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Valorizing Landfill Gas Condensate as an External Carbon Source for Denitrification in Sewage–Landfill Leachate Co-Treatment

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ABSTRACT

Landfill gas condensate (LFGC) is an underused byproduct that could reduce the cost and environmental impact of purchased external carbon for biological nitrogen removal. This study tested LFGC as a denitrification carbon source in sequencing batch reactors (SBRs) co-treating municipal sewage and landfill leachate. Four lab-scale SBRs operated for 170 days with 98.5% sewage and 1.5% landfill leachate, supplemented with methanol, glycerin, sodium acetate, or LFGC. All reactors performed stably, achieving total nitrogen removal efficiencies of 93.1%–93.8% and effluent nitrate below 1 mg N/L. The LFGC reactor reached nitrate removal up to 99.2%, with effluent NO_3–N as low as 0.31 mg N/L. Although LFGC required more external COD (6.6 g COD/g NO_3–N removed) than the conventional carbon sources (3.6–3.7), it also enabled the shortest minimum denitrification time (2.2 h). EEM–PARAFAC and FTICR–MS analyses showed similar dissolved organic matter characteristics in all effluents, indicating that LFGC did not produce distinct fluorescent or molecular DOM signatures compared with conventional carbon sources. Overall, LFGC performed comparably to conventional liquid carbon sources while beneficially reusing a landfill byproduct.

1 | Introduction

Landfill leachate is a complex and highly polluted wastewater formed when infiltrating water percolates through municipal solid waste (MSW), mobilizing organic and inorganic contaminants, heavy metals, and high concentrations of nitrogenous species such as ammonia and organic nitrogen (Mojiri et al. 2020; Wijekoon et al. 2022). Leachate generation can range from 0.1 to 320 million gallons per year per landfill, depending on landfill size, waste composition, climate, and management practices (Masoner et al. 2014; Abunama et al. 2019). In the United States alone, annual leachate production has been estimated at 16.1 billion gallons, with most facilities relying on off-site treatment at publicly owned treatment works (POTWs) (Lang et al. 2017; Gokgoz et al. 2023). The associated treatment cost, typically on the order of 0.04–0.21 USD per gallon, can translate into millions of dollars per year for large landfills, and even higher costs are reported in jurisdictions requiring advanced treatment (Christensen et al. 2014; Gokgoz et al. 2023).

Biological nitrogen removal (BNR) is widely employed to treat the nitrogenous fraction of sewage and landfill leachate (Hu et al. 2023; Mishra et al. 2022). In conventional BNR, ammonium (NH_4^+–N) is first oxidized to nitrate (NO_3^-–N) through

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Summary

- Landfill gas condensate (LFGC) was evaluated as an external carbon source in SBRs.
- LFGC achieved total nitrogen removal comparable to methanol, glycerin, and acetate.
- LFGC required a higher specific COD dose but enabled slightly shorter denitrification time.
- EEM–PARAFAC and FTICR–MS showed no increase in effluent DOM recalcitrance with LFGC.
- Valorizing LFGC can offset purchased carbon and reduce separate condensate treatment needs.

nitrification, followed by reduction of nitrate to dinitrogen gas (N_2) via heterotrophic denitrification (Peng and Zhu 2006; Lin et al. 2009). Denitrification requires an external electron donor, typically an organic carbon source, especially when treating low C/N municipal wastewaters co-mingled with high-strength but often low-biodegradability landfill leachate (Miao et al. 2014; Remmas et al. 2018).

External carbon sources used in practice can be classified into solid, liquid, and gaseous forms (Fu et al. 2022). Liquid carbon sources, such as methanol, ethanol, glycerin, acetic acid, and sodium acetate, are the most commonly applied due to their high bioavailability and operational simplicity (Peng et al. 2007; Cyplik et al. 2013; Chen et al. 2015; Zheng et al. 2019). However, the continuous addition of these commercial chemicals substantially increases operating costs. For example, approximately 4 L of methanol may be required to remove 1 kg of NO_3^-–N at a C/N ratio of 6.0 (Cao et al. 2020). To mitigate these costs, a range of alternative carbon sources has been explored, including natural cellulosic materials, biodegradable polymers, industrial wastewaters, and fermentation liquids from sludge or food waste (Ovez 2006; Fernández et al. 2011; Tokiwa et al. 2009; Zhang et al. 2016; Hu et al. 2020). Although many of these alternatives are effective, their availability, handling requirements, and variability can limit broader implementation.

Landfill gas condensate (LFGC) is a byproduct of landfill gas collection systems, formed when moisture and short-chain volatile organic compounds (VOCs) condense as the gas cools in collection pipes (Zhao et al. 2013). These short-chain VOCs are typically readily biodegradable and can serve as electron donors for denitrifying microorganisms (Maione et al. 2005). LFGC generally consists of an aqueous phase with dissolved organic compounds and a floating hydrocarbon layer (Zhao et al. 2013). Therefore, although LFGC is generally less contaminated than raw landfill leachate, it still requires proper handling, treatment, or disposal due to the presence of dissolved organic compounds, hydrocarbons, and trace organic/inorganic constituents. In many landfills, LFGC is managed as a waste stream requiring separate treatment or disposal, despite production volumes that can reach several thousand liters per day at individual sites. Valorizing LFGC as an external carbon source for denitrification could simultaneously offset the cost of purchased liquid carbon and reduce or eliminate dedicated condensate treatment.

To date, the feasibility of using LFGC as an external carbon source for denitrification has not been systematically evaluated in comparison with conventional liquid carbon sources under controlled BNR conditions. In addition, its impact on effluent dissolved organic matter (DOM) composition has not been thoroughly characterized using EEM–PARAFAC and FTICR–MS.

This study addresses this gap by investigating the performance of LFGC as an external carbon source in lab-scale sequencing batch reactors (SBRs) co-treating municipal sewage and landfill leachate. Specifically, the objectives were to (i) compare nitrogen removal performance, volumetric carbon dose (VCD), specific external COD requirement, and denitrification time among reactors fed methanol, glycerin, sodium acetate, and LFGC; and (ii) assess whether LFGC alters effluent DOM quantity or quality using EEM–PARAFAC and FTICR–MS. The results provide insights into the technical viability and sustainability benefits of LFGC as an alternative denitrification carbon source for sewage–landfill leachate co-treatment.

2 | Materials and Methods

2.1 | Raw Sewage and Landfill Leachate

In lab-scale SBR reactors, raw sewage and landfill leachate were mixed as influent for the BNR process. Raw sewage was collected from the Lexington Sewer Treatment Plant, NC, United States, and sampled every 10–15 days. The sewage was subsequently

TABLE 1 | Sewage and landfill leachate characteristics.

Parameters	Sewage	Landfill leachate
pH	7.5 ± 0.2	8.1
TN (mg/L)	40.0 ± 2.5	3370
TKN (mg/L)	39 ± 2	3350
NO_3^-–N (mg/L)	1.0 ± 0.4	16.3
NO_2^-–N (mg/L)	0.05 ± 0.01	0.84
NH_4^+–N (mg/L)	32 ± 4	2350
COD (mg/L)	260 ± 40	4880
BOD (mg/L)	124 ± 15	1680
BOD/COD ratio	0.48	0.34
Total phosphorous (mg/L)	17 ± 3	1770
Alkalinity (mg/L as $CaCO_3$)	186 ± 12	1950
TS (mg/L)	N/A	12,100
TSS (mg/L)	N/A	250
TDS (mg/L)	N/A	11,800

Abbreviations: TDS = total dissolved solid, TKN = total Kjeldahl nitrogen, TS = total solid, TSS = total suspended solid.

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stored in 5-gal high-density polyethylene buckets at 4°C upon collection. The sewage was allowed to reach 22°C before being fed into the reactors. The raw leachate was obtained from an MSW landfill in Virginia, United States. Upon arrival, the leachate was stored at 4°C and subsequently elevated to 22°C prior to utilization. Table 1 shows the characteristics of the raw sewage and landfill leachate.

