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Integration of autotrophic nitrogen removal, ozonation and activated carbon filtration for treatment of landfill leachate

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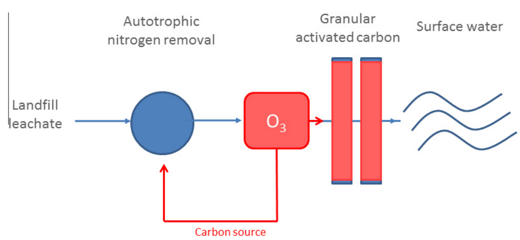
HIGHLIGHTS

- ANR biological system was applied for nitrogen removal from landfill leachate.
- Post-treatment techniques (O_3/GAC and their combinations) were compared.
- Recycling ozonated ANR effluent to the ANR reactor resulted in improved performance.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to determine an appropriate combination for effective and economical COD and nitrogen removal from landfill leachate. Biological (nitrogen) treatment was performed with the autotrophic nitrogen removal (ANR) process. The (post-) treatment performances of (i) ozonation alone, (ii) adsorption to granular activated carbon (GAC) alone, (iii) the combination of O_3 and GAC and (iv) an integrated approach with continuous recirculation of ozonated ANR effluent were investigated. It was observed that ANR post-treatment with ozonation was able to remove only 15.2% of residual COD and 14.0% of total nitrogen in the effluent whereas using activated carbon a 73.6% removal efficiency of COD and 17.3% of total nitrogen was achieved. The best performance was obtained for an ANR post-treatment combination of ozonation and GAC, ensuring a high removal for both COD and total nitrogen. When different ratios of ozonated effluent were mixed with the influent of the ANR, it was found to be possible to reach COD removal as high as 40% in the ANR with a slightly decrease in nitrogen removal of around 70–80% compared to respectively 5.31% (COD) and 85.9% (total nitrogen) when no ozonated leachate was recycled to the ANR reactor. Taking the overall performance and operational expenses into account, a combination of ANR biological treatment and its different (post-) treatments (ANR + ozonation + GAC) results in a good performance and a lower cost compared to traditional treatment.

1. Introduction

Within the last few decades, the continuing expansion of population and industry drove toward the overwhelming solid waste generation [1]. Approximately 95% of this solid waste is landfilled [2], which subsequently leads to the production of leachate. Landfill leachate has been generally known as a high-strength and hazardous wastewater due to its composition characteristics

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(numerous (toxic) pollutants such as ammonium, organic matter, heavy metals, xenobiotic organic substances,...). Its disposal poses an ecological problem and presents a hygienic risk [3–6]. Therefore, the concern about leachate production and environmental protection has made it one of the most challenging topics over the years. Many treatment options (or their integrated technologies) were utilized by landfill operators for economically efficient treatment of landfill leachate [7].

Presently, biological treatments are usually preferred over physical–chemical ones [8] because of its economics, simplicity and efficiency for the removal of ammonium and/or as a first stage of young leachate treatment. However, these techniques can be limited by refractory compounds. In such a situation, one way to overcome such operational problems is to assess the potential of non-biological treatment processes to remove biorecalcitrant material and improve overall leachate treatment. For this purpose, a combination of several treatment methods is usually applied. The most common post-treatments referred to in the literature are reverse osmosis, flocculation–precipitation, evaporation, incineration, activated carbon adsorption and chemical oxidation (such as ozonation) [9]. The latter two are the attractive methods and particularly received an increased interest in recent years.

Granular activated carbon, in combination with biological pretreatment has already been implemented on many landfill leachate sites [6]. In most cases, activated carbon adsorption allows to remove sufficient organic compounds (nonbiodegradable organics and inert COD), color and other toxic substances to acceptable levels for biologically treated landfill leachate. The use of activated carbon is believed to contribute to a synergy effect as it provides an attachment surface for micro-organisms (which contribute to biodegradation) and serves as a nucleus for floc formation [10]. However, high cost caused by high activated carbon consumption during leachate treatment indicates that a pre-treatment would be required to reduce the organic load and/or to enhance the biodegradability in an economical efficient way [6]. Ozone was reported capable of oxidizing the large refractory organic molecules (up to 10^4 g/mol), found in leachates, to smaller more biodegradable molecules even to their highest stable oxidation states (producing water and carbon dioxide) [11]. The generated smaller molecules can be removed more easily in a subsequent process.

