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Landfill leachate treatment strategies: Energy recovery and reduction of environmental impacts

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Abstract. The work commences by addressing characterization and regulatory constraints, underlining the spatio-temporal variability of leachates and the increasingly stringent requirements for their discharge. Following this, treatment technologies are reviewed, covering those from classical physico-chemical treatment schemes (coagulation and struvite precipitation) to advanced oxidation (ozonation and UV-light) and biological treatment approaches (anaerobic digestion treatment and aerobic granular sludge). The authors also look collectively at hybrid systems and focused attention on resource extraction and enhanced recovery, especially from leachates with attention being paid to the adoption of membranes enhanced for sustainability. The heart of the article discusses energy and material recovery pathways from leachates, which is considered the secondary focus of the paper treatment comes first. Attention is paid to biogas production from UASB reactors and co-digestion and nutrients recovered in the struvite form. Environmental impacts highlight the advantages that come through the donation of resource and material recovery from leachates and circularity. The article ends by looking at issues that remain in treatment such as variability of effluents, energy costs and membrane fouling as major issues and propositions for research perspectives encapsulating those areas. The authors conclude by talking about the necessity for progression to combined treatment that can turn storage sites into true biorefineries, and not just sites for storage, with the production of resources and an eye on environmental protection/security as key components.

1 Introduction

Leachates stemming from nonhazardous waste storage facilities (ISDND) are exceedingly complicated because these effluents arise from the percolation of rainwater through the installed waste as well as from the metabolism of its constituents [1]. By far the most significant effluents generally produced within the scope of sanitary reclamation of refuse, these polluted liquids are characterised by more or less acute organic pollution, high ammoniacal nitrogen and heavy metal contents, and composition that frequently changes greatly from day to day [2]. n untreated form, leachates may contaminate acquifers, underground and surface waters and lands [3]. They often produce losses to fish and cause other serious harm to public health and daanger to human beings [4]. Recently, a different point of view has prevailed, according to which leachate is a resource to be exploited to recover energy and nutrient, while processing of natural resources through advanced technology introduces further steps into every recquired resource-value chain, creating an economic habit for every environmental constraint[6]. Is a way of changing things around [7]. Here is presented further the state of the art of technologies for the treatment of landfill leachates with 'energy recovery'/recovery of nutrients and coagulation of the reduced environmental impact through various process integrations and optimizations.

The review article provides a thorough examination of leachate treatment strategies, emphasizing energy recovery and minimizing environmental impacts. Leachate's liquid resulting from the passage of water through waste are a significant challenge due to their complex, variable composition, containing a plethora of organic matter, ammoniacal nitrogen, heavy metals and emerging contaminants.

2 Characterization of leachate and regulatory framework

2.1 Origin and composition

Landfill leachate is derived from rain which percolates through waste and dissolves organic and inorganic constituents [8]. Its chemical composition varies dramatically with the type of waste, climatic and operating conditions [9]. Fresh leachates contain a large proportion of biodegradable organic matter with a high ratio of BOD and COD [10]. Old leachate will have low biodegradability due to the build-up of refractory compounds—especially humic substances [11]. Nutrient concentrations of ammoniacal nitrogen and phosphorus are often so high as to require specific treatment [12]. The occurrence of emerging contaminants such as PFAS further complicates processing [13].

2.2 Critical pollution parameters

Chemical oxygen demand (COD) is considered the most important indicator of organic pollution of landfill leachatess [14]. Ammoniacal nitrogen is another persistent pollutant, which does not disappear even after significant landfill maturation [15]. Heavy metals, especially lead, chromium and nickel, have toxicological risks requiring their effective disposal [16]. Recalcitrant organic compound does not degrade by simple biodegradation processes [17]. Salinity and alkalinity often reach levels that compromise most biological treatment systems. Associated hazardous effects in the pollution mixture require integrated, multi-step treatments [10].

2.3 Regulatory framework

The need to tighten restrictions on the quality of such effluents has urged landfill operators to improve their treatment performance [8]. Old leachates are often not amenable to treat by more conventional treatment technologies to allow them to be exported to modern landfills [9]. Multi-step treatments are a generally applicable strategy to meet such environmental goals [10]. Economic factors also impede the installation of sophisticated systems in many facilities [5]. Variability around leachate composition means that constant regulatory compliance may be unattainable [8]. The discrepancy between what treatment systems can achieve and what can be tolerated on rejection drives the innovation of treatment technologies [13].

