of 120 m^3 day^{-1} and consists of a modular container-mounted system composed of the following process: sand filter, cartridge filters and, three-step RO (Figure 4-1).
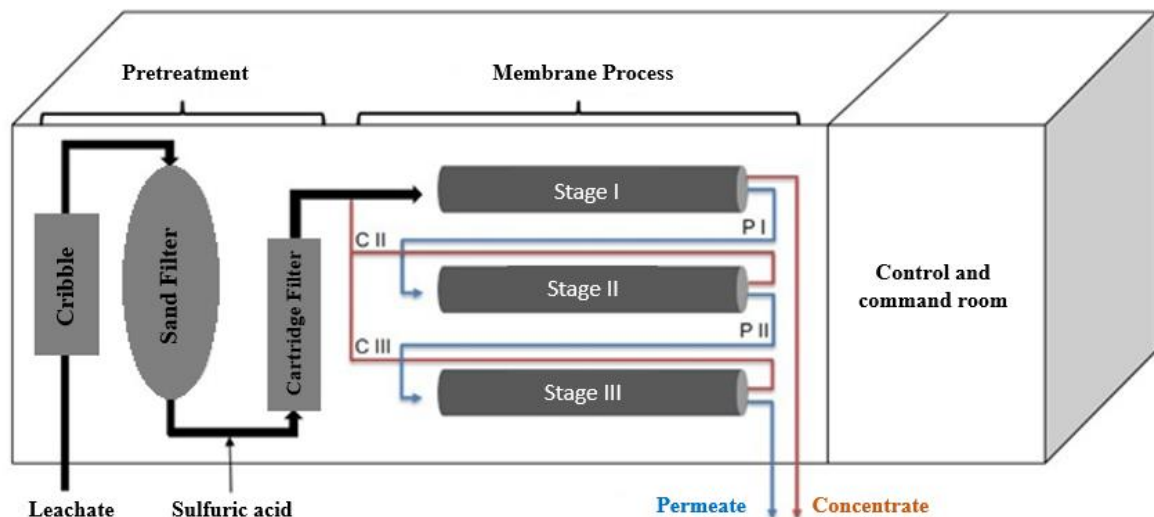
Figure 4-1 Leachate treatment plant set-up. P: permeate; C: concentrate.

The RO system consists of twenty GE Osmonics Desal spiral type membrane modules (GE membranes SC8040F1012). Weekly the membranes are alkaline washed (pH = 12), and monthly are subjected to citrus washing, both for 60 minutes. The reagents are added via pipelines and, pH is monitored on control room. The concentrate from the filtration process generated in stages-II and III returns to stage-I and, the brine produced is stored in two accumulation ponds and sent to three basins for concentrate storage of 10 m^3 and recirculated to the landfill cells. Part of the permeate is used as reuse water for humidification of access roads and landfill sites, and the remainder sent to leachate accumulation ponds.

4.2.2 Cost estimation

The cost estimation was represented by the capital expenses (CAPEX), by the operational expenses (OPEX) and by the specific treatment cost (STC) – total cost normalized per unit volume of treated leachate. For preliminary estimation of the expenses of full-scale RO, the following considerations were made: (i) the leachate treatment plant would operate 365 days a year and would be out of operation only during periods of routine maintenance, chemical cleaning and integrity testing (GUERRA & PELLEGRINO 2012); (ii) the RO process would operate with 71% recovery (R = 71%); (iii) The full-scale RO permeate flux (flow rate per unit membrane area, J = 10 L m^{-2} h^{-1}) was based on manufacturer data (iv) the spiral membrane

modules used would be 40 m^2 in working area per module (BAKER, 2012) and (v) final disposal of the concentrate generated in the process was not considered since this waste is disposed at landfill itself without additional costs for the leachate treatment plant.

4.2.2.1   CAPEX

CAPEX was determined by adding the acquisition costs of (i) membrane modules and housings; (ii) valves, piping, pumps and instrumentation that constitute an RO-plant; (iii) construction costs; (iv) start-up; (v) indirect capital costs; and (vi) RO pre-treatment.

i)   Membranes and housings

Membrane and housings acquisition costs are approximately 25-35% of the total investment value (SALEHI et al., 2014). The cost of membranes and housings was calculated from Equations 3.1, 3.2, 3.3, 3.4, and 3.5 in chapter 3.

ii)   Peripherals and Pumps

The estimated investment cost for peripherals in studies reported in the literature is approximately US\$ 780 m^{-2} membrane, considering the purchase of valves, instruments, equipment, and piping (SALEHI et al. 2014). The cost of RO pumps was estimated at US\$ 12,671.00 stage^{-1} (WESCHENFELDER et al. 2015) and corrected for information referenced in the Chemical Engineering Plant Cost Index to represent updated data for 2019. The correction factor for the pump cost was 1.48.

iii)   Construction costs

The cost of construction was obtained, considering a percentage of 20% to the value of equipment and components (MIERZWA et al. 2008).

iv)   Start-up

The start-up cost is the amount of capital required to start the operation, corresponding to 8% of direct capital cost (AMARAL et al. 2016).

v)   Indirect capital costs

In addition to direct capital costs (directly related to the process and plant construction), there are also indirect costs, which correspond to the cost of land, team training, smaller-scale experiments, state permission to operate, among others. The value of CAPEX can be obtained by multiplying the direct investment cost (sum of capital costs) by the cost factor for indirect costs. For membrane separation process systems, the use of factor 1.67 is recommended (EPA, 2005).

vi)   RO pre-treatment

According to Baker (2012), a reasonable pre-treatment system is essential to achieve a long RO membrane life. All RO units are fitted with a 0.45 µm cartridge filter followed by the high-pressure pump, and a sand filter (BAKER, 2012). RO pre-treatment was estimated at 5% of the total RO system installation cost.

4.2.2.2   OPEX

For the composition of OPEX, the costs of energy consumption for RO operation, membrane exchange, workforce, membrane cleaning, depreciation, and maintenance were considered (SALEHI et al. 2014). Depreciation and maintenance costs incur independently of the operational status of the plant.

According to Samhaber and Nguyen (2014), the OPEX of membrane processes can be attributed to the membrane replacement costs (MRC) (Equation 4.1), which directly depend on the required membrane area and therefore, on the size of the plant that is used for the process. In wastewater treatment, the maximum affordable MRC is less than 10% of the equipment costs (SAMHABER & NGUYEN 2014).

$$OPEX = 6.80 \times MRC \qquad (4.1)$$

Where, OPEX (US$ year^{-1}); MRC (US$ year^{-1}).

4.2.2.3 Specific total cost (STC)

The STC was obtained by Equation 4.2, which accounts for the OPEX normalized by the annual amount of treated effluent and the CAPEX normalized by volume of treated effluent added annually to the time, in years, of operation of the RO plant.

$$STC = \frac{(CAPEX + [n \times OPEX])}{V_t} \qquad (4.2)$$

Where, n: the operating period of the leachate treatment plant (years); Vt: total volume of treated effluent in the period (m^3); CAPEX (US$); OPEX (US$ year^{-1}).

4.3 RESULTS AND DISCUSSION

For the operation of the leachate treatment system, given the considerations made in item 2.2, 405.75 m^2 of membranes arranged in 11 filtration modules are required, as shown in Table 4-1.

The case study full-scale RO of this work has 20 spiral membrane modules. Considering the data provided by the manufacturer for membrane model SC8040F1012 (LENNTECH, 2019), the case study leachate treatment system has 490 m^2 of membrane area. However, it was not informed how many of these are kept in operation for leachate treatment.

Table 4-1 Cost estimation parameters.

Parameters	Unit	Value
Leachate flowrate (Q)	$m^3\ h^{-1}$	5.400
Project flow rate (Qp)	$m^3\ h^{-1}$	3.834
Effective flow rate (Qe)	$m^3\ h^{-1}$	4.058
Recovery (R)		0.71
Permeate flux (J)	$L\ m^{-2}\ h^{-1}$	10
tlq	$h\ d^{-1}$	0.821918
tmr	$h\ d^{-1}$	0.166667
tti	$h\ d^{-1}$	0.333333
tinop	$h\ d^{-1}$	1.321918
Rop		0.94492
Am	m^2	405.75
Modules		11

According to Zirehpour & Rahimpour (2016), a two-stage RO filtration system is owned and operates since 1995 by Pall Corporation for leachate treatment. The membranes used are polymeric (polyamide). A gravel filter for the separation of coarse matter and a cartridge filter are arranged upstream as RO pre-treatment. The treatment system contains 60 modules with about 460 m^2 total membrane surface area; 44 of which are used in the leachate stage, 13 in the first concentrate stage and 3 in the second concentrate stage. The system treats 5 m^3 of leachate per hour, and the permeate recovery is about 94% (ZIREHPOUR & RAHIMPOUR, 2016).

The CAPEX for a full-scale RO was estimated at MUS$ 1.413 (Table S. 4- 1), and specific total cost ranged from US$ 10.09 to US$ 8.58, full-scale RO with R = 71% based on the case study of this paper. Before the implementation of the RO system, the leachate treatment cost was estimated at US$ 21 m^{-3} (PMSG, 2014).

According to Calabrò et al. (2018), the overall treatment cost of landfill leachate (on-site) by RO in Italy is in the range of € 15-40 m^{-3} (US$ 17-44 m^{-3}). Kurniawan & Chian (2006) point out that in German, subject to the complexity of the RO-plant, the landfill leachate treatment ranges from 2 to US$ 30 m^{-3}. On the other hand, Thörneby et al. (2003) conducted the treatment of stabilized leachate from the Hedekosga landfill (Sweden) using RO on a pilot-scale, and the overall cost for a full-scale treatment was

about 30 SEK m^{-3} (US$ 3.10 m^{-3}).

Figure 4-2 (a) shows that capital costs per volume of treated effluent decrease given more extended treatment plant operational time, with 71% recovery and MRC corresponding to 10% of equipment costs. Moreover, CAPEX has highlighted the expenses with indirect capital costs (60%) and peripherals and pumps (26%) (Figure 4-2 (b)). On the other hand, it stands out that peripherals' expenses are related to membrane costs. In this analysis, as can be seen, membranes and housings corresponded to 1% of capital costs.
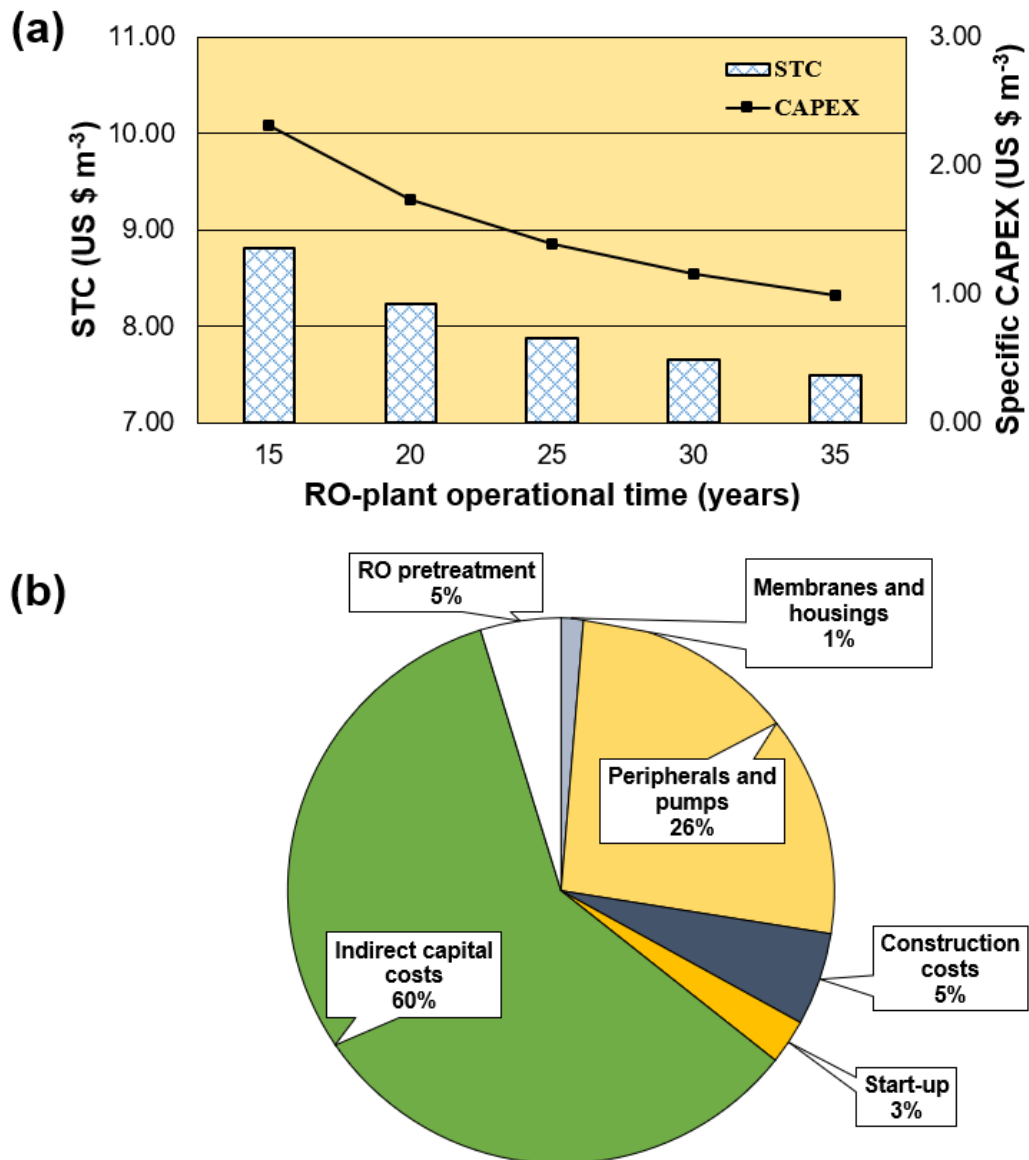
Figure 4-2 CAPEX and specific CAPEX as a function of operational time RO-plant in years (a) and, relative composition, in percentage, of CAPEX (b).

Figure 4-3 shows a sensibility analysis performed to evaluate the impact of an increased % of the equipment costs attributed to MRC (Table S. 4- 2).
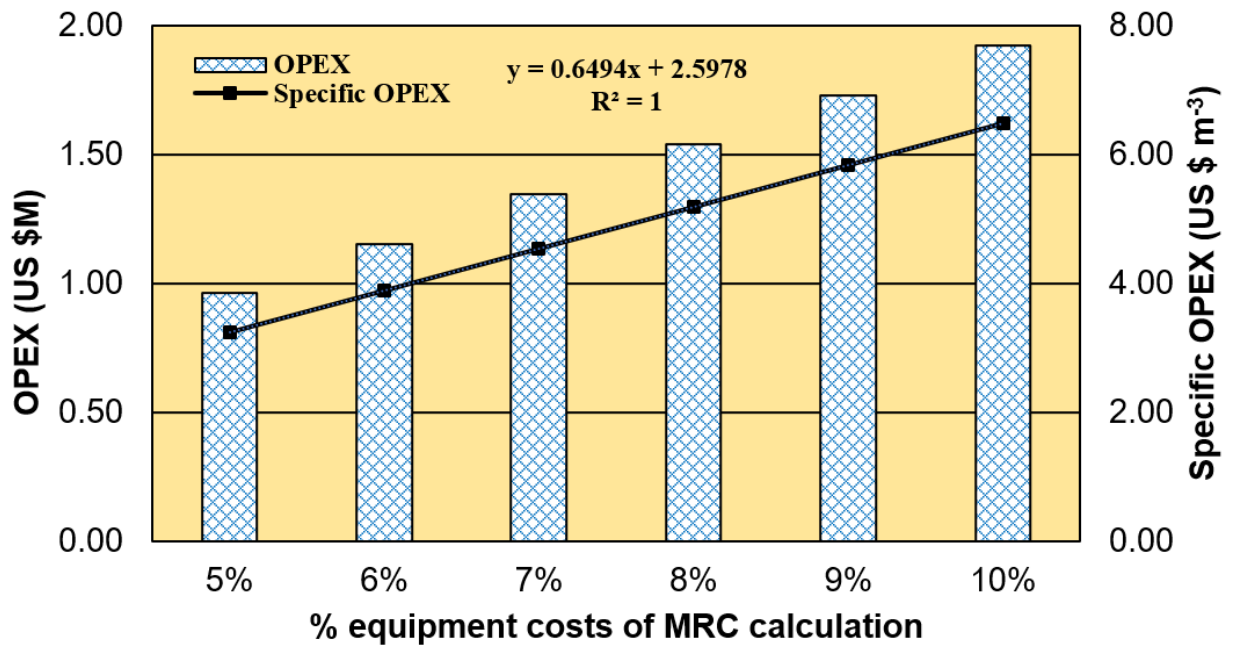
Figure 4-3 OPEX and specific OPEX as a function of the percentage of equipment costs considered in the MCR calculation.

The OPEX for a full-unit RO ranged from MUS$ 0.132 to US$ 0.265. Also, it is observed that the operational expenses per cubic meter of treated effluent can be described as a first-degree equation (Specific OPEX = 0.6494 × "% of equipment costs" + 2.5978). Hence, the higher the value attributed to MRC, the higher the process OPEX will be. On the other hand, operational expenses are incurred over the life of the project and include a variable component that can be managed, for example, periodic cleaning of membranes, may reduce membrane replacement costs and make the RO more economically attractive.

Besides high-performance membranes and appropriate operating conditions, the success of RO plants also depends on smart membrane element and process design to minimize adverse effects from concentration polarization and membrane fouling/biofouling, to meet the requirement of permeate as well as to reduce capital cost and energy consumption (WANG & WANG, 2019). Particularly in relation to the

operation of the RO-unit of this case study, biofouling was reported by the plant technicians as the biggest barrier to the operation of the leachate treatment plant. In a literature review, Guo et al. (2012) state that biofouling of RO would be prevented by a combination of pre-treatment and periodic cleaning of membranes. Therefore, it becomes clear again that performing preventive/curative process may extend membranes lifespan and reduce operational costs. A sensitivity analysis of RO recovery on STC was performed as shown in Figure 4-4.
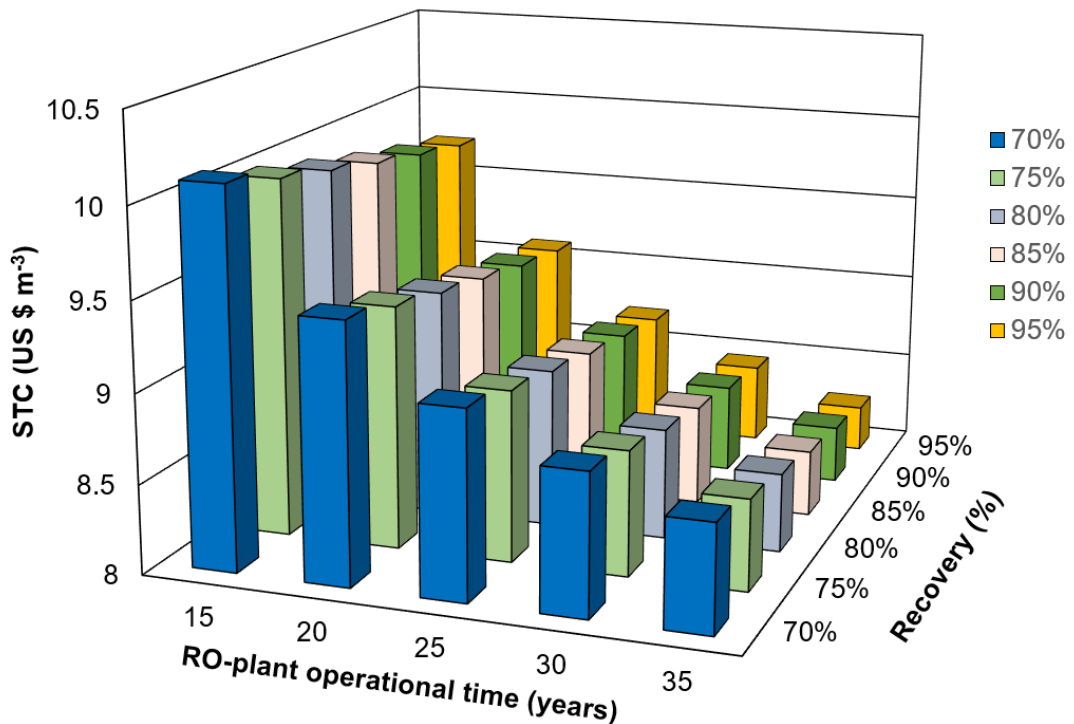
Figure 4-4 STC as a function of operational time of RO-plant considering different percentual of recovery.

The STC of leachate treatment decreases with both increased plant operational time and RO system recovery (Table S. 4- 3). STC also has its reduced value given higher leachate feeding flowrate (Table S. 4- 4). However, the phenomena that occur on the membrane surface (fouling and polarization of concentration) will be even more intense when the quantities of contaminants increase. Nonetheless, in the case of leachate, this increase in membrane process recovery would result in an increase in polarization concentration at the membrane surface, resulting in severe risk of fouling and lower efficiency of removal of effluent pollution parameters (DI MARIA et al., 2018; SCHIOPU et al., 2012).

4.4 CONCLUSIONS

In this work, a procedure by retrieving data from the literature was proposed to estimate the treatment costs of a RO-plant. The CAPEX for a full-scale plant was estimated at MUS$ 1.413, and OPEX range from US$ 0.132 to US$ 0.265 m^{-3} per year. The literature reviewed shows that the cost of leachate treatment by RO range from 2 to US$ 44 m^{-3}. In this case study, the cost of leachate treatment has been estimated at US$ 8.58 m^{-3} considering the operation of RO-unit for 20 years after landfill closure. The theoretical analysis was divided into a set of well-known equations for the cost estimation of RO-plant organized to estimate the cost of this case study leachate treatment, an approach not yet available in the literature. Also, the information presented and discussed contributes to the significant advance of the full-scale RO-plant. Furthermore, the procedure presented may be used as an auxiliary tool to estimate the cost of any membrane processes for wastewater treatment.

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Supporting content

Cost estimation of landfill leachate treatment by reverse osmosis in a brazilian landfill

Table S. 4- 1 CAPEX componentes costs (MRC = 10% equipment costs; R = 71%).

Components	US$
Membranes and housings	16,229.94
Peripherals and pumps	372,743.14
Peripherals	316,483.90
Pumps	56,259.24
Construction costs	77,794.62
Start-up	37,341.42
Indirect capital costs	841,862.23
RO pretreatment	67,298.57
CAPEX	**1,413,269.93**

Table S. 4- 2 MRC at different percentages of equipment costs.

% Equipment costs	MRC (US$ year^{-1})
5%	132,250.85
6%	158,701.02
7%	185,151.19
8%	211,601.36
9%	238,051.53
10%	264,501.70

Table S. 4- 3 STC at different operational time of RO-plant and recovery (MRC corresponding 10% of equipment costs).

	STC (US$ m^{-3})				
n (years)	15	20	25	30	35
R (%)					
70	10.11	9.45	9.05	8.79	8.60
75	10.01	9.36	8.96	8.70	8.51
80	9.93	9.28	8.89	8.62	8.44
85	9.85	9.21	8.82	8.56	8.37
90	9.78	9.14	8.76	8.50	8.32
95	9.72	9.09	8.70	8.45	8.26

Table S. 4- 4 STC, CAPEX and specific OPEX at different leachate flowrate (full-scale RO operating time corresponds to 15 years).

Leachate flowrate (m^3 h^{-1})	STC (US$ m^{-3})	CAPEX (MUS$)	OPEX m^{-3} (US$)
5.40	10.09	1.413	7.44
6.00	9.95	1.547	7.33
7.00	9.76	1.771	7.20
8.00	9.62	1.995	7.09
9.00	9.51	2.219	7.01
10.00	9.42	2.443	6.95
15.00	9.16	3.562	6.75
20.00	9.03	4.681	6.66
25.00	8.95	5.800	6.60
30.00	8.90	6.920	6.56
35.00	8.86	8.036	6.53
40.00	8.83	9.159	6.51
45.00	8.81	10.279	6.49
50.00	8.79	11.398	6.48
55.00	8.78	12.517	6.47

Chapter 5.   A review on Membrane Concentrate Management from Landfill Leachate Treatment Plants: the Relevance of Resource Recovery to Close the Leachate Treatment Loop

As discussed in previous chapters, membrane-based technologies have been used to treat landfill leachate. On the other hand, closing the leachate treatment loop and finding a final destination for landfill leachate membrane concentrate– residual stream of membrane systems – is challenging for landfill operators. This chapter comprehensively reviews the state-of-the-art of current research on membrane concentrate management from leachate treatment plants. Critical insights into leachate concentrate management and future research trends are provided herein.

This chapter is published as a review article in Waste Management & Research Journal.

5.1 INTRODUCTION

Membrane-based technologies, that is, nanofiltration (NF) and reverse osmosis (RO), are considered the most reliable and effective methods for leachate treatment (ALMEIDA et al., 2020; CHAUDHARI & MURTHY, 2010; CHIANESE et al., 1999; CINGOLANI et al., 2017; DE ALMEIDA et al., 2020; LINDE et al., 1995; ŠÍR et al., 2012). In contrast, to close the loop of the landfill leachate treatment and find a final destination for the membrane concentrate (LLMC) — residual stream of the membrane filtration process — is a critical issue, and LLMC management is a challenging task.

Several technologies have been proposed and investigated to manage the membrane concentrate, including recirculation (CALABRÒ et al., 2010; CHAMEM et al., 2020; HE et al., 2015), natural evaporation (COSSU et al., 2018), solidification/stabilization (S/S) (HUNCE et al., 2012), chemical coagulation (LONG et al., 2017), electrocoagulation (EC) (FERNANDES et al., 2019), ozonation (SHAH et al., 2017), advanced oxidation processes (AOPs) (e.g., Fenton, photo-Fenton, anodic oxidation) (FERNANDES et al., 2017; HONG et al., 2017; SOOMRO et al., 2020), and thermal treatment (ZHANG et al., 2019). The recirculation of the LLMC onto landfill waste cells is the conventional approach, similar to leachate recycling to adjust moisture content and degrade organic pollutants in landfills (CALABRÒ & MANCINI, 2012; GROSSULE & LAVAGNOLO, 2020; SOHOO et al., 2019). However, some critical issues like failures of landfill stability and accumulation of pollutants in the leachate treatment facility can emerge as negative impacts linked to this practice. Before the study carried out by (HENIGIN, 1995), the consequences of the reinjection of concentrated leachates into the landfill body were under-discussed. In recent years, there has been an increasing amount of literature on the effects of this procedure (CALABRÒ et al., 2018; CHAMEM et al., 2020; MORELLO et al., 2016; TALALAJ, 2015; TALALAJ & BIEDKA, 2015). Nonetheless, the published studies show contrasting conclusions. Therefore, further research in this area is still of high importance.

Several other reviews already exist, which do excellent work in describing LFL treatment processes (ABUABDOU et al., 2020; COSTA et al., 2019; GAO et al., 2014; LUO et al., 2020; RENOU et al., 2008; WISZNIOWSKI et al., 2006), membrane-based technologies for wastewater treatment (KAMALI et al., 2019), and treatment technologies for membrane concentrate volume minimisation (JOO & TANSEL, 2015; SUBRAMANI & JACANGELO, 2014). Readers are guided toward these contributions for further background information. Recently, Keyikoglu et al. (2021) reviewed the state-of-the-art of technologies for the treatment of LLMCs. Among existing methods, they paid more attention to AOPs. However, these techniques cannot effectively handle the high salinity of the LLMC and, therefore, are mainly applied as a pretreatment step rather than a stand-alone treatment. Besides, AOPs are associated with high installation and operational expenses, and the possible generation of intermediates with higher toxicity during the LLMC treatment also represents a limitation for their consolidation on a full-scale application.

