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Digestate application in landfill bioreactors to remove nitrogen of old landfill leachate

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ABSTRACT

Anaerobic digestion of organics is one of the most used solution to gain renewable energy from waste and the final product, the digestate, still rich in putrescible components and nutrients, is mainly considered for reutilization (in land use) as a bio-fertilizer or a compost after its treatment. Alternative approaches are recommended in situations where conventional digestate management practices are not suitable. Aim of this study was to develop an alternative option to use digestate to enhance nitrified leachate treatment through a digestate layer in a landfill bioreactor. Two identical landfill columns (Ra and Rb) filled with the same solid digestate were set and nitrified leachate was used as influent. Ra ceased after 75 day's operation to get solid samples and calculate the C/N mass balance while Rb was operated for 132 days. Every two or three days, effluent from the columns were discarded and the columns were refilled with nitrified leachate (average N-NO_3^-concentration = 1,438 mg-N/L). N-NO_3^- removal efficiency of 94.7% and N-NO_3^- removal capacity of 19.2 mg N-NO_3^-/gTS-digestate were achieved after 75 days operation in Ra. Prolonging the operation to 132 days in Rb, N-NO_3^- removal efficiency and N-NO_3^- removal capacity were 72.5% and 33.1 mg N-NO_3^-/gTS-digestate, respectively. The experimental analysis of the process suggested that 85.4% of nitrate removal could be attributed to denitrification while the contribution percentage of adsorption was 14.6%. These results suggest that those solid digestates not for agricultural or land use, could be used in landfill bioreactors to remove the nitrogen from old landfill leachate.

1. Introduction

Solid digestate from the anaerobic digestion of organic waste is a semi-stable material that is rich in organic matter and minerals. With or without further treatment, this type of solid digestate is often used as a bio-fertilizer or soil conditioner applied to agricultural fields. In Italy, the digestates from agricultural residues (manure and energy crops) are almost exclusively spread to the land while digestates from organic fraction of municipal solid waste (OFMSW) is classified as wastes, which cannot be directly used in agriculture but need to be further treated. Besides, high transportation costs (Delzeit and Kellner, 2013), the potential for eutrophication (Lukehurst et al., 2010), low agriculture demand during some seasons (Wellinger et al., 2013) and strict regulations in nitrate sensitive zone (Neumann et al., 2016) could also restrict land use of digestate.

When digestate is not qualified for land use or land use is not possible, alternative options for digestate disposal could be based on the concept of Back to Earth Alternatives (BEA), which considers a landfill as a necessary sink to close the material loop (Cossu, 2016; Peng and Pivato, 2017). When solid digestate is applied to a landfill site, it might be possible to combine solid digestate disposal with leachate treatment.

It is generally known that leachate from old landfill sites is usually rich in ammonia but its low content of biodegradable organics make denitrification the rate limiting step for a good nitrogen removal (Kozub and Liehr, 1999). Leachate treatment options to remove nitrogen include conventional denitrification through wastewater treatment technologies (Renou et al., 2008), phytotreatment (Garbo et al., 2017; Lavagnolo et al., 2016) and on-site denitrification by recirculation of nitrified leachate (Berge et al., 2006; Bolyard and Reinhart, 2016; Shao et al., 2008; Shouliang et al., 2008). Either strategy for nitrogen removal includes nitrification and denitrification.

A functional layer of digestate in the temporary top cover of a landfill might facilitate nitrate biological conversion and physical/-chemical removal, when nitrified leachate is recirculated on the top. Several studies tested nitrified leachate denitrification in landfill reactors filled with old waste because of its low cost (Jokela et al., 2002; Zhong et al., 2009; Sun et al., 2014). However, limited organic carbon in old waste could decrease the nitrate reduction

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rate (Wu et al., 2009). On one hand, digestate rich in organic matter could offer a partial external carbon source for nitrate denitrification as digestate contains an amount of total organic carbon (TOC) in the range of 27.5–45.9%-total solid (TS) (Michele et al., 2015; Quina et al., 2015; Zhang et al., 2012). Unlike the high nitrogen content in digestate from agricultural biogas production using manure and energy crops, digestate from OFMSW with less nitrogen content is preferred to be used as carbon source because less ammonia will be transferred into the leachate with leaching. In addition to the heterotrophic denitrification potential by using digestate as an external carbon source, nitrogen in both leachate and digestate might also be removed through autotrophic metabolic pathways (Valencia et al., 2011; Xie et al., 2013). On the other hand, solid digestate might have a nitrate adsorption capacity, as it happened with the municipal solid waste (MSW) in a landfill bioreactor experiment (Fu et al., 2009). Activated carbon, sepiolite, surfactant-modified zeolite and zeolite clinoptilolite has been used as absorbents for nitrate removal (Malekian et al., 2011; Öztürk and Bektaş, 2004), however they might not be suitable in case of leachate because of the high cost (Della Rocca et al., 2007).

