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Organics Recovery from Waste Activated Sludge *In-situ* Driving Efficient Nitrogen Removal from Mature Landfill Leachate: An Innovative Biotechnology with Energy Superiority

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ABSTRACT

The sustainable recovery and utilization of sludge bioenergy within a circular economy context has drawn increasing attention, but there is currently a shortage of reliable technology. This study presents an innovative biotechnology based on free nitrous acid (FNA) to realize sustainable organics recovery from waste activated sludge (WAS) *in-situ*, driving efficient nitrogen removal from ammonia rich mature landfill leachate by integrating partial nitrification, fermentation, and denitrification process (PN/DN–F/DN). First, ammonia ((1708.5 ± 142.9) mg·L^{-1}) in mature landfill leachate is oxidized to nitrite in the aerobic stage of a partial nitrification coupling denitrification sequencing batch reactor (PN/DN-SBR), with nitrite accumulation ratio of 95.4% ± 2.5%. Then, intermediate effluent (NO_2^-–N = (1196.9 ± 184.2) mg·L^{-1}) of the PN/DN-SBR, along with concentrated WAS (volatile solids (VSs) = (15 119.8 ± 2484.2) mg·L^{-1}), is fed into an anoxic reactor for fermentation coupling denitrification process (F/DN-SBR). FNA, the protonated form of nitrite, degrades organics in the WAS to the soluble fraction by the biocidal effect, and the released organics are utilized by denitrifiers to drive NO_x^- reduction. An ultra-fast sludge reduction rate of 4.89 kg·m^{-3}·d^{-1} and nitrogen removal rate of 0.46 kg·m^{-3}·d^{-1} were realized in the process. Finally, F/DN-SBR effluent containing organics is refluxed to PN/DN-SBR for secondary denitrification in the post anoxic stage. After 175 d operation, an average of 19 350.6 mg chemical oxygen demand organics were recovered per operational cycle, with 95.2% nitrogen removal and 53.4% sludge reduction. PN/DN–F/DN is of great significance for promoting a paradigm transformation from energy consumption to energy neutral, specifically, the total benefit in equivalent terms of energy was 291.8 kW·h·t^{-1} total solid.

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1. Introduction

Within the context of a circular economy, future wastewater treatment plants (WWTPs) operation should endeavor to be energy neutral and maximise resource recovery. Sustainability metrics (e.g., energy consumption, carbon emission, and resource recovery) have been integrated to evaluate the performance of WWTPs [1–4]. However, several critical drawbacks limit sustainable development within WWTP systems. For instance, efficient biological nitrogen removal from wastewater is hardly realized due to a lack of organic carbon sources. Additionally, large amounts of waste activated sludge (WAS) and related energy-intensive treatments account for up to 40%–60% of the total operating costs WWTPs [5].

WAS is the metabolic by-product formed by microbial assimilation of contaminants and organics in wastewater treatment process [6]. In China, the yield of WAS has increased significantly, with annual growth of 10.7%, and it is forecasted output will enhance to 103.0 million tonnes in 2025 (80% water content) [7]. Although WAS is a type of pollutant, it also contains rich bioenergy and has great resource utilization potential. In the context of environmental management evolving from waste elimination to valorizing, bioenergy and bioresource recovery from WAS is gaining increasing attention [7–9].

Methane generation from anaerobic digestion is traditionally considered as the sustainable approach to energy-efficient WAS treatment, and has the possibility of decreasing energy

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consumption in WWTPs [10]. To optimize efficient bioenergy recovery and improve sludge digestibility, various pretreatments were applied prior to the anaerobic digestion process, including thermal hydrolysis, ozone, ultrasound, alkali, acid, and mechanical forces [8,11–13]. However, in practice, the current so-called energy recovery technology for WAS treatment is essentially an energy exchange process, that is, the bioenergy contained in WAS is exchanged by inputting valuable physical, chemical, or electrical energy. Unfortunately, the output energy is often lower than that of the input, resulting in a significant deficit in the energy balance [14]. For example, although a pretreatment step of thermal hydrolysis could enhance the performance of anaerobically digested sludge in terms of economic, technical, and environmental benefits, there remains a 1470 $kW{\cdot}h{\cdot}t^{-1}$ total solid (TS) energy deficit [15]. Similarly, Hao et al. also confirmed an energy deficit in the thermal hydrolysis coupling anaerobic digestion (THP-AD) sludge treatment process reached up to 324 $kW{\cdot}h{\cdot}t^{-1}$ TS [16]. Therefore, finding a sustainable and cost-efficient procedure for WAS treatment is one of the most challenging topics, and turn even more significant in the upcoming years.

In addition to methane collection for energy generation, bioresource recovery of inexpensive organic carbon (e.g., carbohydrate, proteins, volatile fatty acids, and lipid, etc.) from WAS also appears to be a promising valorization pathway [17–19]. This approach will not only achieve sludge management (reduction and stabilization), but also efficiently solve the problem of organic carbon source deficit in wastewater biological treatment (resource utilization), facilitating advanced nitrogen removal. Nevertheless, sustainable technology for recovering organics from sludge is currently lacking, and limited studies pay attention to effective combination pathway for wastewater and WAS treatment.

Free nitrous acid (FNA), as the protonated form of NO_2^-–N, can inhibit microbial metabolic activities, including tricarboxylic acid cycle, adenosine triphosphate (ATP) generation, and substrate transport across the cell membrane, then promote short chain volatile fatty acids (SCFAs) release from WAS [20–22]. A previous study demonstrated a 15%–21% enhancement in fermentative efficiency in WAS pretreated with 0.2 $mg{\cdot}L^{-1}$ FNA [23]; while Pijuan et al. [20] observed a 20% decrease in viable microorganisms when sludge was exposed to FNA (2.02 $mg{\cdot}L^{-1}$) for 48 hours. In another application, SCFA concentration increased substantially (720.0–1423.6 $mg{\cdot}L^{-1}$ chemical oxygen demand (COD)) when FNA increased from 0.197 to 0.394 $mg{\cdot}L^{-1}$, followed by a denitrification rate ranging between 0.0875 and 0.1750 $kg{\cdot}m^{-3}{\cdot}d^{-1}$ [24]. In the process, heterotrophic microorganisms in situ capture released organics from sludge, driving denitrification. Importantly, FNA is a renewable green substrate that can be obtained through partial nitrification. Using FNA-based biotechnology for wastewater and WAS treatment is of great significance for promoting a paradigm transformation from energy consumption to energy neutrality in WWTPs. The advantages of this approach include: ① no energy inputting pretreatment is required to realize sludge stabilization and the recovery of organic resources; ② provides a new idea for the sustainable simultaneous treatment of wastewater and WAS in a single system; and ③ organic matter dosing in the biological nitrogen removal process.

In this study, an innovative biotechnology was developed to realize efficient recovery of organics from WAS and advanced nitrogen removal from mature landfill leachate through integrating partial nitrification, fermentation, and denitrification processes (PN/DN–F/DN). The aims of this work were to as follows: ① achieve simultaneous treatment of mature landfill leachate and WAS; ② determine the variation in recovered organics during long-term operation; ③ track the dynamics of dominant organics and the transformation of size-fractionated organics; and ④ analyze the energy balance and environmental benefits.

2. Materials and methods

2.1. System set-up and operational strategy

The PN/DN–F/DN system was continuously operated for 175 d in two sequencing batch reactors (SBRs) (Fig. 1(a)). Firstly, mature landfill leachate (5.0–7.0 L) was introduced to the partial nitrification coupling denitrification reactor (PN/DN-SBR; working volume of 10.0 L), which operated under aerobic/anoxic mode. In a typical cycle, partial nitrification process first proceeded in the aerobic stage and the aeration was stopped once "ammonia valley" emerges in the pH profile [25]. Next, half of the PN/DN-SBR intermediate effluent coupled with external concentrated WAS (2.0 L) was fed to the anoxic reactor (working volume of 8.0 L) for fermentation coupling denitrification process (F/DN-SBR) (Fig. 1(b)). F/DN-SBR reaction duration was set to be half that of the PN/DN-SBR. Finally, organics-contained effluent from two cycles of F/DN-SBR were refluxed to PN/DN-SBR for denitrification process in the post anoxic stage (Fig. 1(c)).

