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Denitrification of leachate using composted domestic waste at different levels of stability: A batch test investigation

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ABSTRACT

This study presents an investigation into the viability of pre-treated general wastes at different degrees of stability as carbon sources for in-situ bio-denitrification at landfills. Domestic waste was composted and stabilised for eight (8) and sixteen (16) weeks within two different mini-landfill cells located at the Bisasar Road landfill, Durban, South Africa. Eight substrate categories were developed using the composted domestic wastes and commercial garden refuse with stand-alone and combinatory approaches. Adopting small-scale dynamic batch tests, landfill leachate, treated with 500 mg/l nitrate concentration level, was used to comparatively assess the denitrification efficiency of the stand-alone and augmented substrates. Results demonstrate that substrates based on domestic waste were unable to independently sustain the denitrification process. Full denitrification was only achieved on the augmented substrates. Kinetic analyses show that a zero-order reaction better describes the denitrification rate independent of the nitrate concentration. These results thus elucidate the benefits inherent in adopting augmented substrates in treating landfill leachate.

Introduction

Developing countries, including South Africa, still send 91% of their waste to landfill sites [2, 5]. Generally, a landfill site generates two main waste streams in form of leachate and biogas [1]. Landfill leachate typically contains high levels of nitrate that exceeds the recommended discharge limits, posing a threat to human health and the environment [19]. For example, an increase in nutrient levels results in a very high risk of eutrophication within water systems [22]. Thus, denitrification of treated leachate is crucial prior to its release into the environment. The denitrification process is defined as the reduction of nitrates (NO_3^-) to dinitrogen gas (N_2), via the intermediates nitrite (NO_2^-), nitric oxide (NO) and nitrous oxide (N_2O) [17].

Several methods are being used for treating landfill leachate. Conventional treatments of landfill leachate include combined treatment with domestic sewage and ex-situ aerobic biological treatment in sequencing batch reactors (SBR). SBRs

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provide increased process flexibility, which is ideal for leachate that exhibits a constant flux in concentration and composition [15]. Aerobic nitrification, however, results in high concentrations of nitrates in the treated effluent, which far exceed limits specified by the law, including the South African legislation. Treated effluent from an SBR at Mariannhill landfill site, Durban, South Africa, typically contains 500 to 2000 mgL $NO_3{}^{-1}$ while the limit set by South African legislation is 66.4 mgL $NO_3{}^{-1}$ [5].

Various physicochemical and biological methods have been explored to denitrify leachate [9]. Physicochemical methods, including chemical denitrification, catalytic denitrification, ion exchange and reverse osmosis have been shown to have limited usefulness in terms of success rates and costs [9]. However, biological denitrification has been proven to be the most reliable and most economically feasible option [8].

A supplemental carbon source supports the denitrification process in ex-situ bioreactors [18]. Many carbon sources are readily biodegradable and widely used in the ex-situ treatment of nitrified leachate. These sources include methanol, ethanol, sucrose, acetic acid and molasses [17]. The high water solubility of these carbon products ensures that they are easily available to microorganisms, thus enabling them to readily fuel microbial metabolism [18]. The denitrification rate is primarily dependant on external carbon sources and the COD/NO_3-N ratio [4]. Studies have shown that high C/N ratios can affect the rate of denitrification due to the dissimilatory reduction of ammonium. On the other hand, a low C/N ratio could lead to nitrate accumulation [12]. Moreover, the high costs of carbon-based sources have given room for consideration of low-cost composted domestic waste as a promising alternative [20], thereby developing an economical and environmentally sustainable process that cannot be overlooked.

Recirculation of treated leachate into the landfill enables the use of existing organic waste as a viable carbon source for denitrification. The idea of leachate recirculation was proposed years ago to enhance waste decomposition [11]. By increasing the moisture content of the refuse, leachate recirculation enables stimulation of microbial activity by improving contact between the microorganisms, soluble nutrients and organic fractions for bacterial growth [14]. A further increase in microbial activity within the wastes engenders several other advantages, including an increase in biogas emissions as well as an opportunity for in-situ denitrification of the leachate [11].