3 Overview of treatment technologies: from remediation to healing

3.1 Conventional physico-chemical treatments

3.1.1 Coagulation-flocculation and precipitation processes

Coagulation-flocculation favours the efficient removal of suspended solids and colloidal organic matter from landfill leachates [16]. pH adjustment in an acid medium enhances the precipitation of humic acids by means of protonation. For example, a gradual decrease in pH below 2.0 [is reported to cause] precipitating of humic acids, thereby permitting removal of 86.1% of the COD in that mechanism alone and a colour reduction on the leachate of 84.7% [20]. The zeta potential which is negative at neutral pH becomes closer to zero during acidification; which was indicative of an efficient destabilization of colloidal structures [21]. The use of aluminum-based EC is particularly promising and has given an apparent colour removal of 92.5% at pH 5 and a further removal of 58.75% COD and 47.24% total coliforms, at an estimated cost of treatment of 3, $00/m3 [22].

Struvite precipitation has now established itself as a two-in-one technology, which allows for the simultaneous removal of nutrients by a slowly released fertilizer [23]. his process is most effective at about pH 9.0-10.5, at which magnesium, ammonium and phosphate ions precipitate as MgNH PO 6H O [24]. crystals. At the optimum condition (pH 9.0, reaction time 30 minutes, AMP molar ratio of 1:1.5:1.5) the struvite precipitation technique should give a nitrogen and phosphorus recoveries of more than 90%. Ammoniacal nitrogen and phosphate removals in the leachate of 70% and 57,1 % respectively from leachate effluent anaerobic digestion [25]. The effects of cadmium and nickel etc. have been shown to have little deleterious effect on the purity of the precipitated struvite so it could be safely recovered from contaminated leachate sources [24].

3.1.2 Adsorption and advanced oxidation processes

Electrochemical oxidation (EO) stands as one of the few methods that can undeniably destroy compounds unsuitable for biodegradation or representing environmental difficulties such as aqueous halogens, even in old landfill leachates. It is based on generation of extremely active hydroxyl radicals, leading to complete removal of organic contaminants and avoiding chemical transport, with de minimis sludge formation [27]. Specific combinations of irradiation with electrochemically activated persulfate demonstrated a synergistic effect ,removal efficiencies showed for COD 55.6%, for UV 68.5% and for color 89.9%, and were dependent on pH3, current 3A, persulfate dose 5g/L, and reaction time 25 minutes [28]. The total energy consumption of this process denominated “UV/Electro-Sep” was ascertained as 27.4kWh/ kg COD, placing the energy use itself into perspective - this input needs to be justified by efficiency of treatment. Magnetite-catalyzed ozonation constitutes a novel process of “selective” oxidation [13]. Magnetite-catalyzed ozonation pretreatment showed enhanced specific methanogenic activity at 0.22g CH -DCO/(gVSS j) in 40 minutes, and thus its optimal coupling point with the subsequent anaerobic biological treatment [31].

3.2 Biological treatments: the heart of organic recovery

3.2.1 Anaerobic digestion for biogas production (CH_4)

Anaerobic digestion constitutes the basic enabling technology for conducing the removal of organic matter accompanied by a simultaneous production of renewable energy [32],[33]. UASB reactors perform well for the treatment of high concentration leachates of different climates [34],[35]. The production of biogas enriched in methane is amenable to energy recovery together with greenhouse gas reduction [36]. Co-digestion improve the stability of the process and increase the amount of biogas produced [37]. Electrochemical pretreatment combined with the application of zerovalent iron increases biomethane potential [38]. The structure of the microbial community influence directly the performance of methanogenesis and stability of the system [39].