2.2 | Biological Reactor Setup and Operation

Four identical SBR reactors, designated as R1, R2, R3, and R4, were used for this study. The reactors were fabricated with polymethyl methacrylate (PMMA) sheets, possessing a total volume of 5 L and an active working volume of 3 L, each with square dimensions measuring 7.1 in. on each side (Figure 1). Each reactor was equipped with an overhead mechanical stirrer, an air disperser, and a water heater; pH and dissolved oxygen (DO) sensors (IQ SensorNet) were also deployed.

The reactors were fed with a blend of 98.5% (v/v) sewage and 1.5% (v/v) landfill leachate to mimic a representative real-world scenario of sewage-leachate co-treatment in a POTW. The reactors were operated for 170 days in three stages: Stage 1 (initial 65 days) to determine the maximum denitrification efficiency for minimum effluent NO_3–N, Stage 2 (subsequent 62 days) to determine the optimal external carbon dosage, and Stage 3 (43 days) to determine the optimal denitrification time with the optimal external carbon dosage. Each reactor operated with a hydraulic retention time (HRT) of 1 day, 2 cycles per day, and a solid retention time (SRT) of 15 days. The average organic loading rate (OLR) over the study period was 0.36 g COD/L/day. The operational sequence consisted of aerobic (6.5 h), anoxic (3.5 h), post-aerobic (1 h), settling, decanting-feeding, and idle (1 h). Airflow was maintained at 1.5 L/min to keep DO level between 1–2 mg/L, and the mixed liquor-suspended solids (MLSS) concentration was kept at around 4000 mg/L. Samples were analyzed daily for the initial 10 days and subsequently every other day, filtered through 0.45 μm glass microfiber prior to analysis. Initially, reactors were supplied exclusively with sewage for 7 days, and upon attaining effective nitrification–denitrification, leachate was introduced to achieve a final concentration of 1.5% (v/v).

2.3 | Carbon Sources

Four carbon sources were introduced into the reactors following the nitrification phase to improve denitrification efficacy. The reactors R1, R2, R3, and R4 were supplemented with methanol (VWR, Radnor, PA, USA), glycerin (Essential Depot Inc., Sebring, FL, USA), sodium acetate (Sigma-Aldrich, Burlington, MA, USA), and LFGC (Wake County MSW Landfill, NC, USA), respectively. Methanol and LFGC were directly introduced into the reactors in their liquid forms without any further preparation. Glycerin, due to its very dense concentration, was diluted by four times with deionized (DI) water. Sodium acetate, initially in granular form, was dissolved by mixing 100 g of sodium acetate into 1 L of DI water to prepare a solution. External carbon addition was expressed using both VCD, to indicate the practical liquid dosing requirement, and specific external COD dose, to normalize COD input by nitrate removal, as defined below:

VCD was defined as the volume of external carbon solution added per unit reactor working volume per day:

$$\mathrm{VCD}\ \left(\mathrm{L\,m^{-3}\,d^{-1}}\right) = \frac{V_{\mathrm{C,day}}}{V_{\mathrm{R}}} \tag{1}$$

where $V_{\mathrm{C,day}}$ is the total volume of carbon solution added per day ($\mathrm{L\,d^{-1}}$) and V_{R} is the reactor working volume ($\mathrm{m^3}$).

The specific external COD dose was defined as the COD supplied by the external carbon source per nitrate–nitrogen removed:

$$\text{Specific external COD dose}\ \left(\mathrm{g\,COD\,/\,g\,NO_3-N}\right) = \frac{M_{\mathrm{COD,ext}}}{M_{\mathrm{NO_3-N,removed}}} \tag{2}$$

with $M_{\mathrm{COD,ext}} = C_{\mathrm{COD,sol}} \times V_{\mathrm{C,day}}$, where $C_{\mathrm{COD,sol}}$ is the COD concentration of the carbon solution ($\mathrm{g\ COD\ L^{-1}}$).

In this study, the COD concentration for methanol (pure), glycerin solution (25%, v/v), sodium acetate solution (100 g/L), and LFGC were 1,184,000, 331,000, 78,000, and 16,000 mg/L, respectively. No additional VOC or hydrocarbon speciation was conducted; therefore, the LFGC composition in this study is reported based on its measured bulk COD, while detailed

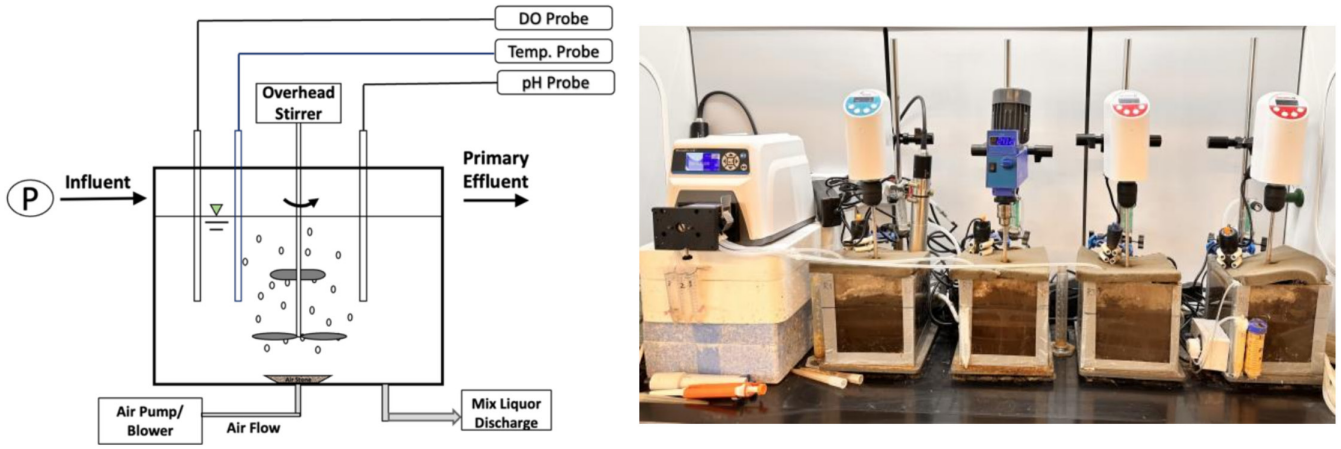


FIGURE 1 | SBR schematic diagram (left) and laboratory setup (right).

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compound-level characterization should be considered in future full-scale applications. The VCD ranged from 0.13 to 3.34 L/m^3 for methanol, 0.35 to 5.34 L/m^3 for glycerin, 0.34 to 6.67 L/m^3 for sodium acetate, and 10 to 40 L/m^3 for condensate.

2.4 | Sample Analysis

All tests followed the standard methods outlined by the American Public Health Association (APHA) (Rice et al. 2012). Physicochemical analyses include pH, DO, temperature, TN, NH_4^+–N, NO_2^-–N, NO_3^-–N, TKN, total phosphorus (TP), total alkalinity, COD, and BOD. pH was measured using the Apera Instruments AI311 Premium Series PH60 tester kit (Columbus, Ohio, USA) and DO was determined using a HACH Intellical LDO101 probe with a HACH HQ440 multi-meter (Loveland, Colorado, USA). The remaining parameters were analyzed using the HACH DR6000 spectrophotometer with HACH TNT 822, 826, 830, 831, 832, 835, 839, 846, and 870 kits (Loveland, Colorado, USA) (Joshi et al. 2021). For BOD testing, the APHA 5210 method was employed (Rice et al. 2012).

Statistical analysis was performed using Minitab to compare effluent water quality parameters among the four reactors under steady-state operating conditions. One-way analysis of variance (ANOVA) followed by Tukey's post hoc test was applied to evaluate differences in effluent NH_4^+–N, TN, NO_3^-–N, COD, and BOD among reactors at a significance level of $p < 0.05$.