As part of South Africa's first pilot project on mechanical-biological pre-treatment of waste prior to disposal, in 2005, five semi-full-scale shallow landfills (cells) were established at the Bisasar Road landfill site in Durban. Of the five cells, two (40 m^3 in volume) contain finely sorted organic fractions only, with wastes treated at different degrees of stability (i.e. composted for 8 and 16 weeks). One of the remaining three cells was filled with unsorted and untreated general municipal solid waste and served as control. The cells were designed to simulate shallow landfills (only 1,5 m deep) and were equipped with leachate and biogas extraction systems. They provide an ideal incubator for testing in-situ bio-denitrification using waste at different degrees of pre-treatment as a carbon source, and at a larger scale than used in other studies.

This study was conducted to understand the potential that exists in using municipal solid waste to treat landfill leachate. Therefore, the study aimed to investigate the efficacy of domestic waste and commercial garden refuse as potential carbon sources to fuel biological denitrification, using the Bisasar landfill, Durban, South Africa – an integrated waste management system as a case study.

Materials and methods

Materials

Nitrate solution (leachate)

The study adopts treated leachate from a holding tank at the Mariannhill landfill site, Durban, South Africa. The treated leachate from the SBR was diluted with distilled water to obtain a concentration of 500 mg/L of NO_3. The initial characteristics of the leachate, upon testing, are presented in Table 3.

Organic wastes

The composted domestic waste used for this research was sourced from test cells located at Bisasar Road landfill site, Durban, South Africa. The domestic wastes were composted and split into two categories, and thereafter stabilized for 8 and 16 weeks, respectively. Following this process, the two categories of composted waste were mechanically sieved through a 50 mm sieve resulting in 'fines' made up of organic fractions of the waste. The fines were then deposited into the cells. The domestic waste, which was composted and stabilised for 8 and 16 weeks, were deposited into two cells, namely Cell 1 and Cell 2, respectively.

Additionally, some fresh commercial garden refuse (CGR_{raw}) and composted commercial garden refuse (CGR_{10}) were sampled from the Mariannhill landfill site. The garden refuse was separated from the main waste stream and is a readily available, low-cost, carbon source. The CGR_{10} consists of CGR_{raw} composted for approximately 10 weeks.

Methods

Characterisation tests

Characterisation tests were conducted on all the substrates and their eluates including the treated leachate using the conventional testing methods of the American Society for Testing and Materials [6]. The analyses done on the solid substrates

are as follows: moisture content (MC), total solids (TS), volatile solids (VS), respiration index (RI_7), which were determined using respirometric system type OxiTop®, total nitrogen (Tot N), total carbon (Tot C) and carbon-to-nitrogen ratio (C/N).

The eluates were tested to obtain pH, TS, VS, chemical oxygen demand (COD), biochemical oxygen demand (BOD_5), ammonia (NH_3), Tot C and Tot N. A 10:1 liquid-to-solid ratio was used in preparing the eluate.

The treated leachate was tested to establish pH, TS, VS, COD, BOD_5, NO_x, NH_3, Tot C and Tot N. All the characterisation tests were conducted in triplicate for accuracy.

Denitrification tests and batch test setup

Batch tests were conducted to assess the suitability of each substrate as a carbon source for denitrification. The denitrification of the treated leachate, diluted to 500 mg/L, was conducted using three replicates (R1, R2 and R3). All the batch tests were conducted using closed-top batch reactors of 1 L, equipped with two airtight silicone septa, which enables the sampling from the vessel without allowing ingress of air. Each substrate (S) was mixed with the treated leachate (L) solution at a ratio of 10:1 (L/S) by weight to ensure full saturation throughout the experiment and an optimal controlled temperature of 25 °C. A control test with distilled water was also carried out for each batch test.