According to the operational strategies, four phases occurred during long-term operation. In phase I (days 1–25), neither reflux nor the post anoxic stage was applied in PN/DN-SBR, which only performed the partial nitrification process (Fig. 1(b)). The volume exchange ratio (VER) of PN/DN-SBR and F/DN-SBR were initially set at 50.00% and 31.25%, respectively, with the sludge dosing ratio (SDR) of 25% for F/DN-SBR (Table 1). In phase II (days 26–65), organics contained effluent from F/DN-SBR were refluxed to PN/DN-SBR for denitrification in the post anoxic stage (Fig. 1(c)). In phase III (days 66–90), the organics recovery in F/DN-SBR was strengthened by prolonging the hydraulic detention time (HRT) (from 38.5 to 76.8 h) (Table 1). In phase IV (days 91–175), the VER of PN/DN-SBR and F/DN-SBR were adjusted to 70.00% and 43.75%, respectively, followed by a gradual shortening of the F/DN-SBR HTR (from 76.8 to 41.2 h). Throughout the operating period, the temperature was maintained at (27 ± 3) °C.

2.2. Wastewater and biomass

Typical mature landfill leachate was collected from San He urban solid waste landfill site (Lang Fang, China) once a week, which has been in stable operation for more than 20 years. The characteristics are as follows: (1708.5 ± 219.4) $mg{\cdot}L^{-1}$ NH_4^+–N, (2370.2 ± 433.1) $mg{\cdot}L^{-1}$ total nitrogen (TN), (2286.3 ± 592.8) $mg{\cdot}L^{-1}$ COD, (205.1 ± 64.9) $mg{\cdot}L^{-1}$ BOD_5, and (87 420 ± 2470.3) mg $CaCO_3$ per liter of alkalinity. The PN/DN-SBR and F/DN-SBR were established in our previous study, which has been operated for more than 300 d [26]. Concentrated WAS collected from the secondary sedimentation tank of Yanjiao sewage treatment plant had average mixed liquor TSs of 23 713.1 $mg{\cdot}L^{-1}$ and mixed liquid volatile solids (VS) of 15 176.4 $mg{\cdot}L^{-1}$, respectively.

2.3. Calculations

2.3.1. Hydraulic detention time

The HRT of aerobic/anoxic stages in PN/DN-SBR and the F/DN-SBR were calculated according to Eq. (1):

$$\mathrm{HRT}\,(\mathrm{h}) = t \times \frac{1}{\mathrm{VER}} \tag{1}$$

where VER is the volume exchange ratio of reactors (%) and t is the actual reaction time (h).

2.3.2. Recovery of organics resource from WAS

Stoichiometric equations of denitrification (NO_3^-–N → N_2 and NO_2^-–N → N_2) were used to evaluate the consumed organics, by the stoichiometry of $\Delta COD/\Delta NO_3^-$–N = 2.86 and

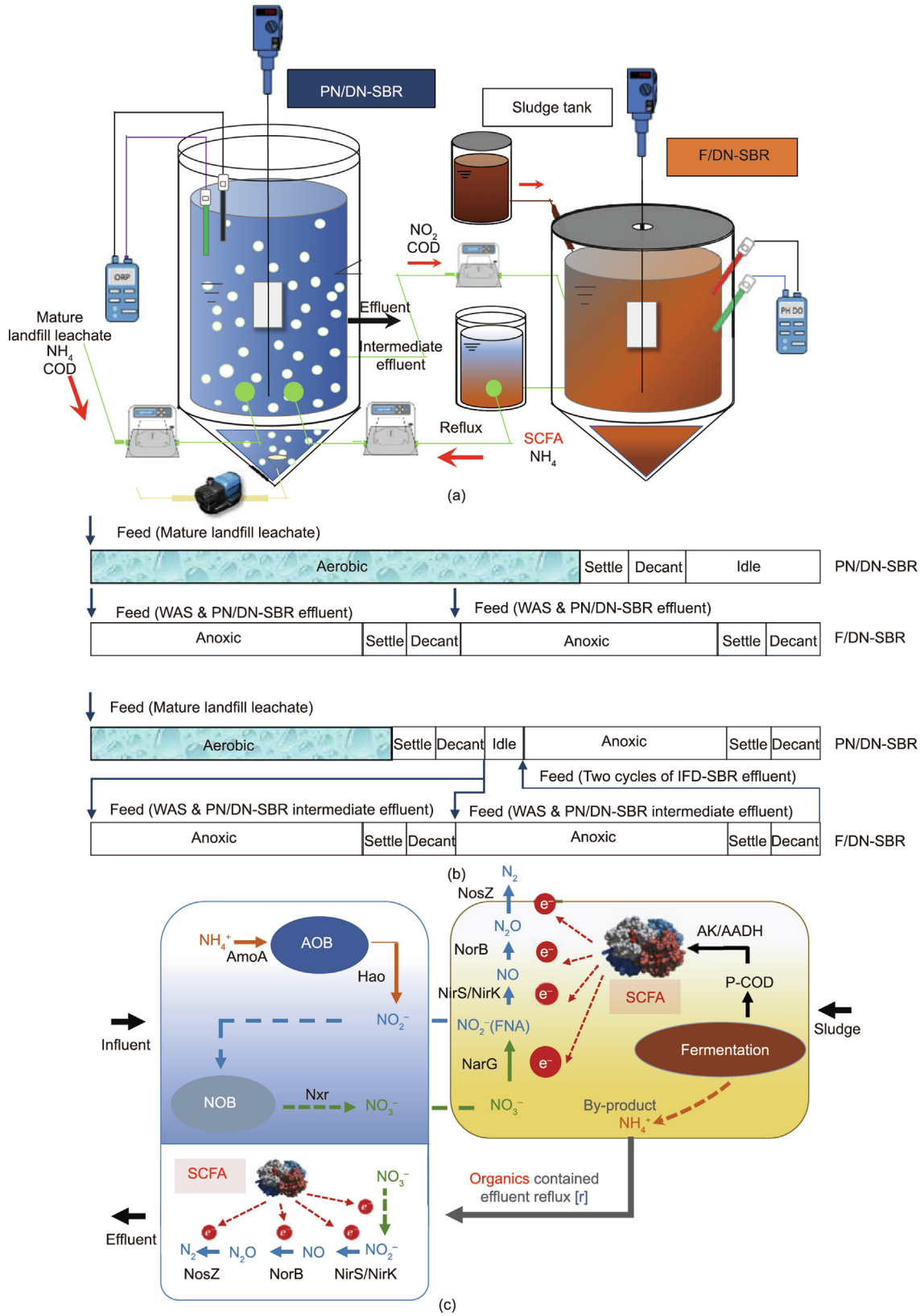

Fig. 1. (a) Experiment device, (b) operational strategies, and (c) schematic diagram of PN/DN–F/DN system for simultaneous treatment of mature landfill leachate and WAS.

$\Delta COD/\Delta NO_2^-\text{–}N = 1.71$ [27]. To determine the mechanism of organics recovery from WAS, consumed organics and the variation in COD between influent and effluent were comprehensively evaluated by Eqs. (2)–(4):

Organics consumed by denitrification in F/DN-SBR ($M_{F/DN}$):

$$M_{F/DN} = (NO_2^- - N_{inf-F/DN} - NO_2^- - N_{eff-F/DN}) \times V_{inf} \times 1.71 + (NO_3^- - N_{inf-F/DN} - NO_3^- - N_{eff-F/DN}) \times V_{inf} \times 2.86 \quad (2)$$

Table 1
Detailed operational procedure of PN/DN–F/DN system over 175 d.

Phase	Days	VER (%)		SDR (%)	HRT (h)			VS[c] ($mg \cdot L^{-1}$)
		PN/DN-SBR	F/DN-SBR	F/DN-SBR	PN/DN-SBR$_O$[a]	PN/DN-SBR$_A$[b]	F/DN-SBR	
I	1–25	50	31.25	25	65	–	38.5	15 176.4
II	26–65	50	31.25		65	20	38.5	15 012.1
III	66–90	50	31.25		65	20	76.8	13 954.6
IV	91–115	70	43.75		62	10	55.0	16 283.5
	116–130				62	10	46.0	14 069.7
	131–175				62	10	41.2	15 369.2

[a] PN/DN-SBR$_O$ represents the aerobic stage of PN-SBR.
[b] PN/DN-SBR$_A$ represents the post anoxic stage of PN/DN-SBR.
[c] VS represents the mixed liquid volatile solid (VS) of additional WAS.