2.5 | DOM Characterization

EEM–PARAFAC and FTICR–MS were used to determine whether LFGC introduced distinct or more refractory DOM signatures in the effluent compared with conventional carbon sources. EEM–PARAFAC is useful in identifying fluorescent organic compounds, while FTICR–MS provides high-resolution molecular characterization (Jiang et al. 2022).

HPLC-grade solvents (Sigma-Aldrich Chemical Co., St. Louis, MO) and solid-phase extraction (SPE) extracts were utilized for FTICR–MS analysis with negative ion electrospray ionization at the National High Magnetic Field Laboratory in Tallahassee, Florida. DOM was analyzed using a custom-built hybrid linear ion trap FT-ICR mass spectrometer featuring a 21 T superconducting solenoid magnet (Hendrickson et al. 2015). Molecular formula assignments and data visualization were conducted using PetroOrg software (Corilo 2014). Molecular formulas were also used to construct the Van Krevelen diagram. Details on the FTICR–MS evaluation of molecular composition are further outlined in Supporting Information S1: Section 1 (S1).

Fluorescence EEM spectroscopy was used to assess effluent DOM (McKnight et al. 2001). Samples were scanned at 20°C using an Agilent Cary Eclipse fluorescence spectrometer (Santa Clara, CA, USA). Excitation and emission wavelengths were scanned at intervals of 5 nm, ranging from 200 to 600 nm and 200 to 700 nm, respectively, with varying slit widths and scan rates. The PARAFAC multivariate statistical method was employed to objectively and accurately characterize the EEM spectrum (Murphy et al. 2011).

3 | Results and Discussion

3.1 | Oxygen Demand Materials Removal and Nitrification

Throughout the study period, the pH levels of the effluents for R1 (methanol), R2 (glycerin), R3 (sodium acetate), and R4 (LFGC) remained stable at 8.0 ± 0.2, 7.9 ± 0.3, 8.0 ± 0.4, and 8.0 ± 0.2, respectively (Figure S1), with no significant fluctuations observed following the addition of different carbon sources. The COD in the effluent was reduced to 182 ± 40, 125 ± 35, 115 ± 30, and 93 ± 30 mg/L from an influent COD of 358 ± 80 mg/L, achieving removal efficiencies of 49.2%, 65.1%, 67.9%, and 74.1% for reactors R1, R2, R3, and R4, respectively (Figure 2a, Table 1, Figure S2). BOD removal efficiencies were 59.2%, 66.7%, 68.1%, and 72.1% for R1, R2, R3, and R4, respectively (Figure 2b, Figure S3, and Table 1). The highest COD and BOD removal efficiencies were achieved in the condensate reactor (R4), demonstrating the SBR's effectiveness with this new external carbon source. Similarly, a separate study (Renou et al. 2008) reported a 75% COD removal from landfill leachate in an SBR process, comparable to the condensate reactor's performance. An additional study demonstrated that an SBR was capable of removing 70% BOD, which is slightly less than the condensate reactor used in this study, further confirming the enhanced efficiency of the SBR (Zouboulis et al. 2001). Effluent BOD exceeded the conventional NPDES limit of 30 mg/L in all reactors, likely due to residual biodegradable or slowly biodegradable organic matter remaining after external carbon addition for near-complete denitrification. The LFGC reactor showed the lowest effluent BOD among the four reactors, indicating that LFGC addition did not cause a greater residual BOD burden than the conventional carbon sources.

The NH_4^+–N removal efficiencies were 99.8%, 99.9%, 99.8%, and 99.8% for reactors R1, R2, R3, and R4, respectively, with effluent NH_4^+–N levels of 0.14 ± 0.03, 0.05 ± 0.02, 0.11 ± 0.05, and 0.10 ± 0.05 mg/L from an influent concentration of 65 ± 7 mg/L (Figure 3, Figure S4, and Table 2). These findings are consistent with those of Chen et al. (2015), who achieved similar NH_4^+–N removal efficiencies using five different external carbon sources in their SBR study. Similarly, a study reported comparable NH_4^+–N removal trends in their study on landfill leachate using the SBR process (Wang et al. 2013). The reactors consistently achieved full nitrification across all carbon sources, with the addition of LFGC not impairing the nitrification process.

The effluent TN levels were found to be 5.1 ± 2.2, 4.9 ± 2.6, 5.2 ± 2, and 5.5 ± 2.3 mg/L for R1, R2, R3, and R4, respectively, compared with the influent value of 80.2 ± 4 mg/L (Table 2). The TN removal efficiencies were therefore calculated at 93.6%, 93.8%, 93.5%, and 93.1%, respectively. The effluent TN primarily consists of refractory dissolved organic nitrogen (rDON). These results are consistent with the findings of Morling (2010), who reported TN removal efficiencies of 93%–95% in the SBR process for leachate treatment. Similarly, Chen et al. (2015) demonstrated that 93% removal efficiency of TN was achieved when acetate was utilized as a carbon source for denitrification of wastewater in the BNR process. Our research shows that the LFGC reactor achieved similar TN removal efficiency with other commonly used carbon

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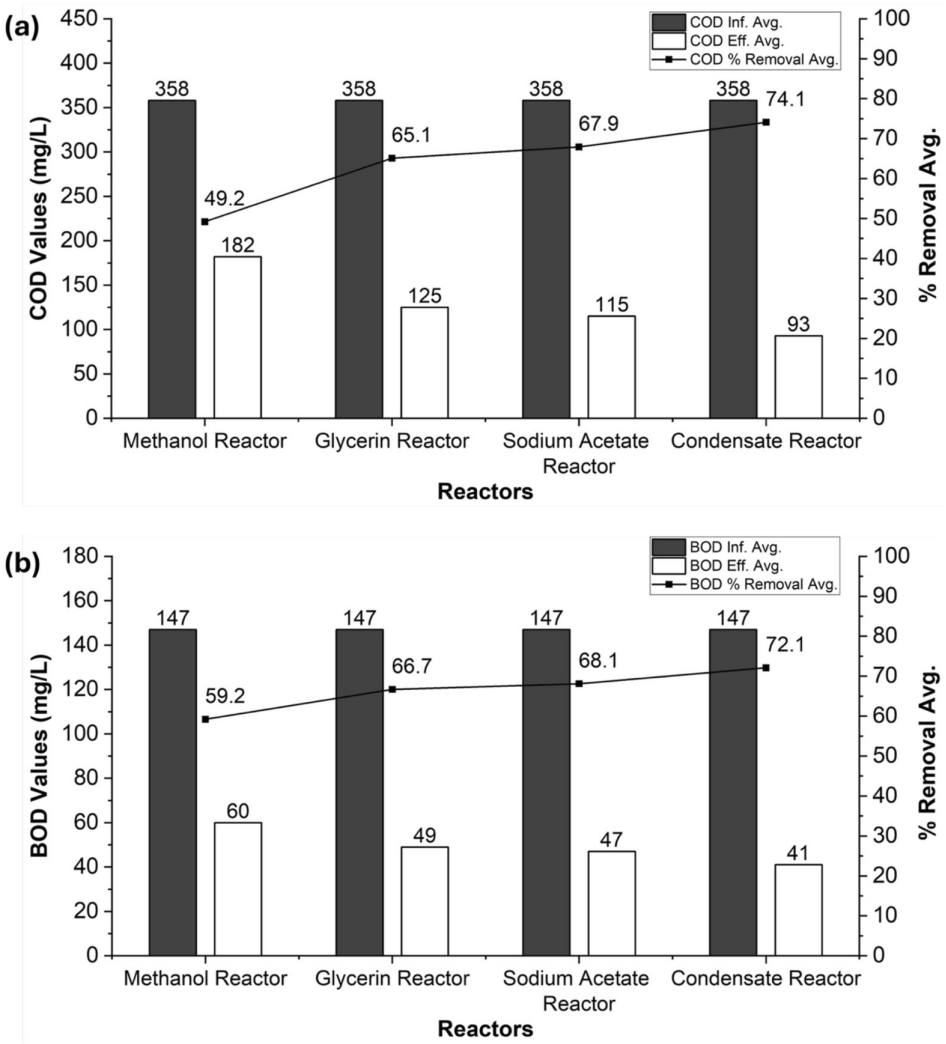


FIGURE 2 | Oxygen demand materials of the influent and effluents of the four reactors.

sources, which further confirms the feasibility of LFGC as a feasible external carbon source for sewage and landfill leachate denitrification.

