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Simultaneous removal of organics and ammonium-nitrogen from reverse osmosis concentrate of mature landfill leachate

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A B S T R A C T

Leachate treatment forms one of the main issues in landfill management. The recirculation of reverse osmosis (RO) concentrate of mature leachate, which is characterized by low biodegradability, into the landfill has not proven itself to be a sustainable treatment method. Hence alternate methods for the treatment of the concentrated leachate products have to be devised. In this work, the treatment of a mature leachate and its RO concentrate using different combinations of physico-chemical and biological treatment methods for the removal of organic carbon and ammonium-nitrogen (N-NH_4^+) were investigated. The coupling of electrocoagulation and ozonation with biological treatment was examined in this study. The combination of biological treatment with pretreatment processes like ozonation, electrocoagulation and adsorption proved to be a successful strategy for the removal of refractory organics as well as ammonium-nitrogen (N-NH_4^+). Ozonation led to a total organic carbon (TOC) removal of up to 12.5% whereas with electrocoagulation, up to 40% removal of TOC was observed. In combination with biological treatment, up to 90% TOC and 65% ammonium-nitrogen removals were observed during the experiments, all of which were performed in batch mode.

1. Introduction

Landfilling is the most widely used procedure to dispose of municipal solid waste [1–3]. A colossal 1.3 billion tons of landfill waste is produced worldwide annually, and is projected to increase to 2.2 billion tons by 2025 [4]. Although landfilling has proved to be a competitive waste management alternative over the decades, the production of a huge quantity of leachate is a matter of great concern. Hence, landfill management and leachate treatment form vital areas of environmental research.

Landfill leachate is the wastewater generated in a landfill as a result of percolation of precipitation, water produced from biochemical reactions within the waste mass, and the inherent water content of the waste material [5]. Landfill leachate is an example of a highly loaded wastewater containing a wide variety of pollutants, including an array of organic compounds, ammonium-nitrogen, heavy metals, and inorganic salts [5–7]. The concentrations of most components in landfill leachates exceed the concentrations of the same in sewage sludge [8].

Initially, large amounts of biodegradable organic compounds are present in leachates from young landfills. When anaerobic conditions develop inside the landfill, these compounds undergo anaerobic fermentation leading to the formation of volatile fatty acids (VFAs). As much as 95% of the total organic carbon in the leachate can be made up of VFAs during this phase [9]. As the landfill gets older, the VFAs get converted into acetate and finally into landfill gas. Once the VFA content in the leachate gets exhausted, the remaining organic matter is predominantly refractory. Leachates from mature landfills contain a high quantity of refractory organic compounds, which are not biodegradable under normal conditions, and a high percentage of the total nitrogen exists as ammonium ions [10,11]. Table 1 shows the typical change in leachate quality with the ageing of a landfill and Table 2 shows the values of different parameters of the RO concentrate of landfill leachate used in this study.

Environmental regulations enforce strict discharge limits for various pollutants contained in a wastewater that is to be released into natural waters like lakes, rivers, or seas. These include limits on the chemical oxygen demand (COD), total organic carbon (TOC) and ammonium-nitrogen (N-NH_4^+) concentrations among many others, which are detrimental to the hydrosphere if present in high concentrations. Selection of suitable processes for the treatment of leachate depends on its composition, which is in turn dependent on the age of the landfill.

In order to achieve high treatment efficiency and to minimize the risk of contamination, usually a combination of several physical-chemical and biological methods is used for treating landfill leachates [10,12,14–16]. High pressure membrane processes have to be utilized to complement biological and physical-chemical methods to achieve

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Table 1
Characteristics of different types of landfill leachates [12,13].

Parameter	Type of leachate		
	Young	Intermediate	Old/Mature
Landfill age (years)	< 5	5–10	> 10
pH	< 6.5	6.5–7.5	> 7.5
Ammonia nitrogen (mg/L)	< 400	n.a.	> 400
COD (g/L)	> 10	4–10	< 4
TOC/COD	< 0.3	0.3–0.5	> 0.5
BOD/COD	0.5–1.0	0.1–0.5	< 0.1
Biodegradability	high	medium	low

n.a. − data not available.

Table 2
Values of different parameters of the RO concentrate from the company on-site measurement data.

Parameter	Value (mg/L)
TOC	4060
Total nitrogen (TN)	3120
Ammonium-nitrogen	3000
pH	7.4

completeness of treatment. Reverse osmosis is widely utilized in combination with other treatment processes, and is in some cases employed standalone [17–19]. Although the quality of the produced permeate is very good that it can be discharged in to nature, reverse osmosis generates a highly polluted retentate that has to be further treated [5].

Biological treatment is known to be effective in removing organics and nitrogen from young leachates with a high BOD/COD ratio. Fluidized bed reactor systems, as an example, have been reported to remove up to 82% COD and 60% DOC in young leachates [20,21]. For methanogenic leachate, the effectiveness of the process is lesser as refractory (non-biodegradable) compounds form the bulk of the organic content [5]. Welander et al. [22] reported nearly 90% total nitrogen removal with a COD removal of only 20% from a mature landfill leachate using a suspended carrier biofilm reactor. In such cases, biological processes can be used in combination with other pretreatment or polishing processes.