A batch test was conducted on the mixture of domestic waste (composted in Cell 1 and Cell 2) and treated leachate (hereafter referred to as "Cell 1″ and "Cell 2″), and the durations and nitrate removal rates observed (Fig. 1(a)). For comparison purposes, Cell 1 and Cell 2 were mixed with fresh CGR_{raw} and CGR_{10} (Fig. 1(a) and (b)), using a ratio of 1:1. This was done to observe if the mixing of Cell 1 with other substrates would increase the kinetics efficiency of the batch tests. The resulting substrates are henceforth referred to as "Cell 1 + CGR_{raw}", "Cell 1 + CGR_{10}", "Cell 2 + CGR_{raw}" and "Cell 2 + CGR_{10}", respectively. In addition, CGR_{raw} and CGR_{10} were separately subjected to a batch test. In all, eight categories of waste comprising domestic and garden wastes were investigated.

Considering that an anaerobic condition is required for the denitrification process to take place, nitrogen gas was used to remove any air inside the batch reactor bottle. After thoroughly flushing the bottle with nitrogen gas, the bottles were sealed with the silicon seal. The sample was thereafter placed on a shaker that was operated at a speed of 150 rpm to ensure a continuous and full contact of the solid with the liquid.

Small samples of approximately 1 to 5 mL were extracted using a gas-tight syringe every hour for the first day and once a day thereafter. The samples were analysed for nitrate and nitrite concentration using a nitrate test stick type Merckoquant (MERCK) by the colorimetric method [10]. At the end of the batch test, the eluates were analysed for pH, nitrates, nitrites, ammonia and COD. The solid matter was also analysed for Tot C and Tot N as well C/N ratio.

Kinetic tests

The reduction in the nitrate concentration was measured and the reaction rate was determined. The zero- and first-order kinetic reaction model was used in modelling the results.

The zero-order equation is expressed generally as:

$$C = C_0 - k_0 t$$

The first-order equation is expressed generally as:

$$C = C_0 e^{-kt}$$

where:

k = zero-order rate constant (mg/min)
C_0 = nitrate concentration at the time *0*
C = nitrate concentration at time t

Results and discussion

Characterisation of substrates and leachate

Tables 1, 2 and 3 present characterisation results of the solid matter, their eluates, and the treated leachate, respectively. As observed in Table 1, Cell 1 displayed a RI_7 value of 2.60 which is higher than that of Cell 2 (1.77). The lower RI_7 value attained for Cell 2 can be attributed to lower carbon content and a higher degree of stability when compared to Cell 1. Table 2 shows that pH values in all the substrates fall between 5.97 and 7.47. The pH values can be considered as favourable for denitrification as the optimum pH typically required for biological denitrification range between 6 and 8 [13]. A low pH could affect the rate of nitrate removal negatively. The substrates analysed in this study produced a favourable pH for denitrification compared to results obtained by Plüg et al. [13] wherein a low pH of 5.45 was reported for the CGR_{raw} and CGR_{10} substrates investigated. Table 1 also shows that Cell 1 + CGR_{raw} had a lower RI_7 value of 2.60 mg O_2/g DM when compared to a RI_7 value of 7.77 mg O_2/g DM reported for CGR_{raw} in Plüg et al. [13]'s study.