The treatment of biologically treated landfill leachate using GAC or ozonation has previously been investigated. In Germany for example systems such as the "Bio–O_3–Bio" has been used for the treatment of leachates [12]. Oloibiri et al. [13] studied the effect of starting pH, initial COD concentration, inlet gas flow rate and ozone generator capacity on ozonation alone of a biologically treated landfill leachate, achieving COD removal efficiencies of 27% and an UV_{254} absorbance decrease of 58%, respectively, when the treatment time was 60 minutes [13]. Derco et al. [14] reported that ozone alone ensured a COD reduction of approximately 60% after about 10 to 11.5 h ozonation time. Morawe et al. [15] showed activated carbon could reduce the concentration of the non-biodegradable organic and chlorinated organic compounds, as well as the color to an acceptable level). Several sites use a combination of biological treatment and activated carbon adsorption [7]. Only limited research was performed however on the possible synergistic coupling of an oxidative process with GAC treatment for treatment of biologically landfill leachate [6,13]. With such combinations both COD as well as nitrogen removal can be achieved.

In this study, biological treatment is performed by autotrophic nitrogen removal (ANR). ANR combines the partial nitrification process (conversion of ammonium to nitrite) with the Anammox process which converts ammonium and nitrite on an almost equimolar basis to nitrogen gas [16,17]. This biological treatment process has future prospects of wide application as the cost of nitrogen removal can be significantly decreased [16]. However, for landfill leachate treatment, complete removal of nitrogen compounds was not possible [16]. Therefore, in the study, several post-treatment methods were evaluated in terms of reduced organic load, decreased amount of nitrogen components and increased biodegradability. The following techniques were applied in the study: ozonation, adsorption to granular activated carbon (GAC), their combination (O_3 prior to GAC) and recirculating ozonated effluent into the ANR system.

2. Materials and methods

2.1. Characterisation of landfill leachate

Raw leachate was sampled from the landfill site of IMOG in Moen, Belgium (www.imog.be). The on-site treatment facility and sampling procedures are described by Chys et al. [6] and Gao et al. [16]. In Table 1, typical pH and pollutant concentrations in the raw leachate used as ANR influent are given. It is a slightly alkaline mixture with a dark brown color. The composition, during the last two years of lab-scale ANR operation, can be characterized with total influent COD concentration between 548.5 and 1826 mg L^{-1}, which, after ANR treatment, was reduced to 250.5–869.1 mg L^{-1}. As such, approx. 40% COD removal was achieved. The total nitrogen concentration, was decreased from 244–819.3 mg L^{-1} to 59.7–406.4 mg L^{-1} resulting in up to 75% nitrogen removal. Other characteristics of the landfill leachate are also given in Table 1.

Tests with post-treatment techniques (ozonisation and GAC filtration) were performed with manually sampled ANR effluent which was stored at 4 °C until use without any pH correction.

2.2. Investigated treatment methods

2.2.1. ANR Biological treatment-recycling of ozonated ANR effluent to the ANR reactor

The ANR biological treatment with the ANR reactor is similar to Gao et al. [16] and its detail description can also be found in Gao et al. [16]. The difference is that in this study, the influent was a mixture of ozonated ANR effluent and raw landfill leachate. The amount of ozonated effluent which was mixed with the raw influent in different proportions, gradually increased from 0 over 1/10 volume ratio up to 1/4 and 1/2 volume ratio during the course of the experiment. The effect of ozonation on ANR effluent and effluent recirculation was studied.

2.2.2. Ozonation of ANR treated leachate

The ozonation batch apparatus consisted of a cylindrical vessel with a working volume of 10 L [18]. Ozone was generated from pure oxygen by a laboratory ozone generator (Ozomat COM-AD-02, Anseros). 10 L of leachate after biological pretreatment was transferred into the reactor and ozonated by continuously bubbling the ozone-oxygen gas mixture through a porous gas

Table 1
Physical–chemical analysis of the used landfill leachate.

Parameter	Range	
	Raw leachate (influent of ANR)	Biologically treated leachate (effluent of ANR)
COD (mg/L)	548.5–1826	427–869
BOD_5 (mg/L)	94.3–281.9	1.15–9.95
N-NH_4^+ (mg/L)	244–627	0.08–186
N-NO_2^- (mg/L)	0–65.6	0.21–145
N-NO_3^- (mg/L)	3.5–17.2	30.9–110
Ec, (μS/cm^2)	5590–9360	11 ± 0.2
pH	7.8–8.2	7.02–9.21