The use of ozonation and electrocoagulation has been widely reported in literature [11,23–29] for the treatment of inert organics in several wastewaters. Ozonation promotes the formation of microflocs of organic matter, in addition to oxidizing recalcitrant and colour-causing humic substances to biodegradable compounds [30]. For a methanogenic leachate with low biodegradability, the pretreatment with ozonation before biological treatment hence has practical importance. Wang and Gamal El-Din [31] reported up to 100-fold increase in the BOD to COD ratio after ozonation of methanogenic leachate.

Electrocoagulation is usually used as a pre-treatment or a post-treatment polishing process, and not as a main treatment method in case of highly polluted wastewaters. Ilhan et al. [25] reported 59% decrease in COD and 14% ammonium-nitrogen removal from a mature leachate during electrocoagulation using aluminum electrodes. Djelal et al. [11] have reported removal of organics with up to 56% COD reduction using a current density of 95 A/m^2 and a contact time of 150 min. Most studies have reported an acidic operating pH as optimum for electrocoagulation [32].

The investigation of a possible synergy of electrocoagulation and ozonation processes in combination with biological treatment has not yet been investigated for the treatment of mature leachates. This work was aimed to be a preliminary evaluation in that direction. The goal of this study was to determine the effect of (i) pretreatment strategies of ozonation, electrocoagulation and a combination of the two and, (ii) the type of reactor system (attached growth vs. suspended growth); on the removal of TOC and ammonium-nitrogen content during biological treatment of a mature landfill leachate. The study investigated aerobic biological treatment of pretreated landfill leachate and its RO concentrate using an up-flow aerobic packed bed reactor (attached growth) and continuously stirred tank reactor (suspended growth). Ozonation and electrocoagulation were used as pre-treatment processes in the study.

2. Materials and methods

2.1. Materials

The leachate used in this work was from the Ihlenberg landfill, located near the north German city of Luebeck. This landfill has been in operation since 1983 and has 113 ha available for landfilling. With a maximum yearly intake capacity of 1 million tons, this landfill has been described as ‘the biggest of its type in Europe’ and is expected to run for many years to come [33].

The leachate generated at this landfill site, methanogenic in nature, exhibits a TOC value of 800 mg/L and an ammonium-nitrogen value of 600 mg/L. The raw leachate is subjected to an on-site multi-step purification. A two-stage reverse osmosis plant, operated at 60 and 120 bar with a total processing capacity of 40 m^3/h, recovers about 80% permeate (clean water) generating about 100 m^3 of highly polluted concentrate per day. This RO concentrate exhibits an electrical conductivity of about 92 mS/cm and TOC and ammonium-nitrogen concentrations of about 4000 mg/L and 3000 mg/L, respectively. Concentrations of other major solutes in the retentate can be found elsewhere [7]. RO retentate was provided from the landfill site for this study. The activated sludge was obtained from the county wastewater treatment plant at Seevetal (Lower Saxony, Germany) for inoculating the aerobic biological degradation trials.

2.2. Methods

The feed samples used in the experiments were different dilutions of the RO concentrate. A 5-times dilution (RO5x) of the concentrate was used as an approximation of the raw leachate (80% water is recovered from raw leachate), while as experiments were also performed with the concentrate itself (RO) and a 3-times dilution of the same (RO3x). The dilutions were done with deionized water. Dilution was done mainly to investigate possible inhibitory effects of contaminants like chloride as the given leachate had a very high chloride content (30,000 mg/L in the RO concentrate).

2.2.1. Ozonation

Fig. 1 shows a schematic of the ozonation setup used in the study. Oxygen (99.99%, Westfalen AG, Germany) was supplied to an ozone generator (Labor-Ozonisator 301.7, Erwin Sander Elektroapparatebau GmbH, Germany) at 1.2 bar (abs) at a flowrate of 120 L/h, producing 2.125 g O_3 per hour. The generated ozone was fed into a coiled tubular reactor (8 mm ID, 18.6 m long, made of Teflon) in which it came into contact (cocurrent) with the leachate. The leachate was pumped using a Masterflex variable speed peristaltic pump (Cole-Parmer Instrument Company, Illinois, USA) at a flowrate of 350–400 mL per minute. Ozone dosages ranging between 0.2–1.3 g/$L_{leachate}$ were used during the experiments, corresponding to ozonation times of 5–30 min. The ozonated leachate was recirculated into the feed flask. The residual ozone in the leachate was removed by bubbling gaseous nitrogen before it was fed for biological treatment.

2.2.2. Electrocoagulation

Fig. 2 shows a schematic of the electrocoagulation setup. For all experiments, 700 mL of raw leachate or its RO concentrate was taken in a HDPE cuboidal vessel (10.5 cm long, 9 cm wide and 15.5 cm high). Aluminum electrodes (EN AW 5754 $AlMg_3$), 10 × 10 cm (thickness