Results presented in Table 2 also show a high level of NH_3_N in the eluate of the Cell 2 substrates. This could be attributed to leaching of the substrate into the system which, in turn, could be detrimental to the denitrifying capacity of the reactor for two reasons. First, ammonia is known to be toxic to many organisms and could, therefore, in a high enough concentration, negatively affect the population density of denitrifying bacteria in the system. Second, micro-pockets of oxygen

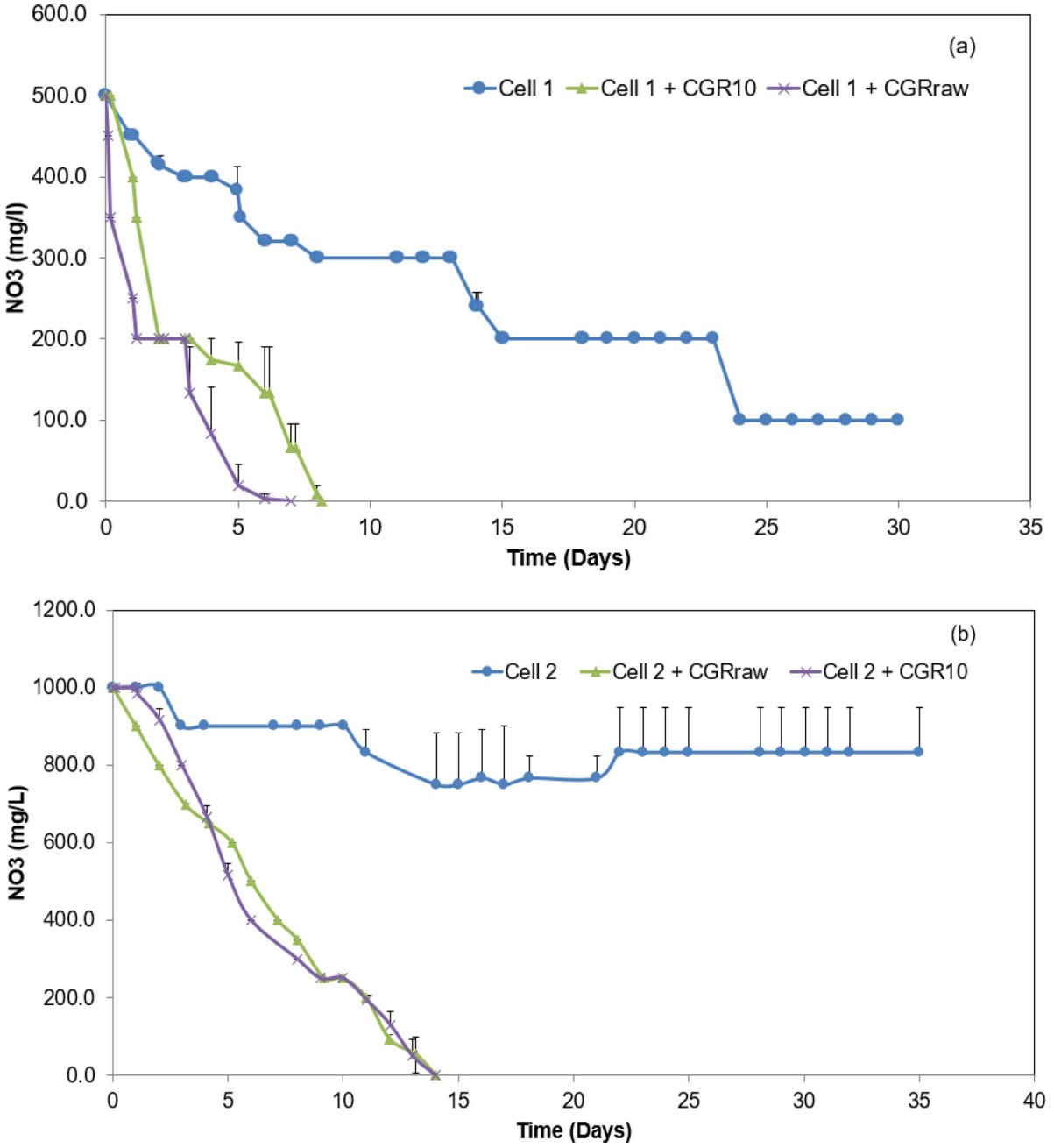

Fig. 1. Evolution of the nitrate concentration for all substrates investigated at 500 mg/L – (a) Cell 1 substrate, (b) Cell 2 substrate.