3.2.2 Aerobic treatments for nitrogen removal

Aerobic granular sludge technology presents a remarkable removal efficiency for organic matter and nitrogen [15]. having this also sorting properties such as biosorption to remove pollutants and metals and likewise bioaccumulation "to recover"[16]. The technique of aerobic granular sludge (BGA), is effective in removing pollutants and metals from landfill leachates. A 2-L sequential batch reactor for 198 days eliminated 94% of COD and 97% total nitrogen, where biosorption mechanisms constituted 53% and bioaccumulation 47% of all mechanisms involved. The desorption of these metals with NaCl 0.1 M does not significantly affect granules' structure, although it diminishes nitrification and denitrification activities, extending heterotrophic activity instead [40] Typical influent for aerobic granular sludge (BGA) would be raw landfill leachate, hence cationic composition is dominated by calcium (1608.7 ± 86.6 mg/L) and magnesium (518.0 + 48.5 mg/L), followed by iron (252.3 ± 44.2 mg/L) and manganese (104.6 ± 6.5 mg/L). Trace metals (chromium, zinc, copper, lead) are presented at concentrations lower than ~3 mg/L **Figure 1**[40] .

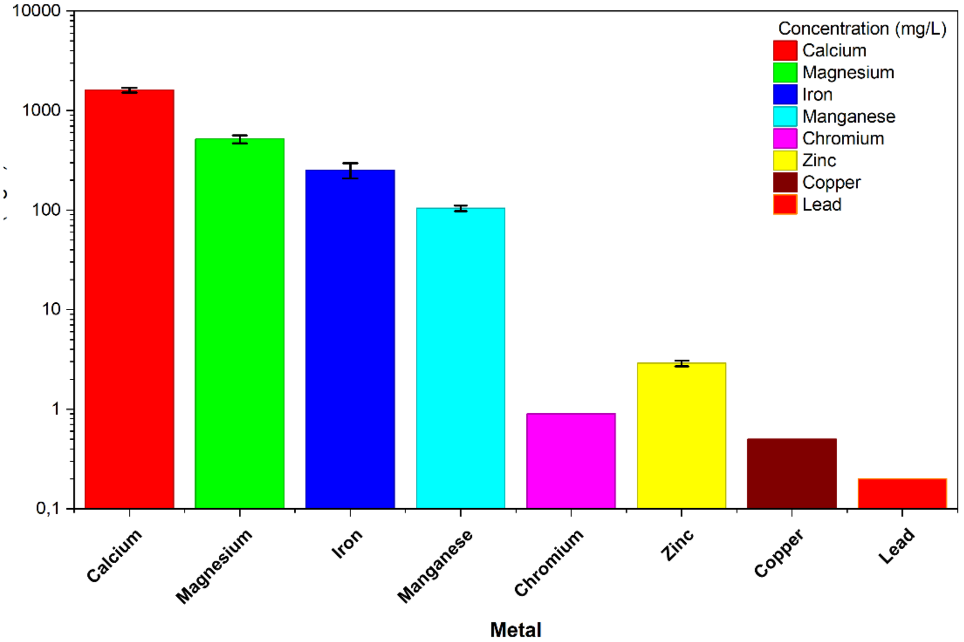

Fig. 1. Concentration of metals in raw landfill leachate used as influent for BGA reactor. Error bars represent standard deviation (n = 3).

The coupling of advanced oxidation and biological nitrogen removal leads to a COD removal rate of around 75% in a UASB reactor, following pre-treatment by magnetite-catalyzed ozonation. However, removal of ammoniacal nitrogen and phosphorus remains deficient, mitigated by further treatment with microalgae, resulting in a 65% decrease in total nitrogen and a 70% decrease in total phosphorus removals [31].

3.2.3 Hybrid and natural technologies

Hybrid bioreactors use aeration and leachate recirculation to improve degradation rates [41]. Partial aeration strategies try to achieve aerobic and anaerobic regions in one reactor. Br-2 reaches 8 pH before reaching the maximum reaction time, while Br-1 does not go above 7. Physico chemical reduction varies from 33% to 98%. Br-2 produces 21.5 L biogas and more than 60% methane while Br-1 produces only 8.75 L biogas with less than 50% methane. [42]. Nanofiltration membranes are indispensable for the optimal selective separation of organic compounds as well as inorganic salts [43], but with lower operating pressures [44]. Though reverse osmosis is relatively inexpensive disposal it takes up too much of the energy [45]. Despite advances, fouling of membranes remains a significant challenge, but scientists have been working on new membrane materials and surface modifications to improve their resistance to this problem [46]. A combination of a hybrid system and integration with biological processes allows to optimize the system and enhance the efficiency [47]. Modified nanofiber membranes, especially those made of polyacrylamide with silver nanoparticles and a green tea extract show promising potential in the elimination of nitrates and achieving an efficiency of up to 73,25%. The membranes show a significant reduction in the diameter of the fibres and an increase porosity. They are also more hydrophilic and manage to decrease the turbidity of wastewater from a value of 32 NTU to 3 NTU. Substrates achieve up to an effectiveness in nitrate removal of 73,25% within contacted period of time. Ultimately offering a galvanic recovery with efficiency and a superior resistance compared to unmodified membranes [32], [47].