Organics consumed by denitrification in PN/DN-SBR post anoxic stage ($M_{PN/DN}$):

$$M_{PN/DN} = [NO_2^- - N_{O-PN/DN} \times (1 - VER) - NO_2^- - N_{eff-PN/DN}] \times V \times 1.71 + [NO_3^- - N_{O-PN/DN} \times (1 - VER) - NO_3^- - N_{eff-PN/DN}] \times V \times 2.86 \quad (3)$$

Organics recovered in the PN/DN–F/DN process ($M_{PN/DN-F/DN}$):

$$M_{PN/DN-F/DN} = M_{PN/DN} + M_{F/DN} + (COD_{eff} - COD_{inf}) \times V_{inf} \quad (4)$$

where NO_2^-–$N_{inf-F/DN}$ and NO_2^-–$N_{eff-F/DN}$ refer to the NO_2^-–N concentration ($mg \cdot L^{-1}$) in influent and effluent of F/DN-SBR, respectively; NO_3^-–$N_{inf-F/DN}$ and NO_3^-–$N_{eff-F/DN}$ are NO_3^-–N concentration ($mg \cdot L^{-1}$) in influent and effluent of F/DN-SBR, respectively; NO_2^-–$N_{O-PN/DN}$ and NO_2^-–$N_{eff-PN/DN}$ are NO_2^-–N concentration ($mg \cdot L^{-1}$) at the end of the aerobic stage and in effluent in PN/DN-SBR, respectively; NO_3^-–$N_{O-PN/DN}$ and NO_3^-–$N_{eff-PN/DN}$ are NO_3^-–N concentration ($mg \cdot L^{-1}$) at the end of aerobic stage and in effluent in PN/DN-SBR, respectively; COD_{inf} and COD_{eff} represent the COD concentration ($mg \cdot L^{-1}$) in the influent and effluent, respectively; and V_{inf} is the influent volume (L) and V is the working volume of PN/DN-SBR, respectively.

2.3.3. FA and FNA

The concentrations of FA and FNA were calculated according to Eqs. (5) and (6):

$$FA\,(mg \cdot L^{-1}) = \frac{C_{NH_4^+-N} \times 10^{pH}}{\exp\left(\frac{6334}{273+T}\right) + 10^{pH}} \quad (5)$$

$$FNA\,(mg \cdot L^{-1}) = \frac{C_{NO_2^--N}}{\exp\left(-\frac{2300}{273+T}\right) \times 10^{pH}} \quad (6)$$

$C_{NH_4^+-N}$ and $C_{NO_2^--N}$ are the concentrations of NH_4^+–N and NO_2^-–N ($mg \cdot L^{-1}$), respectively; temperature stand for T (°C).

2.4. COD fractionation

Organic matter contains subvisible colloidal and particle fractions, which include soluble organics (S-COD), small colloidal organics (SC-COD), large colloidal organics (LC-COD), and particle organics (P-COD). Colloidal and particulate fractions could be gradually degraded to soluble fraction in WAS treatment process. To gain a better understanding of the compositional transformation of complex organics, exploration of different size-fractionated organics was carried out following the method described by [28].

The wastewater firstly passed through 12–25 μm paper filter and measured as the toal organics (T-COD). Then it was passed through a 1.5 μm glass-fiber filter and 0.45 μm nylon membrane filters. The concentration of P-COD was the difference between filtered COD that passed through 1.5 μm filter and T-COD. The concentration of LC-COD was the difference between filtered COD that passed trough 1.5 μm filter and 0.45 μm filter. The S-COD fraction was analyzed by the flocculation-filtration ($ZnSO_4$ as flocculant, 0.45 μm filtration) method [29]. SC-COD was the difference between the S-COD and filtered COD that passed trough 0.45 μm filter.

2.5. Energy balance

To compare the energy balance analysis of THP-AD and PN/DN–F/DN, input and output energy of individual treatment units were calculated. The output energy of THP-AD and PN/DN–F/DN were determined by the generated methane and the recovered organics, respectively. For this, dry sludge of 1 tonne TS was considered as functional unit, and the parameters for energy balance analysis are presented in Table 2 [14,15,30,31]. To ensure a systematic comparison the energy balance between these two cases, the output production moisture content was set at 55%.

2.6. Analytical methods

Samples were collected daily and filtered through 0.45 um disposable millipore filters before analysis. The concentrations of

Table 2
Parameters for energy balance analysis.

Serial number	Items	Value	Unit
1	VS/TS	54[a]	%
2	THP steam demand (12 bar[b]; TS)	860	$kW \cdot h \cdot t^{-1}$
3	Specific enthalpy of saturated steam (12 bar)	2785 [15]	$kJ \cdot kg^{-1}$
4	Average net methane production (CH_4/VS)	0.2439 [14]	$m^3 \cdot kg^{-1}$
5	Methane calorific value	35.6 [15]	$MJ \cdot m^{-3}$
6	Thermal loss in methane burning	7 [30]	%
7	Energy generation associated CO_2 emission	1.05 [31]	$kg \cdot (kW \cdot h)^{-1}$
8	Power price	0.852 [14]	$CNY \cdot (kW \cdot h)^{-1}$
9	Thermoelectric conversion coefficient	1 kW·h	–
10	Price of industrial grade sodium acetate	4200	$CNY \cdot t^{-1}$
11	Purity of industrial grade sodium acetate	50	%
12	Conversion coefficient between sodium acetate and COD	1 g CH_3COONa = 0.78 g COD	–

TS represents the mixed liquid total solids; VS represents the mixed liquid volatile solids.
[a] Mean of 20 sewage plants measured value.
[b] 1 bar = 1 × 10^5 Pa.

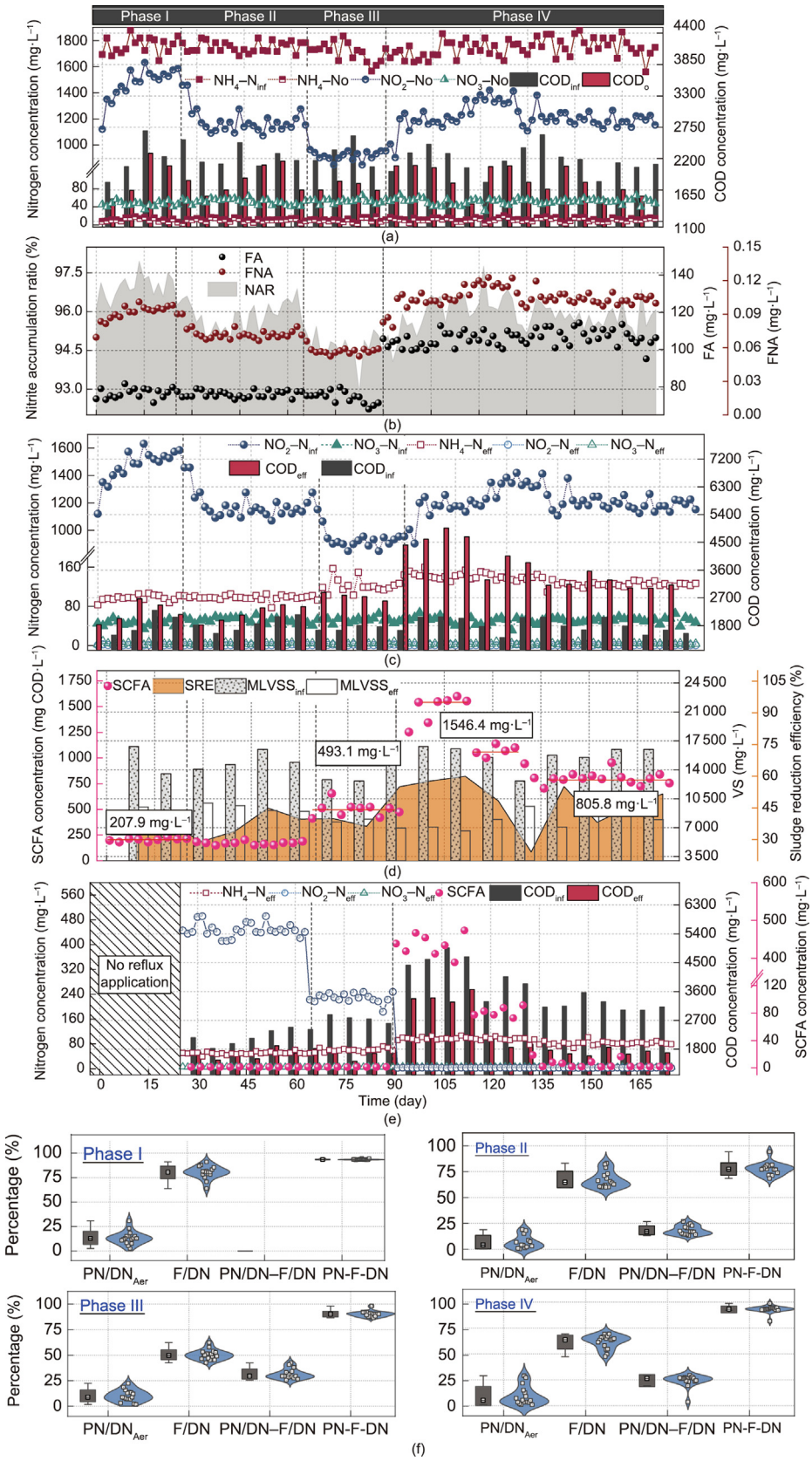

Fig. 2. Long term performance of PN/DN–F/DN system for 175 d. (a) Influent NH_4^+–N and COD, as well as NH_4^+–N, NO_2^-–N, NO_3^-–N, and COD concentrations at the end of PN/DN-SBR aerobic stage (intermediate effluent). (b) Variation free acid concentration at initial of PN/DN-SBR aerobic stage and NAR at the end of PN/DN-SBR aerobic stage, as well as free nitrous acid concentration at initial operation cycle of F/DN-SBR. (c) Influent NO_2^-–N and COD, as well as NH_4^+–N, NO_2^-–N, NO_3^-–N, and COD concentrations in the effluent of F/DN-SBR. (d) Variation of VS, SRE, and the SCFA in F/DN-SBR. (e) Influent COD, as well as NH_4^+–N, NO_2^-–N, NO_3^-–N, COD, and SCFA concentrations in the effluent of PN/DN-SBR. (f) Nitrogen removal efficiency of PN/DN–F/DN, nitrogen removal contribution in F/DN-SBR, and aerobic/anoxic stage of PN/DN-SBR (PN/DN_{Aer} and PN/DN_{Ano}).