Table 1
Characterisation of the solid substrate.

Substrate	MC (%)	TS (%)	VS (%)	RI_7 (mg O_2 /g DM)	Tot C (%)	Tot N (%)	C:N Ratio
Cell 1	25.26	74.74	18.03	2.60	8.84	0.49	18.04
Cell 2	27.18	72.82	15.38	1.77	7.16	0.47	15.23
Cell 1 + CGR_{raw}	34.71	76.47	59.50	19.33	19.84	0.86	23.07
Cell 2 + CGR_{raw}	31.11	68.89	56.85	18.25	11.87	0.77	15.42
Cell 1 + CGR_{10}	23.53	65.29	45.12	15.21	16.55	1.10	15.05
Cell 2 + CGR_{10}	28.72	71.28	39.15	11.14	9.00	0.73	12.33
CGR_{raw}	41.31	58.69	95.25	103.13	44.00	1.98	22.22
CGR_{10}	27.96	72.04	83.03	52.20	25.80	1.54	16.75

Table 2
Results of the eluate tests.

Substrate	TS (%)	VS (%)	pH	COD (mgL^{-1})	BOD ($mg\ L^{-1}$)	NH_3- ($mg\ L^{-1}$)	NO_x ($mg\ L^{-1}$)	Tot C (%)	Tot N (%)	C:N Ratio
Cell 1	11.69	2.84	7.20	1437	39	2.10	0.35	1.69	0.07	24.14
Cell 2	10.60	4.77	7.23	3328	22	11.56	12.60	0.71	0.03	23.67
Cell 1 + CGR_{raw}	8.14	4.99	7.03	3298	137	0.70	0.70	1.69	0.04	42.25
Cell 2 + CGR_{raw}	8.30	3.61	7.03	3249	109	11.20	12.60	0.83	0.04	20.75
Cell 1 + CGR_{10}	15.52	8.47	7.47	4323	148	1.40	1.12	0.82	0.04	20.50
Cell 2 + CGR_{10}	11.48	4.98	7.18	4136	128	16.80	15.40	0.62	0.03	20.67
CGR_{raw}	4.48	3.11	5.97	3471	1720	<1	<1	0.58	0.02	29.00
CGR_{10}	6.92	4.24	7.34	3876	370	14	6.10	0.63	0.05	12.60

Table 3
Characterisation of the input leachate.

Substrate	NO_3 (mg/l)	TS (%)	VS (%)	pH	COD ($mgL^{-1)}$	BOD ($mg\ L^{-1}$)	NH_3 ($mg\ L^{-1}$)	NO_x ($mg\ L^{-1}$)	Tot C (%)	Tot N (%)	C:N Ratio
Leachate	500	1.55	0.25	7.40	203	40.9	7.70	27.30	0.54	0.05	10.8

that may be present in either the solution or the pores of the substrate could result in the conversion of NH_3 to NO_3, thus increasing the concentration of nitrates within the system, thereby limiting its potential to remove nitrates.

The decomposition rate is affected by the C/N ratio which is dependant on the carbon presence. This is because the organisms responsible for decomposition uses carbon as their source of energy. A correct proportion of carbon is required for energy and nitrogen for the production of protein [13]. This protein is essential as food for denitrification agents. Cell 1 + CGR_{raw} was found to have the highest C/N ratio (Table 2) which makes it the most suitable carbon source when compared to the other substrates. Tot C and Tot N as well as C/N ratio within a sample are of extreme importance in terms of substrate denitrifying potential. It is well documented that a high C/N ratio is required for high rates of decomposition to occur as bacteria utilise carbon as an energy source to fuel metabolism [7, 21]. In this way, under anaerobic conditions, substrates containing higher amounts of available carbon to bacteria will result in increased bacterial metabolism, thus increasing the rate of denitrification achieved.

