Towards Energy Neutrality in Municipal Wastewater Treatment: A Systematic Analysis of Energy Flow Balance for Different Scenarios

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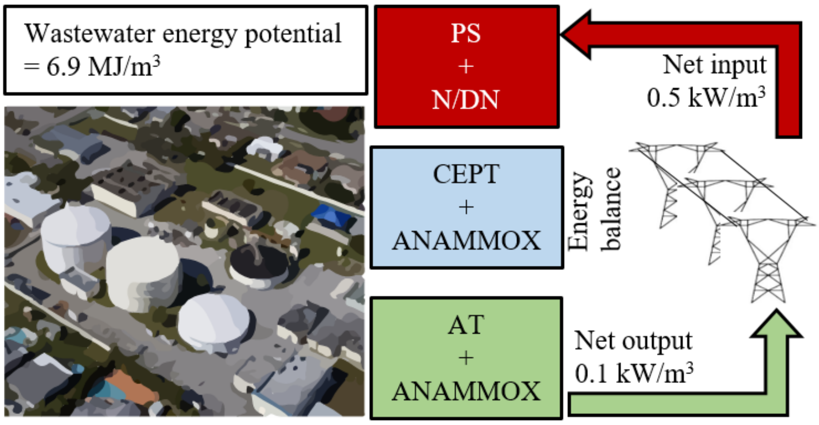
Table of Contents (TOC)



(All of the materials appearing in the TOC belong to the authors; and are 8.25 cm by 4.45 cm in size)

ABSTRACT

To help reduce operational costs and adverse impacts on the environment, particularly those connected with global climate change, the achievement of energy neutrality in municipal wastewater treatment plants (WWTPs) is a highly promising approach. Over 100 nitrification/denitrification (N/DN) and anaerobic ammonia oxidation (ANAMMOX)-based wastewater treatment systems were reviewed in this study. The results showed that the energy consumption rates of N/DN systems ranged from 0.3 to 4 kWh/kg-COD and 5 to 15 kWh/kg-N; while those of ANAMMOX-based systems ranged from 1 to 5 kWh/kg-COD and 0.5 to 1.5 kWh/kg-N. Based on an energy balance analysis, it was found that the conventional N/DN process consumed 1.78 MJ/m^3 more energy than was recovered from biogas and sludge digestate via a combined heat power system. However, if wastewater is pretreated by a chemically enhanced primary treatment (CEPT) or anaerobic treatment (AT), the subsequent ANAMMOX-based wastewater treatment systems may realize WWTP energy autarky or even output electricity at a rate of up to 0.17 kWh/m^3. In such a nexus of energy recovery, the biogas generated from the AT or sludge digestion would be an effective manner of recovering energy, while the incineration of sludge digestates was found to be an energy negative process. A two-part process that includes early-stage COD capture and ANAMMOX-based nitrogen removal is a promising approach to improving the energy performance of WWTPs in the direction of sustainability.

INTRODUCTION

By removing carbon and other nutrients (e.g., nitrogen and phosphorous compounds) from domestic wastewater, wastewater treatment plants (WWTPs) have long played an important role in both protecting the natural environment and maintaining human hygiene. Such plants, however, account for a significant part of the energy consumed worldwide. It has been reported that about 3.4% of the total electricity supply in the United States is consumed by municipal WWTPs,[1] a figure equivalent to 24% of the public energy use of a municipality.[2] In China, the total amount of treated wastewater has increased threefold in the last two decades, with more than 148 million cubic meters of domestic wastewater processed per day,[3] thus amounting to an annual consumption of 14 billion kWh of electricity.[4] The pre-eminent challenges from stringent wastewater discharge standards and rapid urbanization have driven up WWTP energy demands in both developed and developing countries.[5, 6] Hence, the recent developments in wastewater treatment energy recovery systems, such as anaerobic digestion[7] and combined heat and power (CHP) systems,[8] highlight the potential of WWTPs to be self-sufficient in energy.