4 Ways of recovering energy and materials

The methanogenic potential of landfill leachates is related to substrate, community and operational parameters. A two-step process of anaerobic fermentation and methanogenesis leads to the acquisition of biohydrogen and biomethane. With anaerobic fermentation moderate yields of hydrogen are achieved 6.1 to 8.2 NmL H/g volatile dry matter, while with anaerobic digestion more methane is produced 275.2 to 277.5 NmL CH/g volatile dry matter. The system demonstrates a biodegradability of 66% of volatile materials, and a COD removal between 50.8 and 60.1%, proving the greater efficacy of the methanogenesis rather than the hydrogen genesis for the valorisation of the lexiviates [44]. For treatment of leachates and gas production, the innovative coupling of storage and anaerobic fermentation is considered. The anaerobic storage of leachates (starting DCO of 5 353 mg/L) at 23 °C within for 15 days converts organic matter into lactic acid, favouring the production of biohydrogen. As inoculum in the anaerobic fermentation of leachates, although COD is noticeably reduced (93.63%), palm mill effluents do not lead to hydrogen production due to their inhibition by exogenous microorganisms [45].Here we note how, despite the continued issues in hydrogen production, the coupled process treats the leachate well, via degradation of organic matter in **Table 1**.

Table 1 : Anaerobic Digestion Systems for Energy Recovery from Landfill Leachate.

System Configuration	HRT (hours)	COD Removal (%)	CH_4 Yield (%)	Energy Production	Refs

UASB (ambient temp)	06-oct	64±1	86.1±1.1	Not specified	[44]
UASB + Ozone pretreatment	Variable	75	Not specified	Optimized for methanogenesis	[31]
Hybrid bioreactor	Variable	33-98	67	21.5 L biogas (41× increase)	[45]
EC pretreatment + ZVI (10 g/L)	Not specified	Not specified	70.12	0.2805 NL/g sCOD removed	[46]
Co-digestion (leachate + kitchen waste)	Variable	>90% organics conversion	High	>90% gas in 1 day	[47]

Raw biogas from the treatment of landfill leachate by anaerobic digestion typically ranges from 50-67% methane, 35-40% carbon dioxide and hydrogen sulfide, together with other contaminations [42]. [45] . In the case of biomethane to be freed of CO and im-purities up-to-the-mark, it is necessary to proceed with purification technologies membrane separation, pressure swing adsorption, or chemical wash. Membrane systems become the most economical being as well the cost of 5,654$/m3, of which water washers and chemical absorbers are excluded, indubitably peculiar to the valorization of biogas [45].
The recovery of the nutrients contained in landfill leachates corresponds to simultaneously fulfilling prevention of pollution and retaining resources deserving to provide alternatives to a mineral fertiliser [44]. The lightweight capital of crystallization of struvite also provides that phosphorus to be recovered deeper than 60% while the surplus part nitrogen to be stripped off [47].

5 Environmental analysis and challenges of sustainable management

The landfill leachate efficient management, however, greatly minimizes the potential for greenhouse gases, as well as preventing the pollution of groundwater and soil. The valorization of methane from an anaerobic treatment system means that it is possible to mitigate emissions of a greenhouse gas with a global warming potential 25 times greater than that of CO. Life-cycle analysis demonstrates that the integrated treatment approaches with resource valorization have the largest environmental benefits. Moving from a linear waste disposal model to circular resource valorization alters environmental profiles according to the fundamental idea of converting liabilities into assets [44]. From an economic perspective, this option of treatment technology choice can be explained as follows. According to a life cycle cost assessment, fractional foaming (FF) treatment is costed at between 77.4$–110.6$ per 1000m3 (about 0.077–0.111$/m3) depending whether the placement is a single, or comprised of three tiers [65]. Basically, the recovery of energy with biogas capture for use improves the economic viability of landfills, generating revenue for electricity sales, whilst nurturing the reduction of greenhouse gas emissions, but does not provide a figure in such terms. The recovery of nutrients in the form of struvite, or other fertilizer products creates further cash flow whilst assisting in attaining the circular economy objectives. The low implementation cost where advanced treatment facilities are concerned represents a stumbling block to their widespread implementation necessitating in-depth techno-economic analyses and incentive policy frameworks.