As aforementioned, several approaches could be adopted for the management of LLMCs. Considering the demands of efficient water reuse, carbon, and nutrients from LLMCs or from LFL itself, efforts have been focused on extracting add-value products from concentrated leachates, e.g., inorganic salts and biofertilizers (GU et al., 2019; KURNIAWAN et al., 2021; LI et al., 2015). At present, a review dealing with the management of LLMCs focusing on resource recovery has not been published yet. This work comprehensively reviews the state-of-the-art of current research on membrane concentrates management from LLTPs towards a resource recovery approach. Lastly, within a circular bioeconomy context, a general recovery train based on the main LLMC characteristics for implementing the best recovery route is presented.

5.2 MATERIALS AND METHODS

The databases, including Web of Science, Scopus, and Engineering Village, were explored. The following keywords were combined to find the scientific literature: "landfill leachate", "nanofiltration", "reverse osmosis", "membrane concentrate", and "concentrated leachate". The screening was undertaken using the eligibility and exclusion criteria to include the works relevant to the research topic. Eligibility criteria consisted of selecting articles that deal with NF and RO concentrate treatment and management options. In contrast, excluding articles published in a language other than English and papers that do not deal with LLMC management was part of the exclusion criteria.

The present article is structured as follows: first, the main characteristics of NF and RO concentrate from LLTPs are introduced. Second, a critical analysis of LLMC destination practices and treatment systems are presented. Third, resource recovery from LLMCs covering water reuse technologies and material extraction, i.e., organic fertilizers, nutrients, and inorganic salts recovery, is comprehensively reviewed. Current trends and challenges are addressed. Last, a management diagram for the best resource recovery route definition from LLMCs is proposed.

5.3 LANDFILL LEACHATE MEMBRANE CONCENTRATE (LLMC) MANAGEMENT

5.3.1 LLMC characteristics

Different factors can affect the composition of concentrate streams from LLTPs, including leachate characteristics, pretreatment applied, additional chemicals used, i.e., fouling/biofouling prevention chemicals or reagents used for pH control, and treatment configurations (LADEWIG & ASQUITH, 2012; VAN DER BRUGGEN et al., 2003). These influence factors have been found in the analysis of nanofiltration concentrate (NFC) and reverse osmosis concentrate (ROC) from the LLTP in Xiamen (China), where concentrate streams were collected from two different leachate treatment configurations. The NFC contained a high amount of refractory organics; conversely, recalcitrant contaminants in the ROC were lower because most of these were removed by pretreatment processes used in the treatment chain (CHU et al.,

2020). From that, it is presumable that the LLMC composition can vary depending on the LLTP's defined treatment scheme.

As NF and RO are the preferred membrane processes for leachate treatment, the main characteristics of NFC and ROC from LLTPs reported in various studies are summarized in Table 5-1. It must be emphasized that this table intends to highlight the characteristics of the main concentrate streams generated in LLTPs (i.e., NFC and ROC), focusing on the reported values instead of the treatment scheme that generated the concentrate stream. For instance, the values of some parameters like NH_3-N and TKN have a wide range since, in some cases, nitrogen can be removed by biological process in the leachate treatment chain, confirming what was discussed above.

Table 5-1 The main characteristics of NFC and ROC from LLTPs.

Parameters	NFC	ROC	Parameter	NFC	ROC
pH	6.61—8.97	6.17—8.3	TDS (mg L^{-1})	8140—18,910	20,200—66,900
BOD$_5$ (mg L^{-1})	2.6—285	549—17,000	Chloride (mg L^{-1})	1280—10,000	1823—30,768
COD (mg L^{-1})	1281—9500	1646—49,521	Sulphate (mg L^{-1})	53—5252	20—7900
TOC (mg L^{-1})	1347—2365	719—4500	Sodium (mg L^{-1})	890—4206	15,400
UV$_{254}$ (cm^{-1})	20—60	8.29—19.8	Potassium (mg L^{-1})	210—2806	9600
HS (mg L^{-1})	1393—1501	Data not available	Calcium (mg L^{-1})	241—480	150
NH$_3$-N (mg L^{-1})	15—3276	63—8300	Copper (mg L^{-1})	0.01—9.26	0.022—3.71
TKN (mg L^{-1})	260—1,000	1,820—9100	Zinc (mg L^{-1})	0.079—6656	<0.008—2663
BOD$_5$/COD	< 0.09	0.01—0.40	Lead (mg L^{-1})	0.006—56.97	0.05—22.79
Conductivity (µS cm^{-1})	16,130—79,400	10,500—98,000	Nickel (mg L^{-1})	0.08—3.182	0.20—1.59

BOD$_5$: five-day biochemical oxygen demand. COD: chemical oxygen demand. HS: humic substance. NFC: nanofiltration concentrate. NH$_3$-N: ammonia nitrogen. pH: potential hydrogen. ROC: reverse osmosis concentrate. TDS: total dissolved solid. TKN: total kjeldahl nitrogen. TOC: total organic carbon. UV$_{254}$: absorbance at 254 nm.

Source: Adapted from Calabrò et al. (2018), Chamem et al. (2020), Fernandes et al. (2019), Hendrych et al. (2019), Hunce et al. (2012), Long et al. (2017), Shah et al., (2017), Talalaj (2015), Talalaj & Biedka (2015), Top et al. (2011), Varank et al. (2020), Wang et al. (2016, 2020), Xiong et al. (2014), Xue et al. (2020), Wang et al. (2016).

As can be seen from Table 5-1, NF/RO concentrates are rich in some heavy metals such as copper, zinc, lead, and nickel (up to 9.26 mg Cu L^{-1}, 6,656 mg Zn L^{-1}, 56.97 mg Pb L^{-1}, and 3.182 mg Ni L^{-1}), which can cause environmental pollution and potential bioaccumulation in living organisms and human body resulting in adverse effects (BRIFFA et al., 2020). Besides, priority pollutants such as toluene,

ethylbenzene, chlorobenzene, and dibutyl phthalate were also identified in LLMC samples. These chemicals are highly toxic pollutants, representing an environmental hazard (ZHANG et al., 2013).

The COD concentration in NFC and ROC ranges from 1281 to 9500 mg L^{-1} and 1646 to 42,000 mg L^{-1}, respectively. In addition, the BOD_5/COD ratios are in the range of 0.03–0.40, indicating low biodegradability. The differences between LFL and LLMC, such as non-biodegradable content and organic matter composition, were investigated in three LLTPs. Humic substances (HSs), including humic acid (HA) and fulvic acid (FA), accounted for the highest fraction of organic matter in LLMCs, ranging from 61.7 to 69.2% (ZHANG et al., 2013). Chan et al. (2007) stand out that high-molecular-weight and non-biodegradable compounds are removed mainly by membrane processes and accumulate in the residual stream. He et al. (2015) reported that concentrates from a full-scale NF-plant had an average COD of 5357 mg L^{-1}, a value about two-fold higher than that found in the landfill leachate (2623 mg L^{-1}).

A COD of 6200 mg L^{-1} was found in concentrates from an NF-full scale plant in Odayeri Sanitary Landfill, Istanbul (Turkey). From the same site, concentrations of ammonia (110 mg L^{-1}), TKN (1000 mg L^{-1}), and chloride (10,000 mg L^{-1}) were recorded (TOP et al., 2011). Similar concentrations were found in NFCs from landfills in Shenzhen and Beijing (China) (3450 mg COD L^{-1}, 80 mg TNK L^{-1}, and 2519 mg Cl^- L^{-1}) (LI et al., 2016). Previous studies have also confirmed that NFC and ROC are heavily polluted by organic and inorganic compounds (MOJIRI et al., 2017; XIONG et al., 2014; XU et al., 2017).

Similarly, as shown in Table 5-1, a high concentration of salts (evaluated by conductivity) is found in ROCs – values range from 10,500 to 98,000 µS cm^{-1}. (Kallel et al., 2017) reported that ROCs from an LLTP located in Tunisia contained high levels of TDS (66,900 mg L^{-1}), chloride (30,768 mg L^{-1}), sodium (15,400 mg L^{-1}), and potassium (9600 mg L^{-1}). Similar findings have been reported by (Hendrych et al., 2019). ROCs from Erzurum landfill (Turkey) were also characterized by high levels of BOD_5 and COD, reaching values up to 4800 mg L^{-1} and 8882 mg L^{-1}, respectively (HUNCE et al., 2012).

5.3.2 Conventional management of LLMCs

Conventional membrane concentrate management from LLTPs can be categorized into two main groups: (1) disposal and (2) treatment. The former includes natural evaporation and recirculation to the landfill body, and the latter involves processes aiming at pollutants removal. These two approaches are critically discussed in the following items.

5.3.2.1 Disposal

The disposal of LLMC streams into the landfill body is the simplest and cheapest method. In general, LLMCs are accumulated in lagoons where physicochemical processes occur, and depending on climatic conditions; there is a moderate reduction of the concentrate volume. Afterwards, the concentrate is injected onto the waste mass by vertical and/ or horizontal drains (CALABRÒ et al., 2018; ISWA, 2019; ROBINSON, 2005). Before the work of (HENIGIN, 1995), the consequences of concentrate injection were under-discussed. A body of scientific research was published in the last two decades. However, literature findings have revealed contrasting conclusions; therefore, this practice's sustainability is not a consensus.

Robinson (2005) presented monitoring data of a German landfill that operated a RO system for one year, returning the concentrate to the landfill. The study showed that ROC infiltration increased COD, ammonia, and conductivity of the generated leachate, which immediately affected the RO performance. Similar results were found by (TALALAJ, 2015) and (TALALAJ & BIEDKA, 2015). On the other hand, 15-years monitoring data of an Italian landfill revealed a moderate change in leachate composition (slight increase in NH_4^+, Cl^-, and SO_4^{-2}) and leachate quantity (i.e., leachate volume increased). However, RO treatment performance was not impacted (CALABRÒ et al., 2018). In a previous study at the same Italian site, Calabrò et al. (CALABRÒ et al., 2010) observed a moderate rise in COD, nickel, and zinc concentrations; on the other hand, no significant change in leachate quantity was identified in this case. Table 5-2 summarises the main findings of LLMC infiltration monitoring studies found in the relevant literature.

Table 5-2 Main findings of LLMC infiltration studies.

Scale	Salient features	Main findings	Reference
Lab	German landfill Experimental landfill cells 20 months of monitoring	The LFL quality remained equal with and without infiltration of LLMCs	HENIGIN (1995)
Full	German landfill 12 months of monitoring	ROC infiltration increased COD, ammonia, and conductivity of the leachate Changes in leachate composition affected the RO performance	ROBINSON (2005)
Full	Italian landfill Infiltration of 20 m^3 d^{-1} 30 months of monitoring	No significant change in the leachate quantity Moderate increase in COD, nickel, and zinc of the generated leachate Possible reduction of methane content from the biogas stream	CALABRÒ et al. (2010)
Full	Brazilian landfill Infiltration of 9.6 m^3 h^{-1} Four months of monitoring	No significant change in the methane content from the biogas stream	ZANON et al. (2013)
Full	Polish landfill Infiltration of ~375 m^3 per month Eight months of monitoring	ROC infiltration increased COD, ammonia, conductivity, and sulphates of the leachate The increase of leachate conductivity affected the COD removal via RO treatment	TALALAJ & BIEDKA (2015)
Lab	Italian landfill Infiltration of 17 LLMC litres during the study period 123 days of monitoring	No change in the LFL quantity No consistent changes in COD emissions and methane production LLMC infiltration increased NH_4^+ of the generated leachate	MORELLO et al. (2016)
Full	Landfill in Bosnia-Herzegovina 83 days of monitoring	Increase in conductivity and decrease of pH values Increased the landfill gas flow and methane content in the short-term	DZOLEV & VUJIC (2016)
Lab	Chinese landfill Simulated landfill cells filled with 1, 5, and 15 yr age wastes Infiltration of 72 ROC litres during the study period 24 days of monitoring	Increase of organics and ammonia in leachates from the 1 yr waste landfill cell Moderate increase of organics, salinity, and heavy metals in leachates from the 5 yr waste landfill cell	WANG et al. (2017)

Scale	Salient features	Main findings	Reference
Full	Italian landfil 15 years of Monitoring The infiltrated LLMC corresponded to 30% of the generated leachate	LFL quantity increased of 10 years of LLMC infiltration monitoring	CALABRÒ et al. (2018)
		Moderate increase in NH_4^+, Cl^-, and SO_4^{-2} of the generated leachate	
		Reduction of heavy metals concentration	
		Changes in leachate composition did not affect RO treatment performance	
Full	Tunisian landfill Infiltration of 8 m^3 ROC d^{-1} during 5 yr 10 yr of monitoring	Reduction of biogas generation and methane content	CHAMEM et al. (2020)

In sum, the literature shows that the consequences of concentrate disposal to landfills are site-specific. In Rio de Janeiro State (Brazil), according to the recently sanctioned law number #9055/2022, landfill managers can recirculate to the landfill body up to 1/3 of waste streams from the LLTP, that is, the byproducts of the leachate treatment chain (e.g., sludge and LLMC) disposed of into the landfill body cannot exceed 1/3 in volume or mass (RIO DE JANEIRO STATE, 2020). However, it should be stated that the concentrate recirculation may be only a temporary solution resulting in never-ending re-introduction of pollution as concentrate contaminants may eventually accumulate in LLTPs. Moreover, this practice should be no longer acceptable within a circular and sustainable wastewater management system, if resources from LLMC streams can be extracted and recovered.

5.3.2.2   Treatment

LLMC treatment options include physicochemical processes (e.g., C/F, EC, adsorption, AOPs, ozonation, and S/S) (AN et al., 2012; CHEN et al., 2019; HE et al., 2021; HONG et al., 2017; KALLEL et al., 2017; REN et al., 2021; TOP et al., 2011; WANG et al., 2020), biological methods (YANG et al., 2018), and thermal processes, including membrane distillation (MD) – a thermally driven membrane filtration technique (CHEN et al., 2021; YUE et al., 2007; ZHANG et al., 2020, 2019) or even combinations of them (DING et al., 2021; WOO et al., 2019). Some of LLMC treatment procedures and their salient features are summarised in Table 5-3.

Table 5-3 LLMC treatment technologies and their salient features.

Treatment technology	Important parameters	Salient features	Reference
	C/F	Chemicals and pH	Low cost Insufficient removal efficiencies Requires addition of chemicals Requires sludge management	LONG et al. (2017)
	EC	Electrode type, current intensity, and time	Good removal efficiencies High-tech and automated system No chemical needed Requires energy input	TOP et al. (2011)
	Adsorption	Adsorbent dose and time	Low cost Insufficient removal efficiencies Adsorbent regeneration is needed	HONG et al. (2017)
Physicochemical	Fenton oxidation	Chemicals, pH, and time	Low cost Requires addition of chemicals Possible change in ecotoxicity Requires sludge management	YAZICI GUVENC & VARANK (2021)
	Photo-Fenton	Chemicals dose, radiation intensity, pH, and time	Increase concentrate biodegradability Requires addition of chemicals Requires sludge management Possible change in ecotoxicity	LI et al. (2016)
	Ozonation	Ozone dose and time	Increase concentrate biodegradability High cost Possible changes in ecotoxicity	CHEN et al. (2019)
	S/S	Mixing ratio (LLMC/aggregate)	Low cost Time-consuming process Non-destructive technique Volume of treated concentrate increases	KALLEL et al. (2017)

	Important parameters	Important parameters	Salient features	Reference
Biological	Co-bioevaporation	Mass ratio and aeration	Efficient removal of water and organics Requires energy input Time-consumingprocess Gaseous emissions	YANG et al. (2018)
	Algal treatment	Culture, aeration, and light intensity	High nutrients removal Requires low energy Low cost Pretreatment required Time-consuming process	WOO et al. (2019
Thermal	Incineration	LLMC's properties, residence time, temperature, and turbulence	High reduction of concentrate volume High energy demand Equipment corrosion Requires flue gas treatment system and management of the residual stream	REN et al. (2019); Tow et al. (2021)
	Submerged combustion evaporation	Energy input and time	High reduction of concentrate volume High energy demand Equipment corrosion Requires management of the residual stream	ZHANG et al. (2019)
	MD	Energy input and permeate flux	High water quality High reduction of concentrate volume High energy demand Equipment corrosion Membrane fouling susceptibility	CHEN et al. (2021)

Physicochemical processes are among the most investigated treatment route. C/F, EC, and adsorption are low-cost techniques with good removal efficiencies. However, they are primarily applied as a pretreatment step rather than a stand-alone technology because of their low salinity removal. In contrast, although AOPs and ozonation can produce a high-quality treated concentrate, these processes have high installation and operational costs, limiting full-scale implementation. Similarly, thermal processes are associated with expensive operating costs, which contributes to few large-scale projects.

Biological techniques stand out in terms of simplicity and low cost. However, due to the high salinity and poor concentrate biodegradability, they are not recommended for the treatment of LLMCs. A novel biological method for LLMCs treatment has been investigated in a laboratory study. Yang et al. (2018) evaluated a

named co-bioevaporation (coBE) process mixing LLMCs with food waste (FW). In coBE, the concentrate is evaporated by the metabolic heat released from aerobic microbial degradation of organic compounds. Under optimal operating conditions (1:1.1 (m/m) mixing ratio of LLMC and FW and 0.035 m^3 h^{-1} airflow per kg TS), 96.7% of water was removed by consuming 96.5% of VS contained in the mixture during five cycles of operation (YANG et al., 2018). However, coBE is time-consuming and requires energy input for aeration, increasing the footprint and operational costs. Recently, the authors proposed intermittent aeration as an energy-saving strategy and found that at a regime of 10 min on/20 min off, more than 50% of energy could be saved with similar treatability results (LIU et al., 2021). Even though the coBE process seems promising, more research is needed to optimise the technique and assess its techno-economic feasibility in large-scale applications. Besides, air pollutants emissions from this process are a significant source of pollution; therefore, carbon footprint and related environmental impacts should also be considered in future investigations.

5.3.3 Resource recovery options

The development of sustainable and cost-effective methods for LLMCs treatment combining resource recovery processes is a promising field of research. LLMC components that would be infiltrated on the landfill body can be extracted and transformed into valuable products such as organic fertilizer and solid salts with commercial value or used for energy purposes (Figure 5-1). This section summarises the literature on concentrated leachate treatment techniques and their application in LLMCs' resource recovery.
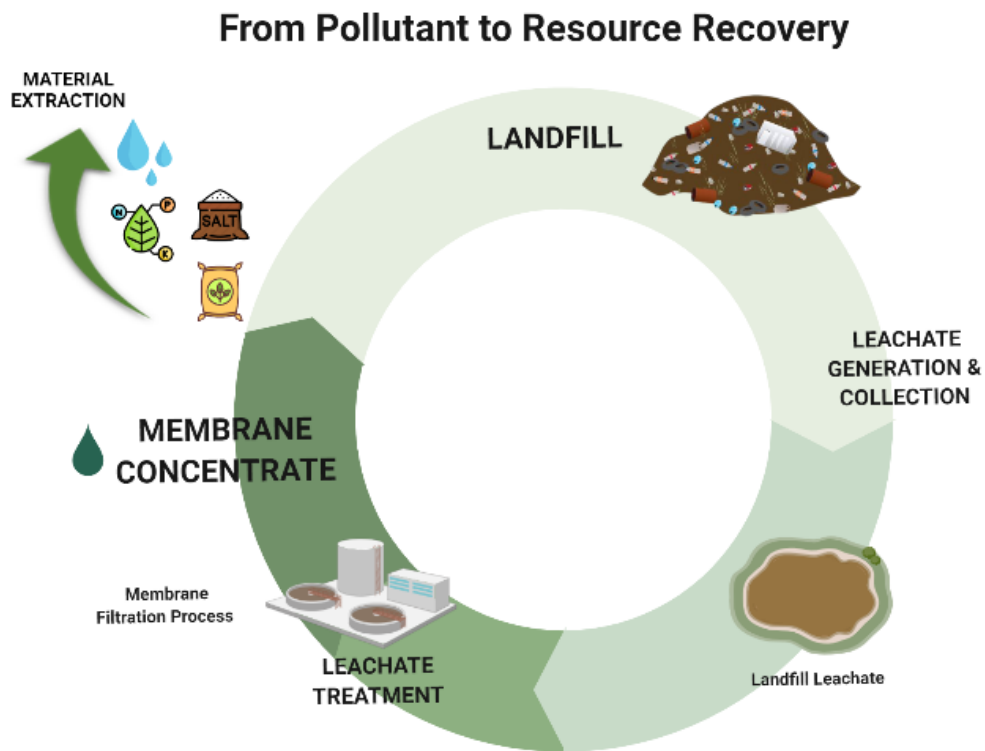
Figure 5-1 From pollutant to resource – perspectives to close the leachate treatment loop.

5.3.3.1   Reclaimed water

Typically, 50–80% of the NF/RO feed is recovered as water. The maximisation of high-quality water recovery during the LFL treatment through membrane processes can reduce the concentrate volume and guarantee high reclaimed water production (ISKANDER et al., 2017). As mentioned early, several patented technologies to

improve feed water recovery based on MLD and ZLD strategies is available. However, these systems are high capital and energy-intensive, which hampers their implementation in LLTPs. For example, Panagopoulos & Haralambous (2020) assessed two different scenarios considering the MLD and ZLD framework. The MLD scheme comprises membrane-based technologies, and the ZLD is membrane and thermal-based. At the freshwater recovery of 84.60% (MLD system) and 98.15% (ZLD system), the energy consumption of MLD and ZLD systems was estimated at 5.40 and 10.43 kWh m^{-3}, respectively.

On the other hand, multi-stage RO has proved to improve permeate recovery and reduce specific energy demand when less than five stages are used (JUDD, 2017). Cingolani et al. (2018) proposed a three-stage RO system (RO1, RO2, RO3) to maximise water recovery of LFL treatment and reach standards for water discharge or reuse. The water recovery was optimised to >90% following RO1 and RO2 stages, while the RO3 step was needed to achieve boron and nitrogen local requirements for reuse.

Another approach beyond concentrate minimisation would be treating LLMC streams to accomplish minimal concentrate disposal producing high-quality water. In this regard, a microbial desalination cell (MDC) was studied to treat the concentrated leachate from a forward osmosis (FO) system. The FO concentrate was desalinated in the MDC and the treated concentrate was returned to the FO for further water extraction. FO water recovery increased from 51.5% to 83.5% when operated along with the MDC system at the hydraulic retention time of 10 h (ISKANDER et al., 2018).

It should be highlighted that FO technology has received increased attention for water recovery due to its low energy requirements and the low fouling propensity of FO membranes (LI et al., 2019). The FO technology uses the osmotic pressure gradient to transport freshwater through the membrane. In this process, a draw solution (DS) with high salinity creates an osmotic pressure difference, leading water to flow from the feed effluent across the semipermeable FO membrane. Further separation of the diluted DS is required after the FO process to obtain reclaimed water as a product (SHAFFER et al., 2015).

CaCl$_2$, Ca(NO$_3$)$_2$, NaCl, and thermolytic solutes based on ammonia and carbon dioxide, similar to NH$_4$HCO$_3$, are the most DS employed for FO applications (Achilli et al., 2010). At the end of the FO filtration, the DS is recovered and recycled back to the FO unit while the permeate may need further treatment for its direct discharge or reuse (Wu et al., 2018). The energy consumption of FO systems without DS regeneration is notably lower (0.2—0.9 vs. 10—14 kWh m^{-3}) (VOUTCHKOV & KAISER, 2020); therefore, from an energy-efficient and resource recovery point of view, the selection of a fertilize draw solute for FO application can be an attractive strategy. Li et al. (2017) investigated a FO system using NH$_4$HCO$_3$ (3 mol L^{-1}) as the DS to treat landfill leachate. FO recovery was higher than 90%, and the water product met the regulatory standards for agricultural fertigation. In another study, Qin et al. (2016) proposed to use the recovered NH$_4$HCO$_3$ (2 mol L^{-1}) as DS in a hybrid-FO system for water recovery from landfill leachates.

5.3.3.2   Humic substances

As discussed in item 5.3.1, LLMCs are rich in refractory organic compounds, mainly consisting of humic substances — high molecular weight compounds (300—10,000 Da) with phenolic, carboxylic, and alkoxy groups along with the occasional presence of esters and quinones (GU et al., 2019; LIMA et al., 2017; XU et al., 2017). In agriculture, HSs are used as organic fertilizer and play a key role in improving soil proprieties (e.g., soil physical structure, nutrient retention, and water holding capacity), increase soil organic content and microbial diversity, and boost fertilizer efficiency. Due to its high salinity, the LLMC cannot be applied directly as an HS-containing liquid fertilizer (YE et al., 2019). Thus, the enrichment and extraction of HSs from LLMCs have aroused strong interest.

To date, the main methods for HS extraction from landfill leachates are chemical precipitation and membrane filtration. Chemical precipitation requires low pH (pH < 2), and therefore, the recovered product is not suitable for direct soil applications. Membrane processes have been used for the efficient extraction of HSs (GU et al., 2019). Overall, UF and NF membranes with MWCO ranges of 200—1000 Da and 1—10 kDa are the primary techniques to fractionate these compounds (XU et al., 2017; YE

et al., 2020, 2019). However, tight NF membranes can jeopardize the purification process for producing liquid fertilizer. The high salt rejection of NF membranes can result in excessive salts in the target product (i.e., concentrate stream) when the NF process is applied for desalination. On the other hand, UF membranes have wide pore sizes and enable almost unrestricted passage of inorganic salts, failing to efficiently fractionate salts and humic compounds due to inadequate rejection of substances of low molecular weight (300–3500 Da) such as fulvic acids (YE et al., 2019).

Xu et al. (2017) employed two-stage tight ultrafiltration (MWCO = 1000 Da) for extracting HSs from leachate concentrates. At the end of the concentration process, organic content was 45,370 mg HS L^{-1}. With the addition of nutrient macroelements in the obtained liquour, the recovered stream could be used as liquid fertilizer. The authors estimated the economic benefit of the HS-containing soluble fertilizer production at 4672 USD m^{-3}. This value could offset production costs, including operating and purchase costs of macronutrients and generates an attractive margin profit.

To efficient fractionating and desalting of NF concentrates, Ye et al. (2019) propose to use loose nanofiltration membrane with MWCO of 860 Da, which takes the merits of both NF and UF membranes. At a concentration factor of 9.6, the HS content was enriched from 1765 to 15,287 mg L^{-1} with about 86% desalting efficiency. As a water-soluble fertilizer, the recovered liquour stimulated the seed germination and enhanced the growth of green mungbean plants, presenting no phytotoxicity. In recent work, the same research group proposed an integrated bio-inspired self-polymerisation procedure to tailor loose nanofiltration proprieties for efficient fractionation of HSs and desalination. Using the modified loose nanofiltration (298 Da), the LLMC was preconcentrated by a factor of 10.0 without permeate recirculation into the feed. Subsequently, a diafiltration step was performed to demineralize the pretreated LLMC. The bio-inspired membrane showed superior selectivity between HSs and inorganic salts. The concentration of HSs was enriched from 1779.4 to 17,247.1 mg L^{-1} and desalting efficiency of 99.5% was achieved, resulting in high HS purity (i.e., 98.3%) for potential liquid fertilizer applications (YE et al., 2020). Table 5-4 depicts recovering schemes for organic components extraction of concentrated leachates.

Table 5-4 Recovery schemes for humic substances extraction from concentrated leachates.