One-way ANOVA followed by Tukey's post hoc test showed no statistically significant differences among the four reactors for effluent NH_4^+–N ($p = 0.095$), TN ($p = 0.078$), NO_3^-–N ($p = 0.192$), COD ($p = 0.116$), or BOD ($p = 0.081$). These results indicate that the LFGC-fed reactor achieved effluent quality statistically comparable to the reactors supplemented with methanol, glycerin, and sodium acetate, further supporting the feasibility of LFGC as an alternative external carbon source for denitrification. Details can be found in the Supporting Information S1: Section 5 (S5).

3.2 | Denitrification

After achieving complete nitrification, external carbon sources—methanol, glycerin, sodium acetate, and condensate—were introduced to the reactors to drive denitrification with initial dosages of 0.13, 0.35, 0.34, and 10 L/m^3 for reactors R1, R2, R3, and R4, respectively. The initial doses were selected based on the COD contents of the carbon sources (Section 2.3). After 14 cycles (7 days of operation), the reactors achieved complete denitrification with sewage alone. Then, landfill leachate was gradually introduced to all four reactors, beginning with 0.3% (v/v) of the reactor's working volume. Over the next 15 days, the leachate concentration was increased incrementally, reaching the target concentration of 1.5% (v/v). Simultaneously, the dosages of external carbon sources were adjusted to optimize denitrification performance. After the nitrification phase in each SBR cycle (before the start of the anoxic phase for denitrification), the NO_3–N concentrations were measured to be 34.5 ± 6, 37.4 ± 5, 36.4 ± 6, and 37.5 ± 4 mg/L for reactors R1, R2, R3, and R4, respectively. Although the feed contained 98.5% sewage and 1.5% landfill leachate by volume, the calculated COD contribution was approximately 77.8% from sewage and 22.2% from leachate. After nitrification, COD was reduced by approximately 85%, leaving 53.7 mg/L before external carbon addition. Depending on the operational phase, the COD after dosing ranged from 369.4–2027.0 mg/L for methanol, 230.2–936.4 mg/L for glycerin, 105.7–573.7 mg/L for sodium acetate, and 213.7–587.0 mg/L for LFGC. The higher specific COD dose for LFGC suggests a lower readily biodegradable COD fraction than conventional carbon sources. After the anoxic phase, the NO_3–N levels in the final effluents of all reactors dropped to below 1 mg/L,

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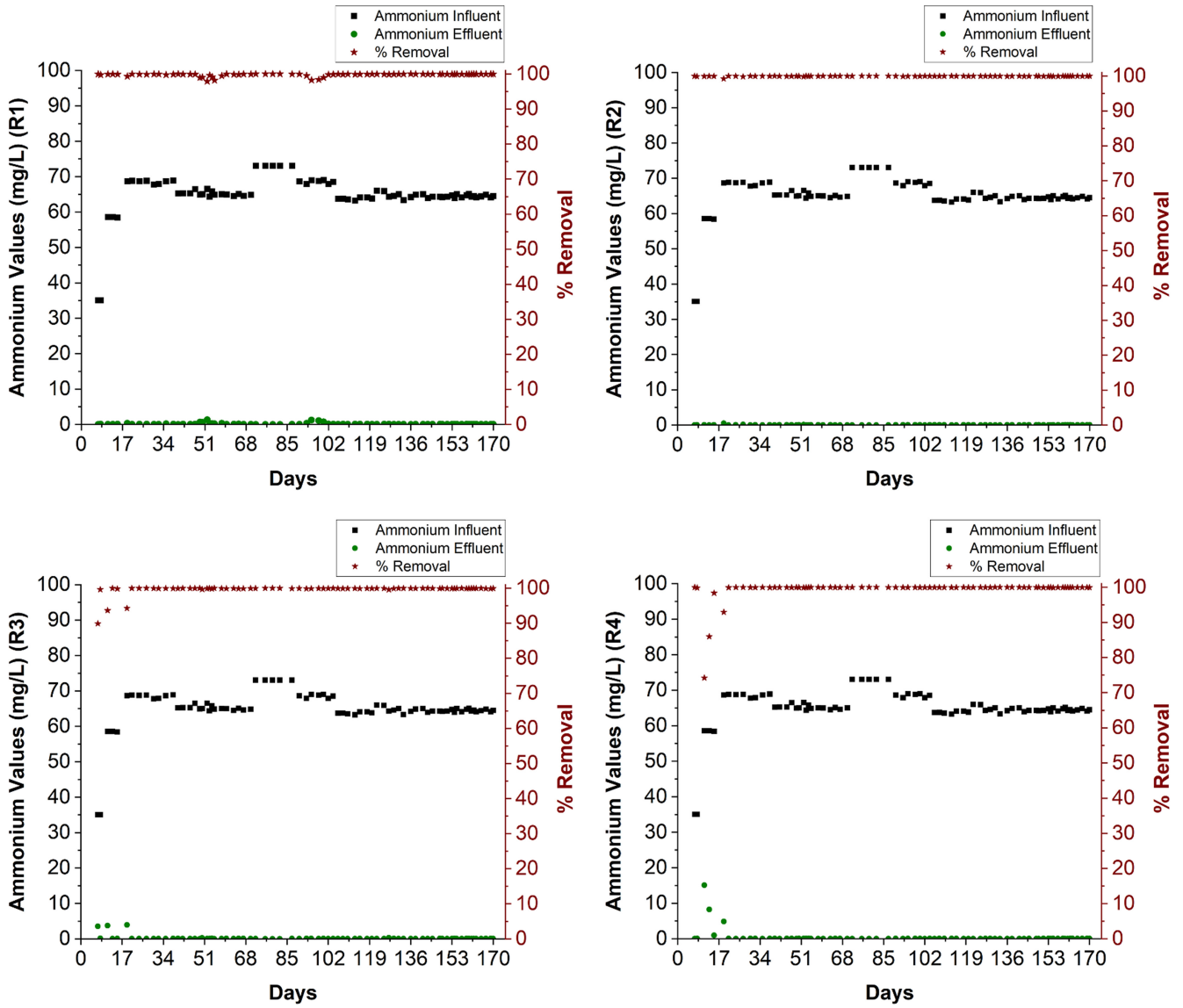


FIGURE 3 | The influent and effluent NH_4^+–N levels for reactors: R1 (methanol reactor), R2 (glycerin reactor), R3 (sodium acetate reactor), and R4 (LFGC reactor), and corresponding removal efficiencies throughout the study period.

TABLE 2 | The characteristics of the influent and effluents of the four reactors.

Parameters	Influent (98.5% sewage and 1.5% leachate)	R1 effluent (methanol)	R2 effluent (glycerin)	R3 effluent (sodium acetate)	R4 effluent (condensate)
pH	7.7 ± 0.4	8 ± 0.2	7.9 ± 0.3	8 ± 0.4	8 ± 0.2
TN (mg/L)	80.2 ± 4	5.1 ± 2.2	4.9 ± 2.6	5.2 ± 2	5.5 ± 2.3
TKN (mg/L)	79.3 ± 4.4	—	—	—	—
NH_4^+–N (mg/L)	65 ± 7	0.14 ± 0.03	0.05 ± 0.02	0.11 ± 0.05	0.10 ± 0.05
NO_3–N (mg/L) (before denitrification)	—	34.5 ± 6	37.4 ± 5	36.4 ± 6	37.5 ± 4
NO_3–N (mg/L) (after denitrification)	—	0.7 ± 0.4	0.7 ± 0.3	0.8 ± 0.3	0.7 ± 0.4
NO_2–N (mg/L)	0.050 ± 0.006	BRL	BRL	BRL	BRL
COD (mg/L)	358 ± 80	182 ± 40	125 ± 35	115 ± 30	93 ± 30
BOD (mg/L)	147 ± 20	60 ± 20	49 ± 30	47 ± 25	41 ± 20

Abbreviation: BRL: below reporting limit.