In this study nitrate removal from nitrified leachate was investigated, using a digestate layer in two reactor columns. The nitrate removal pathway and the effect of solid digestate on nitrate adsorption were evaluated.

2. Materials and methods

2.1. Solid digestate and leachate

The solid digestate was obtained from a full scale thermophilic two-stage wet anaerobic digestion facility (Camposampiero, Padua, Italy) for the treatment of sewage sludge and source-segregated biodegradable waste. This plant has a total reactor volume of 3300 m^3 and operates at a hydraulic retention time of 22 days. After the end of anaerobic digestion, the digestate was separated by centrifugation. The solid digestate was composted to stabilize the material prior to land use whereas the liquid was treated internally at the wastewater treatment plant. The average characteristics of the solid digestate samples were: Total Solids (TS) = 25.6%, Volatile Solids (VS) = 64.1% TS, TOC = 314g-C/kg-TS, Total Kjeldahl Nitrogen (TKN) = 48.5g-N/kg-TS.

Raw leachate was collected from a MSW landfill located in Northern Italy where untreated MSW were disposed of during the 1980s. The physical-chemical characteristics of the raw leachate and nitrified leachate are reported in Table 1. The raw leachate was nitrified in a nitrification tank described below.

2.2. Experimental setup

The experimental setup includes two parts: leachate nitrification reactors (Fig. S1a) and landfill simulation columns (Fig. S1b). The experiment schematic is shown in Fig. 1. For complete nitrification, the dissolved oxygen in the aeration tank was kept around 4 mg O_2/L. For every liter of raw leachate, 2 g of sodium bicarbonate was added to provide alkalinity and to buffer the pH. Temperature for nitrification was maintained at 25 ± 2 °C. The landfill simulation reactors were operated using two identical reactors (Ra and Rb) by using polymethyl methacrylate columns (height 100 cm and inner diameter 10 cm). A gravel layer (10-cm thickness) was placed at the bottom and at the top of each column for leachate drainage. 3.0 kg of raw solid digestate was added to each reactor, resulting an average density of 764 kg/m^3. Before the start-up of the two columns, pure nitrogen gas was used to flush the columns to maintain anaerobic conditions. Every two or three days, 0.50 L or 0.75 L of nitrified leachate (equivalent to 0.76 g-N/kg-VS day) produced from the leachate nitrification reactor were added to the landfill bioreactors and the effluent collected from columns was sampled and discharged at the same time. Every day effluent from the landfill bioreactors were recirculated to the digestate body three times. The nitrified leachate volume was set according to Xie et al., 2013 and based on preliminary experiments, which indicate that a loading rate around 0.73 g-N/kg-VS/day was necessary to maintain a stable nitrate removal.

Landfill reactor Ra's operation ceased after 25 feeding cycles (approximately 75 days) to get solid samples and calculate the C/N mass balance while the operation of reactor Rb was extended until 48 feeding cycles (132 days). Each feeding cycle lasted 2–3 days. At the end of the experiments, the total liquid-to-solid ratios (L/S) were equivalent to 17 ml/g and to 32 ml/g, respectively, for Ra and Rb.

Nitrate removal efficiency was calculated by Eq. (1) and was used to evaluate the denitrification performance of the landfill columns:

$$\text{Nitrate removal efficiency} = \sum_{i=1}^{n} \frac{C_{inf,i}V_{inf,i} - C_{eff,i}V_{eff,i}}{C_{inf,i}V_{inf,i}} \times 100\% \quad (1)$$

where:

$C_{inf,i}$ and $C_{eff,i}$ = influent and effluent concentrations of nitrate in the landfill columns at feeding cycle i;
$V_{inf,\,i}$ and $V_{eff,\,i}$ = influent and effluent volumes at i feeding cycle;
n = total feeding cycles; n equal to 25 and 48, for Ra and Rb, respectively.

Table 1
Raw and nitrified leachate characteristics.

Parameters	Raw Leachate		Nitrified leachate[a]	
	Average	Range	Average	Range
pH	8.95	8.85–9.05	7.08	5.69–8.30
TOC (mg-C/L)	264	252–276	415	273–565
TC (mg-C/L)	1600	1580–1620	553	366–725
BOD (mg-O_2/L)	60	58–62	30	15–45
COD (mg-O_2/L)	1014	987–1041	952	814–1090
TKN (mg-N/L)	1723	1698–1748	90	35–236
N-NH_4^+ (mg-N/L)	1703	1665–1741	71	7–208
N-NO_2^- (mg-N/L)	1.9	1.6–2.2	1.8	0.0–62.3
N-NO_3^- (mg-N/L)	0	0	1438	605–1791
SO_4^{2-} (mg-N/L)	206	201–211	188	184–192

[a] Unlike the stable characteristics of raw leachate, the characteristics of nitrified leachate depend on the performance of the nitrification reactor, which slightly changed over time.