Recovery scheme	Target resource	Product applications	Main findings	Reference
UF	Humic substances	Liquid fertilizer	High-fractioning of HS, salts, and heavy metals	YUE et al. (2011)
Coagulation + centrifugation + sun-drying	Humic and fulvic acids*	Soil conditioning	Recovered material enhanced germination and growth of soybeans	YANG & LI (2016)
Two-stage tight ultrafiltration	Humic substances	Liquid fertilizer	Economic benefit/ attractive margin profit	XU et al. (2017)
Loose nanofiltration	Humic substances	Water-soluble fertilizer	Liquid fertilizer application promoted plant growth with no phytotoxicity	YE et al. (2019)
FO + chemical precipitation	Humic acid	Fertilizer component	Possible application as soil stabilizer or fertilizer component	ISKANDER et al. (2019)
Bio-inspired loose Nanofiltration + diafiltration	Humic substances	Liquid organic fertilizer	High HS concentration and desalting efficiency	YE et al. (2020)

*Recovery from the dewatering effluent of thermally treated sludge (raw effluent: 2180 ± 82 mg HA L^{-1} and 1317 ± 96 mg FA L^{-1}).

It should be underlined that membrane concentrates may have a certain level of toxic pollutants (e.g., heavy metals and xenobiotic organic compounds), which may hinder the direct reuse of the recovered HS as organic fertilizer; thus, further purification will be needed. An interesting work investigated the use of encapsulated HAs in alginate beds extracted from anaerobic sewage sludge to allow controlled and slow-release of humic substances in the soil. The agronomic tests showed that the dry biomass of the treated plants was remarkably higher than that for non-treated plants. The encapsulation of HAs within alginate beads could immobilize toxic compounds as well as reduce the amount of added product, ensuring a proper dose of HAs in the soil at lower operational costs (CRISTINA et al., 2020).

Following another recovery approach, the recalcitrant substances in LLMC streams could be reused for energetic purposes. Ben Hassen-Trabelsi et al. (2020) investigated the co-pyrolysis of ROC and sewage sludge to recycle organics as biofuels. At optimum conditions (mixture of 30:70 ROC/sewage sludge, 550ºC, and 10ºC min^{-1} as heating rate), the process produced high-energy gas (12.29 MJ kg-1), which could be at least employed as an energy source for pyrolytic conversion. Thermochemical processes (e.g., torrefaction, pyrolysis, and gasification) seem to be an exciting strategy to produce value-added products from leachate concentrates, including biochar and bio-oils.

5.3.3.3   Nutrients

Nitrogen (N) and phosphorus (P) are the major nutrients that are present in LFLs. As mentioned in section 2, ammoniacal nitrogen is one of the main landfill leachate pollutants and therefore most of LLTPs are designed to remove N. As contaminants, N and P stimulate excessive plant and algal growth, leading to waterbody eutrophication and associated adverse impacts. N-NH$_3$ removal of landfill leachates has been extensively studied (ANTWI et al., 2020; DE ALMEIDA et al., 2019; GENETHLIOU et al., 2021). As resources, N and P are critical macronutrients for crops and hence are key components for fertilizer production. Besides, phosphate shortage linked to food security has intensified interest in P recovering of wastewaters. In NFC and ROC, the ammonia nitrogen concentration is generally high, reaching levels of 3273—8300 mg L^{-1} (Table 5-1); such levels are high enough to shift the focus from removal to recovery.

Stripping technology and chemical precipitation are the main methods for nutrients recovery from wastewaters. Stripping technology is based on the ammonia gas-liquid equilibrium in an aqueous solution. In an alkaline environment, usually pH from 10.5 to 11.5 at 25ºC, the balance of ammoniacal nitrogen in leachates tends to produce more ammonia than ammonium (NH_4^+ $_{(aq)}$ + $OH^-_{(aq)}$ $\leftrightarrow$ $NH_{3(g)}\uparrow$ + $H_2O_{(l)}$) (CAMPOS et al., 2013). Ammonia is a water-soluble gas. Thus, passing an air stream through the wastewater, the ammonia concentration reaches the gas-liquid equilibrium in the system, and ammonia can be recovered from the stripped gas. The primary

factors affecting ammonium stripping are pH, airflow, and temperature. High-temperature water vapour can be used as the air stream to boost the ammonia mass transfer. The stripping gas must be allocated into an acid solution for recovery purposes, so ammonia is recovered as an ammonium salt like NH_4Cl and $(NH_4)_2SO_4$. The recovered salt can be reused in different industrial and commercial applications (CAMPOS et al., 2013; KURNIAWAN et al., 2021; XIANG et al., 2020).

However, full-scale applications of ammonia stripping may be costly, making nitrogen recovery from membrane concentrates unfeasible. Dos Santos et al. (2020) estimated the total cost of ammonia recovery from concentrated leachate at 51.64 USD m^{-3}. This cost was mainly dependent on the price of the tower (in which the mass transfer occurs), chemicals for absorption, and electricity consumption (KURNIAWAN et al., 2021). In each scenario, a detailed techno-economic evaluation should be performed, considering the process energy demand, system robustness, product quality, and local market demands.

In the precipitation method, nutrients are recovered via struvite precipitation. Struvite ($MgNH_4PO_4{\cdot}6H_2O$) is a phosphate mineral and can be used as a slow-release fertilizer or raw material for the chemical industry. In struvite precipitation, an alkaline solution is obtained either by the addition of alkali solution or aeration stripping of carbon dioxide, followed by the introduction of magnesium salts for $MgNH_4PO_4{\cdot}6H_2O$ formation, which has a 1:1:1 molar ratio of ammonium (NH_4^+), phosphate (PO_4^{-3}), and magnesium (Mg^{+2}). When the concentrations of NH_4^+, PO_4^{-3}, and Mg^{+2} exceed their solubility limit, struvite formation recovers both N and P from leachates (LI et al., 2019). The main drawback is that, in general, struvite precipitation requires external magnesium and phosphorus to promote struvite crystallization. As leachates contain less magnesium and phosphate ammonium, a large amount of chemicals may be required. Besides, P recovery is ideal at a pH higher than 9.5, requiring the addition of an alkaline solution (KURNIAWAN et al., 2021).

Moreover, struvite precipitation is susceptible to interference by heavy metals and some inorganic ions like Ca^{+2}, K^+, Fe^{+3}, and CO_3^-, which also reduces the purity of the recovered mineral. Therefore, a compromise between process control and cost-

effectiveness is needed to ensure the product quality and the process sustainability with more economic benefits (LI et al., 2019; XIANG et al., 2020). According to (KURNIAWAN et al., 2021), P recovery from waste streams is hardly carried out because the cost of the recovered P is higher than that of natural rock-phosphate and the current technologies have a long cost recovery time of up to seven years.

Recently, techniques such as microbial electrolysis, MD, and FO have been proposed for nutrients recovery of LFLs (QIN et al., 2016; XIE et al., 2016; ZICO et al., 2021). Considering the demerits of each technology, the hybridisation of these processes with existing precipitation methods could benefit nutrients reclamation from waste streams. Hybrid systems could improve nutrient recovery efficiency and integrate different resource platforms, making nutrient recovery cost-effective and more attractive to be an option for valorisation of LLMC streams (XIE et al., 2016).

A submerged FO process linked to struvite precipitation was proposed to focus on both water and nutrient extraction. Three arrangements were tested to determine the optimal configuration about effects of struvite recovery on the FO performance: FO — calcium pretreatment — struvite precipitation (A1), calcium pretreatment — FO — struvite precipitation (A2), and calcium pretreatment — struvite precipitation — FO (A3). The A2 system was the optimum arrangement in terms of FO performance. Calcium pretreatment mitigated FO membrane fouling and improved the purity of the obtained struvite. The submerged FO system efficiently recovered water from the leachate and reduced its volume by 37%. The recovered mineral in chemical precipitation had a similar crystal structure and composition to that of standard struvite. The proposed system recovered about 4.34 kg struvite and 366 kg of water per m^3 of treated leachate in optimum conditions. The net profit was estimated at 0.80 USD m^{-3} (WU et al., 2018).

5.3.3.4   Inorganic salts

NFC and ROC from LLTPs contain valuable inorganic ions such as Na^+, K^+, and Cl^-, often located at levels higher than 890, 210, and 1280 mg L^{-1}, respectively (Table 5-1). Some of these inorganic species like potassium ions are scarce and therefore could exist a driving force behind their possible extraction. Other elements like chloride

ions could be recovered as mixed salts or even transformed into high add-value products. Therefore, in this scenario, mineral reclamation of waste streams could make leachate treatment more sustainable and potentially economical (HUANG et al., 2020; LE DIRACH et al., 2005; LI et al., 2015).

At present, few published studies focus on mineral recovery from concentrated leachates. Most of the researches has investigated mineral extraction from sea-water brines. For example, Mohammadesmaeili et al. (2010) applied RO and isothermal evaporation to manage lima-soda pretreated concentrates from desalination plants. The proposed system recovered freshwater and produced mixed solid salt (i.e., Na_2SO_4 and NaCl) with potential resale value. However, it should be noted that the current energy demand of thermal-based technologies (7.7—72 kWh m^{-3}) can hinder field applications (PANAGOPOULOS, 2021). Therefore, more studies are still needed to evaluate the process economics considering the influence factors of each situation.

A combined process composed of cation-exchange membrane electrolysis and chemical precipitation was developed to simultaneously treat NF concentrates and recover K^+ and Cl^- ions as commercial by-products. The combined system exhibited excellent treatability results and allowed efficient recovery of gaseous chlorine and potassium-containing struvite. However, a preliminary economic analysis showed that the net profit of products resale would not offset the system's operating costs, which were most associated with the high electricity consumption of the electrochemical process (LI et al., 2015).

In another work, through a simple chemical precipitation method, antichlors of Bi(III) containing oxides with quantum dots (QDs) or two-dimensional (2D) structures were synthesized and then mixed with spinel ferrites ($M-Fe_3O_4$) and titanium dioxide (TiO_2) to combine with Bi_2O_3 for magnetic recycling and photocatalysis improvement. The constructed antichlor was then used to treat concentrated leachates. Under optimum conditions, Cl^- removal efficiencies ranged from 60 to 90%. The treatment of concentrated leachates with the antichlors led to the formation of Bi-precipitates that contained coated BiOCl on the residual Bi_2O_3/TiO_2 structure. This by-product was recovered and further used to treat the dechlorinated leachate. It was concluded that

due to the excellent photocatalytic activity of the recovered material under UV—vis-near-infrared irradiation, better than that of commercial BiOCl in the mineralisation of methyl orange, they could be used as a photocatalyst for the degradation of organic compounds of dechlorinated wastewaters (HUANG et al., 2020). This research provides insights into chlorine removal techniques and the potential production of commercial photocatalytic materials. Further research in this field could help find alternatives for the valorisation of membrane concentrate and other chlorinated effluents.

5.4 FUTURE PERSPECTIVES AND MANAGEMENT DIAGRAM FOR RESOURCE RECOVERY

Environmental concerns and resources depletion is expected to accelerate greener and sustainable practices. Most of the literature findings are based on laboratory studies, showing that LLMC resource recovery systems are at the embryonic stage. At present, few technologies appear to be techno-economically applicable at a commercial scale, and some critical aspects (e.g., energy demand and process robustness) still need to be solved. Considering that future researches are required, we propose a recovery train to select the best recovery route. The proposed diagram can be helpful to define and test the overall performance of the selected recovery arrangement. In general, a sequential scheme for concentrate valorisation should include volume reduction with the recovery of clean water, followed by extraction of add-value materials. As discussed earlier, different types of LFLs and applied membrane treatment will result in different organic, salt, and nutrient contents in the LLMC. Figure 5-2 shows the recovery train for implementing the best management route based on the main characteristics of LLMCs.

The recovery scheme's first step incorporates FO for water reclamation. The FO process appears the most promising technology for volume reduction and water recovery from membrane concentrates. As FO permeate stream is not clean water but a diluted DS, a regeneration step is required. The Hybrid FO-RO process is more energy-efficient than standalone FO; therefore, coupling these processes helps save energy and reduce operational costs (SINGH et al., 2021).
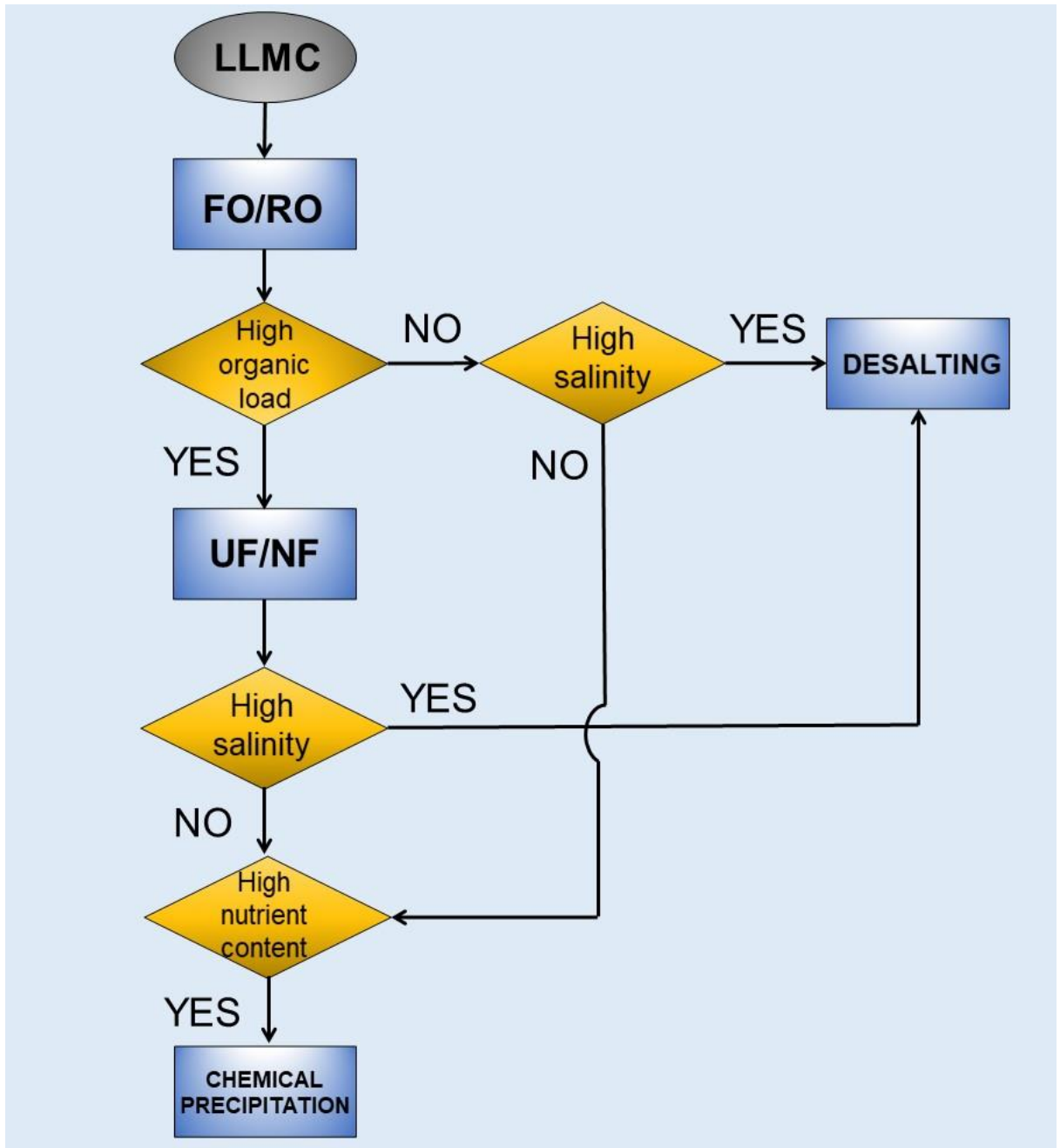
Figure 5-2 General recovery train for the selection of resource recovery route.

After volume reduction, a membrane-based process, i.e., tight UF or loose NF, can fractionate heavily organic streams, recovering HSs as liquid fertilizer. A desalting process should be employed if either FO concentrate stream or UF/NF permeate has high salinity. Thermal-based technologies like thermal evaporation and crystallization are the preferred systems for the valorisation of saline effluents (PANAGOPOULOS, 2021). However, high energy consumption and operational expenses can hamper its implementation in LLTPs. A renewable energy source such as biogas, commonly

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available in landfills, could produce energy input for desalting systems. Additionally, mixed salts (e.g., NaCl, Na_2SO_4, $CaCl_2$, and $MgCl_2$) resale can generate net profit and make them more economically attractive.

The tertiary step is optional if the quality of the concentrate stream is acceptable to be recycled back to the LLTP head or sent to the FO-RO system for further water recovery. On the other hand, high nutrient loads imply that instead of being moved back to the facility head, struvite could be precipitated out and sold as fertilizer. The treated effluent could then be recycled to the head of LLTP, reducing the effluent's nutrient load. The proposed recovery train intends to recommend LLMC management based on resource recovery approaches rather than treatment and disposal. Recommended technologies in the diagram are not limited to those described here. For example, emerging resource recovery systems have been tested in laboratory studies, and further research on pilot-scale is expected for its implications and economic feasibility. Bioelectrochemical systems (BESs) have grabbed attention for simultaneous nutrient reclamation and energy production from leachates (ISKANDER et al., 2018; QIN et al., 2016). Iskander et al. (2017) demonstrated that a BES producing 0.123 kWh m^{-3} could treat LFLs. Hybrid processes involving BESs coupled to FO might be promising to recover both water and chemicals, improving FO efficiency with less energy consumption. Low-cost phytoremediation systems can be implemented as a polishing step for nutrients and heavy metals removal depending on the concentrate composition. Harvested biomasses are useful for bioenergy production (biogas, biofuels, combustion for energy recuperation and heating) (GOMES, 2012; WIJEKOON et al., 2021). Thus, energy recovery can further improve the proposed system's overall economic and commercial viability.

The following aspects are recommended during the assessment of the defined resource recovery framework: 1) Material balance aiming to describe the resource recovery route in a quantitative way before its implementation; 2) detailed analysis of product quality, applicability, and local market demand for the recovered material; and 3) evaluation of potential impacts of the management route through a life cycle analysis perspective.

5.5 CONCLUSIONS

The state-of-the-art of current research about membrane concentrates from LLTPs was critically examined in this article. Although LLMC recycling into the landfill body is the convenient management option, a more sustainable strategy is recommended. A general recovery train for implementing the best LLMC recovery route was proposed within this context. Low energy demand and membrane fouling propensity have made FO indispensable for volume reduction and water reclamation of concentrate streams. Material extraction (e.g., fertilizers and inorganic salts) from the FO treated effluent could generate net profit and increase the system's economic feasibility. However, few technologies appear to be techno-economically applicable at a commercial scale, and some critical aspects (e.g., energy requirements and process robustness) still need to be solved. Future studies should focus on developing novel integrated systems combining benefits of each recovering technology, scale-up, techno-economic evaluation of recovering processes, and assessment based on a life cycle perspective (i.e., environmental impacts and carbon footprint). Besides, the extraction of non-conventional value-added products (e.g., catalysts and bio-fuels) via existing or novel technologies is a promising area for future investigations.

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Chapter 6.   Monitoring of Experimental Landfill Cells with Membrane Concentrate Infiltration: a Systematic Assessment of Leachate Quality and Treatment Performance

This chapter presents the findings of an experimental investigation that aimed to assess the impacts of membrane concentrate infiltration on the leachate quantity, quality, and treatment performance. Simulated landfill cells were constructed and monitored for 420 days. Concentrate samples collected from the full-scale leachate treatment facility identified in chapter 4 were used in the proposed assessment.

A final version of this chapter will be submitted as a research article in a specialized journal.

6.1 INTRODUCTION

Landfill leachate (LFL) is an inherent consequence of waste disposal in landfills. Due to its pollution potential, this complex wastewater must be adequately treated before discharging into a receiving environment (RENOU et al., 2008). Following the most restrictive standards for wastewater disposal over the years, advanced technologies have been added to several LFL treatment facilities. Among them, nanofiltration (NF) and reverse osmosis (RO) are used to achieve contaminants removal beyond conventional treatments and set the most stringent legislation standards (DE ALMEIDA et al., 2020). In 2018, the treatment capacity of NF/RO treating landfill leachates was more than 60k m^3 per day in China (CHEN et al., 2021). In European countries, due to the change of leachate quality linked to the ban on the landfilling of organic waste, membrane systems are expected to continue to play a key role in the leachate treatment chain since conventional processes (e.g., biological processes) are not suitable to handle methanogenic leachates (DI MARIA et al., 2018; WARWICK et al., 2018).

On the other hand, a heavily contaminated stream named leachate concentrate is produced during the membrane technology operation. Concentrate streams account for up to 30% of the incoming leachate (WANG et al., 2017). Landfill leachate membrane concentrate (LLMC) streams have high organic and salt content (total organic carbon (TOC): 2365—4500 mg L^{-1}, total dissolved solids (TDS): 18,910—66,900 mg L^{-1}) (KEYIKOGLU et al., 2021). Besides, a significant concentration of heavy metals and micropollutants not detected in the fresh leachate can also be found due to the membrane concentration effect (KEYIKOGLU et al., 2021; LI & LIU, 2021; WANG et al., 2016). Therefore, LLMC management is a crucial aspect to be considered to close the leachate treatment cycle.

Due to its convenience and simplicity, LLMC recirculation onto the landfill body is a commonly adopted management tactic (ISWA, 2019). However, the negative experience of concentrate recirculation in some sites started to grow the debate about its long-term sustainability. In the last decade, scientific interest emerged, and some important studies have been published. 15-year monitoring data of an Italian landfill

revealed a moderate change in leachate composition (slight increase in NH_4^+—N, Cl^-, and SO_4^{-2}) and leachate quantity. However, RO treatment performance was not impacted (CALABRÒ et al., 2018). In a previous study at the same Italian site, the authors observed a moderate rise in organic matter content and heavy metals (i.e., Ni and Zn). However, no significant change in LFL quantity was identified in this case (CALABRÒ et al., 2010).

Other research efforts were conducted to elucidate the impacts of LLMC infiltration. TALALAJ & BIEDKA (2015) assessed the infiltration of 375 m^3 per month of RO concentrate in a Polish landfill. An increase in mean values of sulphate, conductivity, ammonia, and chemical oxygen demand was reported after eight months of monitoring. DZOLEV & VUJIC (2016) observed a similar increase in leachate conductivity during 83 days of monitoring in a landfill in Bosnia-Herzegovina. In another work, WANG et al. (2017) simulated landfill cells filled with 1, 5, and 15-yr age waste to analyse the infiltration process in a 24-day experiment. The main findings indicated an increase in organics and ammonia in leachates from the one-year cells and a moderate increase in salinity and heavy metals in the five-year cells. Morello et al. (2016) evaluated the quantity and quality of leachates generated by lab-scale columns with membrane concentrate recirculation for 123 days. No consistent leachate quantity changes and organic content were noticed in this case.

Despite the associated limitations of the studies above, that is, time of monitoring, scale, or no assessment of leachate treatment performance, it has been argued that concentrate recirculation sustainability is site-specific and depends on several factors associated with the landfill site and its operational conditions. The literature's contrasting conclusions justify this management option, especially in emerging economies. For instance, in Rio de Janeiro State (Brazil), law number #9055/2020 permits the return to the landfill body of up to 1/3 of the leachate treatment byproducts, including sludge, biomass in excess, and membrane concentrate. To the best of our knowledge, LLMC reinjection has been adopted in other Brazilian landfills, where membrane systems are used in the leachate treatment chain.

Adopting such a practice without fully understanding its consequences can lead to unprecedented impacts in the LFL treatment chain and limit the development of greener management strategies. Within this context, the current study extended and complemented previous work by assessing the impacts of RO concentrate infiltration using simulated landfill cells in a 420-day experiment. The specific objectives are as follows: 1) compare the leachate quantity in the two operation modes, that is, with and without concentrate infiltration, 2) analyse the quality of the generated LFLs in terms of specific physicochemical parameters, and 3) evaluate RO treatment performance along the cells monitoring experiment.

6.2 MATERIALS AND METHODS

6.2.1 Research programme

Six experimental landfill cells were constructed and operated in different modes, i.e., without and with RO concentrate infiltration. The cells were planned considering the statistical assessment is carried out in triplicate; therefore, data were grouped and named control (C_0) and infiltration (C_i) groups. RO concentrate collected from full-scale facilities was infiltrated at rate of 900 mL per week, while LFLs were drained at least twice a month. Samples were taken in plastic bottles and transported to the laboratory for physicochemical characterisation. The quantity of leachate was recorded, and the local pluviometry index was monitored during the experiment. Once a quarter, integrated samples from C_0 and C_i groups were treated using a lab-scale RO system. As we are interested in leachate behaviour, gasses emission monitoring has not been taken into account in this work.

6.2.2 Leachate monitoring

6.2.2.1 Experimental landfill cells

Experimental landfill cells were constructed using 0.24 m^3 containers (74.5 × 59.5 × 100 cm) of high-density polyethene material. Valves were placed under each cell to drain and collect the generated leachates (Figure S.6- 1). Landfill cells were operated in two operational modes: without concentrate infiltration (control group, C_0) and with concentrate recirculation (infiltration group, C_i). Operating conditions were assessed in triplicate.

6.2.2.2 Household solid waste sampling

Household solid waste (HSW) used to fill the landfill cells was provided by Rio de Janeiro municipality's public waste management company. About one ton of sample was sieved and analysed following the procedure of ABNT (2004). The undersieved fraction (< 80 mm) was loaded into waste cells. Undersieved HSW composition was

characterized as one from an emerging economy, as follows: 48% of organic matter (mainly food waste), 16% of plastics, 13% of paper and cardboard, 3% of glass, 2% metals, and 18% inert and others (Alfaia et al., 2017; de Almeida et al., 2021). Each landfill cell was filled with the same waste composition and density (ca. 600 kg m^{-3}). Moisture content, total solid (TS), and volatile solids (VS) of the HSW sample were about 40%, 76%, and 82%TS, respectively.

6.2.2.3   RO concentrate samples

RO concentrate (ROC) samples were collected from full-scale leachate treatment plants in Rio de Janeiro State (Figure S.6- 2). Three campaigns were carried out between 2019 and 2021 to characterize the concentrate samples considering different seasonal and operational landfill conditions, leading to possible changes in the composition of the generated membrane concentrate. The full chemical characterisation is provided in Table 6-1.

The LLMC infiltration was performed at a rate of 900 mL per week. The concentrate infiltration rate was based on a hydraulic balance to mimic a local full-scale landfill that operates at LLMC infiltration mode. We monitored the pluviometry precipitation using a pluviometry device. The recorded value during the 420-day experiment was 1313.5 mm. Considering the experimental cell scale, the system's evapotranspiration was neglected. The input and output of water were not considered in our assessment. The comparative analysis of leachate production considered only the input of membrane concentrate on the cells of the infiltration group.

Table 6-1 Characterisation of leachate concentrate samples used in the infiltration experiment (n = 3).