NH_4^+–N, NO_2^-–N, NO_3^-–N, S^{2-}, TS, and VS were measured by standard methods. A gas chromatograph (GC; Agilent 7890, USA) was used to analyze the SCFA. A pH/Oxi 3420 analyzer (WTW company, Germany) was used to monitored pH and dissolved oxygen. A fluorescence spectrometer (Hitachi F-7100, Japan) was used to conduct excitation-emission matrix (EEM) measurements.

3. Results and discussion

3.1. Long-term simultaneous treatment of mature landfill leachate and WAS

Long term performance of PN/DN–F/DN system is shown in Fig. 2. Mature landfill leachate was fed to PN/DN-SBR with NH_4^+–N concentration of 1565.5–1851.4 mg·L^{-1} (Fig. 2(a)). Throughout the experiment, nitrite accumulation ratio (NAR) ranged from 92.9% to 97.8% (Fig. 2(b)), and the average NO_3^-–N concentration of 52.1 mg·L^{-1} in intermediate effluent. Notably, satisfied partial nitrification was realized by real-time control parameter application and high free acid concentrations (68.1–112.4 mg·L^{-1}) (Fig. 2(b)). Owing to the presence of various refractory and/or non-biodegradable organic matter contained in mature landfill leachate [32], there was a slight decrease in COD, with effluent concentrations of 1542.5–2290.8 mg·L^{-1} (Fig. 2(a)).

In phase I, concentrated external WAS (2.0 L; VS = 13 539.7–16 813.1 mg·L^{-1}) coupled with nitrite rich PN/DN-SBR effluent (1483.8–1630.7 mg·L^{-1}) was introduced to F/DN-SBR for fermentation coupling denitrification process. FNA, the protonated form of NO_2^-–N, can inhibit microbial metabolic activities, including tricarboxylic acid cycle, ATP generation, and substrate transport across the cell membrane, and then promote WAS fermentation process and release of organics [20–22]. The average FNA concentration was 0.09 mg·L^{-1} at initiation of F/DN-SBR (Fig. 2(b)), and the strong biocidal effect facilitates fermentative efficiency. A 37.3%–43.5% reduction in sludge was obtained with 5056.6–7314.1 mg·L^{-1} VS consumption in phase I (Fig. 2(d)). Different from "energy input" pretreatment (e.g., THP, ozone, ultrasound, alkali, acid, and mechanical forces), in combination with anaerobic digestion for WAS treatment, FNA based biotechnology can realize efficient WAS disposal without pretreatment. Importantly, FNA is a renewable green substrate that can be obtained through partial nitrification. The organics released from fermentation process were subsequently used as an efficient alternative electronic donor for the denitrification process, replenishing deficient organics for biological nitrogen removal (NO_x^-–N → N_2). Concentrations of NO_2^-–N and NO_3^-–N in F/DN-SBR effluent were 1.7 and 3.2 mg·L^{-1} (Fig. 2(c)), respectively, indicating effective nitrogen removal performance was obtained without external carbon source addition. Significantly, the effluent of F/DN-SBR contained 2163.4 mg·L^{-1} COD and 207.9 mg·L^{-1} SCFA. As is, if the F/DN-SBR effluent is directly discharged, it will not only waste valuable organic resources, but also cause adverse impacts on the following treatment and the environment [33].

To fully utilize the recovered organics, F/DN-SBR effluent was refluxed to PN/DN-SBR in phase II (days 26–65), and then the post anoxic stage was carried out for secondary denitrification (Fig. 1(b)). Concentrations of SCFA and COD subsequently dropped to 1.3 mg·L^{-1} and 1639.4 mg·L^{-1} in the effluent of PN/DN-SBR (Fig. 2(e)). Most biodegradable organics were consumed efficiently, while the nitrogen removal performance was restricted when residual organics contained in F/DN-SBR effluent were further used as electron donors for denitrification in the post anoxic stage of PN/DN-SBR. As a result, 440.4 mg·L^{-1} NO_2^-–N and 4.1 mg·L^{-1} NO_3^-–N were present in the effluent of PN/DN-SBR (Fig. 2(e)), with only 17.9% of nitrogen removal contribution in the post anoxic stage of PN/DN-SBR (Fig. 2(f)). This indicates an incomplete denitrification process. Consequently, in phase III (days 66–90), a strategy of prolonging F/DN-SBR duration was applied to strengthen the fermentation process and supply more organics for denitrification (Table 1). This method contributed to the SCFA peak of 493.1 mg·L^{-1} in the effluent of F/DN-SBR (Fig. 2(d)), followed by a drop in NO_2^-–N to 221.9 mg·L^{-1} in the effluent of PN/DN-SBR (Fig. 2(e)). A previous study reported that SCFA concentrations can be enhanced to 720 mg·L^{-1} when the reaction duration is increased from 30 to 70 h, while a further increase in the reaction period did not lead to a noticeable SCFA increase [24]. As the high-quality organics and intermediate of WAS fermentation, SCFA plays a decisive role in denitrification performance. With deficient organics, NO_x^-–N cannot be completely reduced to N_2. Therefore, a further optimization strategy is required to remove overshooting NO_2^-–N in the PN/DN-SBR effluent.

In phase IV (days 91–175), VER of PN/DN-SBR and F/DN-SBR were adjusted to 70.00% and 43.75%, respectively, to promote the performance: one elevated the initial FNA concentration in F/DN-SBR typical cycle to enhance WAS fermentative efficiency and organics recovery, and the other alleviated NO_x^-–N removal requirements in the post anoxic stage of PN/DN-SBR. Thereafter, an advanced sludge reduction efficiency of 53.4% was obtained, with the initial FNA increasing to 0.105–0.120 mg·L^{-1} in phase IV (Fig. 2(b)). Along with promotion of WAS fermentation, denitrification in F/DN-SBR and post anoxic stage of PN/DN-SBR was correspondingly favored, with 79.8 mg·L^{-1} NH_4^+–N, 2.5 mg·L^{-1} NO_2^-–N, and 5.3 mg·L^{-1} NO_3^-–N in effluent (Fig. 2(e)), and nitrogen removal efficiency progressively increasing up to 95.2% (Fig. 2(f)). This innovative PN/DN–F/DN biotechnology realized efficient WAS reduction, with simultaneous recovery of organics and in-situ driving nitrogen removal from mature landfill leachate. This method is of great significance for promoting paradigm transformation from energy consumption to energy neutrality in practical applications.

3.2. Investigation of an energy-efficient contaminant removal mechanism

A typical operation cycle was investigated (day 163) to evaluate the nutrient transformation mechanism of the PN/DN–F/DN system (Fig. 3). In the aerobic stage of PN/DN-SBR, NH_4^+–N decreased from 1231.4 to 11.8 mg·L^{-1}, accompanied by alkalinity consumption and pH decrease (Fig. 3(a)). The "Ammonia valley" emerging in the pH profile indicated completion of partial nitrification, and aeration is supposed to stop to avoid excess aeration [25]. Owing to the coexistence of nitrite oxidizing bacteria (NOB), poor partial nitrification performance is a common issue [34]. A real-time control strategy effectively solved the problem of NOB over proliferation, with nitrite accumulation of 95.4%.