Batch test: nitrate removal

Different substrates based on Cell 1 and Cell 2 were used during the investigation with an initial nitrate concentration of 500 mg/L in all batches. Fig. 1(a) and (b) shows a summary of the evolution of nitrate concentration during the Cell 1 and Cell 2 batches, respectively.

Fig. 1 shows that Cell 1 batch tests exhibited a rapid initial nitrate removal. Samples of approximately 1 to 5 mL, extracted every hour on the first day and once a day thereafter, showed that nitrate concentration did not drop within the first day. The acclimatisation phase was not evident, and this could be attributed to the presence of readily biodegradable carbon. However, for Cell 2 batches, a plateau phase that signifies the acclimatization phase could be noted for three (3) days upon initialisation of the batch test. As reported by Trois et al. [17], this phase involves buffering the pH level and the competition between nitrifiers and denitrifiers, including microbial competition. This phase occurs until the environment within the batch becomes more stable for denitrification to take place.

Fig. 1(a) and (b) show that augmenting Cell 1 and Cell 2 substrates with commercial garden refuse (CGR_{raw} and CGR_{10}) decreases the time of the acclimatisation phase. Notably, the addition of CGR increased the denitrification process substantially in both cells. Furthermore, it can be observed that full denitrification was achieved for tests done on the augmented substrates (i.e. Cell 1 + CGR_{raw}, Cell 1 + CGR_{10}, Cell 2 + CGR_{raw} and Cell 2 + CGR_{10}). Fig. 1(b) shows that the Cell 2-based substrates released a large initial amount of nitrate concentration of 500 mg/L into the eluate, thereby increasing the initial input concentration from 500 mg/L to 1000 mg/L. Results from this study depict that full denitrification was achieved in Cell 1 + CGR_{10} and Cell 2 + CGR_{raw} between 7 and 8 days. The period required to reach full denitrification is comparable to the period reported by Plüg et al. [13] wherein full denitrification was achieved after 7 days on CGR_{10}.

Batch test: output characterisation

Characterisation tests were conducted on the endpoint (T_{end}) of batch tests relating to the solid and eluates, irrespective of the nitrate removal efficiency (Table 4). These tests were intended to provide a clear insight into the release and presence of carbon, whereas the ammonia test gives insight into nitrogen leaching.

From Table 4, the pH levels in all the batches range between 7.15 and 7.86 which is within the optimal range for denitrification (6.0 – 8.0) as advised by Buckley and Naidoo [3]. The increase in pH from the initial eluate pH is expected as it indicates the occurrence of denitrification and the release of hydroxyl ions (OH^-) during the denitrification process [18].

Table 4
Variation of COD, pH, NH_3-N and Tot C of the eluates.

Substrate	NO_3 (mg/L)	COD (mg/L) Initial	Final	pH Initial	Final	NH_3-N(mgN/L) Initial	Final
Cell 1	0	1437.26 ± 73.69	–	7.20	–	2.10	–
	500	–	18,164.72 ± 168.51	–	7.86	–	22.00 ± 2.65
Cell 2	0	3328.45 ± 186.93		7.23	–	11.55	–
	500	–	7251.45 ± 83.03	–	7.15	–	28.00 ± 3.05
Cell 1 + CGR_{raw}	0	3298.60 ± 818.52	–	7.03	–	0.70	–
	500	–	5408.50 ± 12.50	–	7.43	–	12.67 ± 2.08
Cell 2 + CGR_{raw}	0	3249.98 ± 244.51	–	7.03	–	11.20	–
	500	–	9455.42 ± 164.44	–	7.37	–	8.40 ± 1.40
Cell 1 + CGR_{10}	0	4323.94 ± 815.28	–	7.47	–	1.40	–
	500	–	5649.18 ± 50.76	–	7.84	–	32.43 ± 4.27
Cell 2 + CGR_{10}	0	4136.24 ± 487.48	–	7.18	–	16.80	–
	500	–	8252.00 ± 37.34	–	7.70	–	41.77 ± 5.82
CGR_{raw}	0	3471	–	5.97	–	<1	–
	500	–	4839	–	5.74	–	35.00
CGR_{10}	0	3876	–	7.34	–	14.00	–
	500	–	15,301	–	7.20	–	20.00

Table 5
Zero- and first-order kinetic parameters indicating nitrate removal rate.