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which is more than 97% NO_3–N removal, indicating almost complete denitrification.

After complete denitrification was achieved, the reactors were operated in three stages to assess three extreme denitrification scenarios: Stage 1. Minimum effluent NO_3^-–N; Stage 2. Minimum carbon source dose; Stage 3. Minimum denitrification time. The operation data are shown in Figures S5–S8.

3.2.1 | Minimum Effluent NO_3^-–N

To achieve the lowest effluent NO_3–N concentrations, the carbon source doses were gradually increased to 3.34 L/m^3/day (methanol), 5.34 L/m^3 (glycerin), 6.67 L/m^3 (sodium acetate), and 40 L/m^3 (condensate) until Day 65 (end of the first operational stage). The lowest effluent NO_3–N concentrations were 0.44, 0.41, 0.51, and 0.31 mg/L for R1, R2, R3, and R4, respectively, with removal efficiencies of 98.7%, 98.9%, 98.6%, and 99.2%, respectively (Figures 4 and S5–S8). The corresponding external carbon dosages for the lowest effluent NO_3–N were 1.14 L/m^3/day (methanol), 4 L/m^3/day (glycerin), 4.67 L/m^3/day (sodium acetate), and 33.34 L/m^3/day (condensate). A study on landfill leachate treatment using the SBR process reported 86% NO_3–N removal (Miao et al. 2014). Another study on SBR for sewage treatment using sludge fermentation products as a denitrification carbon source resulted in an effluent NO_3–N concentration of 10.2 mg/L (Yuan et al. 2016). Additionally, Yan et al. (2019) reported an effluent NO_3–N concentration of 1.85 mg/L after treating synthetic wastewater in an SBR. This study demonstrated that using condensate for denitrification in sewage and landfill leachate was highly effective, with similar or better performance as previous studies on different wastewater types. It underscores the potential of LFGC as a promising carbon source for future landfill leachate denitrification studies.

3.2.2 | Optimal External Carbon Doses

After achieving the lowest effluent NO_3–N concentrations and maximum denitrification efficiency, the optimal external carbon dosage was determined in Stage 2 (following 62 days of operation) (Figures 4 and S5–S8). During this operational period, the external carbon dosages for each reactor were gradually reduced until complete denitrification was hindered, as evidenced by elevated effluent NO_3–N concentrations (Figure 4). The denitrification performance remained stable

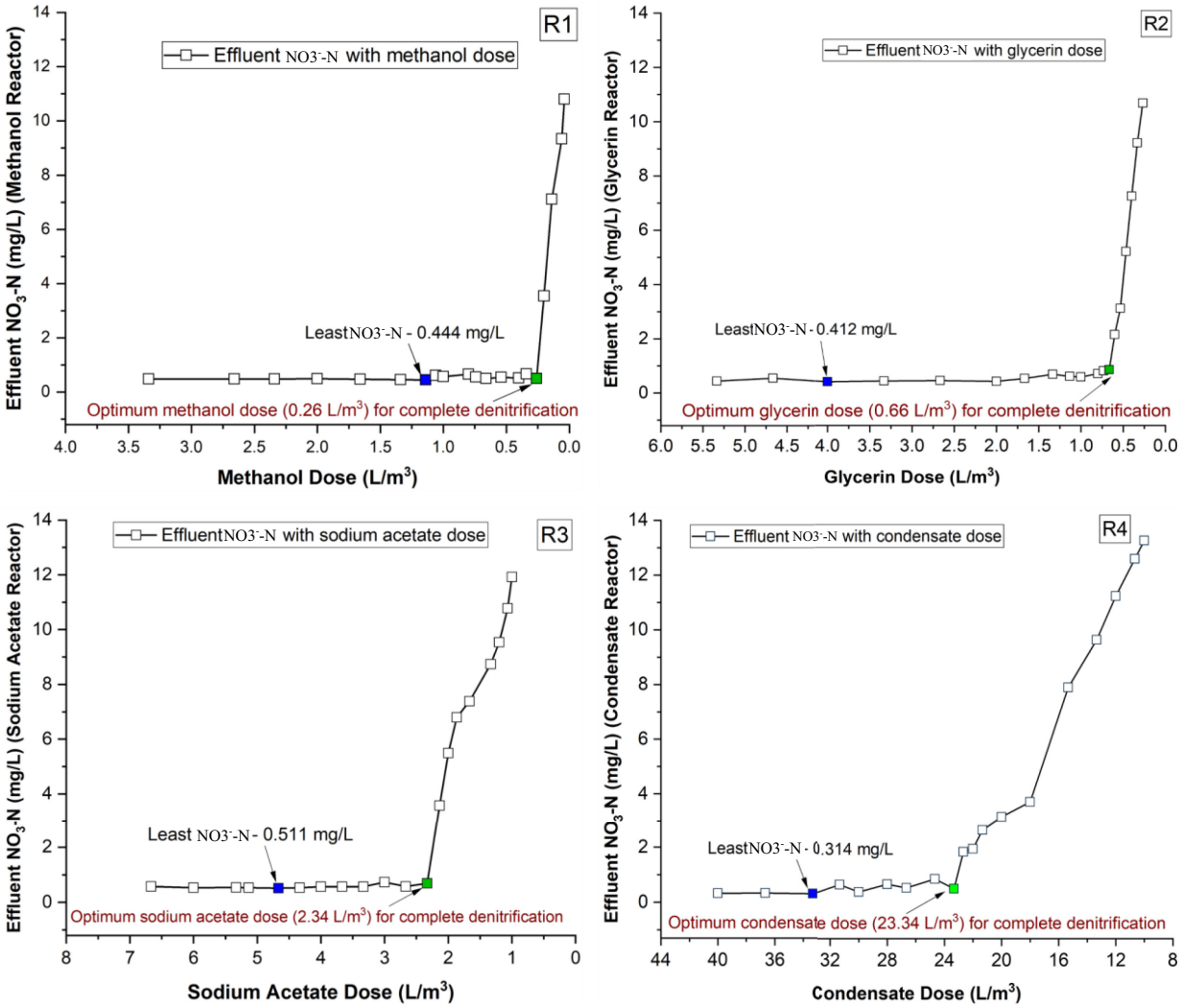


FIGURE 4 | The variation in effluent NO_3–N concentration with different carbon source dosages for reactors: R1 (methanol reactor), R2 (glycerin reactor), R3 (sodium acetate reactor), and R4 (LFGC reactor).

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until the methanol dosage decreased to 0.26 L/m^3/day, glycerin to 0.66 L/m^3/day, sodium acetate from 2.34 L/m^3/day, and condensate 23.34 L/m^{3}day. Any further reduction in external carbon source dosage resulted in incomplete denitrification, leading to increased effluent NO_3–N concentrations. Thus, the above dosages were identified as their optimal doses. At optimum, based on the COD contents of carbon sources solutions used in this study, the corresponding normalized carbon doses were 3.7 g COD/g NO_3–N for methanol, 3.7 g COD/g NO_3–N for glycerin, 3.6 g COD/g NO_3–N for sodium acetate, and 6.6 g COD/g NO_3–N for condensate, as detailed in Table S1. At these optimal dosages, the effluent NO_3–N concentrations were measured at 0.503, 0.667, 0.691, and 0.521 mg/L for R1, R2, R3, and R4, respectively. It appears the LFGC requires a higher specific external COD dose compared with other carbon sources. The elevated COD/NO_3–N ratio for LFGC is more likely attributable to a lower fraction of readily biodegradable COD available during the anoxic period and to COD measured on constituents that are ultimately oxidized via non-denitrifying pathways or removed through sludge sorption.