Parameters	Min–Max	Mean
pH	7.6–8.8	8.0
BOD_5 (mg L^{-1})	345–3402	1541
COD (mg L^{-1})	3678–11,730	7480
DOC (mg L^{-1})	4556-5098	4682
UV_{254} (cm^{-1})	56.71–244.22	122.01
NH_3–N (mg L^{-1})	2172–13,633	7817
Alcalinity (mg L^{-1})	1140–2344	1320
True colour (mg Pt-Co L^{-1})	7828–8542	8185
Turbidity (NTU)	508–1628	1068
EC (mS cm^{-1})	40.11–72.25	54.17
Cl^- (mg L^{-1})	4567–5938	5127
Na^+ (mg L^{-1})	9974–11,543	10,689
K^+ (mg L^{-1})	3065–3400	3377
Ca^{+2} (mg L^{-1})	309–397	378
Mg^{+2} (mg L^{-1})	1189–1267	1209
SO_4^{-2} (mg L^{-1})	4567–8150	6792
Cu (mg L^{-1})	1.06–2.06	1.67
Ni (mg L^{-1})	0.98–1.45	1.24
Pb (mg L^{-1})	0.33–0.76	0.55
*Ecotoxicity (*Vibrio fischeri*) [EC50(%); TUa]	—	0.1745; 573

pH – potential hydrogen. BOD_5 – 5-day biochemical oxygen demand. COD – chemical oxygen demand. DOC – dissolved oxygen demand. EC – electroconductivity. EC50 – effective concentration. NH_3–N – ammonium nitrogen. TUa – toxicity unit acute. UV_{254} – absorbance at 254 nm. *Ecotoxicity was performed for one of the collected membrane concentrate sample.

6.2.2.4 Monitoring of landfill cells

LFLs were drained at least two times per month. The collected leachate was tested for pH, BOD_5, COD, Absorbance at 254 nm (UV_{254}), ammonium nitrogen (NH_3–N), electroconductivity (EC), total dissolved solids (TDS), chloride (Cl^-), sulphate (SO_4^{-2}), metals (Na^+, K^+, Ca^{+2}, Mg^{+2}), and heavy metals (Cu, Ni, and Pb). pH, COD, UV_{254}, NH_3–N, TDS, and EC have been monitored in all leachate collections. Table 6-2 shows the conditions and monitored parameters during the monitoring period.

Table 6-2 Conditions of landfill cells and monitoring parameters of leachates.

Groups	HSW composition	HSW density (kg m^{-3})	HSW moisture (wt%)	Total solid (TS) (%wt)	Volatile solid (wt%)	Concentrate volume weekly infiltrated (mL)	Monitored leachate parameters		Monitored parameters during RO treatment assessment	
							Weekly	Once a quarter	Before treatment (compost sample)	After treatment (permeate)
C_0 (Cells 1, 2, and 3)	Organic matter (food waste) – 48%, 16% of plastics – 16% paper and cardboard – 13%, glass – 3% metals – 2%, inert and others – 18%	600	40	76	82%TS	0	pH, COD, UV_{254}, TDS, conductivity, and NH_3─N	BOD_5, SO_4^{-2}, Cl^-, metals (Na^+, K^+, Ca^{+2}, Mg^{+2}), heavy metals (Cu, Pb and Ni)	pH, COD, UV_{254}, TDS, conductivity, and NH_3─N	pH, COD, UV_{254}, TDS, conductivity, NH_3─N
		600	40	76	82%TS	0				
		600	40	76	82%TS	0				
C_i (Cells 3, 4, and 6)	Organic matter (food waste) – 48%, 16% of plastics – 16% paper and cardboard – 13%, glass – 3% metals – 2%, inert and others – 18%	600	40	76	82%TS	900	pH, COD, UV_{254}, TDS, conductivity, and NH_3─N	BOD_5, SO_4^{-2}, Cl^-, metals (Na^+, K^+, Ca^{+2}, Mg^{+2}), heavy metals (Cu, Pb and Ni)	pH, COD, UV_{254}, TDS, conductivity, and NH_3─N	pH, COD, UV_{254}, TDS, conductivity, NH_3─N
		600	40	76	82%TS	900				
		600	40	76	82%TS	900				

pH – potential . BOD5 – 5-days biochemical oxygen demand. COD – chemical oxygen demand. DOC – dissolved oxygen demand. EC – electroconductivity. EC50 – effective concentration. NH_3─N – ammonium nitrogen. UV_{254} – absorbance at 254 nm. *INMET (2022)

6.2.3 Treatment of LFLs

Once a quarter, integrated samples from C_0 and C_i groups were treated using a lab-scale RO system. Compost samples were named campaigns 1, 2, and 3. Stirred Cell (PAM Selective Membranes Inc.) was used to carry out the filtration in dead-end-filtration mode providing an active membrane area of 63.59 cm^2 (A_m). The system has a total volume capacity of 1.5 litres. A nitrogen gas manometer and pressure regulators apply the desired pressure inside the cell (Figure 6-1).
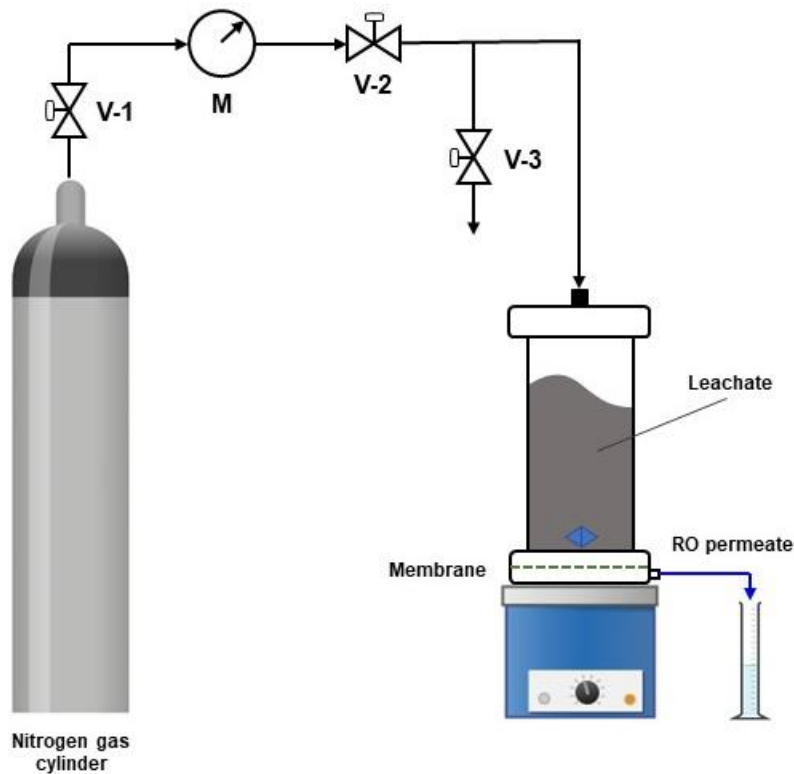
Figure 6-1 RO experimental setup. M - manometer; V-1 – nitrogen gas cylinder valve, V-2 – nitrogen gas control, V-3 – nitrogen exhaust valve.

The leachate treatment was performed with a polymeric commercial membrane of polyamide (BW30-4040, Dow Filmtec Membranes). It has sodium chloride rejection of 99.5% and works at a maximum operating pressure of 41 bar. BW30-4040 membrane was supplied by Dow Filmtec Membranes. Membrane hydraulic permeability (L_p) was determined to be 1.019 L m^{-2} h^{-1} bar^{-1} (L_p = 0.9934–1.488 L m^{-2} h^{-1} bar^{-1}, n = 7).

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The leachate samples were treated at an operating pressure of 20 bars. Before filtration experiments, the RO membrane was compacted with ultrapure water at 5 bar for 2 hours to stabilize the permeate flux. Thereafter, the RO system was fed with 500 mL of leachate and conducted in the concentration mode for 48 hours. Leachate samples were pretreated using 0.45 µm cellulose filters to mimic RO pretreatment by cartridge filters applied at full-scale facilities. The permeate samples were carefully collected and stored at 4°C for physicochemical characterisation. Treatment performance assessment was based on permeate flux along the filtration process, fouling resistance, and permeate quality (i.e., removal efficiencies of pollution parameters). RO treatment of leachate samples was performed in duplicate.

The permeate flux (J) was calculated by Equation 6.1. According to the Darcy equation, membrane flux at any time can be expressed by Equation 6.2 (LIU et al., 2021).

$$J = \frac{Q_p}{A_m} \tag{6.1}$$

$$J = \frac{\Delta P}{\mu(R_m + R_f)} \tag{6.2}$$

Where Q_p, ΔP and μ are the permeate flowrate (L h^{-1}), operating pressure (Pa), and permeate viscosity (Pa s), respectively. R_m and R_f represent the membrane resistances induced by membrane material (m^{-1}) and deposited foulants (m^{-1}). It was assumed permeate viscosity (μ) value of water (1.002×10^3 Pa s). R_m is estimated using ultrapure water as RO feed at a treatment operating pressure of 20 bars. The fouling resistance (R_f) was calculated to compare the fouling propensity of membranes treating leachate samples from C_0 and C_i groups.

6.2.4 Analytical methods

BOD_5, COD, NH_3–N, TDS, SO_4^{-2}, Cl^-, Na^+, K^+, Ca^{+2}, Mg^{+2}, and heavy metals were performed following the Standard Methods (APHA/AWWA/WEF, 2012). pH and EC were measured on-site using probes.

6.2.5 Statistical and data analysis

Although some parameters had a trend for normal distribution verified by Shapiro-Wilk's test, the Mann-Whitney U test was preferred for two independent groups, checking the significance difference by employing the hypothesis test. All statistical analyses were performed using Statistica 10.0 software (*Stat Soft*) at a significance level (α) of 5% (α=0.05).

6.3 RESULTS

6.3.1 Monitoring of landfill cells

Figure 6-2 shows the data of leachate production and median values in the 420-day monitoring experiment.

Values of leachate production for C_0 and C_i groups ranged from 228 – 35,147 mL and 278 – 5491 mL per week, respectively (Table S.6- 1). Statistical analysis using the Mann-Whitney U test showed that the quantity of LFLs produced in the two operational conditions was insignificant during the experimental investigation (p-value = 0.7509). In total, about 138 litres were recirculated onto the infiltration group. However, the leachate quantity generated in the control group was higher than that of the Ci group (194 versus 130 litres). Interestingly, the accumulated leachate volume of Ci group can be described as a first-order linear equation until 177-day (y = 804.43, R^2 = 0.995).
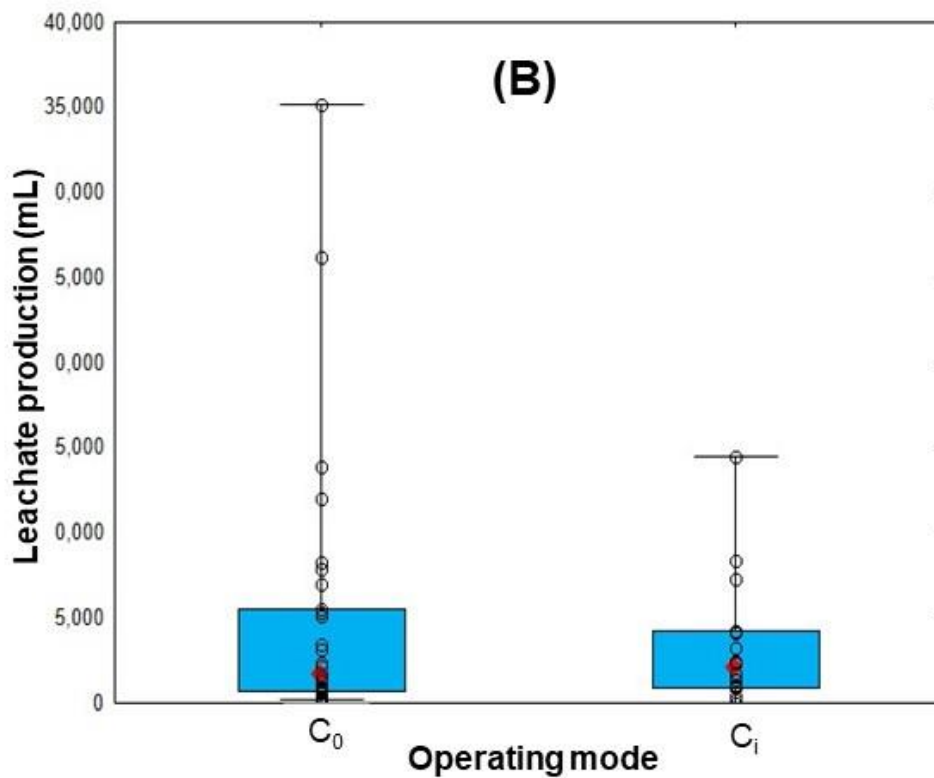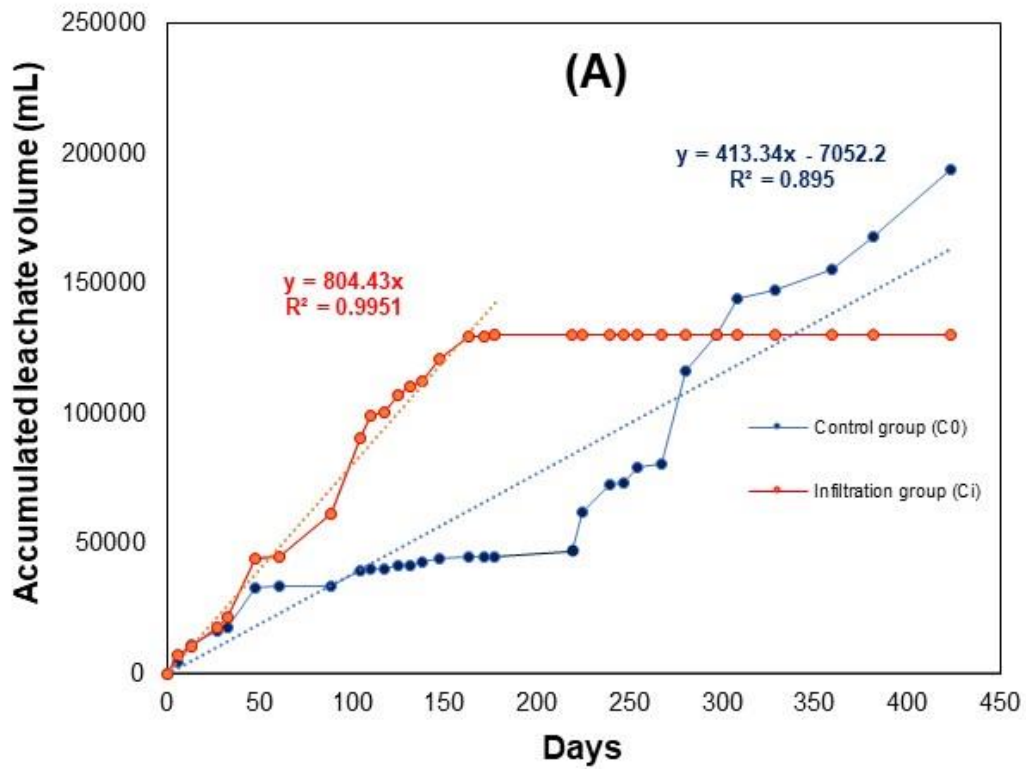
Figure 6-2 Leachate production of landfill cells in 420-day monitoring experiment.
A) Accumulated leachate volume during the investigation and B) Median values of C_0
and C_i groups' leachate production in 420-day monitoring experiment.

Figure S.6- 2 shows the physicochemical characterisation of leachates generated by experimental landfill cells in different operating conditions during the 420-day monitoring. It is important to note that the comparative data of LFL generation in the two operating conditions were registered until the 254-day. Therefore, physicochemical characterisation of leachates is shown considering 254 days, which provides a comparative assessment. Median values of pH, NH_3–N, COD, UV_{254}, conductivity, and TDS in the 254-day experiment are illustrated in Figure 6-3.

In a 254-day experiment, median values of pH, NH_3-N, conductivity, and TDS between leachates from C_0 and C_i groups were statistically insignificant (p-value > 0.05). On the other hand, COD and UV_{254} parameters showed a significant difference (p-values = 0.0286 and 0.0041, respectively). The median values were higher in the infiltration group.

BOD_5/COD ratio, salts and heavy metals analyses, performed once a quarter during the monitoring of the simulated landfill cells, are shown in Table 6-3. The biodegradability of leachates from both simulated cell groups had a similar trend. BOD_5/COD values ranged from 0.69 – 0.09 and 0.70 – 0.07 for C_0 and C_i groups, respectively. Similar behaviour was also reported for salt concentrations such as Na^+, K^+, Ca^{+2}, Mg^{+2}, and Cl^-. In this sense, sulphate was an exception since it was observed with higher values in leachates from the Ci group from the first to the last week of monitoring. This result is analysed in the discussion section. Heavy metals (i.e., Ni, Pb, and Cu) were below local discharge limits (Conama #430/2011) for both cell groups from the 177-day.
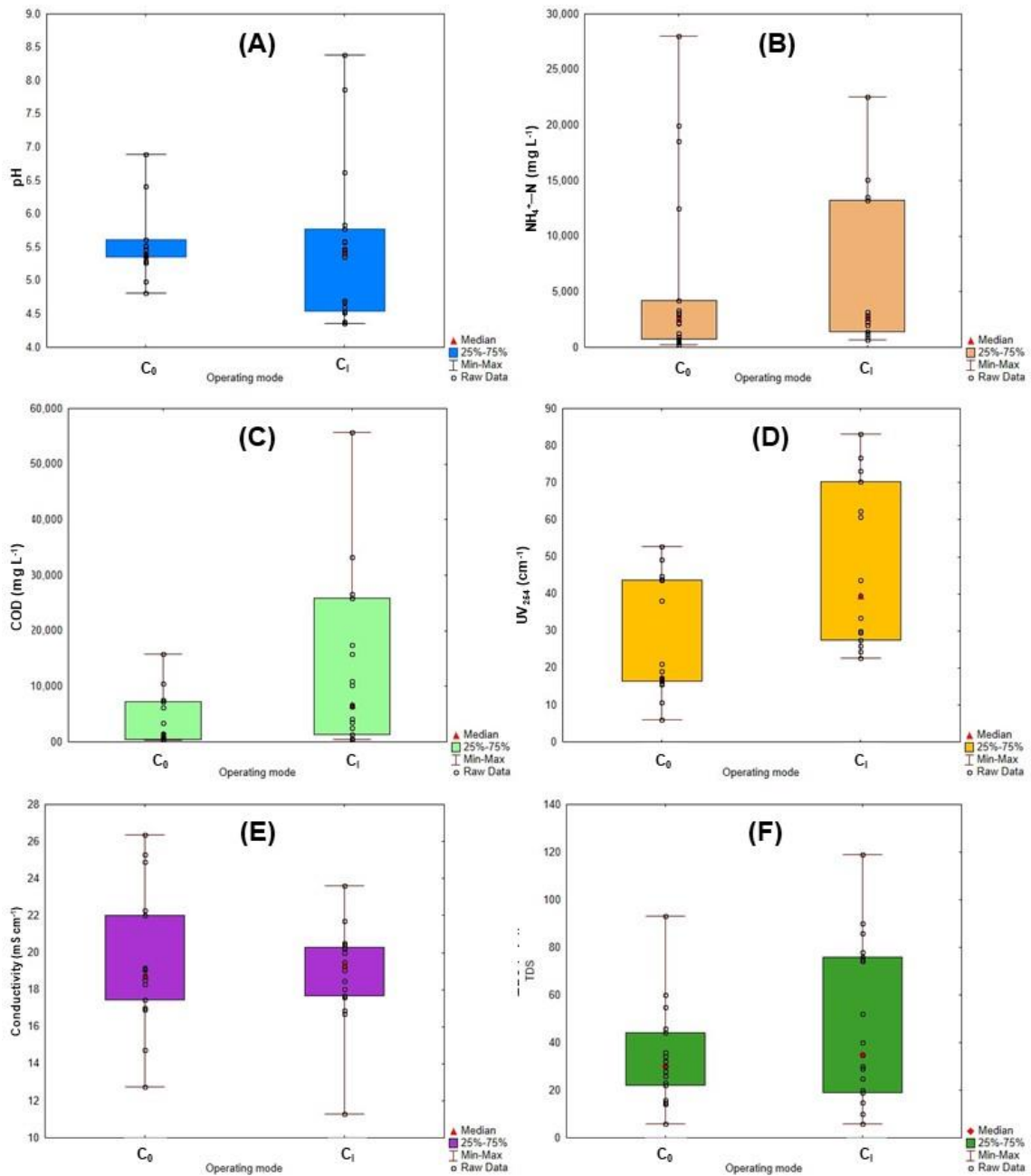
Figure 6-3 Median values of leachate parameters in 254-day monitoring. A) pH; B) NH_4^+-N; C) COD; D) UV_{254}; E) conductivity, and F) TDS.

Table 6-3 Once a quarter analysis of chemical parameters for leachate samples ($n \leq 3$).

Date	Days	BOD_5/COD		Na^+ (mg L^{-1})		K^+ (mg L^{-1})		Ca^{+2} (mg L^{-1})		Mg^{+2} (mg L^{-1})		Sulphate (SO_4^{-2}) (mg L^{-1})		Chloride (Cl^-) (mg L^{-1})		Nickel (Ni) (mg L^{-1})		Lead (Pb) (mg L^{-1})		Copper (Cu) (mg L^{-1})	
		C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci	C_0	Ci
03/2021	6	0.68—0.69	0.69—0.70	10,783	9888	2098	3098	150	166	343	267	2335	7890	5988	5339	0.87	0.98	0.91	0.96	0.85	1.55
06/2021	89	0.27	0.12	5789	7899	1390	1278	76	88	123	222	1025	5331	2991	3443	<0.1	0.49	0.33	0.86	<0.1	1.45
09/2021	177	0.20	0.09	2897	8994	1209	1456	13	27	145	208	210	5443	1056	2334	n.d	<0.1	n.d	1.78	<0.1	0.33
11/2021	254	0.09	0.07	3090	5880	1067	1767	55	77	108	199	345	4222	977	1560	n.d	n.d	n.d	n.d	n.d	n.d

n.d: not detected. Limit of quantification = LOQ = 0.01 µg L^{-1}.

6.3.2 Treatment of LFLs

Table 6-4 shows the treatment results of leachate compost samples collected from C_0 and C_i groups. It should be highlighted that three campaigns were assessed because, with the reduction of leachate production in the C_i group, we could not collect a fourth integrated sample. The behaviour of permeate fluxes and fouling resistances during the treatment of samples by RO is shown in Table 6-5.

Table 6-4 Treatment results of leachate compost samples from C_0 and C_i landfill cells groups.

Parameters	Campaign 1, raw leachate (Mean)		After RO treatment (Removal efficiency, %)[*], [a] (mean)		Campaign 2, raw leachate (Mean)		After RO treatment (Removal efficiency, %)[*], [a] (mean)		Campaign 3, raw leachate (Mean)		After RO treatment (Removal efficiency, %)[**], [a] (mean)	
	C_0	C_i	C_0	C_i	C_0	C_i	C_0	C_i	C_0	C_i	C_0	C_i
pH	5.10	4.68	4.81	4.29	5.34	5.36	5.09	5.12	7.22	6.92	7.7	7.10
COD (mg L^{-1})	114,700	132,600	68	47	3500	9880	25	74	300	1133	20	34
UV$_{254}$ (cm^{-1})	39.99	85.92	83	85	12.05	24.50	75	89	5.53	22.77	42	67
TDS (mg L^{-1})	10,717	10,728	15	10	10,195	13,461	43	58	3221	5376	38	46
EC (mS cm^{-1})	26.12	26.63	25	23	20.89	26.03	39	53	6.45	7.25	35	45
NH$_3$-N (mg L^{-1})	10,525	10,517	93	91	723	823	13	30	261	267	12	16

COD = chemical oxygen demand. EC = electroconductivity. UV$_{254}$ = absorbance at 254 nm. TDS = total dissolved solids. NH$_3$–N = ammonium nitrogen. [*]Results are shown for RO treatment after 48 h of system operation. [**]Results are shown for RO treatment after 24 h of system operation. [a]Except pH values, which are shown the mean values of raw leachate and RO permeate.

Table 6-5 Permeate flux and fouling resistance during the RO treatment of leachate compost samples.

Time (h)	Campaing 1				Campaing 2				Campaing 3			
	C_0		Ci		C_0		C_i		C_0		Ci	
	J (L m^{-2} h^{-1})	R_f (m^{-1})	J (L m^{-2} h^{-1})	Rf (m^{-1})	J (L m^{-2} h^{-1})	Rf (m^{-1})	J (L m^{-2} h^{-1})	Rf (m^{-1})	J (L m^{-2} h^{-1})	Rf (m^{-1})	J (L m^{-2} h^{-1})	Rf (m^{-1})
0.25	0.9435	0.73	0.2516	2.82	0.1510	4.61	0.1573	4.42	4.8435	0.1100	0.2048	3.32
1	0.3460	2.04	0.1651	4.32	0.2831	2.44	0.8177	0.82	13.2096	0.0154	0.2359	3.01
2	0.3460	2.04	0.1573	4.54	0.1887	3.68	0.7863	0.86	10.6935	0.0252	0.2201	3.23
3	0.2202	3.23	0.1127	6.34	0.3145	2.19	0.7234	0.94	11.9516	0.0280	0.2417	3.54
4	0.2202	3.23	0.1127	6.34	0.3460	1.99	0.6919	0.98	7.5484	0.0568	0.3246	2.21
5	0.3774	1.87	0.2359	3.01	0.3774	1.82	0.4892	1.32	6.2903	0.0712	0.3775	1.81
6	0.0629	1.14	0.2202	3.23	0.4089	1.68	n.r	n.r	4.0887	0.1290	0.3458	1.98
7	0.1573	4.54	n.r	n.r	0.3774	1.82	n.r	n.r	4.0887	0.1290	0.3333	2.01
8	0.1139	6.27	n.r	n.r	0.2949	2.34	n.r	n.r	4.0887	0.1290	0.3229	1.85
24	0.1573	4.54	0.0472	15.20	0.1887	3.58	0.3460	1.89	0.7077	0.8290	0.3450	1.84
25	0.1415	5.04	n.r	n.r	0.1887	3.58	0.3774	1.73	—	—	—	—
26	0.1258	5.68	n.r	n.r	0.3460	1.94	0.4089	1.59	—	—	—	—
27	0.2359	3.01	n.r	n.r	0.1573	4.31	0.4089	1.59	—	—	—	—
28	0.0157	4.57	n.r	n.r	0.2202	3.07	n.r	n.r	—	—	—	—
29	0.1415	5.04	0.2516	3.54	0.1739	3.91	n.r	n.r	—	—	—	—
30	0.1415	5.04	0.1110	8.06	0.2044	3.31	n.r	n.r	—	—	—	—
48	0.1415	5.04	0.1258	7.11	0.1997	3.32	0.4023	1.58	—	—	—	—

n.r = not registered. Data not registered means that the RO filtration system did not produce sufficient permeate volume to estimate permeate flux and fouling resistance at t time.

Looking at the pH values of the generated wastewaters in both landfill cell groups, we can infer that collected leachates from campaigns 1 and 2 are in the acidogenic stage, while those from campaign 3 are in the methanogenic phase. Concerning the permeate quality and comparing parameters statistically different, i.e., COD and UV_{254}, removal efficiencies ranged from 25 – 68% (C_0) and 47 – 74% (Ci) for COD and 75 – 83% (C_0) and 85 – 89% for UV_{254} at acidogenic phase, respectively. In the methanogenic phase, removal efficiency values of these pollution parameters were below 70%. In this case, removal efficiencies were higher when the RO system was used to treat leachate samples from the Ci group.

Removal efficiencies decrease as low as NH_3–N concentration in the RO feed for the ammonia parameter. The ammonia nitrogen removal was higher than 90% with an affluent concentration >10,000 mg L^{-1} and decreased to below 20% when the fed concentration was < 300 mg L^{-1}. Figure 6-4 illustrates raw leachate and permeates from the RO filtration bench system.