In the F/DN-SBR, the biocidal effect of FNA facilitated gradual degradation of organics contained in WAS to soluble fraction in the fermentation process, and the released organics could be captured by heterotrophic microorganisms, thereby driving denitrification. As expected, NO_2^-–N decreased from 523.3 to 1.1 mg·L^{-1} within the initial 12 h (Fig. 3(c)). During the process, an ultra-fast sludge reduction rate of 4.89 kg·m^{-3}·d^{-1} and a nitrogen removal rate of 0.46 kg·m^{-3}·d^{-1} were obtained, accompanied by vigorous nitrogen gas spilling out (Fig. 3(d)). In this period, the SCFA generation rate was slower than that consumed by heterotrophic denitrifiers, leading to a constant SCFA concentration. As reported earlier, 2.86 and 1.71 g COD would be consumed to reduce 1 g NO_3^-–N and 1 g NO_2^-–N in the denitrification process [27]. In the denitrification process, 1 4287.4 mg COD organics were consumed. The breakpoint of "nitrite apex" in the pH profile denoted complete NO_x^-–N depletion [26]. The WAS fermentation

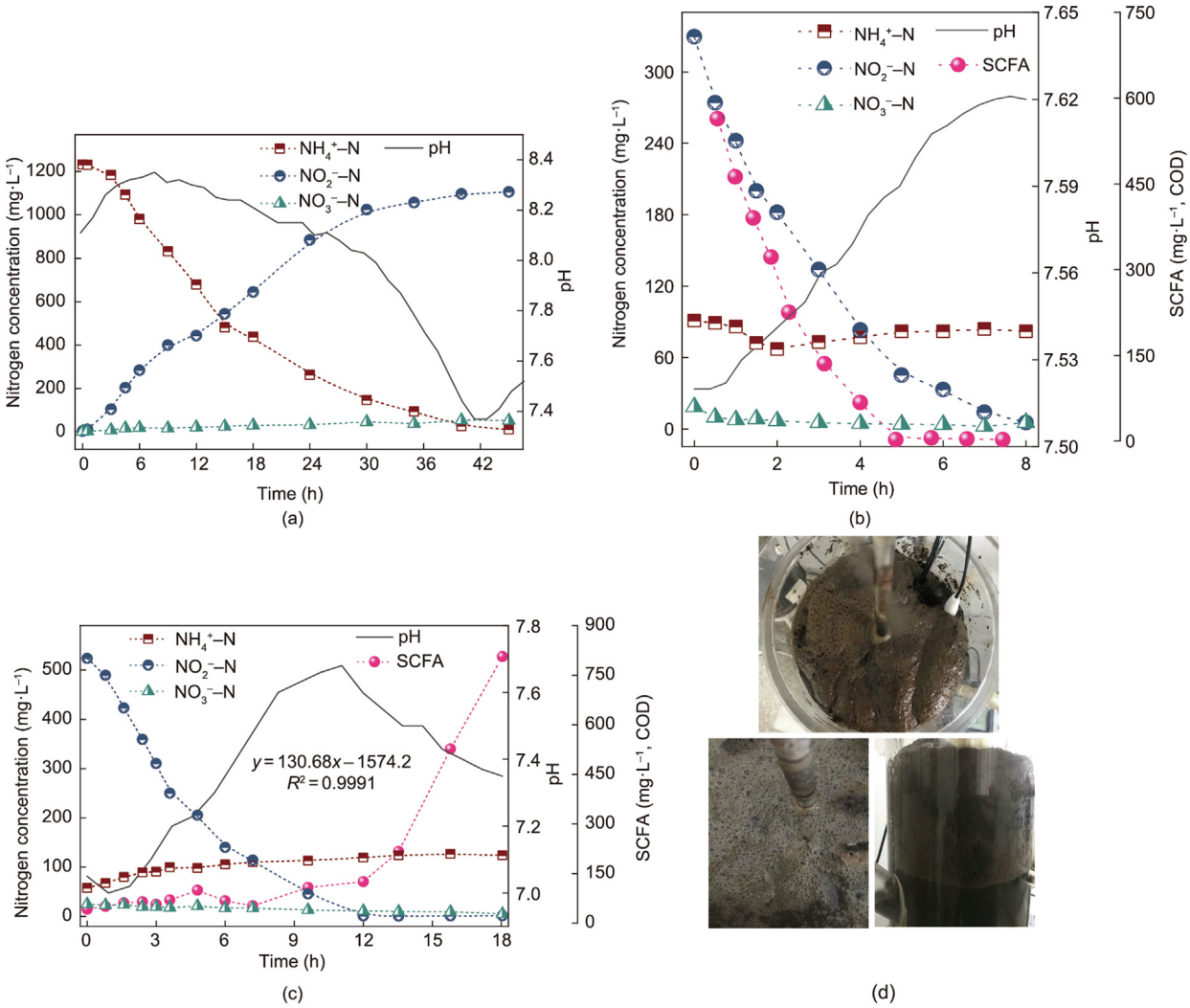

Fig. 3. (a) Variation of compounds transformation in the aerobic stage. Anoxic stage of (b) PN/DN-SBR and (c) F/DN-SBR. (d) Actual diagram of vigorous nitrogen gas spilling in F/DN-SBR.

proceeded continuously after 12 h, accompanied by an increase in SCFA to 805.8 mg·L^{-1} (Fig. 3(c)).

In the post anoxic stage of PN/DN-SBR, there was a significant decrease in NO_2^-–N (Fig. 3(b)). Organic matter generated from F/DN-SBR was used as a reliable electron donors since there were almost no biodegradable organics left after PN/DN-SBR aerobic stage. Coupled with the denitrification process, SCFA correspondingly decreased from 546.1 to 2.4 mg·L^{-1} (Fig. 3(b)). According to the calculation, organics consumed in the PN/DN-SBR post anoxic stage was 5543.8 mg COD. Alkalinity was produced by the denitrification process and the completion of denitrification was represented by a "nitrite apex" in the pH profile [25]. Owing to the efficient effect of organics recovery, nitrogen removal contributions were 63.7% for F/DN-SBR and 26.6% for PN/DN-SBR post anoxic stage (Fig. 2(f)). The established PN/DN–F/DN process achieved a 95.2% nitrogen removal efficiency, a 53.4% reduction in external WAS, and saved 100% additional external organics for biological nitrogen removal.

3.3. Analysis of organics recovery

Sufficient organics is the prerequisite for advanced nitrogen removal, but the refractory organics contained in mature landfill leachate cannot be utilized by nutrient removal microorganisms [32]. Consequently, nitrogen removal entirely depends on the recovery of organics from WAS. Due to the bactericidal effect of FNA at the ppm level, microbial metabolic activities (e.g., tricarboxylic acid cycle, ATP generation, and substrates transport across cell membrane) are inhibited, and particulate organics contained in sludge are released to the aqueous phase. These released organics subsequently serve as preferred electron donors for the denitrification process (NO_x^-–N → N_2). To reveal the mechanism of organics recovery by PN/DN–F/DN, the variation of COD between influent and effluent, as well as the consumed COD by denitrification were explored during the long-term operation.

As shown in Fig. 4, an average of 10 084.7 mg COD organics were recovered in phase I, with a recovered organics per gram VS reduction of 0.59 g COD per gram VS. In phase II, the concentration of recovered organics was similar to that of phase I, ranging between 8597.3 and 12 752.6 mg COD (Fig. 4(a)). With the extension HRT of F/DN-SBR (Table 1), recovered organics correspondingly increased to 1 4881.2 mg COD in phase III. This was comparable to previously published increases in SCFA of 21.1–871.6 mg·L^{-1} with nonlinear fitting (y = 62.023x – 611.82, R^2 = 0.997) after complete depletion of NO_x^-–N [35]. In phase IV, recovered organics reached 28 726.1 mg COD in days 91–115, with FNA concentration of 0.105 mg·L^{-1} and HRT of 55.0 h. However, there is 435.5 mg·L^{-1} SCFA resided in the effluent of PN/DN-SBR

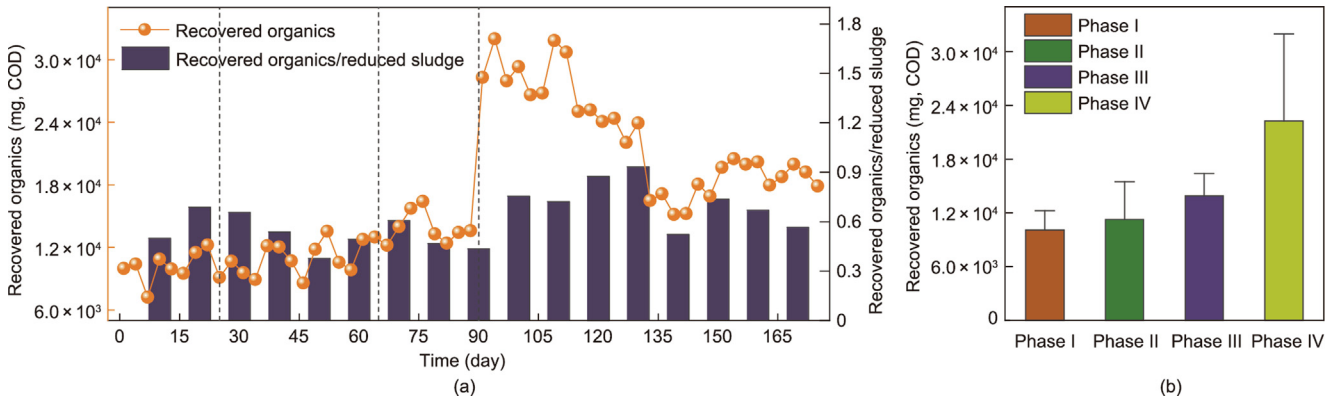

Fig. 4. (a) Variation of recovered organics and the ratio of recovered organics to reduced volatile solids. (b) Box diagram of recovered organics in different operational phases.