3.2.3 | Optimal Denitrification Time

After determining the optimal carbon source dosages, the reactors were operated for an additional 43 days to establish the optimal denitrification time (Figures 5 and S5–S8). During this stage, the optimal carbon dosages were maintained without further adjustments. Initially, the reactors operated with a denitrification time of 3.5 h, which was then gradually reduced to 1 h at a rate of 0.06 h per day. Denitrification performance remained stable until the time was reduced to 2.4 h for methanol, 2.6 h for glycerin, 2.75 h for sodium acetate, and 2.2 h (shortest among all) for the condensate reactor. Further reductions negatively impacted the denitrification performance, increasing effluent NO_3–N levels. Therefore, the optimal denitrification times were 2.4, 2.6, 2.75, and 2.2 h for methanol, glycerin, sodium acetate,

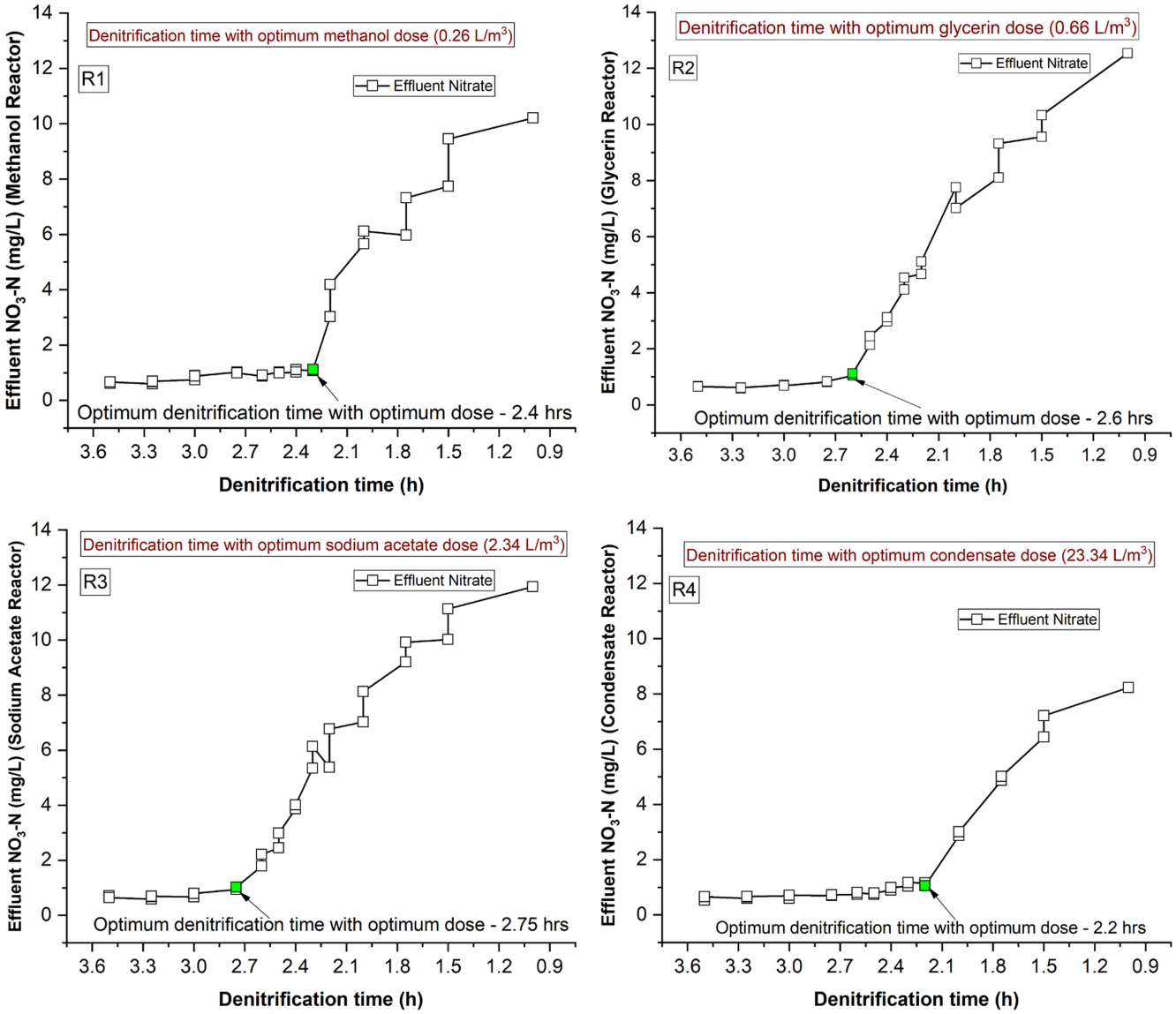


FIGURE 5 | The denitrification time of the reactors with the optimal carbon source dosage for reactors: R1 (methanol reactor), R2 (glycerin reactor), R3 (sodium acetate reactor), and R4 (condensate reactor).

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and condensate reactor, respectively, at their optimal dosages. The shorter denitrification time for LFGC should be interpreted together with its higher optimized COD dose, rather than as evidence of greater intrinsic biodegradability. The higher LFGC dose likely supplied enough readily usable carbon during the anoxic phase to support rapid nitrate reduction, while the larger overall COD requirement reflects the lower readily biodegradable fraction of the condensate. The effluent NO_3–N concentrations recorded at these optimal conditions were 1.02 mg/L (methanol), 1.04 mg/L (glycerin), 1.02 mg/L (sodium acetate), and 1.06 mg/L (condensate). The overall optimal operational conditions are summarized in Table 3.

Three previous SBR studies using a 3-h anoxic/anaerobic phase reported 70% TN removal for landfill leachate (Remmas et al. 2017), 90% TN removal for sewage (Khursheed et al. 2018), and 80% TN removal with an effluent NO_3^-–N of 5.04 mg/L for sewage (Huang and Goel 2015). In comparison, this lab-scale condensate–fed SBR achieved > 90% TN removal and substantially lower NO_3^-–N concentrations with only 2.2 h of denitrification, demonstrating that condensate is a highly effective alternative carbon source that shortens denitrification time while maintaining high nitrogen removal efficiency in sewage and landfill leachate co-treatment.

3.3 | Effluent DOM Molecular Characteristics

3.3.1 | Fluorescent Components of the Effluent DOM

Figure 6 presents the EEM for all effluent samples from reactors R1, R2, R3, and R4. The EEM plots were generated using the same excitation wavelengths (220–450 nm) and emission wavelengths (280–600 nm). The EEM analysis successfully identified multiple fluorophores in the effluent samples from all reactors. Across all effluents, two prominent peaks were observed at Ex/

TABLE 3 | The summary of the optimal denitrification conditions of SBR reactors for landfill leachate denitrification.