Median permeate fluxes were 0.1533 L m^{-2} h^{-1} (C_0) and 0.1466 L m^{-2} h^{-1} (Ci) for the campaign 1 (p-value > 0.05). Fouling resistance was 4.4441 m^{-1} and 4.5791 m^{-1} for C_0 and Ci, respectively (p-value > 0.05). On the other hand, J and R_f obtained during the treatment of leachate samples from campaigns 2 and 3, collected from C_0 and Ci groups, were statistically significant (p-value < 0.05). At the latter stage of leachate monitoring (i.e., methanogenic phase), J and R_f median values were 6.9200 L m^{-2} h^{-1} (C_0) and 0.3238 L m^{-2} h^{-1} (Ci) (p-value = 0.000182) and 0.0906 m^{-1} (C_0) and 2.11 m^{-1} (C_i) (p-value = 0.000179), respectively.
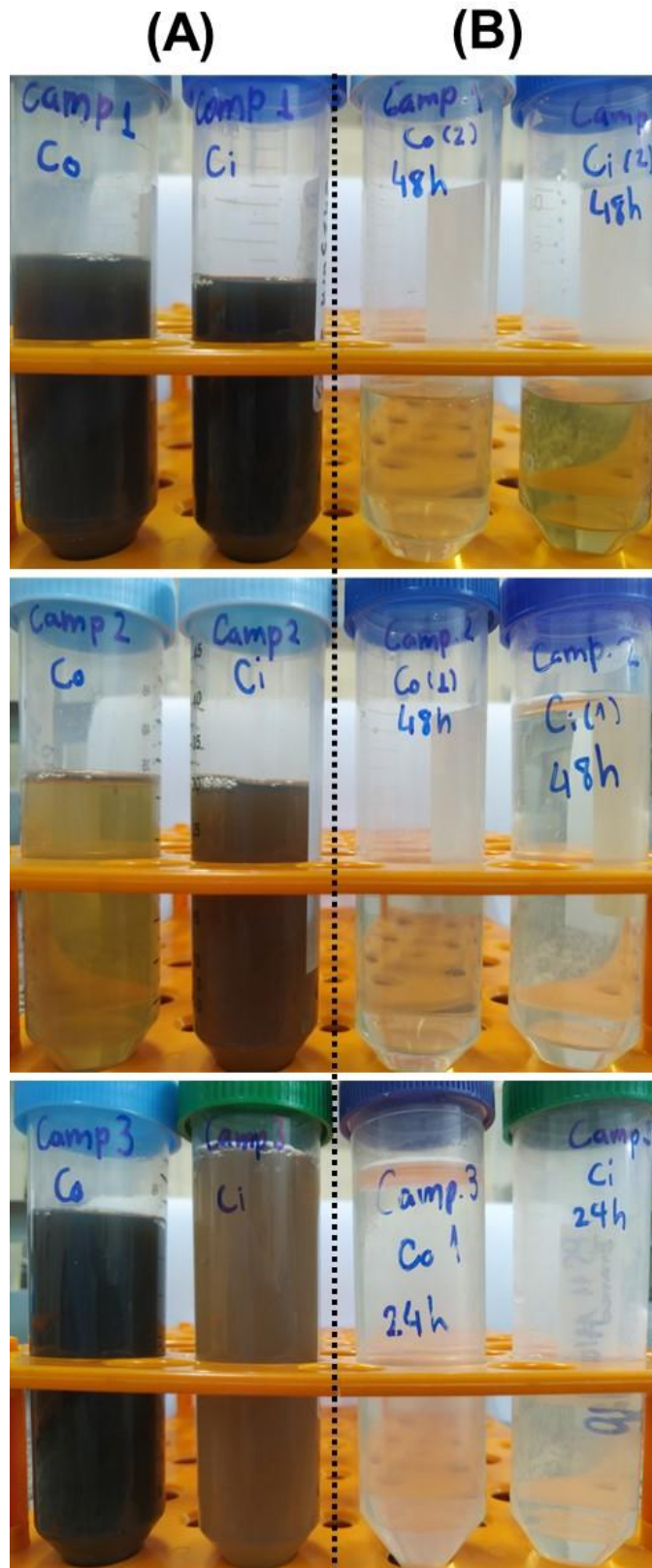
Figure 6-4 The visual aspect of raw and treated leachates. A) Raw leachate from campaigns 1, 2 and 3. B) Respective permeates obtained in the RO treatment.

6.4 DISCUSSION

6.4.1 Landfill cell monitoring

No statistically significant change in the leachate quantity was observed in this study. Even though our analysis provides a start-point to reflect on the impacts of concentrate recirculation on the leachate production in landfills that use such practice, this result should be carefully interpreted since from a 219-day experiment, the leachate production of C_i cells was diminished. The invariability of wastewater generation happened after a working incident. Consequently, the cells were rebuilt, and from there on, leachate generation was not uniform among C_0 and Ci cells. We hypothesized that short-circuits of the percolation ways and/or blockage by incrustation inside the cells were responsible for hampering the leachate draining. An autopsy of experimental cells may be helpful to elucidate such behaviour in future investigations.

Concerning leachate production in landfills that operate at membrane concentrate recirculation mode, contrasting conclusions are reported in the literature. Data from 123-day monitoring performed by Morello et al. (2016) showed that RO concentrate infiltration in simulated cells did not change the leachate quantity. In another work, Calabrò et al. (2010) assessed the concentrate reinjection practice in an Italian landfill. The results from this 30-month experiment showed that infiltration practice did not change the leachate produced in the full-scale landfill. Later, the same research group published a 10-year monitoring assessment with a different conclusion.

The pollution parameters analysis revealed that no significant change in pH, NH_3–N, conductivity, and TDS values was observed after 254-days of monitoring. In contrast, RO concentrate infiltration altered the leachate's organic content reported as COD and UV_{254}. The latter parameter is related to the aromatic content of organic substances (LIMA et al., 2017). Higher COD amount after concentrate recirculation was also reported in leachates analysed by Robinson (2005), Calabrò et al. (2010), and Talalaj & Biedka (2015). This phenomenon could be associated with the high organic input from the concentrate recirculation process. Since the UV_{254} parameter showed an increasing trend, microorganisms could be unable to degrade complex

organic carbon leading to the accumulation of organic matter in the drained leachate. BOD_5/COD ratio values at the later stage of landfill cells (0.07 – 0.09) show that the leachate biodegradability is low. Therefore, anaerobic digestion inside the landfill site is hampered due to the recalcitrant content of organic matter. Besides, considering the high toxicity of injected concentrate (i.e., TUa > 500), biological inhibition effects could also play a key role in this process.

On the other hand, Robinson (2005) and Talalaj & Biedka (2015) observed an increase in leachate's ammonia and conductivity in their analysis. However, it must be noted that these authors performed no statistical assessment. In the present study, although both Ci median values were higher than those for leachate from C_0 group (2675 vs 2432 mg L^{-1} and 19.72 vs 18.50 mS cm^{-1}), they were not statistically significant. Similar analysis occurred for TDS (37.5 vs. 30.0 g L^{-1}, p-valor > 0.05). One of the first study on this topic hypothesized that inorganic chemical reactions (e.g., precipitation and crystallization) cause the fixation of salts onto the landfilled waste, which can explain such results (HENIGIN, 1995).

Sulphate was detected in a higher quantity in leachates from Ci cells than that in C_0 group. This result can be justified by the high quantity of SO_4^{-2} ions in RO concentrate injected into landfill cells. We collected the RO concentrate from a full-scale facility that employs sulfuric acid in leachate pretreatment steps, which contributes to the high sulphate dose found in the concentrate (4567–8150 mg L-1, Table 1). Besides, sulfides can form precipitates with Ni, Pb, and Cu. The literature shows that in a landfill environment, heavy metals precipitate as sulfides and carbonates. Precipitates are fixed by adsorption onto the landfill waste, which often justifies the low concentration of heavy metals in the municipal solid waste leachate (KJELDSEN et al., 2002).

Finally, it is worth mentioning that other parameters, including dissolved organic carbon, turbidity, true colour, and ecotoxicity assays using *Vibrio fischeri* bacteria were performed, but not analysed yet. Data will be included in future examinations to expand this work's contribution.

6.4.2 Treatment of LFLs

The median of permeate fluxes was higher for leachate treatment from C_0 than that from the Ci group in all analysed campaigns. Values ranged from 0.9415 – 0.1415 and 0.2516 – 0.0472 L m^{-2} h^{-1} for C_0 and Ci in campaign 1, respectively. A similar trend was recorded during the treatment of leachates from campaign 2. However, the statistical analysis showed no significant difference between the groups. In contrast, permeate results were statistically significant for the leachate collected from a long-term monitoring period. The permeate flux was up to 858-fold higher treating the methanogenic leachate from the control group. This result is explained by comparing the membrane fouling resistances. Median values from infiltration and control groups were 0.0906 and 2.11 m^{-1} during the treatment of leachates, respectively. Given the increase in leachate organic matter, it can be inferred that organic fouling plays a major role during the treatment of leachate samples from Ci landfill cells. Overall, our findings suggest that the concentrate infiltration condition impacts the leachate treatment performance by RO. Our analysis is following Robinson (2005), who evaluated data from a German full-scale landfill in a 12-month monitoring experiment, concluding that the change in leachate composition affected the RO performance. According to the author, the landfill managers operated the membrane system for one year, after which it was removed from the site and replaced by a biological treatment system.

6.5 CONCLUSIONS

The present work assessed the impacts of concentrate infiltration practice on leachate quantity, quality, and RO treatment performance using simulated landfill cells. Major findings showed no significant change in pH, ammonia nitrogen, conductivity, and TDS after 254-day monitoring. In contrast, concentrate infiltration increased the leachate's organic content reported as COD and UV_{254}. Biochemical degradation inside the landfill cells was insufficient to reduce the organic load, specifically recalcitrant substances, leading to their accumulation in leachate solution. High organic content in Ci group's leachates greatly impacted RO treatment performance. Fouling resistance was 6-fold higher when the RO system was used to treat the methanogenic leachates generated by cells operating at concentrate infiltration mode. This result may be linked to higher operational expenses to clean and replace membrane modules at the full-scale facility.

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Supporting Content

Monitoring of experimental landfill cells with membrane concentrate infiltration: a systematic assessment of leachate quality and treatment performance
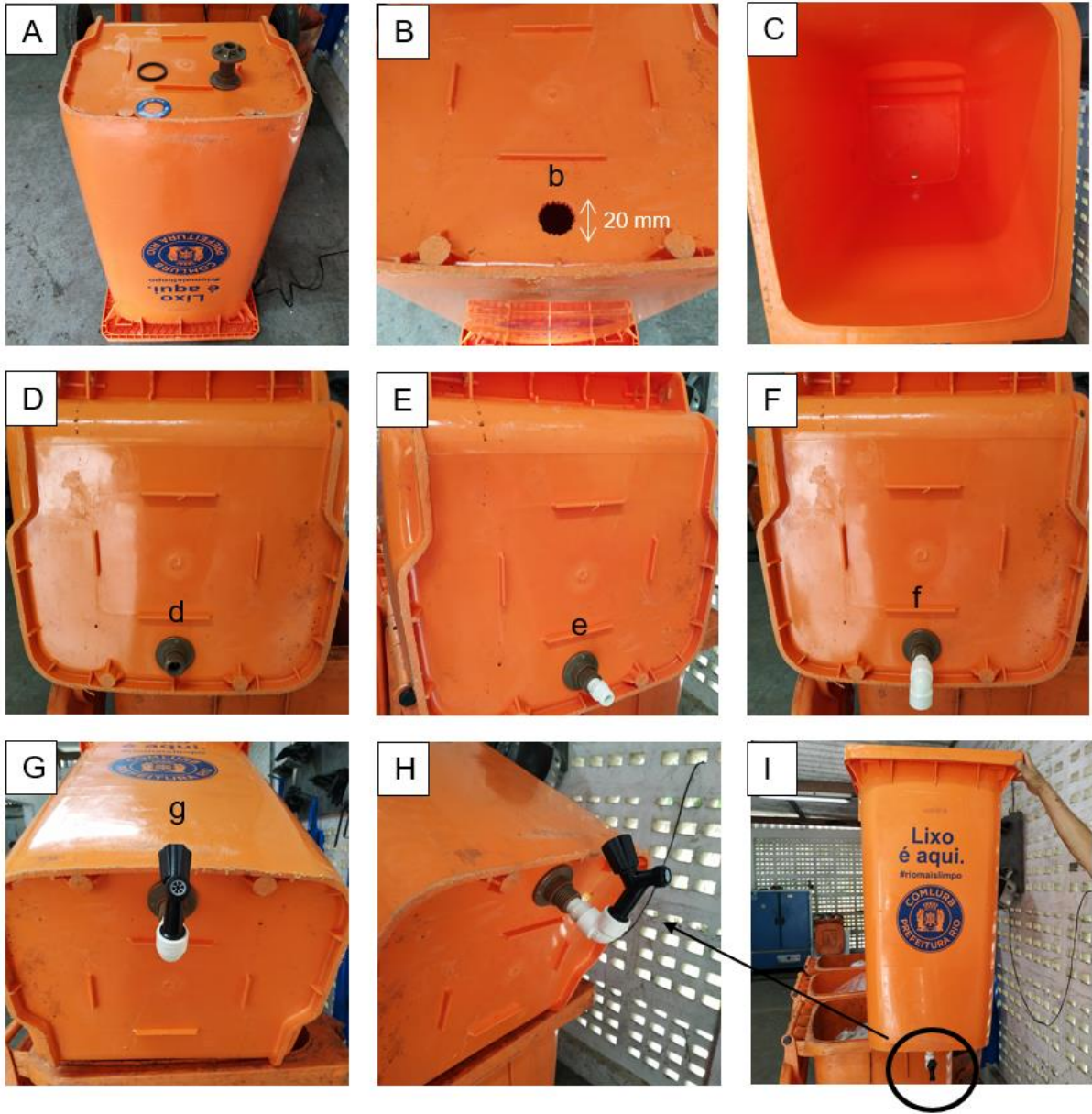
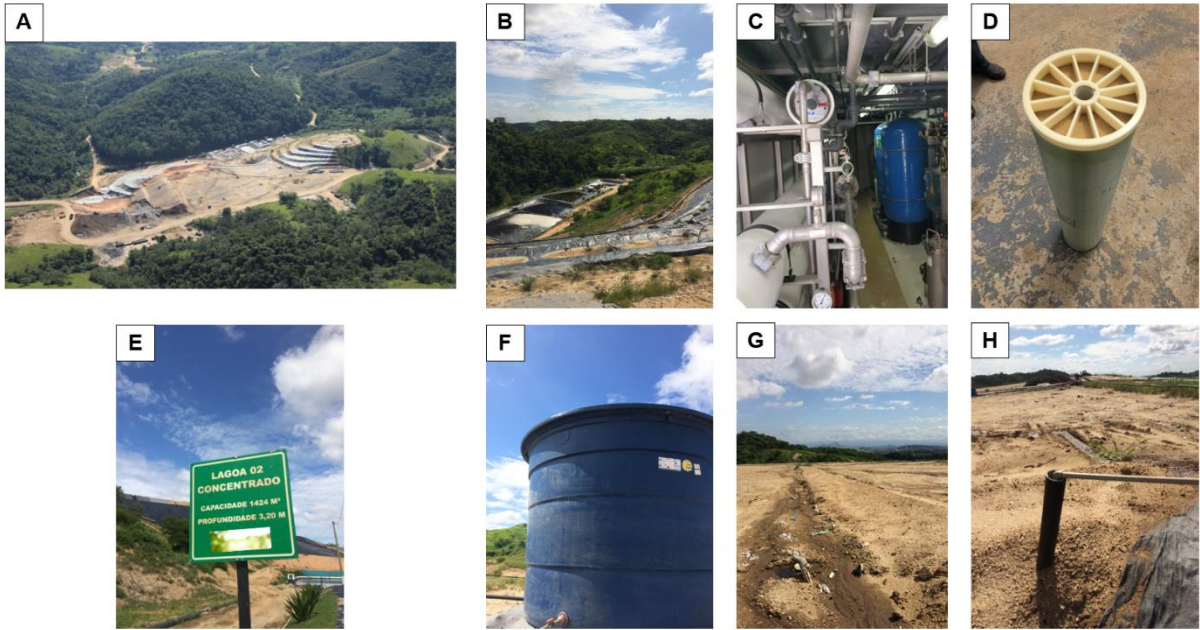**Figure S.6- 1** Experimental landfill cells used in this research.

Figure S.6- 2 Brazilian case study landfill.A) Landfill landscape. B) leachate pounds. C) RO facility. B) membrane module. E) Membrane concentrate pounds. F) Reverse osmosis concentrate container. G, H) pipes for reverse osmosis concentrate infiltration.

Table S.6- 1 Leachate quantity of C_0 and C_i groups from the experimental cells monitoring (n=3).

Days	C_0 (mL)		C_i (mL)	
	Min–Max	Mean	Min–Max	Mean
6	1926 – 2762	2344	20 – 5660	2403
13	756 – 3520	2203	25 – 2339	1079
27	19 – 4000	1722	270 – 4358	2489
33	405 – 1035	720	315 – 2065	1432
48	124 – 11,089	5146	992 – 10,948	7314
61	—	278	158 – 246	213
89	—	415	4895 – 11,950	8423
104	—	5491	9012 – 20,009	14,511
110	—	700	3410 – 4925	4168
117	—	400	663 – 1043	853
125	—	810	3288 – 3298	3293
131	—	648	1729 – 1752	1741
138	—	907	926 – 983	954
147	—	1285	3340 – 5073	4207
163	—	650	3220 – 5310	4265
177	—	330	390 – 400	395
219	—	1485	—	—
224	46 – 15,788	7917	—	18
239	10 – 10,565	5288	—	—
247		228	—	—
254	18 – 6216	3117	—	32
267	—	1497	—	—
280	—	35,147	—	—
297	—	13,864	—	—
308	12 – 13,911	6962	—	—
329	—	3488	—	—
359	—	8286	—	—
382	—	12,040	—	—
423	—	26,180	—	—

Table S.6- 2 Physicochemical characterisation of leachates generated by experimental landfill cells in different operating modes (C_0, control group; C_i, infiltration group).

Date	Days	pH C_0 Min–Max	Mean	pH C_i Min–Max	Mean	COD (mg L^{-1}) C_0 Min–Max	Mean	COD C_i Min–Max	Mean	UV$_{254}$ (cm^{-1}) C_0 Min–Max	Mean	UV$_{254}$ C_i Min–Max	Mean	Conductivity (mS cm^{-1}) C_0 Min–Max	Mean	Conductivity C_i Min–Max	Mean	TDS (g L^{-1}) C_0 Min–Max	Mean	TDS C_i Min–Max	Mean	NH$_3$–N (mg L^{-1}) C_0 Min–Max	Mean	NH$_3$–N C_i Min–Max	Mean
15/03/2021	6	4.48–6.05	5.27	4.31–4.41	4.36	7467–7700	7583	8900–11400	10150	39.5–50	44.75	76.60–89.5	83.05	20.19–24.40	22.30	19,83–22,29	19.25	91–95	93	90–132	119	479; 796	538	398;674; 592	655
22/03/2021	13	4.52–6.16	5.61	4.35–4.42	4.38	1400–15466	10444	14,466–16300	15889	28.38–54.30	43.61	76.70	76.70	14.92–25.81	18.74	15,53–19,46	16.87	43–44	44	85–92	90	2648	2648	–	–
05/04/2021	27	5.07–6.16	5.62	4.38–4.67	4.52	3010–17870	10440	12,500–48,850	33283	32.75–43.75	38.25	68.25–72.50	70.34	17.74–35.02	26.38	17,06–18,20	17.66	46–87	60	83–87	86	17,320–22,637;	19,979	11;435–16,137	13,513
12/04/2021	33	5.08–6.16	5.62	4.37–4.68	4.52	–	7233	22,200–29525	26637	38.77–48.91	43.84	68.20–78.23	73.22	17.52–33.09	25.31	18.21–27.20	23.63	48–60	55	69–76	74	14,072–23,077	18,575	13,167–16,938	15,053
27/04/2021	48	4.81–6.24	5.53	4.38–4.74	4.54	–	15867	–	26583	35.70–78.45	52.77	61.40–63.30	62.35	17.59–32.17	24.88	16.61–23.74	19.48	44–50	46	70–90	75	10,174–16,769	12,463	9,776–14,932	13,213
10/05/2021	55	4.47–5.14	4.82	4.49–5.02	4.68	2975–12125	6200	9175–55,100	25900	49.21	49.21	60.78	60.78	14.14–26.68	18.70	15.53–17.57	16.72	40–47	44	80–89	78	28,047	28,047	21,541–23,306	22,548
07/06/2021	61	4.91–5.06	4.99	4.43–4.96	4.70	5888–8488	7188	2700–17,550	10958	16.51	16.51	43.73	43.73	–	22.00	17.25–22.57	20.20	–	36	70–84	76	1261	1261	1294–1515	1405
22/06/2021	89	–	5.38	4.56–4.66	4.61	–	958	51,150–60,416	55783	17.15	17.15	39.52	39.52	–	17.47	15.25–19.94	17.60	–	34	40–70	52	955	955	1179–1204	1192
28/06/2021	104	–	5.52	4.70–6.45	5.58	–	1475	1767–33118	17442	17.15	17.15	33.39	33.39	–	17.01	18.04–18.94	18.49	–	32	29–62	40	483	483	–	–
05/07/2021	110	–	5.41	4.80–6.16	5.48	–	3433	1625–6608	4117					–	18.30	21.16–22.27	21.72	–	30	28–45	35	700	700	890	890
12/07/2021	117	–	5.29	4.74–5.96	5.35	–	1367	3217–3833	3525	15.73	15.73	29.53	29.53	–	19.07	19.66–20.33	20.00	–	30		30	2216	2216	2013	2013
20/07/2021	125	5.35	5.35	4.80–6.04	5.42	1367	1367	1408–12050	6729	15.43	15.43	27.45	27.45	–	19.17	20.09–20.52	20.31	–	28	–	29	3358	3358	2275	2275
27/07/2021	132	5.40	5.40	4.88–6.03	5.46	692	692	1300–11375	6338					–	19.14	19.54–20.38	19.96	–	26	24–29	25	2325–3847	3086	3189	3189
09/08/2021	147	5.47	5.47	5.18–5.99	5.59	410	410	1080–1558	1319	10.73	10.73	22.56	22.56	–	16.91	20.31–20.71	20.51	–	23	19–23	20	2675	2675	1960–3767	2854
25/08/2021	163	5.37	5.37	5.74–5.94	5.84	402	402	537–717	627	21.12	21.12	26.03	26.03	–	18.50	20.29–20.53	20.41	–	22	17–20	19	2959	2959	1910–3441	2676
08/09/2021	177	5.39	5.39	5.58–5.95	5.77	298	298	819–4365	2592	19	19	29.92	29.92	–	14.76	17.31–18.73	18.02	–	16	18–22	19	2163	2163	1793–3556	2675
28/09/2021	197	5.37	5.37	–	–	413	413	440	440	16.91	16.91	24.35	24.35	–	12.76	–	–	–	15	–	–	4225	4225	3460	3460
25/10/2021	224	5.35–7.49	6.42	6.63	6.63	180–2183	1182	567	567	12.61–22.1	17.39	25.50	25.50	3.08–10.14	6.61	–	19.03	16–12	14	–	15	711	711	1103	1103
24/11/2021	254	5.64–8.07	6.89	7.87	7.87	107–513	310	543	543	4.43–8.05	5.90	24.54	24.54	3.13–5.40	4.27	–	11.28	5–8	6	–	10	256	256	562	562

$DP = \sqrt{\sum_{i=1}^{n}(M - \bar{M})^2 / n - 1}$. C_0 = Control group; C_i = Infiltration group; SD = Standard deviation. M = mean value of leachate from specific landfill cell (Cn). $\bar{M}$ = mean value of leachate from landfill cell group. n = number of experimental cells in each analysed group.

To be continued...

Date	Days	pH C0 Min–Max	pH C0 Mean	pH Ci Min–Max	pH Ci Mean	COD (mg L⁻¹)s C0 Min–Max	COD C0 Mean	COD Ci Min–Max	COD Ci Mean	UV₂₅₄ (cm⁻¹) C0 Min–Max	UV C0 Mean	UV Ci Min–Max	UV Ci Mean	Conductivity (mS cm⁻¹) C0 Min–Max	Cond C0 Mean	Cond Ci Min–Max	Cond Ci Mean	TDS (g L⁻¹) C0 Min–Max	TDS C0 Mean	TDS Ci Min–Max	TDS Ci Mean	NH₃–N (mg L⁻¹) C0 Min–Max	NH₃ C0 Mean	NH₃ Ci Min–Max	NH₃ Ci Mean
20/12/2021	280	5.72	5.72	6.78	6.78	164—187	172	655	655	5.78 – 9.76	7.70	23.45	23.45	5.64—5.66	5.65	—	11.01	5 – 6	5.5	–	6	352	352	435	435
06/01/2022	297	7.45—7.47	7.46	8.38	8.38	83—87	84	641	641	11.85-11.93	11.89	22.65	22.65	4.67—4.75	4.71	—	10.78	3.8 – 4.2	4	–	6	270	270	515	515
17/01/2022	308	7.10—8.25	7.68	—	—	101—416	259	—	—	13.33	13.33	—	—	2.28—3.09	2.69	—	—	—	3	–	–	160	160	—	—
07/02/2022	329	7.20	7.20	—	—	188	188	—	—	7.55	7.55	—	—	3.69	3.69	—	—	—	2.5	–	–	213	213	—	—
09/03/2022	359	7.04	7.04	—	—	99	99	—	—	4.29	4.29	—	—	2.34	2.34	—	—	—	1.6	–	–	167	167	—	—
01/04/2022	382	6.62	6.62	—	—	2550	2550	—	—	1.69 – 5.80	3.75	—	—	2.45	2.45	—	—	—	< 1	–	–	99—112	112	—	—
12/05/2022	423	6.68	6.68	—	—	1158	1158	—	—	8.88	8.88	—	—	3.82	3.82	—	—	—	< 1	–	–	225	225	—	—

$DP = \sqrt{\sum_{i=1}^{n}(M - \overline{M})^2 / n - 1}$. C_0 = Control group; C_i = Infiltration group; SD = Standard deviation. M = mean value of leachate from specific landfill cell (Cn). $\overline{M}$ = mean value of leachate from landfill cell group. n = number of experimental cells in each analysed group..

Chapter 7.   Pyrolysis of Spent Coffee Ground Using Landfill Leachate Concentrate as a Pyrolytic Additive

This chapter focuses on the slow pyrolysis of spent coffee grounds using leachate concentrate residue as a pyrolytic additive. Biochars were characterized in terms of physicochemical parameters, morphology, and elemental composition to discuss potential agronomic and environmental benefits. Based on the ideas presented in chapter 5, the present chapter aims to provide insights into alternative approaches for managing membrane concentrate streams within a circular bioeconomy context.

This research was conducted at Università degli Studi di Padova from november/2021 to April/2022 under supervision of Professors Maria Cristina Lavagnolo, Roberta Bertani, and Paolo Sgarbossa. The research work was continued at Universidade Federal do Rio de Janeiro.

A final version of this chapter will be submitted as a research article in a specialized journal.