(Fig. 2(d)). Nitrogen removal performance of this innovative system mainly depended on the denitrification process. As a heterotrophic process, an optimal C/N ratio is a significant factor. With deficient organics, NO_x^-–N cannot be completely reduced to N_2, and excessive organics would result in a substantial environmental risks that need to be further disposed of. By optimizing the operational strategy, high-efficiency denitrification performance was finally realized, with HRT of 41.2 h (days 131–175) and at this time, the SCFA in effluent was almost completely consumed. Significantly, the average amount of recovered organics was 19 350.6 mg COD in the stable period of phase IV from day 131 to 175 (FNA = 0.105 mg·L^{-1}, HRT = 41.2 h), which is 1.8 times greater than that of phase II (FNA = 0.07 mg·L^{-1}, HRT = 38.5 d). This indicates that FNA efficiently enhances the recovery of organics from WAS (Fig. 4(b)). It is reasonable to indicate that, regardless of the reaction duration, FNA is the most significant factor to facilitate WAS fermentation and biochemical processes involved in organics transformation.

3.4. Size-fractionated organics transformation

As shown in Fig. 5, discrepancies of size-fractionated organics transformation affinities were observed in the process. The proportion of P-COD in intermediate effluent of PN/DN-SBR was unexpectedly lower than that in the influent. This finding agrees with a previous study that large molecular organic carbon sources are more easily degraded in aerobic conditions and are utilized to synthesize extracellular polymeric substances (EPSs) [36,37]. In contrast, small-size organics were preferentially utilized by denitrifiers in the post anoxic stage of PN/DN-SBR, leading to the proportions of SC-COD and S-COD being lower than those in F/DN-SBR effluent from phase II to phase IV (Fig. 5).

Recovered organics were derived from the sludge fermentation process. After undergoing series of steps (e.g., solubilization, hydrolysis, acidification, acetogenesis, and methanogenesis), the large particle solid organic matter contained in sludge is gradually degraded into small molecular organics. By FNA enhancement, concentrations of organics in F/DN-SBR effluent were correspondingly increased, with the highest increase in S-COD from 2006.7 mg·L^{-1} (phase III) to 2284.9 mg·L^{-1} (phase IV) (Table 3). Furthermore, the generation of small-size organics also benefitted from the longer SRT, as the proportions of SC-COD (23.2%) and S-COD (73.1%) in phase III were higher than those in phase II (SC-COD of 17.4% and S-COD of 69.2%) (Fig. 5). This indicates that the degradation of particles and colloids was promoted, thus realizing the recovery of organics and the provision of preferred electron donors for the denitrification process.

3.5. EEM revealing fluorescent organics transformation

To gain a better understaning of resource recovery and utilization in PN/DN–F/DN, organics transformation was also investigated using EEM fluorescence spectroscopy on day 168. Organics characterized as humic-like substances, protein and tyrosine-like products (microbial by-products), and fulvic acid-like substances were readily identified in intermediate effluent of PN/DN-SBR, effluent of F/DN-SBR, and effluent of PN/DN-SBR. The relative changes in the concentration of each component and their transformations were accurately determined via EEM fluorescence spectroscopy (Fig. 6).

Comparing the organics between PN/DN-SBR intermediate effluent and F/DN-SBR effluent, four dominant peaks were detected at much stronger fluorescence intensities in the F/DN-SBR effluent (Fig. 6). As a significant component of WAS, tyrosine/tryptophan (peak A) can potentially promotes SCFA generation and could be converted to SCFA in the fermentation liquid [38]. This agrees with the higher concentration of organics in F/DN-SBR effluent (Fig. 2(c)). Humic acids (peaks C and D) are generally produced from dead organic matter biodegradation, which is recognized as the principal component of humic substances [39]. The degradation degree of tyrosine/tryptophan (peak A) and tyrosine/tryptophan protein (peak B) were observed in the post anoxic stage of PN/DN-SBR. This finding concurs with a previous study in that the tyrosine-like component was easily utilized by heterotrophic denitrifiers, thus contributing to the efficient nitrogen removal performance [40]. In the treatment process, intracellular and extracellular constituents in WAS can be released via sludge floc disintegration, and these constituents can be further degraded to biodegradable organics [41]. Once released, these compounds stimulate heterotrophic denitrification. Consequently, FNA has a crucial role in the recovery of organics from WAS without energy input, providing preferred electron donors for denitrification and promoting an efficient nitrogen removal performance.

3.6. Energy balance and carbon footprint analysis

To comprehensively understand the energy balance of THP-AD and PN/DN–F/DN for sludge treatment, a systematic analysis was conducted by considering both indirect and direct environmental impacts. In addition to the bioenergy generated in form of methane and organics, energy consumed for heating, pumping, reagent addition, and mechanical mixing were also calculated in the energy balance and carbon footprint (Fig. 7). In this analysis, a functional unit of dry sludge containing 1 t TS was considered, and the parameters for the energy balance analysis are presented in Table 2.

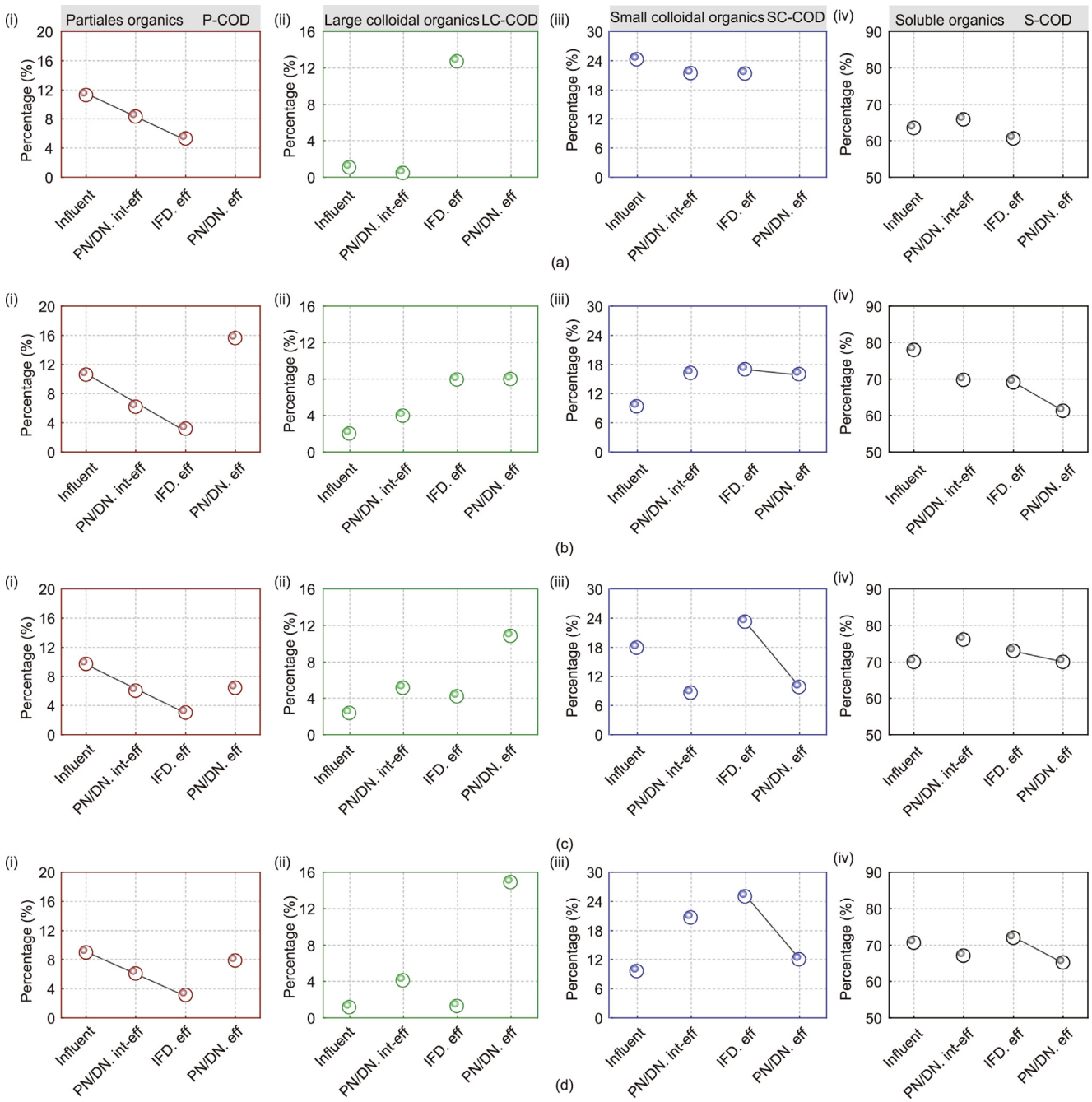

Fig. 5. Evolution of COD fractions in the mature landfill leachate (Influent), PN/DN-SBR intermediate effluent (PN/DN. int–eff), F/DN-SBR effluent (F/DN. eff) and PN/DN-SBR effluent (PN/DN. eff). Note: a, b, c, and d represented variation of COD fractions in phase I, phase II, phase III, and phase IV, respectively.