Reactors	External carbon sources	Optimal volumetric carbon dose ($L/m^3/day$)	Specific external COD dose (g COD/g NO_3^-–N)	Optimal denitrification time (hours)	Effluent NO_3–N (mg/L)
R1	Methanol	0.26	3.7	2.4	1.02
R2	Glycerin	0.66	3.7	2.6	1.04
R3	Sodium acetate	2.34	3.6	2.75	1.02
R4	Condensate	23.34	6.6	2.2	1.06

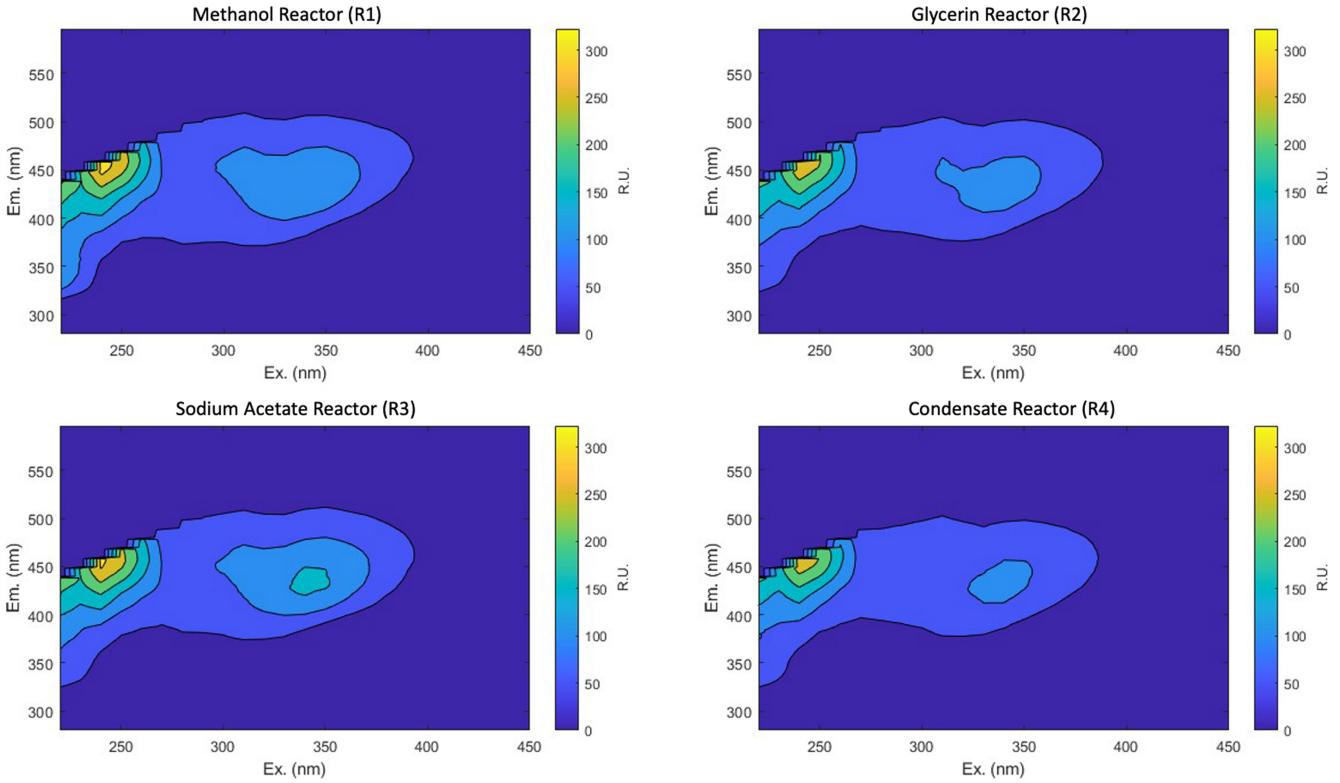


FIGURE 6 | The EEM fluorescence spectra of the effluents from all four reactors: R1 (methanol), R2 (glycerin), R3 (sodium acetate), and R4 (LFGC).

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TABLE 4 | PARAFAC model output: fluorophore components and corresponding intensities.

Effluents	Component 1 humic-like compounds (Ex/Em = 340/450 nm)	Component 2 humic-like compounds and fulvic-like compounds (Ex/Em = 220, 330/430 nm)	Component 3 aromatic proteins II (lignin) (Ex/Em = 220/350 nm)	Component 4 humic-like compounds (Ex/Em = 230/450 nm)
R1 (methanol)	93.70	55.66	86.74	17.81
R2 (glycerin)	79.36	48.84	42.11	16.51
R3 (sodium acetate)	110.42	48.26	36.21	18.12
R4 (condensate)	74.55	45.83	41.69	14.87

Em = 330/430 nm and Ex/Em = 230/450 nm. The peak at Ex/Em = 330/430 nm is associated with humic and fulvic-like compounds (Deng et al. 2018), hydrophobic acids (Xiao et al. 2018), and fulvic-like substances (Wang et al. 2022). Meanwhile, the peak at Ex/Em = 230/450 nm is linked to humic-like compounds (Duan et al. 2024) and fulvic-like compounds (Duan et al. 2024; Lu et al. 2013) (Table S2). This indicates that the same organic matter fractions are present in the effluent across all reactors, suggesting that the addition of condensate showed comparable DOM signatures to conventional carbon sources.

The PARAFAC model identified four distinct fluorophores in reactors R1, R2, R3, and R4. Table 4 provides a detailed summary of the PARAFAC results for each reactor, specifying the wavelengths, components, and intensities of the identified fluorophores. For all four reactors, the model identified four fluorophores with specific peaks at the following excitation/emission (Ex/Em) wavelengths: C1 at 340/450 nm, C2 at 220, 330/430 nm, C3 at 220/350 nm, and C4 at 230/450 nm. The compound C1 is aligned with humic-like compounds (Deng et al. 2018) and is further characterized as hydrophilic substances (Xiao et al. 2018). C2 represents humic and fulvic-like compounds (Deng et al. 2018) and is also described as hydrophobic acids (Xiao et al. 2018). C3 is associated with aromatic proteins II (Deng et al. 2018). Lastly, C4 is associated with humic-like compounds (Li and Peleato 2021; Xu et al. 2016) while other researchers have identified C4 as fulvic-like compounds (Duan et al. 2024; Lu et al. 2013).

Table 4 illustrates the PARAFAC model's capacity to identify and compare component intensities across the reactor effluents. The effluents from all reactors display similar component intensities, although the condensate reactor effluent (R4) shows slightly lower intensities for C1, C2, C3, and C4 (74.55, 45.83, 41.69, and 14.87, respectively) compared with the other reactors. This suggests that the addition of condensate did not cause significant changes in the effluent composition and did not introduce any residual organic compounds, further supporting the viability of condensate as an effective carbon source for denitrification.

3.3.2 | Molecular Composition of Effluent DOM by FTICR–MS

FTICR–MS, a high-resolution mass spectrometry technique, was employed to analyze the molecular characteristics of DOM in the effluents of SBR reactors R1, R2, R3, and R4. This analysis provided insight into the molecular composition and interactions of the DOM. The FTICR–MS identified 7849, 6723, 9027, and 7900 molecular formulas associated with DOM in the effluents from R1, R2, R3, and R4, respectively. Using van Krevelen plots, the molar H/C (hydrogen:carbon) and O/C (oxygen:carbon) ratios were plotted for individual peaks, grouping the elemental formulas according to their functional groups (Shakeri Yekta et al. 2012). Figure 7 and Table 5 illustrate the H/C versus O/C molar ratios for elemental compositions with relative abundances exceeding 0.25% in the effluent samples (Mallick et al. 2024), revealing a complex mixture of DOM molecules. Each plot differentiates DOM compounds by color. All reactors displayed similar DOM molecular compositions in their effluents.

The identified molecular groups across all the reactors were lipids, proteins/amino sugars, carbohydrates, lignin, tannins, and condensed aromatics. There were no unsaturated hydrocarbons present in any of the effluents. This result aligns with the findings of Hu et al. (2020) (Hu et al. 2020), where the effluent from sewage BNR similarly showed the absence of unsaturated hydrocarbons, indicating a consistent composition pattern between different BNR processes. The relative abundance of the components of all four reactors followed the order of lignin > proteins/amino sugars > carbohydrates > tannins > condensed aromatics > lipids, as summarized in Table S3. Compounds with higher concentrations of proteins/amino sugars indicate more readily available DOM, whereas lignin-like substances are classified as refractory due to their potential aromaticity and resistance to degradation (Hertkorn et al. 2006). Across all reactors, lignin-like compounds were more prevalent than protein/amino sugar-like compounds, suggesting that the effluent DOM has a more refractory nature.