7.1 INTRODUCTION

Membrane systems remove organic and inorganic compounds and provide high-level of leachate purification (DE ALMEIDA et al., 2020; DE ALMEIDA et al., 2020). However, closing the leachate treatment loop and finding a final destination for the landfill leachate membrane concentrate (LLMC) — residual stream of membrane systems – is challenging (KEYIKOGLU et al., 2021). Generally, LLMCs are high-saline streams (16,130–98,000 μS cm^{-1}) and, depending on the leachate composition and its treatment layout, refractory organic pollutants such as lignins-like, lipids/proteins-like and unsaturated hydrocarbons are presented on it in high-level (XIANG et al., 2022). LLMC recirculation onto the landfill body is typical, but contaminants can accumulate in the leachate, making this approach critical. Indeed, this practice is prohibited in developed nations like Germany (MEIER et al., 2021). On the other hand, treatment techniques (e.g., coagulation-flocculation, adsorption, ozonation, and incineration) can be costly and/or ineffective in handling highly polluted streams (CHEN et al., 2019; HONG et al., 2017; LONG et al., 2017; TOW et al., 2021).

Considering the current global resource depletion and climate change scenario, material extraction and carbon sequestration strategies are increasingly in demand (HUANG et al., 2020; KURNIAWAN et al., 2021; LI & LIU, 2021). Pyrolysis (Py) has emerged as an option for valorisation and sustainable management of different kinds of biomasses (e.g., sewage sludge, agroindustrial residues, and urban/industrial wastes) (MANYÀ et al., 2018; PELLERA et al., 2021; SEO et al., 2022) and, therefore, could play a key role in this sense. Py is defined as the thermochemical decomposition of carbon-based feedstock in an absence/low oxygen environment at a temperature >400ºC. Solid carbonaceous material named biochar (BC) and py-gasses, including volatile organic substances, which can be condensed to liquid phase (bio-oil), and mixed non-condensable gasses (CO, CO_2, CH_4, and H_2) (syngas) are produced in this process (TRIPATHI et al., 2016). Py-technology can be categorised as fast, intermediate, and slow depending on heating rate, peak temperature, and residence time. Slow pyrolysis is the most employed method for BC production because it offers the highest recovery of solid carbonaceous product (MANYÀ, 2012). Due to its physicochemical proprieties (e.g., porous structure and water retention capacity), BC

can be used as a soil amendment for agronomic benefits. Besides, soil application of this stable carbon-rich material can imply a net carbon removal from the atmosphere since the organic waste conversion to long-term stabilized soil carbon acts as a C-sink.

Recently, co-pyrolysis of LLMC and sewage sludge for recovery of liquid and gas products was tested as an advanced technology for recycling organics in biofuels, which could be exported and/or used as an energy source for the thermochemical process itself (HASSEN-TRABELSI et al., 2020). In this sense, we attempted to expand the use of Py-technology, focusing on biochar production using RO leachate concentrate (ca. 5 g L^{-1} of total organic carbon). However, our preliminary findings showed that obtained pyrolysed material had low porosity and was mainly composed of salts (e.g., Na^+, K^+, and Cl^-) arranged in clusters with irregular shapes. By examining SEM/EDS for its chemical elemental composition, the C/O ratio was lower than 0.01 in all investigated pyrolitic temperatures (see supporting content of this chapter). Therefore, applying leachate concentrate in mono-Py may not be the most sustainable way to recycle organics as biochars. On the other hand, literature shows that inorganic compounds can play catalytic effects in the pyrolytic reaction of biomasses, increasing char yield and porosity of the produced carbonaceous material. In that direction, we hypothesized that leachate concentrate residues could be used as an additive to boost the yield and/or improve the quality of the BC produced in the thermochemical conversion of biomasses.

This research focuses on the slow pyrolysis of spent coffee ground (SCG) using leachate concentrate residue as Py-additive. Biochars were characterized in terms of physicochemical parameters, morphology, and elemental composition to discuss potential agronomic and environmental benefits. SCG is a solid waste by-product from the coffee processing industry. It is usually landfilled, open-burned with other coffee residues and/or mixed with animal fodder. Therefore, the carbon footprint and environmental burdens associated with the existing management solutions are very concerning (MARTINEZ et al., 2021). Thus, our study aims to provide insights into alternative approaches for managing solid wastes within a circular bioeconomy context.

7.2 MATERIALS AND METHODS

7.2.1 Research programme

In order to investigate the pyrolytic conversion of SCG and LLMC, a synthetic leachate concentrate (SLC) was prepared and oven-dried. At a lab-scale pyrolizer under a nitrogen atmosphere, SCGs were treated at 600ºC for 1 h. Experimental conditions were defined based on preliminary Py-tests of oven-dried SLC samples and surveyed literature. Py-tests were conducted for SCG and SCG + oven-dried SLC (1:1 wt%). Proximate and ultimate analyses of biochars were performed.

7.2.2 Biomass and additive

The SLC was prepared following the procedure proposed by Grossule et al. (2022) (Table S.7- 1). Hereafter, the sample was oven-dried at 105ºC for 24 h. The oven-dried SLC presented water content and VS/TS (volatile solids/ total solids) ratio of 2% and 51%, respectively. This residue was powdered and used in the subsequent pyrolytic tests. For practical applications, the LLMC could be evaporated using the heat from the Py-process. The SCG (100% Arabica blend) was provided by an Italian company. The sample was oven-dried overnight at 110ºC and stored in glass bottles.

It should be noted that thermal technologies such as submerged combustion evaporation and mechanical vapour recompression have been used to handle leachate concentrate streams (ZHANG et al., 2019). In these systems, a dried residue is generated and commonly landfilled. Likewise, evaporation pounds used in warm regions for concentrate volume reduction generate sludges that must be properly managed (KEYIKOGLU et al., 2021). In that way, this research could help expand the options to valorize waste streams from available options used to manage the LLMC.

7.2.3 Py-process

Slow pyrolysis experiments were conducted using a lab-scale pyrolizer at a heating rate of 45ºC min^{-1}. The lab-pyrolizer was operated at atmospheric pressure and room temperature.

Py conditions were temperature of 600ºC, the inert gas flow of 100 cm^3 N$_2$ min^{-1}, and residence time of 1 h. Below 500ºC, pyrolysis of biomass may produce biochar with low structural stability, while at temperature >800ºC the quantity of carbon left on char is minimum (AHMAD et al., 2014; PELLERA et al., 2021). Obtained chars were washed with deionized water on a paper filter and left drying at ambient temperature (20ºC). The biochar yield was determined as a mass fraction of the initial feedstock (Equation 7.1). BCs were stored for physicochemical and morphological characterisation.

$$Y(\%) = \left(\frac{BC\ mass\ (mg)}{initial\ mass\ of\ feedstock\ (mg)} \right) \times 100 \qquad (7.1)$$

7.2.4 Characterisation

Proximate analyses, that is, moisture content (MC), volatile matter (VM), ash content, and fixed carbon (FC), were performed. Briefly, MC was measured by heating 1 g of a sample at 105±5ºC for 1 hour in an oven; VM was determined by heating the left residue at 950ºC for 6 min. Finally, ash content was obtained by heating the samples in a furnace at 750ºC for 6 h. FC was calculated by difference according to ASTM method D1762-84. pH, electroconductivity (EC), and total dissolved solids (TDS) were determined for physicochemical characterisation. Physicochemical parameters were measured after mixing the sample with deionized water with a solid/liquid ratio of 1:20 g mL^{-1} for 1.5 h.

Scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM/EDS) micrographs of biomass, blending, and BCs were portrayed by an FEI-QUANTA200 instrument (Milan, Italy). In this work, proximate and ultimate analyses were selected according to focal points for biochar application as a soil amendment, summarised by IGALAVITHANA et al. (2017).

Thermogravimetric (TG) analysis and decomposition profile of feedstocks, i.e., SCG, oven-dried SLC, SCG+oven-dried SLC (1:1 %wt) (blend), and the produced biochars were carried out using a TA Instruments equipment, model SDTQ600. The samples were weighed to around 5 mg. Then, they were heated from 20 to 1000ºC (in an alumina pan) at a rate of 20 °C min^{-1} under an air gas flow rate of 100 mL min^{-1}.

7.3 RESULTS

7.3.1 Biochar characterisation: proximate, ultimate, and physicochemical analyses

Proximate and ultimate analyses are presented in Table 7-1. Table 7-2shows the physicochemical characterisation of SCG, oven-dried SLC and blending before and after pyrolysis.

Table 7-1 Proximate and elemental composition of SCG, oven-dried SLC feedstock samples, and their respective biochars after pyrolysis at 600ºC for 1 hour.

Feedstock			Biochar	
	SCG[a]	SCG+oven-dried SLC (1:1 wt%)	SCG-biochar	Biochar (SCG+oven-dried SLC)
Proximate analysis				
Yield (%)	—	—	23.9±0.2	17.4±1.2
MC (wt%)	3.78	9.10	2.75	28.20
VM (wt%)	94.91	61.83	58.40	44.40
Ashes (wt%)	1.26	24.70	5.81	6.96
FC (wt%)	1.25	4.37	33.04	20.44
Ultimate analysis [b,c]				
Carbon (wt%)	47.26±0.46	45.53±4.59	46.06	63.97±0.59
Oxygen (wt%)	46.13±1.36	33.01±2.66	45.26	20.16±0.27
Sodium (wt%)	n.d	9.84±1.56	n.d	4.32±0.04
Potassium (%wt)	0.73±0.05	4.70±1.57	3.87	4.14±0.02
Calcium (%wt)	1.26±0.27	0.62±0.05	n.d	2.87±0.02
Magnesium	n.d	0.35±0.04	0.40	1.00±0.01
Chloride (%wt)	n.d	6.11±3.80	n.d	0.47±0.01
Sulfur (%wt)	0.73±0.05	1.11±0.13	0.52	1.43±0.01

[a]Oven-dried overnight at 110ºC
[b]On a dry basis
[c]The mean ± standard deviation for two determinations
[d]Values obtained for one determination
n.d: not detected

Table 7-2 Physicochemical characterisation of biomass, addictive and blending before and after pyrolysis (600ºC, 1 h, 100 cm^3 N min^{-1}).

	pH		EC (mS. cm^{-1})		TDS (g L^{-1})	
	Start	Post-Py	Start	Post-Py	Start	Post-Py
SCG	6.4±0.1	10.7±0.1	0.660±0.050	2.8±0.1	0.442±0.014	1.2±0.1
Oven-dried SLC	10.5±0.1	12.8±0.1	39.3±0.1	37.6±0.1	28.3±0.1	26.7±0.1
SCG + oven-dried SLC (1:1 wt%)	10.0±0.1	10.4±0.1	26.3±0.5	1.327±0.026	18.7±0.1	0.895±0.043

Images of feedstocks and biochars produced by pyrolysis at 600ºC for 1 hour is illustrated in Figure 7-1.
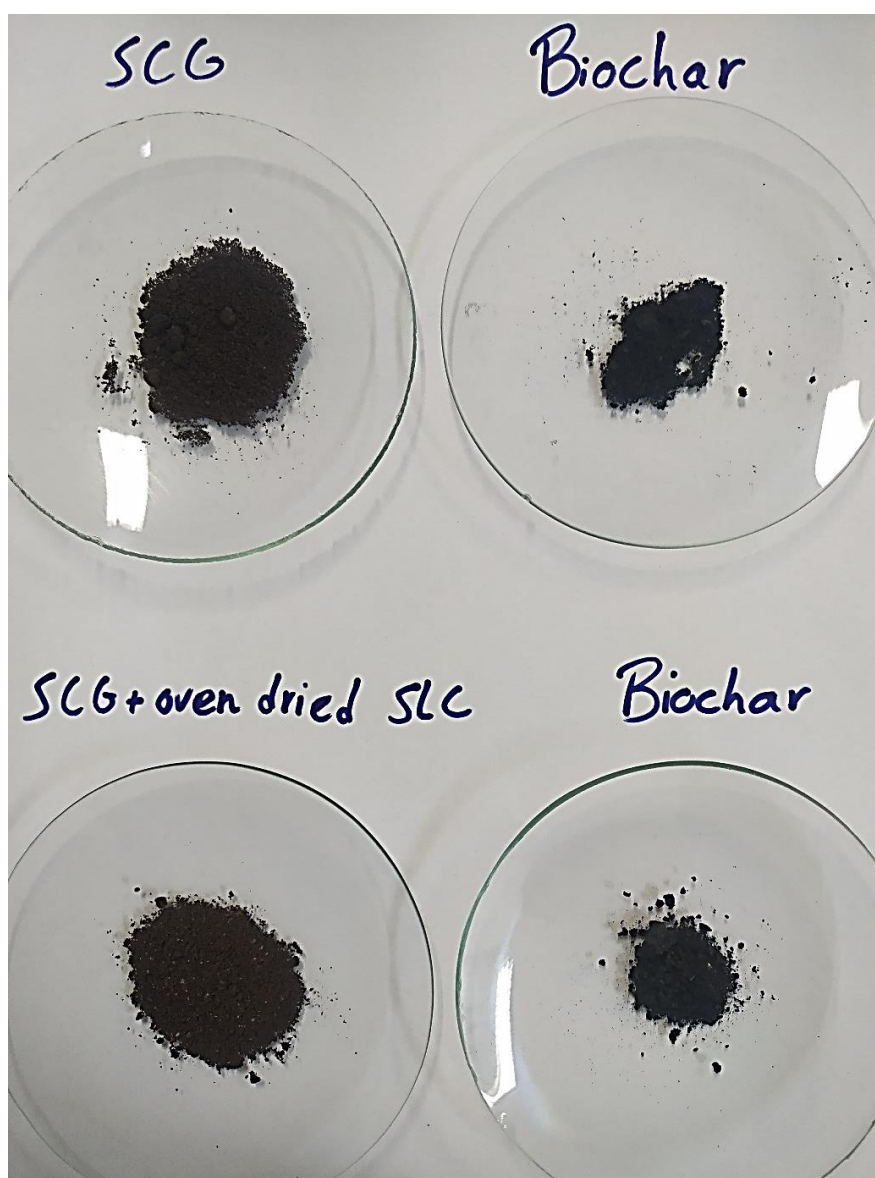
Figure 7-1 The visual aspect of feedstocks (SCG and SCG+oven-dried SLC) and biochars produced in pyrolysis at 600ºC and 100 cm3 N2 min-1 for1 hour.

7.3.2 Thermal analysis

Figure 7-2 shows the TG, derivative thermogravimetry (DTG), and differential scanning calorimetry (DSC) analyses for SCG and the produced biochar.
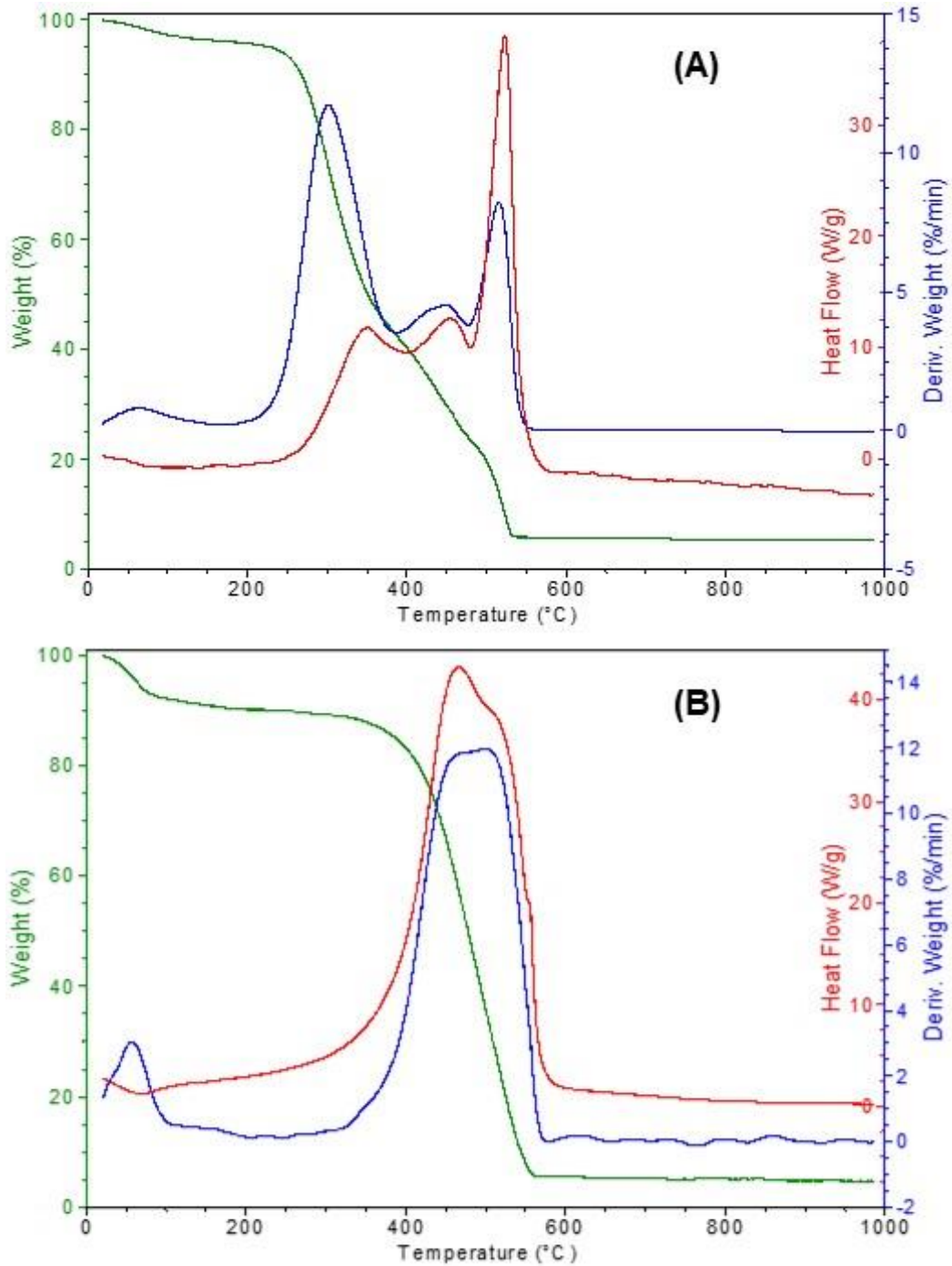
Figure 7-2 TG, DTG, and DSC analyses for SCG (A) and the produced biochar (B).

Thermal analysis for SCG showed that free water loss occurred from 20 to 150°C, followed by combustion of organics at around 580°C. The first DTG peak and its respective endothermic DSC at 150°C are linked to the free water vaporization. DTG and exothermic DSC peaks from 150°C to 580°C are connected to the combustion of SCG organic compounds. Similar behaviour was observed for the produced biochar from that biomass. However, a DSC peak from 150 to 390°C is not presented for biochar, which can be explained by VM loss during the biomass pyrolysis. From TG analysis, water content is estimated at 3.9 and 9.2 wt% for SCG and biochar, respectively.

TG, DTG, and DSC analyses of biomass and additive (SCG+oven-dried SLC, 1:1 wt%) and the produced biochar from that blending is illustrated in Figure 7-3
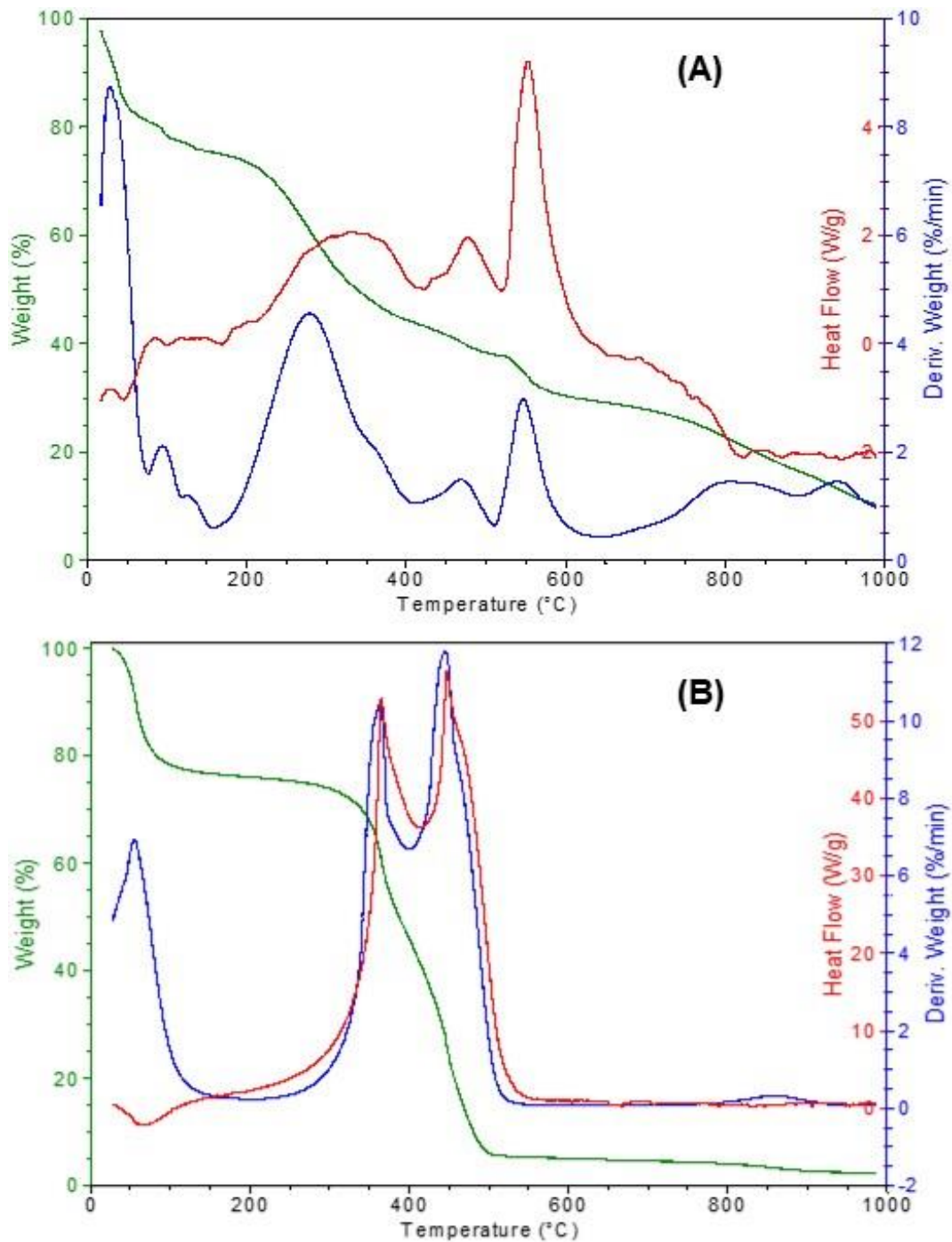
Figure 7-3 TG, DTG, and DSC analyses for blending (SCG+oven-dried SLC, 1:1 wt%) (A) and the produced biochar (B).

For SCG + oven-dried SLC blend, water loss occurred from 20 to 160°C. Organics were combusted from 160 to 650°C. Inorganic salts, mostly present in the oven-dried SLC, melt and vaporise above 650°C. From thermal analysis, the water content in the produced biochar is estimated at 23.25 wt%.

7.3.3 SEM/EDS Analysis

SEM micrographs for SCG, SCG+oven-dried SLC, and the produced biochars are shown in Figure 7-4. SEM (100✕) of biochar from SCG+oven-dried SLC pyrolysis and its EDS spectra are illustrated in Figure 7-5. Overall, the SEM micrograph of SCG+oven-dried SLC shows high microporosity. By EDS spectra, biochar is C-rich (ca. 64 wt%) and has essential soil minerals such as Na, K, Ca, and Mg.

(A) SCG (500✕)

(B) oven-dried SLC (200✕)

(C) SCG biochar (400✕)

(D) SCG+oven-dried SLC biochar (500✕)
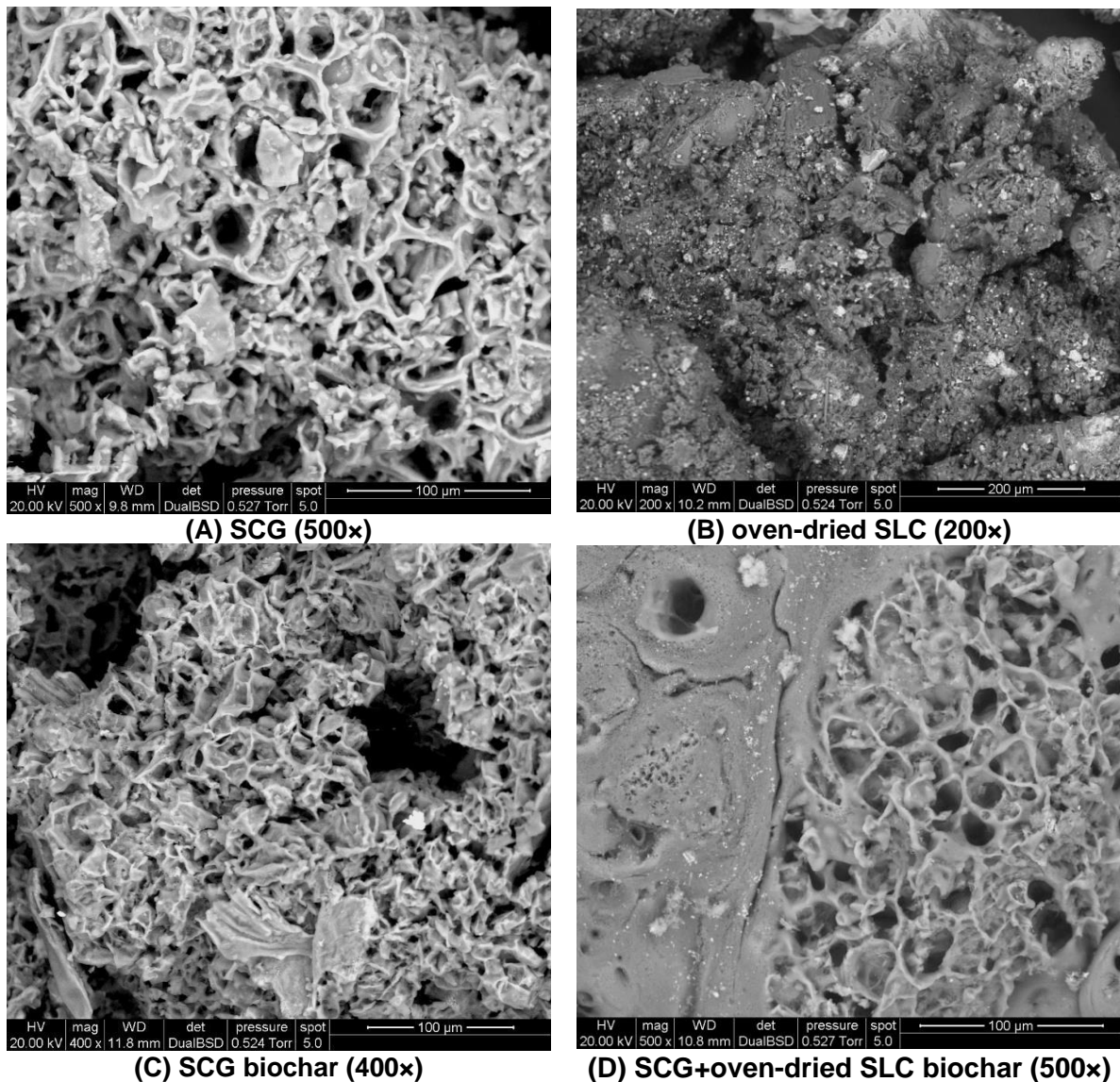
Figure 7-4 SEM micrographs of SCG and oven-dried SLC at 500✕ 200✕ of magnification and biochars from SCG (400✕) and SCG+oven-dried SLC (500✕) (1: 1 wt%) pyrolysis.