In the THP-AD process, dewatering creates a considerable decrease in sludge volume. The energy consumption of mechanical dewatering ranges between 15 and 179 kW·h·t^{-1} TS [42]. As the most extensively applied method in China, belt press filtering energy consumption is 60 kW·h·t^{-1} TS [16], which could reduce the moisture content from 99% to 80%. The thermal hydrolysis process can effectively enhance conversion efficiency of organic matter and methane generation in the anaerobic digestion step. According to the demand of thermal hydrolysis steam (12 bar) for one tonne TS, inputting energy is 860 kW·h·t^{-1} TS in the process [43]. In the AD step, an average 37% of organics contained in digested sludge are reduced, with an average net methane generation of 0.2439 m^3 CH_4 per kilogram VS [14]. The generated methane can be burned directly, and oxidized into H_2O and CO_2 with a calorific value of 35.6 MJ·m^{-3} [15]. Of course, the actual value is weaker than the calculated recovered energy, because of factors such as low temperature off-gases, heat loss from incinerators, and incomplete combustion of methane, etc. (all leading to inefficient thermal energy recovery) [43]. The actual engineering data indicates a total thermal loss of 7% [30]. Following this, the thermal energy was further used for electricity generation, and 1211.3 kW·h·t^{-1}TS power is recovered in methane burning. In the whole THP-AD process chain, an energy deficit of 1503.7 kW·h·t^{-1}TS was obtained. Assuming that the efficiency of energy production related to carbon emission is 1.05 kg CO_2·kW^{-1}·h^{-1} [31], the global warming potential of THP-AD is 1578.9 kg CO_2 equivalent per tonne TS, with a negative environmental impact.

Table 3
Variation of the COD fractions in the innovative process.

Items	Organics	Phase I	Phase II	Phase III	Phase IV
Influent ($mg \cdot L^{-1}$)	P-COD	254.58	233.27	216.21	196.52
	LC-COD	23.94	44.40	53.05	24.92
	SC-COD	547.12	207.43	398.99	209.86
	S-COD	1432.85	1723.02	1558.07	1543.32
PN/DN. int-eff ($mg \cdot L^{-1}$)	P-COD	155.63	112.30	102.73	107.30
	LC-COD	8.55	72.19	88.15	72.12
	SC-COD	400.88	295.87	147.49	362.35
	S-COD	1235.06	1270.63	1305.12	1178.53
F/DN. eff ($mg \cdot L^{-1}$)	P-COD	118.74	69.59	82.20	98.52
	LC-COD	283.56	174.85	115.46	41.31
	SC-COD	475.57	374.36	637.77	794.50
	S-COD	1353.04	1519.44	2006.77	2284.98
PN/DN. eff ($mg \cdot L^{-1}$)	P-COD	–	261.14	103.93	136.01
	LC-COD	–	133.92	176.26	258.81
	SC-COD	–	267.84	159.64	208.44
	S-COD	–	1024.49	1138.67	1132.52

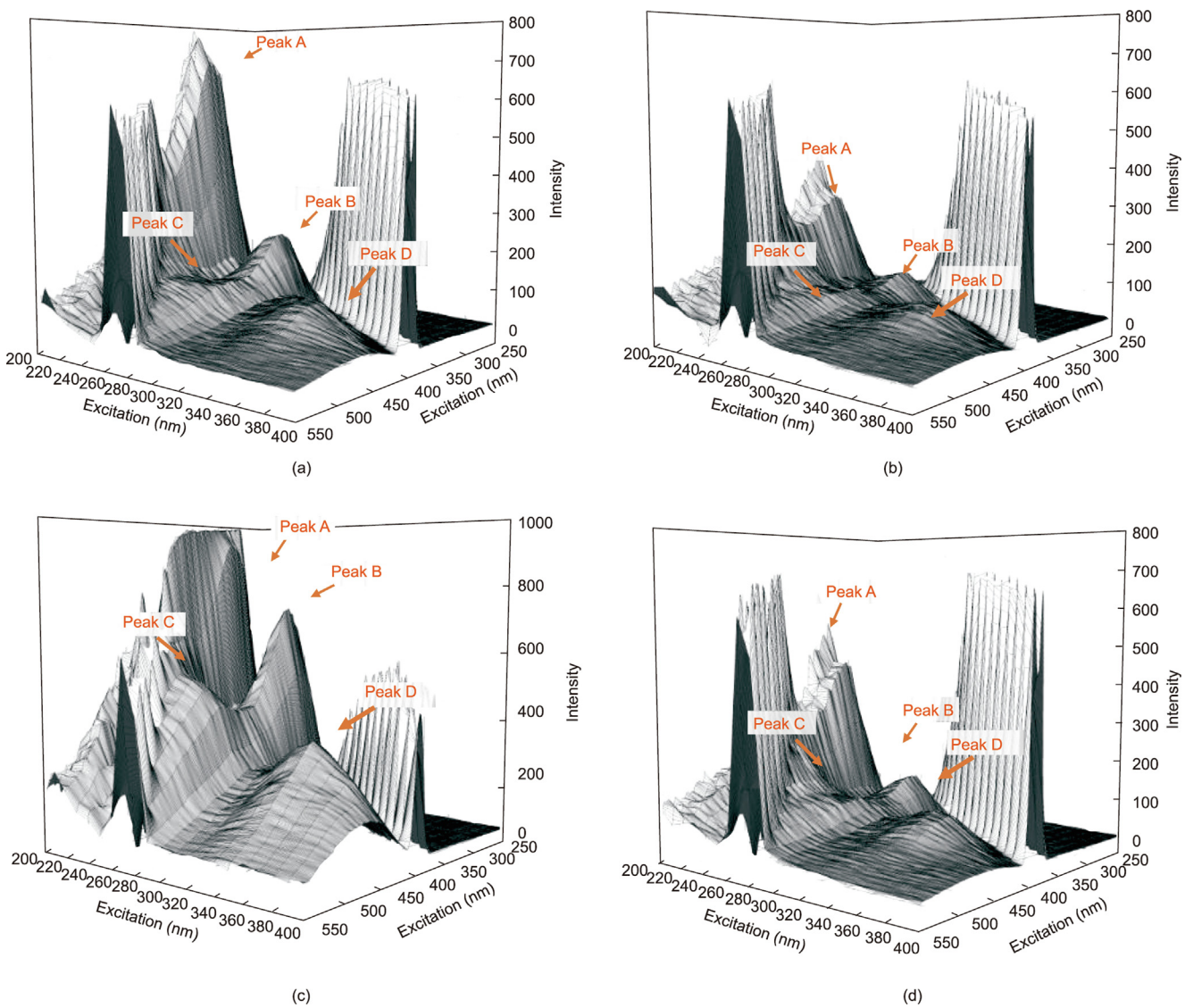

Fig. 6. (a) EEM fluorescence spectroscopy in the mature landfill leachate, (b) PN/DN-SBR intermediate effluent, (c) F/DN-SBR effluent, (d) PN/DN-SBR effluent. Ex: excitation wavelength, Em: emission wavelength.