Overall, the relative abundance and molecular composition of effluent DOM were similar among the four reactors, and all effluents displayed a comparable refractory DOM profile. These results indicate that LFGC addition did not produce distinct fluorescent or molecular DOM signatures compared with the conventional carbon sources. However, EEM–PARAFAC and FTICR–MS provide compositional information and do not directly measure long-term persistence, biodegradation rate, or microbial degradability. Therefore, the results should be interpreted as evidence of comparable effluent DOM characteristics among carbon sources, rather than direct proof that all

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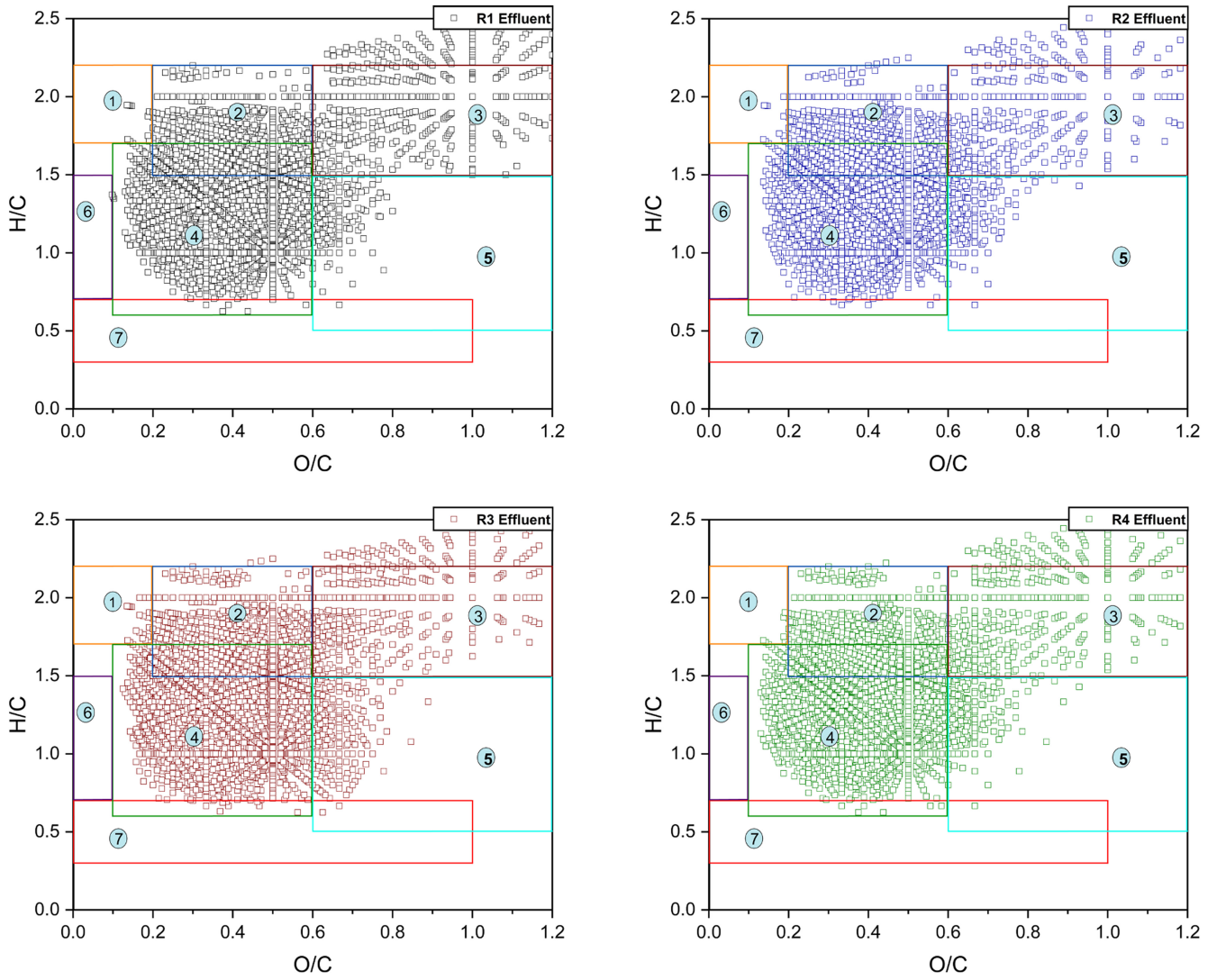


FIGURE 7 | The van Krevelen diagrams of effluent DOM for R1 (methanol), R2 (glycerin), R3 (sodium acetate), and R4 (LFGC). The labeled circles in each diagram represent seven distinct DOM molecular formulas: (1) lipids, (2) proteins/amino sugars, (3) carbohydrates, (4) lignin, (5) tannins, (6) unsaturated hydrocarbons, and (7) condensed aromatics (Hu et al. 2020; Zou et al. 2024).

TABLE 5 | FTICR–MS elemental ratios for different chemical compound groups (Hu et al. 2018).

DOM elements	O/C and H/C ratio
1. Lipids	O/C = 0–0.2, H/C = 1.7–2.2
2. Proteins/amino sugars	O/C = 0.2–0.6, H/C = 1.5–2.2
3. Carbohydrates	O/C = 0.6–1.2, H/C = 1.5–2.2
4. Lignin	O/C = 0.1–0.6, H/C = 0.6–1.7
5. Tannins	O/C = 0.6–1.2, H/C = 0.5–1.5
6. Unsaturated hydrocarbons	O/C = 0–0.1, H/C = 0.7–1.5
7. Condensed aromatics	O/C = 0–1, H/C = 0.3–0.7

residual organics are biodegradable or non-persistent. Future work should include biodegradation assays, respirometric tests, or long-term persistence studies to directly evaluate the microbial degradability and fate of residual LFGC-derived organic compounds.

4 | Conclusion

This study demonstrated that LFGC can serve as an effective external carbon source for denitrification in SBRs co-treating municipal sewage and landfill leachate. LFGC provided total nitrogen removal and effluent NO_3^-–N levels comparable to those achieved with methanol, glycerin, and sodium acetate. Although LFGC required a higher specific external COD dose than the conventional liquid carbon sources, it enabled a slightly shorter denitrification time and produced effluent DOM characteristics comparable to those of the conventional carbon-source reactors, as indicated by EEM–PARAFAC and FTICR–MS analyses. These findings indicate that LFGC is a technically viable and more sustainable denitrification carbon source that can offset purchased external carbon and reduce or eliminate the need for separate condensate treatment.

Future work should evaluate full-scale variability in LFGC composition, potential impacts on nitrous oxide emissions, long-term process stability under fluctuating loading and operating

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conditions, and the biodegradability and persistence of residual LFGC-derived organic compounds.

Author Contributions

Md. Ashik Ahmed: data curation, writing – original draft, visualization. **Brian Brazil:** resources. **Lifeng Zhang:** supervision. **Renzun Zhao:** conceptualization, methodology, validation, investigation, resources, writing – review and editing, supervision, project administration, funding acquisition.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Figure S1:** Effluent pH values over the 170 days study period for all four reactors. **Figure S2:** Influent and effluent COD values with the respective removal efficiencies for all the reactors over the 170-day study period. **Figure S3:** Influent and effluent BOD values with the respective removal efficiencies for all the reactors over the 170-day study period. **Figure S4:** Average influent and effluent NH_4^+-N values with the respective removal efficiencies for all the reactors over the 170-day study period. **Figure S5:** Nitrate values before and after denitrification over the study period with respective methanol dose for R1 (methanol reactor). **Figure S6:** Nitrate values before and after denitrification over the study period with respective glycerin dose for R2 (glycerin reactor). **Figure S7:** Nitrate values before and after denitrification over the study period with respective sodium acetate dose for R3 (sodium acetate reactor). **Figure S8:** Nitrate values before and after denitrification over the study period with respective condensate dose for R4 (condensate reactor). **Table S1:** Calculation of the specific COD dose of all carbon sources. **Table S2:** Detected peaks from EEM and their description. **Table S3:** Relative abundances of organic compounds from the Van Krevelen diagram for FTICR–MS analysis.

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