HV	mag	WD	det	pressure	spot	⎯⎯⎯⎯ 500 µm ⎯⎯⎯⎯
20.00 kV	100 x	11.3 mm	DualBSD	0.524 Torr	5.0	
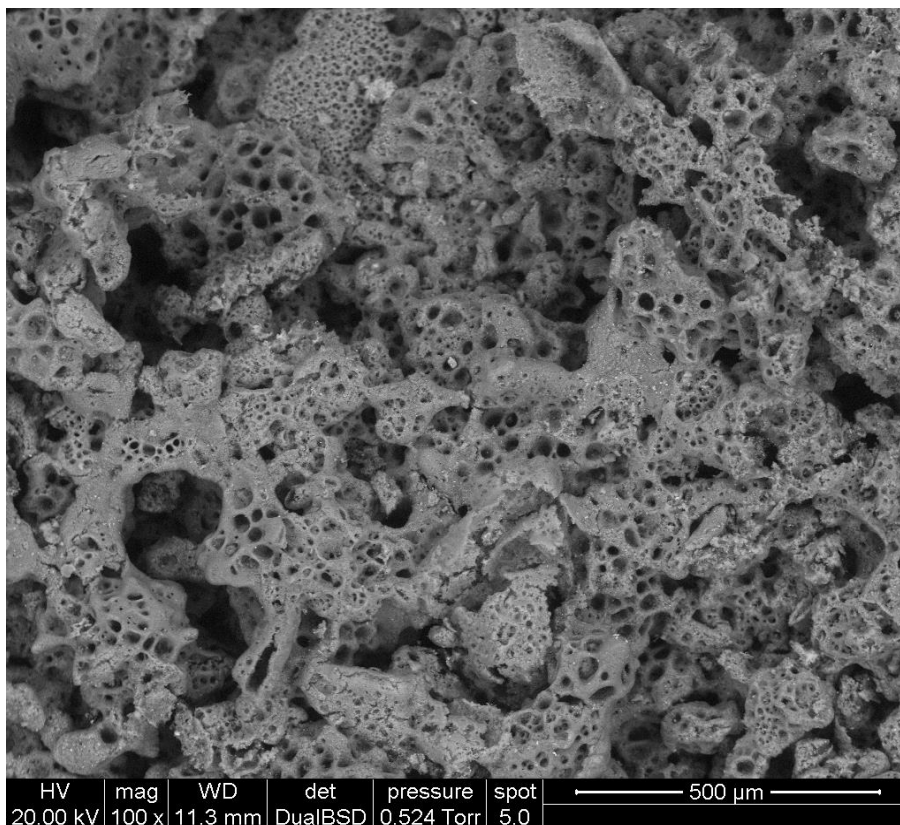
(A) SCG+oven-dried SLC (1:1 %wt) biochar (100×)

Element	Weight %
C	63.97
O	20.16
Na	4.32
Mg	1.00
Al	0.50
Si	0.71
P	0.16
S	1.43
Cl	0.47
K	4.14
Ca	2.87
Fe	0.27
Total	100
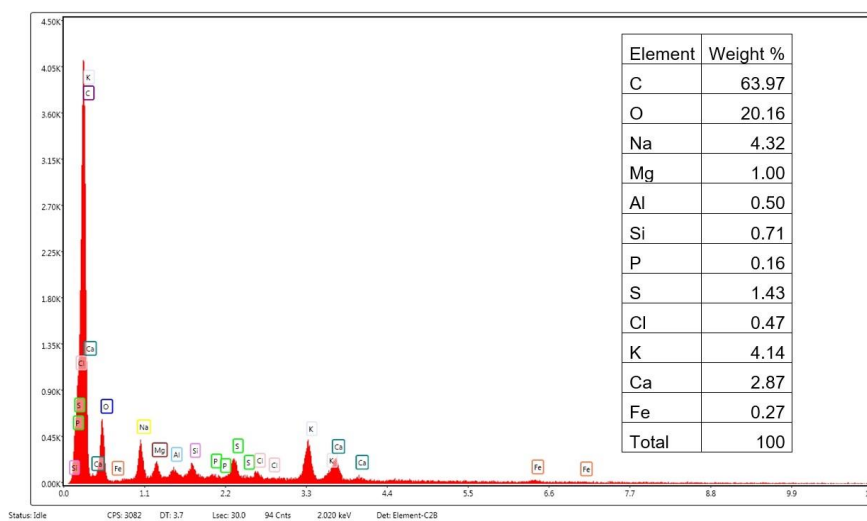
(B) EDS spectra

Figure 7-5 SEM micrograph (100×) of the obtained biochar from SCG+oven-dried
SLC (1: 1 wt%) pyrolysis (A) and its EDS spectra (B).

7.4 DISCUSSION

By thermal analysis, water content was about 2.5-fold higher in SCG+oven-dried SLC biochar than in SCG biochar. Based on the Proximate analysis, this value was 10-fold higher. Therefore, it was indicated that SLC's alkali metals could catalyze the carbonization of organics in the biomass, producing a more porous material able to retain more water. From elemental composition analysis, biochar from SCG+oven-dried SLC is C-rich and has essential soil minerals such as Na, K, Ca, and Mg.

Chen et al. (2008) investigated the effects of eight inorganic additives, including NaOH, NaCl, Na_2CO_3, and Na_2SiO_3, which remarkably increased char yield and made Py-gasses give off earlier. In another work, Wang et al. (2006) showed that sodium compounds promote char formation and make pyrolysis more exothermic. To the best of my knowledge, this is the first study to investigate the effects of leachate concentrate on pyrolysis of an agroindustry residue. Thus, recent literature is not available for a reasoning comparative analysis.

7.5 CONCLUSIONS

This research focused on the slow pyrolysis of SCG using leachate concentrate as a pyrolytic additive. Our preliminary findings suggest that inorganic elements in leachate concentrate could catalyse the pyrolysis process of carbon substrates, increasing biochar porosity and its mineral content. In this sense, pyrolysis can play an essential role in membrane concentrate valorisation for producing a material that can be used as a C-sink and for mineral recycling. However, it must be highlighted that this investigation is at an embryonic stage. Even though it was suggested that co-pyrolysis of leachate concentrate and other substrates could be an exciting way to handle and valorise membrane concentrate streams, our findings cannot be generalized. This research will include the C-balance of the pyrolysis process and phytotoxicity analysis of biochars. Most important, pyrolysis should be performed using actual leachate concentrate samples in future investigations.

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Supporting Content

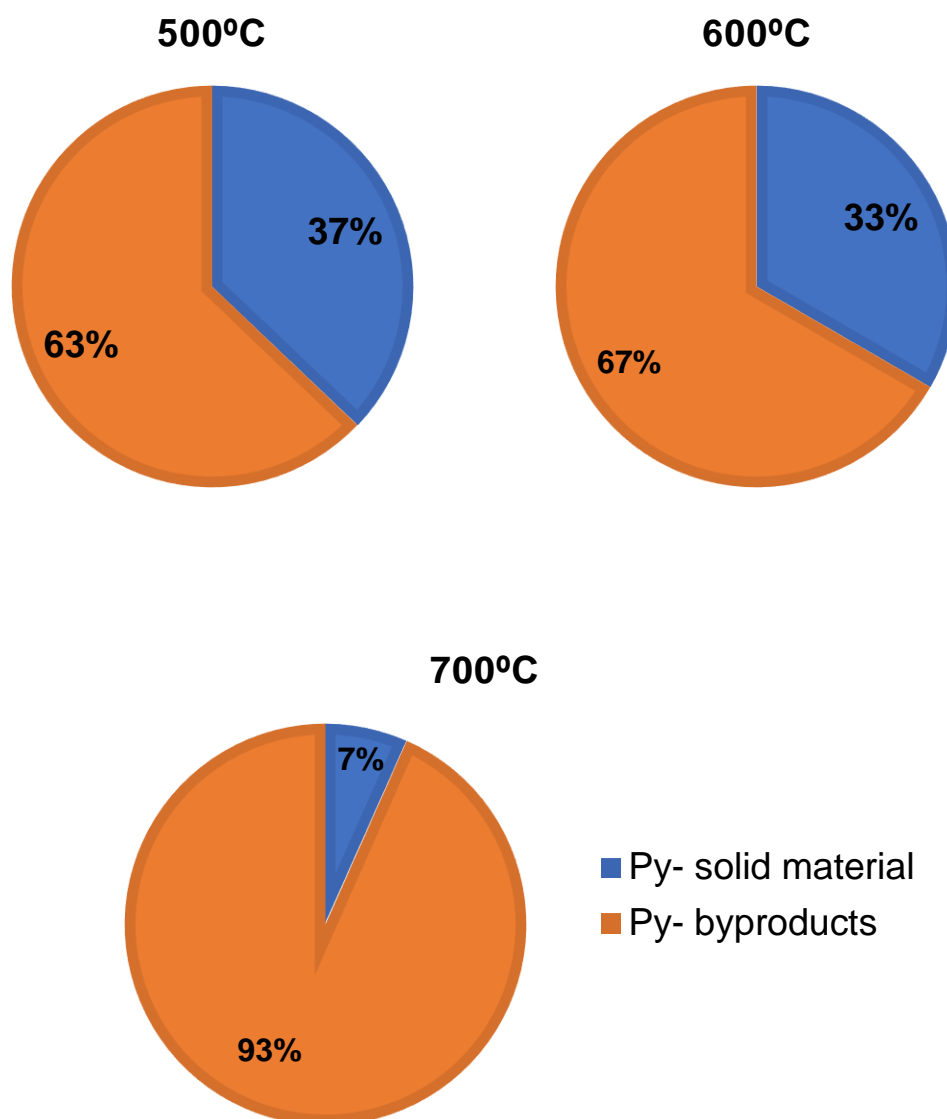**Co-pyrolysis of spent coffee ground with landfill leachate concentrate additive**

500ºC

600ºC

37%		33%
63%		67%

700ºC

7%

93%

■ Py- solid material
■ Py- byproducts

T (ºC)	Py- solid material (g)	Yield (%wt)	Py-byproducts (100 – Y (%wt))
500	0.3871	37.11%	62.89%
600	0.3395	33.35%	66.65%
700	0.0667	6.63%	93.37%

Figure S.7-1 Products distribution and yield (wt%) of pyrolytic conversion of 1.0 gram of oven-dried SLC at 500, 600, and 700ºC. Note: yields were cslculated without wash the samples with deionized water.

Oven-dried SLC

a) 500ºC

b) 600ºC

c) 700ºC
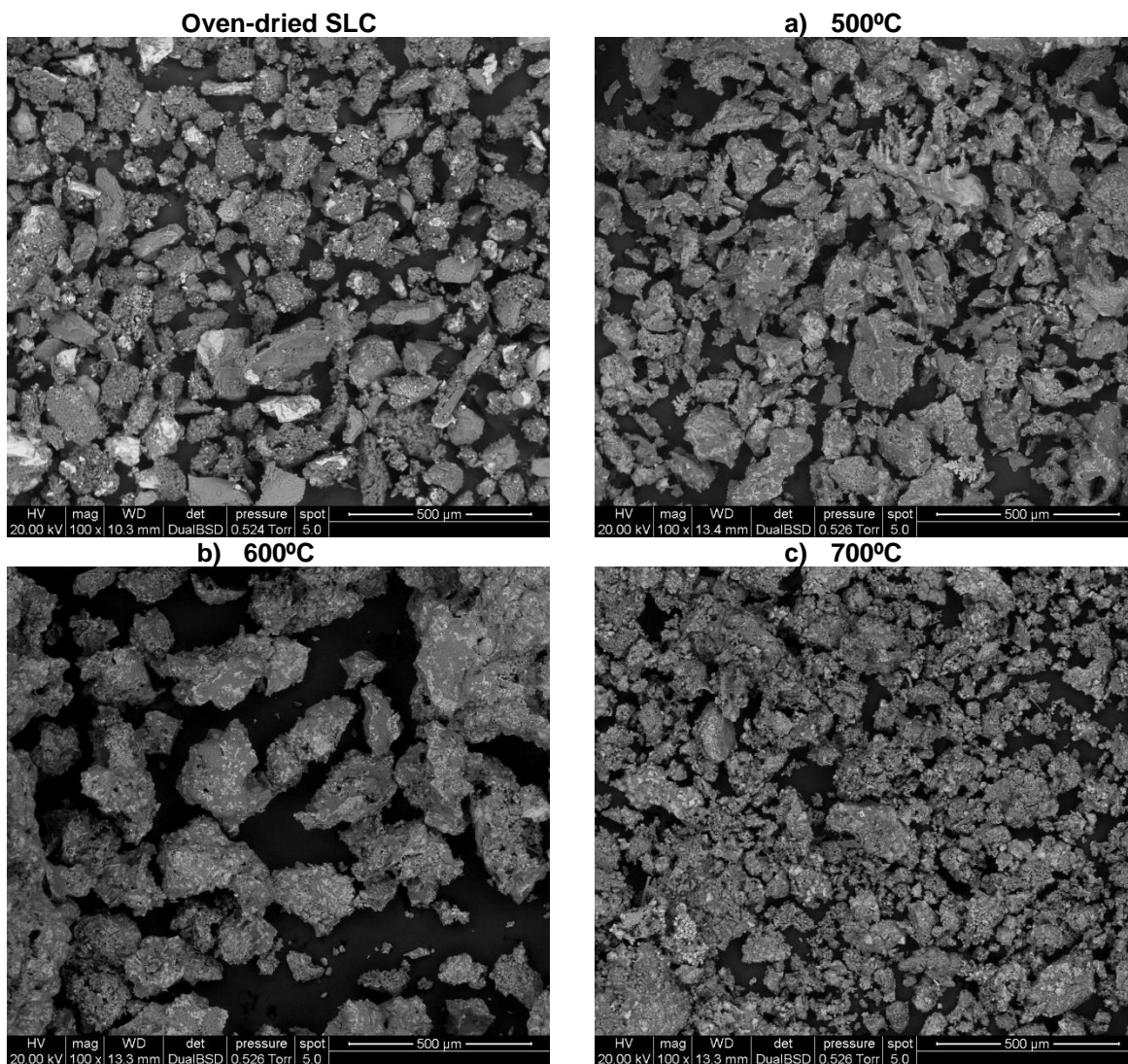
Figure S.7-2 SEM micrographs at 100× magnification of oven-dried SLC and solid Py-materials obtained at residence time of 1 h, 100 cm^3 N$_2$ min^{-1}, temperature conditions of 500, 600, and 700ºC.
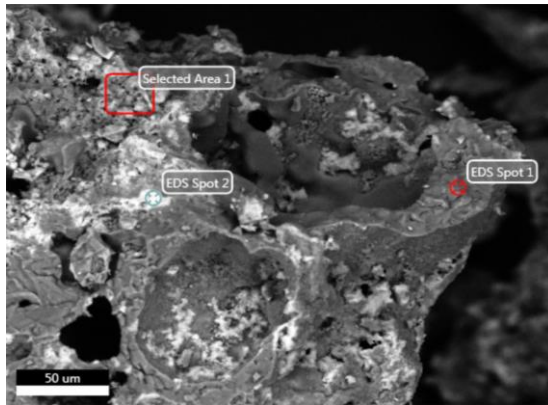
500ºC
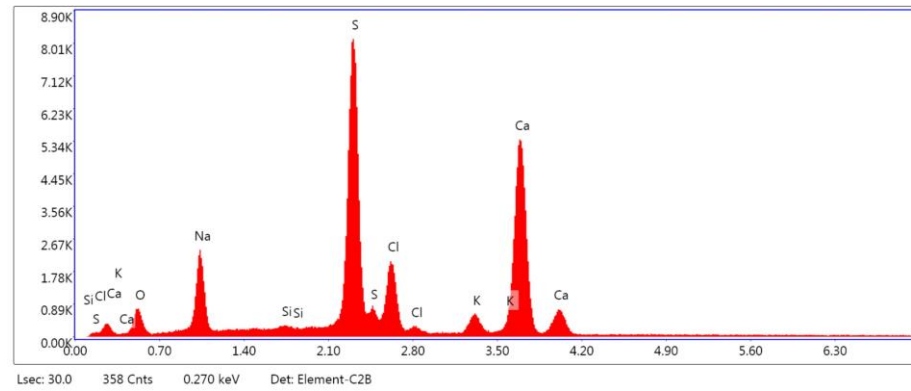
EDS spectra

O (wt%): 12.00; Na (wt%): 11.52; K (wt%): 2.80; Ca (wt%): 36.14; Cl (wt%): 8.96 ; S (wt%): 28.27

Elemental composition (spot 2)
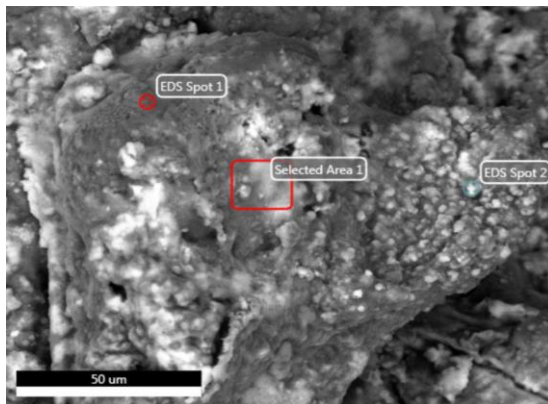
600ºC
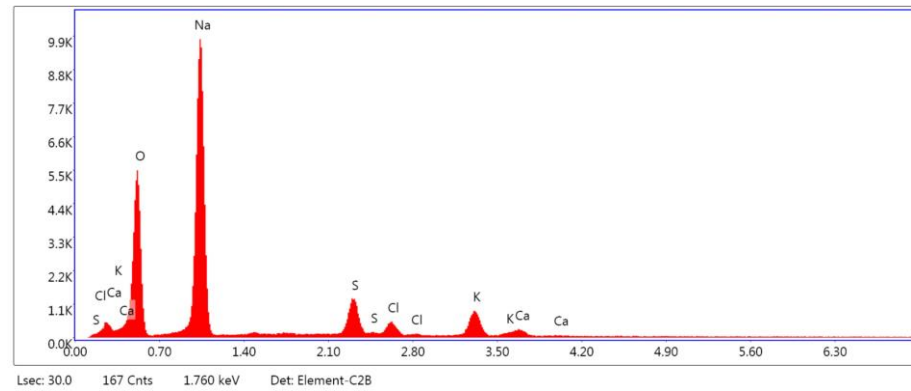
EDS spectra

O (wt%) 34.89: Na (wt%): 51.45; K (wt%): 4.95; Ca (wt%): 1.35; Cl (wt%): 2.10 ; S (wt%): 5.66

Elemental composition (spot 1)
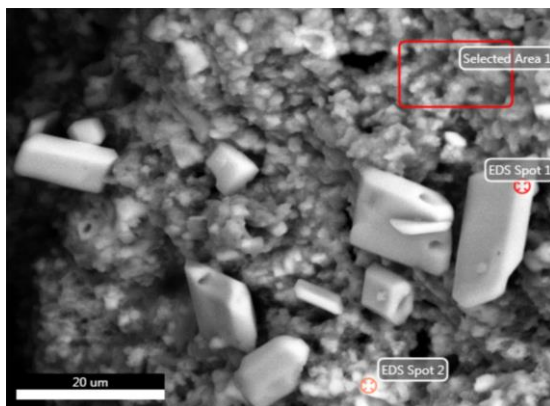
700ºC
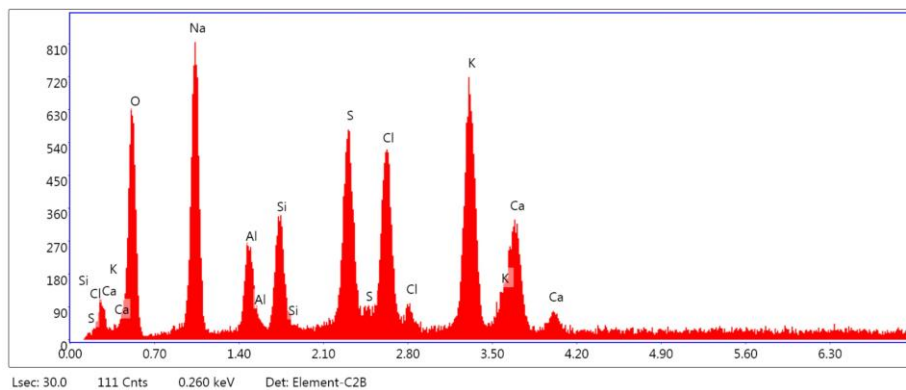
EDS spectra

O: (wt%): 30.74; Na (wt%): 17.97; K (wt%): 15.89; Ca (wt%): 1.35; Cl (wt%): 9.66 ; S (wt%): 9.37

Elemental composition (spot 1)

Figure S.7-3 SEM micrographs and corresponding EDS spectra of Py-solids obtained at 500, 600, and 700ºC, 1 h, and 100 cm^3 N_2 min^{-1}.

Table S.7- 1 Chemical parameters of SLC samples used in this study

Parameters	Value (mg L^{-1})	Real LC* [min–max] (mg L^{-1})
pH	8.3	7.9–8.7
TOC	5000	4680–7350
VS/TS	51%	40–55%
NH$_3$–N	2001	2172–7647
Conductivity (mS cm-1)	57.30	40.11–72.25
TDS (g L^{-1})	22.8	20.2–30.9
SO$_4^{-2}$	8221	4567–8150
Cl$^-$	5254	4567–5938
Na$^+$	9035	9974–11,543
K$^+$	3392	3065–3400
Ca+	237	309–397
Mg^{+2}	110	200–270

TOC – total organic carbon; VS – volatile solids; TS – total solids; NH$_3$-N – ammonium nitrogen; TDS – total dissolved solid; SO$_4^{-2}$ – sulphate; Cl$^-$ – chloride; Na$^+$ – sodium; K$^+$ – potassium; Ca^{+2} – calcium. *Values of RO concentrate from three different Brazilian treatment plants located in Rio de Janeiro State.

Chapter 8.   Concluding Remarks

In sum, this thesis explored the membrane application to treat landfill leachate and its related issues, precisely, the management of the concentrated leachate. The project was undertaken to design experimental landfill cells and assess the impacts of concentrate recirculation on the leachate quantity, quality, and treatment performance. The following conclusions are drawn:

i.   Landfill leachate is a polluting and complex wastewater, and its treatment has demanded the application of advanced technologies. In this sense, NF and RO are applied to achieve leachate contaminants removal beyond conventional systems such as biological and chemical oxidation processes. Membrane techno-economic benefits for leachate purification, i.e., production of high-quality water at competitive costs, were confirmed in Chapters 3 and 4 of this thesis.

ii.   One of the significant issues of membrane application is the management of concentrate streams. In the landfill leachate treatment chain, membrane concentrate recirculation to the landfill body is commonly adopted as a management strategy. This study showed that concentrate infiltration increased the leachate's organic content, which impacted the RO performance during the treatment of methanogenic leachates. In contrast, for NH_3-N, conductivity, and TDS, insignificant statistical differences were calculated between the leachate collected from cells that operated with and without concentrate infiltration. Taken together, these findings suggest that organic fouling plays a critical role in membrane permeability declining. In full-scale facilities, membrane fouling increases treatment operating expenses by demanding intensive cleaning and replacement of membrane modules.

iii. Membrane concentrate infiltration did not impact the leachate quantity produced by the landfill cells. However, the generalisability of this result is subject to certain limitations.

iv.   Despite its limitations, the study certainly adds to our understanding of the impacts related to the leachate concentrate infiltration practice. If leachate concentrate reinjection into the landfill waste mass is adopted as a management strategy, proper engineering design must be done by monitoring site conditions, leachate quantity, quality, and treatment performance.

v.   Membrane concentrate streams offer water, energy, and material extraction opportunities, considering the current global scenario of resource depletion and climate change. However, for commercial applications, several critical issues need to be addressed. In this sense, the pyrolytic process using leachate concentrate as an additive showed promising results, which can contribute to the development of greener management solutions.

vi.   Future studies should focus on the development of processes considering their techno-economic aspects. Their analyses based on a life cycle assessment and carbon footprint are recommended for possible full-scale implementation.

APPENDIX

Landfill Leachate Treatmento by Membrane-based Technologies: Cost-benefit Analysis, Membrane Concentrate Management, and Perspectives

Appendix I. De Almeida et al. A review on membrane concentrate management from landfill leachate treatment plants: The relevance of resource recovery to close the leachate treatment loop. ***Waste Manag. Res. J. a Sustain*. Circ. Econ**, v. -, 1─22, 2022. (IF = 4.432)

A review on membrane concentrate management from landfill leachate treatment plants: The relevance of resource recovery to close the leachate treatment loop

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Bianca Ramalho Quintaes[3], Daniele Maia Bila[4],
Maria Cristina Lavagnolo[2] and Juacyara Carbonelli Campos[1]

Abstract

Membrane filtration processes have been used to treat landfill leachate. On the other hand, closing the leachate treatment loop and finding a final destination for landfill leachate membrane concentrate (LLMC) – residual stream of membrane systems – is challenging for landfill operators. The re-introduction of LLMC into the landfill is typical; however, this approach is critical as concentrate pollutants may accumulate in the leachate treatment facility. From that, leachate concentrate management based on resource recovery rather than conventional treatment and disposal is recommended. This work comprehensively reviews the state-of-the-art of current research on LLMC management from leachate treatment plants towards a resource recovery approach. A general recovery train based on the main LLMC characteristics for implementing the best recovery scheme is presented in this context. LLMCs could be handled by producing clean water and add-value materials. This paper offers critical insights into LLMC management and highlights future research trends.

Keywords

Concentrated leachate, landfill leachate, membrane concentrate, nanofiltration, reverse osmosis, resource recovery

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Appendix II. De Almeida et al. Nanofiltration applied to the landfill leachate treatment and preliminary cost estimation. ***Waste Manag. Res. J. a Sustain. Circ. Econ***, 38(10), p. 0734242X2093333, 2020. (IF = 4.432)

Original Article

Nanofiltration applied to the landfill leachate treatment and preliminary cost estimation

WM&R

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Abstract

The leachate treatment by nanofiltration (NF) process has received much attention over the last two decades. Previous studies focused on the investigation of the technical feasibility of NF. However, there are a limited number of works that examined leachate treatment costs. On the other hand, in a landfill management system, the economic component is an imperative factor for the decisions of all operations. Thereby, this work aimed to investigate the technical feasibility and economic parameters involved in a full-scale NF plant for the treatment of leachate previously treated by the physicochemical process. The average quality of the pre-treated leachate was $2258 \pm 230\,mg\,L^{-1}$ chemical oxygen demand (COD) and $821 \pm 86\,mg\,L^{-1}$ humic substances (HS). NF was performed using a bench-scale filtration module with commercial polymeric membranes SR100 and NP030. At the end of each filtration, a cleaning protocol was applied to recover the initial membrane permeability. The concentration of recalcitrant compounds, expressed as HS, was reduced to $84 \pm 8\,mg\,L^{-1}$ in the permeate, and COD complies with the wastewater discharge standards imposed by local legislation. The capital costs for a full-scale NF was estimated at MUS\$ 0.772, and specific total cost, treated leachate per volume unit, has been estimated at US\$ $8.26\,m^{-3}$.

Introduction

Landfill leachate treatments are expensive, multi-processed, and different from each other due to the complex composition, high loading, and seasonal variation (Liu et al., 2015). Several technologies and treatment schemes are available for the treatment of landfill leachate (De Almeida et al., 2019; Di Maria et al., 2018; Renou et al., 2008; Talalaj et al., 2019): (a) co-treatment of sewage and leachate on the site or transfer to the central wastewater treatment plant; (b) recirculation into the bioreactor landfill body; (c) physicochemical treatment (coagulation–flocculation, precipitation, chemical oxidation, activated carbon adsorption); (d) biological processes (anaerobic treatment, aerated lagoons, activated sludge plants, membrane bioreactor, biofilms in rotating biologic contractors, and trickling filters); and (e) membrane processes (main nanofiltration (NF) and reverse osmosis (RO)). Despite this, to provide a treated effluent that can be discharged into natural water streams or reused, one single treatment technology is not available and a combination of treatment technologies is essential.