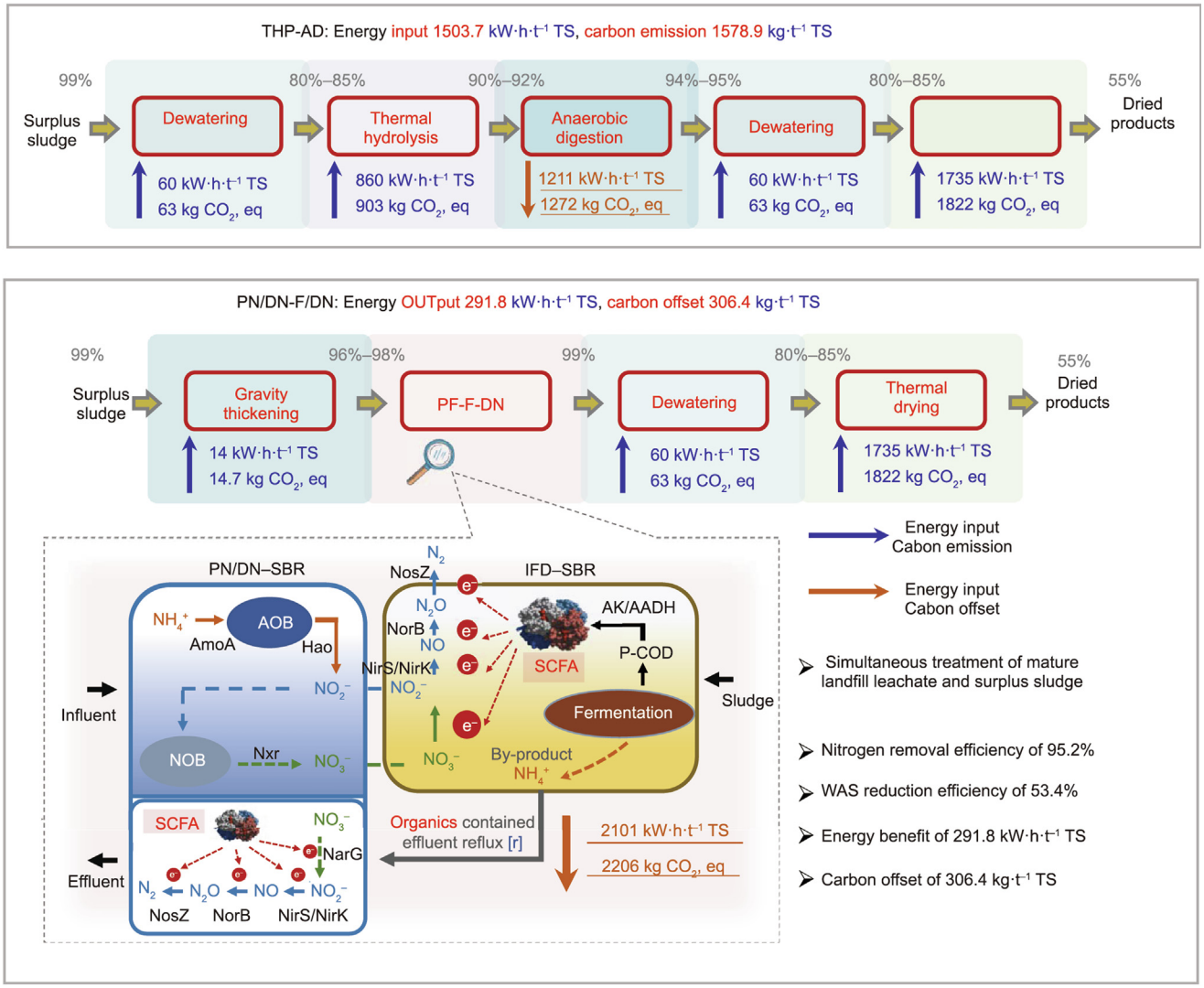

Fig. 7. Comparation of energy balance between THP-AD and PN/DN–F/DN processes. eq: equivalent.

The key benefit of PN/DN–F/DN is that organics resources could be recovered from sludge without an "energy input" pretreatment step (e.g., thermal hydrolysis, ozone, ultrasound, alkali, acid, or mechanical forces). This technology enables positive net energy recovery. The sludge moisture content decreased from 99% to 96%–98% in the gravity thickening. Then, the concentrated sludge coupled with nitrite-rich intermediate effluent of PN/DN-SBR was introduced to F/DN-SBR. Macromolecular organics contained in WAS were gradually degraded into SCFA through a series of fermentation steps, which facilitated NO_x^-–N reduction in the F/DN-SBR and post anoxic stage of PN/DN-SBR. In the stable period of phase IV, VS reduced from 15 712.2 to 7317.9 mg·L^{-1} in F/DN-SBR, which corresponded to 33.57 g VS consumption. When the organics consumed by denitrification, as well as the variation of COD between influent and effluent were comprehensively considered, an average of 19 350.6 mg COD organics was recovered in the stable period of phase IV (day 135–175), with organics recovered by per unit VS reduction of 0.576 g CODper gram VS. These results demonstrate that the PN/DN–F/DN biotechnology is significantly efficient at recovering organics resources from sludge, indicating it can significantly save energy consumption of sludge treatment and reagent dosing in subsequent wastewater treatment. Assuming that all recovered organics are provided by sodium acetate (4200 CNY·t^{-1}, 50% purity), the economic benefit of the recovered resource is 1789.8 CNY·t^{-1} TS, based on the current market price in China. When converted to the equivalent electricity, the actual energy recovery is 2100.8 kW·h·t^{-1} TS at a cost of 0.852 CNY·kw^{-1}·h^{-1}. As a result, the total benefit in terms of energy is 291.8 kW·h·t^{-1} TS. In other words, PN/DN–F/DN has excellent environmental benefit attributes, with 306.4 kg CO_2 per tonne TS carbon negative emission. Compared with the widely applied THP-AD configuration, PN/DN–F/DN is of great significance for promoting a paradigm shift from energy consumption to energy neutrality.

3.7. An innovative method for sludge and wastewater simultaneous treatment by PN/DN–F/DN: Prospects and application

Bioenergy and bioresource recovery from WAS are the principal pathways for achieving carbon neutrality in the field of environmental engineering; therefore, the application of WAS treatment technologies with low or negative carbon emissions is an inevitable choice to fight climate change and global warming. However, in reality, the current so-called energy recovery technology for WAS treatment is only an energy exchange process, whereby the bioenergy contained in WAS is exchanged by inputting valuable energy in

the form of physical, chemical, or electrical inputs [1,16,44]. Significantly, the output energy is much lower than that of the input, resulting in a large deficit in the energy balance [14–16].

PN/DN–F/DN was proven to simultaneously satisfy the demands for contaminant removal and carbon emission by considering the indirect and direct environmental effects. Compared with the widely applied THP-AD configuration, this innovative biotechnology is energy self-sufficient according to energy balance analysis. The prolonged reaction period and energy deficit were the pivotal bottlenecks in application of anaerobic digestion, severely limiting its utilization. The sludge retention time and sludge reduction rate of conventional AD applied in WWTPs were $\geq$ 20 d and 0.75–3.25 $kg \cdot m^{-3} \cdot d^{-1}$ based on previous publications [45–48]. This situation was reversed by PN/DN–F/DN. An ultra-fast sludge reduction rate (4.89 $kg \cdot m^{-3} \cdot d^{-1}$) was obtained, with an average sludge reduction efficiency of 53.4%. Importantly, recovered organics can be in situ utilized by denitrifiers, which further drives advanced nitrogen removal and correspondingly relieves indirect carbon emission from the organic substances dosed in wastewater treatment. As the mature landfill leachate contains various refractory and/or nonbiodegradable organic matter, 1635.6–1837.1 $mg \cdot L^{-1}$ COD remains in the effluent. To meet the discharge standards, application of PN/DN–F/DN in the future needs to be combined with fenton, electrohydrolysis and other tertiary sewage treatment technologies. Overall, PN/DN–F/DN provides a significant reference for the development of sludge and wastewater simultaneous treatment that incorporates high operation efficiency, good economic performance, and environmental benefits.

4. Conclusions

- An innovative PN/DN–F/DN process was firstly developed to realize organics recovery from WAS and nitrogen removal from mature landfill leachate through integrating partial nitrification, fermentation, and denitrification;
- After long-term operation (175 d), an average of 19 350.6 mg COD organics were recovered in PN/DN–F/DN per operational cycle, with 95.2% nitrogen removal and 53.4% sludge reduction;
- PN/DN–F/DN is of great significance for promoting paradigm transformation from energy consumption to energy neutrality, specifically, the total benefit in equivalent terms of energy was 291.8 $kW \cdot h \cdot t^{-1}$ TS.
- Compared with the conventional bioprocess, PN/DN-F/DN provides enhanced engineering potential by reducing oxygen supplementation by 25%, saving 100% of external carbon source dosing, and enabling external WAS reduction.

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Compliance with ethics guidelines

Fangzhai Zhang, Shang Ren, Haoran Liang, Zhaozhi Wang, Ying Yan, Jiahui Wang, and Yongzhen Peng declare that they have no conflict of interest or financial conflicts to disclose.

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