Substrate	Nitrate Concentration (mg/L)	Nitrate Removal Time (Days)		Removal Percentage (%)	Zero-order		First-order	
		100%			k (mg/L day)	R^2	k (1/day)	R^2
Cell 1	500	–	30	80	12.25	0.94	5.300E-02	0.92
Cell 2	500	–	35	6.67	4.14	0.39	5.000E-03	0.36
Cell 1 + CGR_{raw}	500	7.0	–	100	63.82	0.84	–	–
Cell 2 + CGR_{raw}	500	14.0	–	100	70.27	0.99	–	–
Cell 1 + CGR_{10}	500	8.2	–	100	51.34	0.86	–	–
Cell 2 + CGR_{10}	500	14.0	–	100	76.26	0.97	–	–
CGR_{raw}	500	3.08	–	100	176.03	0.79	–	–
CGR_{10}	500	3.84	–	100	156.31	0.84	–	–

Results presented in Table 4 show an increase in the COD level and a decrease in Tot C. The bioleaching of carbon could cause an increase in the COD from the solid to the eluate. An external carbon source (from the substrate) is required to fuel bacterial metabolism, thus driving the denitrification process [17]. This is evident by the initial Tot N and Tot C decrease at the end of the batch test for Cell 1.

Fig. 1 and Table 5 thus indicate that batch tests conducted on the non-augmented substrates (Cell 1 and Cell 2) could not achieve full denitrification.

Kinetic tests

Kinetic studies were conducted to describe the denitrification process, and this was done by applying two different kinetic models. The kinetic constants and R^2 values obtained are illustrated in Table 5. Both the zero- and first-order models were implemented. However, the zero-order reaction provides a better description of the process, producing a higher R^2 value. The kinetic studies indicate that the Cell 2 substrate performed poorly compared to Cell 1. It can be observed that the augmentation of Cell 1 and Cell 2 with CGR_{raw} and CGR_{10} increased the process performance and efficiency, resulting in full denitrification. The zero- and first-order rates in this study were in the range of those observed by other authors [13, 16].

Conclusions

This study has investigated the efficiency of pre-treated general wastes as carbon sources for the in-situ bio-denitrification in landfills. Two categories of waste were analysed, comprising (i) domestic waste, combined with treated leachate and composted at 8 and 16 weeks, and (ii) fresh and composted commercial garden wastes. Stand-alone and combinatory investigative approaches were adopted, resulting in eight categories of substrates. The combinatory approach entailed the use of the commercial garden wastes to augment the pre-treated domestic wastes. Using different degrees of stability in small-scale dynamic batch tests, the viability of the stand-alone and augmented substrates in sustaining denitrification of landfill leachate was examined comparatively. Results show that nitrate removal was evident in all the substrates that were investigated. Although a nitrate removal efficiency of 80% was observed in substrates that fall into the category

of non-augmented domestic wastes (implying a degree of efficacy as a carbon source for denitrification), it is important to note that full denitrification was not achieved due to a higher C/N ratio.

On the other hand, substrates formulated from the augmentation of domestic wastes with commercial garden refuse showed a significant improvement in nitrate removal efficiency resulting in full (100%) denitrification. This study, therefore, suggests that the augmentation of domestic waste with commercial garden refuse offers excellent potential in treating landfill leachate and as a concrete step towards attaining the goal of an environment-friendly society. Future studies could assess the evolution of different nitrogen forms, such as changes in Tot N and NO_2- concentrations during the denitrification process, to better comprehend the denitrification pathway.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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