The selection of the best treatment scheme depends on the characteristics of the leachate, extent of treatment required, and economic parameters. Reported studies have shown that although

biological processes are widely applied for leachate treatment (Fernandes et al., 2017), this is not sufficient in reaching the existing wastewater discharge standards due to the recalcitrance, that is, the presence of non-biodegradable and/or low biodegradable compounds (Campos et al., 2013; Talalaj et al., 2019), characteristic of the mature landfill leachate.

The refractory organic is mainly humic substances (HS). According to Lima et al. (2017), HS can increase the solubility of hydrophobic compounds and solubilize metals such as nickel, cadmium, and zinc by complexation, modifying the bioavailability and biotoxicity of landfill pollutants. The presence of the refractory organic matter in the leachate illustrates the complexity

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Appendix III. De almeida et al. Cost estimation of landfill leachate treatment by reverse osmosis in a Brazilian landfill. *Waste Manag. Res. J. a Sustain. Circ. Econ*, 38(10), 0734242X2092841, 2020. (IF = 4.432)

Original Article

WM&R

Cost estimation of landfill leachate treatment by reverse osmosis in a Brazilian landfill

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Bianca Ramalho Quintaes[3] and Juacyara Carbonelli Campos[1]

Abstract
The reverse osmosis (RO) process has been increasingly applied to landfill leachate treatment. The published literature reports several studies that investigated the technical feasibility of RO. However, information about process costs is scarce. Also, companies that run leachate treatment plants do not provide actual costs. To fill this gap, this study aimed to evaluate the treatment costs of a full-scale RO for the treatment of landfill leachate located in Rio de Janeiro State, Brazil. A procedure was proposed to estimate the capital expenses (CAPEX), operational expenses (OPEX), and specific total treatment cost, the total cost per m^3 of treated leachate, of the leachate treatment by membrane process, and the results obtained are discussed. The CAPEX for this full-scale RO was estimated at MUS\$ 1.413, and OPEX ranged from US\$ 0.132 to US\$ 0.265 m^{-3} per year. The cost of leachate treatment has been estimated at US\$ 8.58 m^{-3} considering the operation of the RO-unit for 20 years after landfill closure.

Introduction

Landfill leachate is a highly contaminated wastewater generated by the decomposition of substances contained in solid waste, which has large amounts of contaminants measured in terms of chemical oxygen demand (COD), biochemical oxygen demand (BOD$_5$), ammonia, and inorganic salts (Kjeldsen et al., 2002). The leachate treatment processes are expensive and different from each other due to the complex composition, also, their composition is variable from different landfills (Talalaj et al., 2019). For this reason, the treatment methods have not been standardized so far. In this context, landfill leachate management is one of the major environmental and economic issues faced by landfill owners.

Reverse osmosis (RO) seems to be one of the most promising and efficient methods among existent processes for landfill leachate treatment used as the downstream stage of integrated treatment or integral treatment (Yao, 2013). Several studies have been using RO in the landfill leachate treatment (Cingolani et al., 2018; Di Maria et al., 2018; Dolar et al., 2016; Schiopu et al., 2012; Talalaj et al., 2019). In the early 2010s, Šír et al. (2012) reported that RO-plants were installed in more than one hundred landfill sites in northern Europe, North America, and the Far East. Besides, membrane filtration processes have been used at landfills in Brazil, for example, at the landfills of Seropédica, São Gonçalo, Gramacho and Campos in Rio de Janeiro State, and Rio Claro and Osasco in São Paulo State (Costa et al., 2019; De Almeida et al., 2019; De Almeida et al., 2020).

The published literature reports studies that evaluated the technical feasibility of RO in the treatment of leachate. In general, the process performance in terms of removal of pollution parameters, for example, COD, ammonia nitrogen, and chloride, were investigated (Košutić et al., 2014; Kuusik et al., 2014; Šír et al., 2012; Smol et al., 2016; Talalaj, 2015). This is due to the ability to retain contaminants dissolved in leachate with high efficiency. Cingolani et al. (2018) reported that RO can remove pollutants from leachate with an efficiency up to >99%. For this reason, it is preferred over other technological options. However, to the best of our knowledge, few researchers have examined in detail the landfill leachate management costs. Moreover, companies that run leachate treatment plants do not provide the actual process costs.

Within this context, the main objective of this study was to estimate the cost of landfill leachate treatment from a full-scale RO plant located in Rio de Janeiro State, Brazil.

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Appendix IV. Technical and economic aspects of a sequential MF + NF + zeolite system treating landfill leachate. ***J. Environ. Sci. Heal. Part A***, 57 (8), 675-684, 2022. (IF = 2.582)

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Technical and economic aspects of a sequential MF + NF + zeolite system treating landfill leachate

Ronei de Almeida, Tairini Joyce Pimenta de Oliveira, Rosana Maurício Gouvea, and Juacyara Carbonelli Campos [iD]

School of Chemistry, Universidade Federal do Rio de Janeiro, Rio de Janeiro, Brazil

ABSTRACT

This work explores the techno-economic aspects of landfill leachate treatment by an integrated scheme composed of microfiltration (MF), nanofiltration (NF), and zeolite application for carbon and nitrogen removal. In bench-scale experiments, MF and NF were investigated, and zeolite batch tests were carried out to determine optimum conditions. A preliminary economic analysis is presented for a $200\,m^3\,d^{-1}$ full-scale treatment facility based on the data obtained from experimental tests and literature surveys. The maximum removals of 92%, 94%, and 79% for chemical oxygen demand (COD), absorbance at 254 nm, and ammonium nitrogen (NH_4^+-N) were achieved in bench experiments, respectively. It was possible to reach the local discharge standard for COD ($200\,mg\,L^{-1}$), but it was not possible to reach the Brazilian disposal requirement for NH_4^+-N ($20\,mg\,L^{-1}$). The total cost of the integrated MF + NF + zeolite system was estimated at 19.89 US$m^{-3}. In this study, the costs of the zeolite application account for around 70% of the total costs of the integrated scheme. Membrane process integration was an adequate strategy for removing organic compounds at low operating costs; However, further NH_4^+-N depuration is needed to meet discharge requirements.

Introduction

Landfill leachate is one of the most complex and polluting wastewaters; hence, its management requires an efficient treatment to avoid negative eco-environmental impacts and ensure the quality of water resources. Despite different contaminants found in waste leachates, organic matter and ammonium-nitrogen (NH_4^+-N) remain the primary target pollutants for removal in the treatment chain due to their adverse impacts, including eutrophication and ecotoxicity.[1]

Several technologies are applied for landfill leachate treatment, from biological to physicochemical processes or combining these methods in hybrid systems or sequential steps. Membrane filtration techniques (e.g., microfiltration [MF], nanofiltration [NF], and reverse osmosis) have been demonstrated to be indispensable for leachate treatment, and many landfill managers have been operated membrane systems.[2,3] MF and NF processes have removed organic compounds and provided high-quality treated effluents at low operating costs.[4,5] Advantages of membrane techniques include modular design, automation, scale-up, and reduced carbon footprint.[2,6] MF and NF are consolidated technologies in full-scale leachate treatment plants (LTPs). However, membrane fouling is a major issue since it increases cleaning requirements, energy consumption and membrane replacement, directly impacting the field application of membrane technology. For instance, it is estimated that fouling costs

range from 11 to 24% of operating expenses in water treatment plants using reverse osmosis or NF systems.[7] Besides, MF/NF alone cannot result in treated leachate that meets the NH_4^+-N disposal standards set by legislative authorities.[8–10] Therefore, additional treatment is required for ammonia removal. The ion exchange method has proven to be an attractive process among treatment options. Low-cost minerals like zeolites can be utilized as an ion exchanger material, providing ammonia removal efficiencies greater than 97% at NH_4^+-N initial concentration of 5-200 mg L^{-1}.[9] Other benefits of zeolite application include high sorption and ion exchange capacity, easy-to-operate, and material regeneration, which permits its use in multiple operation cycles.[11,12]

Apart from the technical efficiency of each technology, economic performance plays a significant role, and the decision-making should be based on the cost-effectiveness of the treatment scheme. On the other hand, companies that run leachate LTPs do not provide actual costs associated with landfill leachate management, and only a few studies were published about this theme.[13–16] In this sense, our work contributes to research on techno-economic aspects of LTPs by demonstrating the technical characteristics of a sequential leachate treatment scheme and its related expenses.

Our previous work confirmed that zeolites are cost-effective sorbents and could be used as a polishing step for

Appendix V. Almeida, R. et al. Lessons and challenges for the recycling sector of Brazil from the pandemic outbreak of COVID-19. ***Waste Disposal & Sustainable Energy***, 3,145-154, 2021.

ARTICLE

Lessons and challenges for the recycling sector of Brazil from the pandemic outbreak of COVID-19

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Abstract

The 2019–2020 coronavirus pandemic imposed unprecedented challenges in Brazilian governance sectors, mostly in the waste management system. Herein, we analyse the main challenges of the recycling sector in Brazil to cope with this scenario. Understanding difficulties during the pandemic outbreak of COVID-19 can help improve waste recycling in the post-pandemic period in Brazil and other developing nations that face similar issues. The current pandemic exposed the deficiencies of this system, and some important lessons can be learned. Recommendations are drawn to advance the proper management of recyclables in the country. The Government must increase total investments in the recycling industry's infrastructure and support local recycling initiatives during a public health crisis and beyond. In sum, this paper strengthens the idea that waste segregation at the source and selective collection will not be sufficient without massive investments in the recycling sector's infrastructure. The future challenge includes strengthening the economic markets for recycled materials.

Graphic abstract
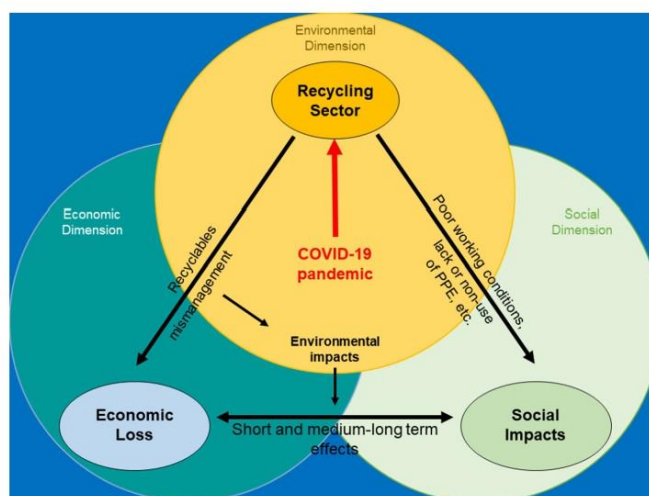
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Appendix VI. Campos, J. C., De Almeida et al. **Monitoramento dos parâmetros físico-químicos e microbiológicos dos lixiviados de células experimentais de resíduos domiciliares com infiltração do concentrado de osmose inversa**. In: 31º Congresso da Abes, 1–11, 2021. [In Portuguese]

III-765 – MONITORAMENTO DOS PARÂMETROS FÍSICO-QUÍMICOS E MICROBIOLÓGICOS DOS LIXIVIADOS DE CÉLULAS EXPERIMENTAIS DE RESÍDUOS DOMICILIARES COM INFILTRAÇÃO DO CONCENTRADO DE OSMOSE INVERSA

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RESUMO

O presente estudo tem como objetivo avaliar os parâmetros físico-químicos e microbiológicos dos lixiviados gerados em células experimentais de aterro de resíduos domiciliares (RDOs) com infiltração de concentrado de osmose inversa (COI). Foram montadas seis células experimentais de RDOs. Três células foram utilizadas como células controle (grupo controle, C_0) e as demais, foram utilizadas para infiltração do COI (grupo infiltração, C_I). A massa específica dos RDOs dispostos nas células foi de cerca de 600 kg/m³. Semanalmente, foram infiltrados 900 mL de COI proveniente de um aterro sanitário de grande porte localizado no Estado do Rio de Janeiro. A caracterização dos lixiviados gerados nas células experimentais foi realizada com base nos seguintes parâmetros: potencial hidrogeniônico (pH), Demanda Química de Oxigênio (DQO), nitrogênio amoniacal (N-NH₃) e condutividade. As populações de coliformes totais e *Escherichia coli* foram expressas em Números Mais Prováveis por 100 mililitros de amostra (NMP/100 mL). O monitoramento ocorreu durante 117 dias. As análises estatísticas foram realizadas utilizando o programa *STATISTICA* 10 (licenciado pela *Stat Soft*). No período monitorado, o valor médio de pH do lixiviado das células C_I foi menor que das células C_0 (4,62 vs. 5,40) e a concentração média de DQO foi maior para as células C_I (18,45 vs. 7,70 g O₂/L) (p-valor < 0,05). Em contrapartida, não houve diferença estatística significativa em relação aos parâmetros condutividade e N-NH₃ (p-valor > 0,05). Além disso, observou-se que as características físico-químicas dos lixiviados foram inibitórias para a proliferação dos microrganismos investigados. Recomenda-se o monitoramento dos lixiviados gerados por um maior período e a inclusão de outros parâmetros de poluição na caracterização desse efluente em avaliações futuras.

PALAVRAS-CHAVE: Aterro sanitário, Concentrado, Lixiviado, Osmose inversa, Resíduos domiciliares

Appendix VII. De Almeida et al. Techno-economic evaluation of landfill leachate treatment by hybrid lime application and nanofiltration process. *Detritus*, 10, 170-181, 2020. (IF = 0.0)

TECHNO-ECONOMIC EVALUATION OF LANDFILL LEACHATE TREATMENT BY HYBRID LIME APPLICATION AND NANOFILTRATION PROCESS

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ABSTRACT
Leachate treatment is a major issue in the context of landfill management since solutions have not been yet developed, resulting in more satisfactory technical and economic results concerning leachate treatment. In this paper, the technical and economic factors concerning lime application and nanofiltration for the treatment of leachate from the Seropédica landfill (Rio de Janeiro State, Brazil) were evaluated. The results indicate that the application of 30 g lime L^{-1}, under optimum conditions, followed by air stripping, was able to place the effluent within the ammonia nitrogen discharge standard imposed by local legislation. The use of nanofiltration produced a clear and colorless permeate and has proved to be very effective at removing all pollutants. Regarding cost estimates, considering a means leachate generation flow of 1000 m³, recovery of 60% and average permeate flux of 12 L m^{-2} h^{-1}. The total cost per m³ of treated effluent was estimated in two scenarios, using different types of membrane and therefore different membrane costs per m². Considering that the landfill would operate for 25 years and after closing, the leachate treatment station would maintain its activities for another 15 years, totaling 40 years, the cost to treat leachate would be of US$ 10.54 and US$ 11.33 m³. In both evaluated scenarios, with regard to process operation costs, the percentage value relative to membrane exchange was emphasized. It is noteworthy that, a treated effluent at a lower cost to that currently presented by the landfill was obtained through the applied hybrid process.

1. INTRODUCTION

Considering the high concentration of pollutants and varying composition influenced by the type of waste, landfill age, and geological conditions, leachate treatment is undeniably one of the most challenging tasks in municipal solid waste management (Kjeldsen et al., 2002; Zhang et al., 2019). Landfill leachate contains high loads of organic matter, inorganic salts (sulfates, carbonates, and sodium chloride), ammonia, and halogenated and heavy metals that must be treated before being released into the environment (Kjeldsen et al., 2002). According to the literature, critical parameters for most landfills are chemical oxygen demand and ammoniacal nitrogen (NH_3-N) (Ehrig and Robinson, 2010). Although many methods may apply, the most appropriate leachate treatment choice will depend on its features, technical applicability, cost-effectiveness, and other factors related to the quality requirements of the effluents.

Main classification of the technical solution and

technology classification for leachate management and treatment on the site or at the leachate treatment plant could be divided into following groups: leachate treatment on the site or transfer to the central wastewater treatment plant (leachate lagoons and recirculation into the landfill body or at the surface, combined leachate with the domestic sewage system and treatment at the wastewater treatment plant), biological processes (different combination of the aerobic and anaerobic processes), physicochemical processes (chemical oxidation, adsorption on activated carbon, chemical precipitation, coagulation-flocculation, air stripping) and membrane processes (main reverse osmosis and nanofiltration) (Ehrig and Robinson, 2010; Schiopu et al., 2012; Serdarevic, 2018; Yao, 2013). The conventional biological process could be effective for the removal of organic biodegradable substances, suspended solids and nutrients (Metcalf et al., 2003; Zhao and Zyyang, 2019). With time, the major presence of refractory compounds (mainly humic and fulvic acids) contribute to limit

Appendix VIII. Almeida, R.; De; Campos, J. C. Análise Tecnoeconômica Do Tratamento de Lixiviado de Aterro Sanitário. *Revista Ineana* 2020, 8 (1), 6-27.

Análise tecnoeconômica do tratamento de lixiviado de aterro sanitário

Techno-economic analysis of the treatment of landfill leachate

> Ronei de Almeida; Juacyara Carbonelli Campos

> ### Resumo

Um dos principais problemas relacionados ao gerenciamento dos resíduos sólidos é o efetivo tratamento dos líquidos lixiviados gerados em aterros sanitários. Até o momento, não foram desenvolvidas soluções que tragam resultados técnicos, ambientais e econômicos satisfatórios no tratamento desse tipo de efluente. Nesse contexto, foram avaliados os fatores técnicos e econômicos da aplicação de cal e nanofiltração (NF) no tratamento do lixiviado do aterro sanitário de Seropédica (RJ). Os resultados indicaram que a aplicação de 30 g cal.L^{-1}, seguida de arraste por fluxo de ar em vazão de 1,33 L.min^{-1} e tempo de retenção hidráulica de 24h, removeu matéria orgânica recalcitrante do lixiviado e adequou o efluente ao padrão de descarte de 20 mg.L^{-1} para nitrogênio amoniacal estabelecido pela Resolução CONAMA nº 430/2011. Posteriormente, a utilização da NF, em pressão operacional de 8 bar, adequou o efluente ao padrão de descarte de Demanda Química de Oxigênio estabelecido pela legislação estadual. Considerando que o aterro sanitário operaria por 25 anos e que, após o encerramento das atividades, a estação de tratamento de lixiviado manteria sua operação por mais quinze anos, totalizando um período de quarenta anos, o custo total de tratamento do lixiviado seria de US$ 10.54.m^{-3}.

> ### Abstract

One of the main problems related to solid waste management is the effective treatment of leachate liquids generated in landfills, since no solutions have been developed, to date, resulting in more satisfactory technical and economic results concerning leachate treatment. In this context, the technical and economic factors concerning lime application and nanofiltration (NF) for the treatment of a leachate from the Seropédica (RJ) sanitary landfill were evaluated. The results indicate that the application of 30 g lime.L^{-1} followed by stripping, at a 1.33 L.min^{-1} flow and hydraulic retention time of 24 h, removed organic matter from the leachate, and to placing the effluent within the 20 mg.L^{-1} ammonia nitrogen discharge standard established by CONAMA Resolution 430/2011. Subsequently, the use of a NF process, 8 bar operating pressure, placing the effluent within the COD standard of 250 mg.L^{-1} established by local legislation. Considering that the landfill would operate for 25 years and after closing, the leachate treatment station would maintain its activities for another 15 years, totaling 40 years, the total cost to treat leachate the m^3 of leachate would be of US$10.54.m^{-3}.

Palavras-chave

Amônia. Avaliação Tecnoeconômica. Cal. Coagulação-floculação. Lixiviado.

Keywords

Ammonia. Coagulation-flocculation. Leachate. Lime. Techno-economic Evaluation.

7

Appendix IX. Evaluation of coagulation–flocculation and nanofiltration processes in landfill leachate treatment. *J. Environ. Sci. Heal. Part A*, 54 (11), 1091-1098, 2019. (IF = 2.582)

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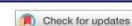
Evaluation of coagulation–flocculation and nanofiltration processes in landfill leachate treatment

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ABSTRACT

Landfill leachate consists mostly of a high content of refractory organic matter, ammonia and toxic compounds. All these compounds, regardless of their nature, have a potential pollution effect on local ground and surface waters. In this context, the purpose of the present study was to evaluate a treatment process (coagulation–flocculation with lime coupled with nanofiltration) applied to landfill leachate from Seropédica, Rio de Janeiro (Brazil). Jar tests were conducted to determine the optimum dosage of lime (ranged from 0 to $10\,g\,L^{-1}$) and, at optimum lime dose, ammonia nitrogen was removed during coagulation–flocculation process due to high pH. The process was settled for 6 h with slow stirring (50 rpm) to promote air entrainment and NH_3-N stripping before using the final treatment step of nanofiltration at 8 bar. After ammonia stripping, NH_3-N was reduced from $1,236\,mg\,L^{-1}$ to $353\,mg\,L^{-1}$ (71% removal efficiency). At the end of the combined treatment, TOC (total organic carbon), HS (humic substances) and COD (chemical oxygen demand) removals were 89%, 80% and 94%, respectively. The results showed that the combined process was effective in the removal of recalcitrant compounds and NH_3-N.

Introduction

One of the main problems related to solid waste management is the effective treatment of leachate liquids produced in landfills. Leachate is an aqueous matrix potentially polluting and extremely complex, because of its high amount of inorganic matter, such as ammonia nitrogen, and organic matter, such as humic substances (HS), among others. These substances are refractory and recalcitrant to biodegradation. Moreover, the composition of landfill leachate can exhibit considerable spatial and temporal variations depending upon site operations and management practices, refuse characteristics, and internal landfill processes.[1]

Regarding landfill leachate treatment, there is not a simple and universal solution. Literature reports that the variations in the composition of leachates strongly affect the efficiency of treatment processes.[2–4] Conventional treatments, derived from the treatment of wastewater, have, so far, mainly consisted of biological degradation of leachate, either by lagooning with activated sludge or by fixed or fluidized bed reactors. This biological degradation is mainly intended to remove chemical oxygen demand (COD), biochemical oxygen demand (BOD_5) and ammonia nitrogen (NH_3-N). Depending on the quality of leachate and the local regulatory constraints, this biological treatment has often been coupled with other physicochemical treatments, specifically adapted to the pollution to be treated, such as chemical oxidation, adsorption on active carbon, coagulation–flocculation (C-F) or chemical precipitation.[5]

According to Renou et al.[5] and Lima et al.,[6] biological treatments are particularly effective for young leachates whose organic fraction is mainly composed of volatile fatty acids. However, in Brazil and also in Europe, America and Asia, most municipal waste landfills are 10 years old or even more and their leachates are increasingly stabilized.[6] The concentration of HS is high, namely humic and fulvic acids – refractory organic compounds of high molecular weight – and cannot be adequately treated by biological degradation processes any longer, even if coupled with physicochemical treatments.

According to Lima et al.,[6] the presence of refractory organic compounds in high concentrations in raw leachate causes various problems concerning its treatment, such as the formation of potentially toxic products and more complex substances. Since these traditional processes do not accomplish the increasingly strict regulations for leachate discharge, it has become necessary to design new treatment methods.

C–F may be successfully used in treating stabilized and old landfill leachates. It is widely used as a pretreatment, prior to biological or nanofiltration (NF) step, or as a final polishing treatment step in order to remove non-biodegradable organic matter.[7,8] Moreover, the application of C–F with lime in the treatment of leachate reduced the salinity of this effluent from 15% to 30% and decreased the concentration of HS through the precipitation of humic acids. In

Appendix X. INEA Award 2019 (in Portuguese, 1º Prêmio INEA de Meio Ambiente).

CERTIFICADO

A Universidade do Ambiente certifica que

Ronei de Almeida, participou do 1º Prêmio de Meio Ambiente do INEA, conquistando o 1º lugar, com o trabalho: Análise tecno- econômica do tratamento lixiviado de aterro sanitário.

18 de novembro de 2019

Carlos Henrique Netto Vaz
Presidente do Instituto Estadual do Ambiente

Renan Guimarães Escopeli Gomes
Diretor de Gente e Gestão

Appendix XI. Menção Honrosa em Sessão de Apresentação do Centro de Tecnologia [CT] na XLII Jornada Giulio Massarani de Iniciação Científica, Tecnológica, Artística e Cultural [JICTAC 2020 – Edição Especial] [In Portuguese].
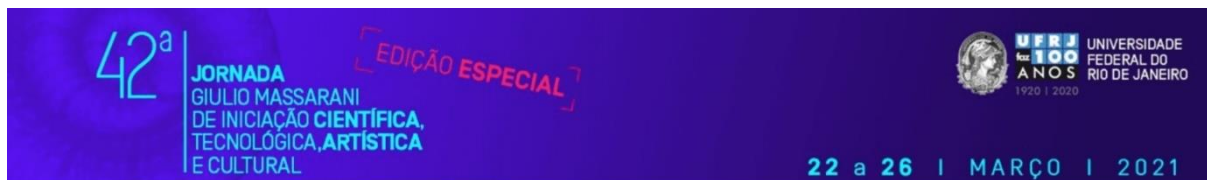
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CERTIFICADO

Certificamos que o trabalho **CARACTERIZAÇÃO FÍSICO-QUÍMICA DO CONCENTRADO PROVENIENTE DE TRATAMENTO POR OSMOSE INVERSA DE LIXIVIADO DE ATERRO SANITÁRIO,** de autoria de **Raphael Ferreira Porto, RONEI DE ALMEIDA e Juacyara C. Campos**, recebeu **Menção Honrosa em Sessão de Apresentação do Centro de Tecnologia (CT)** na XLII Jornada Giulio Massarani de Iniciação Científica, Tecnológica, Artística e Cultural (JICTAC 2020 - Edição Especial), realizada de 22 a 26 de março de 2021.

Rio de Janeiro, 04 de maio de 2021

Profa. Denise Maria Guimarães Freire
Pró-reitora de Pós-graduação e Pesquisa / UFRJ

Profa. Gisele Viana Pires
Pró-reitora de Graduação / UFRJ

Prof. Leonardo Holanda Travassos Corrêa
Coordenador da JICTAC / UFRJ
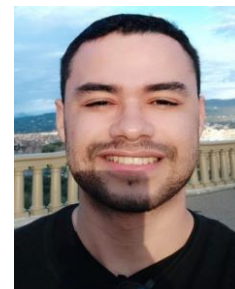
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Curriculum vitae

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08/2010 – 09/2016	**B.Sc. in Chemical Engineering**
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03/2018– 12/2018	Employee, Assistant Professor, Biochemical Engineering Department, School of Chemistry, Federal University of Rio de Janeiro
	Undergratuate courses: Environmental Engineering, Environmental Biotechnology, Environmental Control in the Food Industry, and Fundamentals of Biochemical and Chemical Processes

Scholarships, Grants & Awards

♦ Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), 2018, Doctorate - GD – Grant #165018/2018-6.

♦ Fundação de Amparo a Pesquisa do Estado do Rio de Janeiro (Faperj), 2021. International Mobility – Grant #E-26/200.065/2020.

♦ 1st Environment State Institute's Award, Solid Waste Disposal in Rio de Janeiro: Innovative Solutions, 2019.

♦ Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), 2016, Master - GM – Grant number: 147737/2016-8.

♦ Fundação de Amparo a Pesquisa do Estado do Rio de Janeiro (Faperj), 2015, Undergratuate – Grant #E26/200.731/2015. Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), 2013 and 2014, Undergraduate – Grants #800339/2013-4 and #147676/2013-4.

Hobbies

♦ Gym Workout, Sunbathing, Drink & Chat