6 Research perspectives

In recent advances in landfill leachate treatment several priorities for future research remain in order to improve energy recovery and mitigate environmental impact [46]. These perspectives refer to improvement of existing processes, hybrid and novel technologies, and the systematic coupling of a circular economy approach.

- **Reduction of energy consumption and optimisation of advanced oxidation processes:** Despite the capability of advanced oxidation processes (AOP) like electrochemical oxidation and catalysed ozonation to degrade refractory pollutants [28]. they require high energy consumption. In the future research should be orientated toward the optimisation of operation parameters and the production of more effective and cheaper electrodes or catalysts, as well as coupling these processes with renewable energy. An interesting avenue of improving the energy gain is the coupling of targeted physico-chemical pre-treatments for reduction of the pollutant load before the POA [9],[10], [11], [12],[13].
- **Targeted treatment of emerging pollutants and heavy metals:** The treatment of leachate is complicated by the presence of emerging contaminants, like perfluoroalkyl and polyfluoroalkylated substances (PFAS), and persistent heavy metals [13], [15], [16]. Research should focus on technologies of destruction or selective extraction, via adsorption on new materials (modified biochars, nanostructured materials) or advanced electrochemical processes, capable of puting to sleep these specific pollutants without generating sub-toxic products [41], [46].
- **Intensification and robustness of biological processes:** The optimization of microbial communities within anaerobic (UASB) and aerobic (granular sludge, slime flocs) reactors is important to improve the stability and efficiency of the treatment, particularly if variable loads or temperatures are expected [34],[39], [40]. Research on microbiota, substrate pretreatment and co-digestion with other organic waste will increase biogas production and energy recovery.

- **Development of sustainable membrane materials and processes :** To overcome the main challenge of membrane fouling [46], research should focus on toxic tolerant resistant, more hydrophilic and selective membranes, based on surface modification or nanomaterials. The intelligent design and coupling of these membranes into hybrid treatments schemes aided biologically or via physico-chemical processes will improve their efficiency and sustainability [45],[47].

- **Systemic integration and sustainability analyses:** Encompassing environmental, economic and social performance, a holistic assessment will benefit from the deepening of life cycle analyses (LCA) and life cycle costing (LCC), to be able to systematically compare the scenarios from decontamination to valorization [47].

7 Conclusion

Landfill leachates are a two-fold difficulty and potential resource, in the context of a circular economy. This review evidenced that their sustainable management cannot be dependent only on a single technology, but on an intelligent mix of physico-chemical, biological and advanced separation processes, fitting the specificity of each effluent. Physico-chemical treatments, like coagulation-flocculation and struvite precipitation removal, allow effective removal of suspended solids, nutrients and some metals. AOP, albeit costly in terms of energy, accept advantageously to destruct refractory pollutants. Biological systems, in the form of anaerobic digestion and aerobic granular sludge are at the core of the organic valorization, allowing for pollution abatement and renewable energy generation, under the form of biogas. The sensible tendency is to mix and match hybrid systems, coupling oxidizing pretreatments influx bioreactors, or even combining membranes and biological processes, in order to optimize efficiency and reduce energy consumption. Recovery of resources – energy, nutrients (struvite), water, and metals – appears as an important further step to change storage places into true biorefineries, making life a bit less landfill heavy. Nevertheless, the quest for a fully sustainable, and circular, management of leachates is still fraught with challenges related to variability of effluents, costs of advanced technologies and their integration. Future work therefore remains to energy optimisation, development of robust materials and processes and the development of decision-making tools around life cycle analyses. So that we might finally achieve the two folds aim of leachate management: defence of the environment and resource safety.

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