The energy potential of domestic wastewater stems from its organic content; therefore, the focus of this study was on analyzing the energy flow of wastewater treatment processes through its organic COD mass balance (**Fig. 1**). A previous study showed that 13.9 MJ/kg COD of internal energy (heat value) in domestic wastewater could be released by an adiabatic bomb calorimeter.[9] This indicates that a mere 20% (net) recovery rate from influents containing organics could theoretically lead to energy self-sufficient WWTPs, given that in conventional WWTPs 3.2 MJ of electrical energy is empirically consumed by the removal of one kilogram of COD.[10] This further indicates that, based on the proper combination of wastewater treatment and recovery technologies, energy neutrality in WWTPs can be achieved.

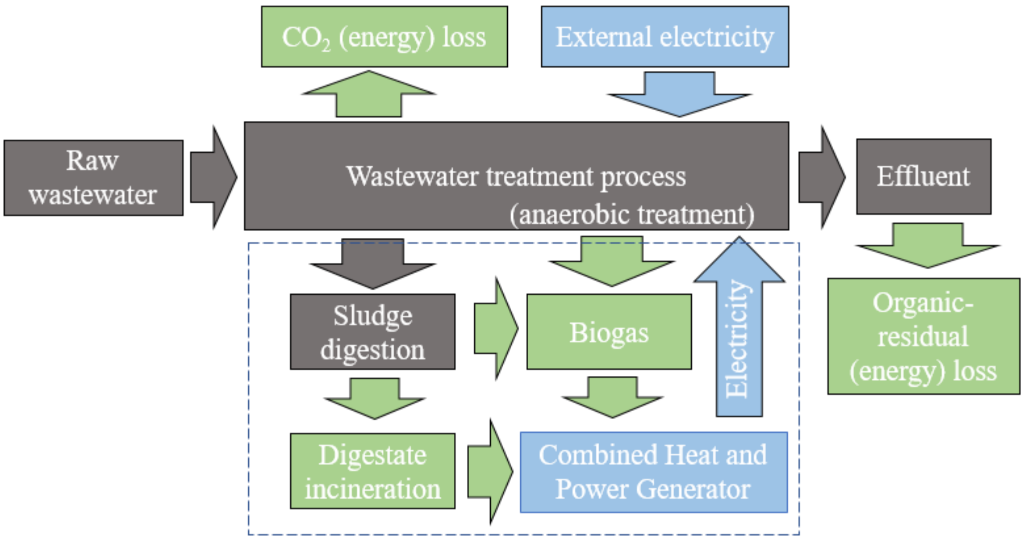


Fig. 1 Scope of the study analyzing the energy balance, including an examination of the wastewater treatment flow (dark grey), energy potential flow (green), and electricity input/output (blue), in wastewater treatment plants

However, there is a lack of studies involving a comprehensive review of the energy consumption of different wastewater treatment processes, coupled with a quantitative analysis of the electricity that could be produced from the energy recovered from wastewater. To address this important knowledge gap, the energy-use performances of more than 100 WWTP systems were evaluated in this study. From assessing the literature on the energy use of WWTPs, in this study mainstream treatment plants were classified into nitrification/denitrification (N/DN) and ammonia anaerobic oxidation (ANAMMOX)-based treatment processes. The major aims of this study were: i) to quantify the key performance indicators (KPIs) of the WWTPs regarding the removal of organic carbon or nitrogen; ii) to compare the KPIs in relation to system configurations and treatment capacities; and iii) to evaluate the energy recovered from domestic wastewater via different treatment approaches. The results that were obtained were utilized to characterize a nexus of energy flows from wastewater influent to effluent/sludge in two stages of treatment. The concept of "net energy balance" is proposed as a new paradigm for constructing sustainable mainstream WWTPs in the future.

MATERIALS AND METHODS

Data Collection and Evaluation of Energy Consumption. Data related to energy consumption/gain, COD loss/capture, and nitrogen removal rates from different treatment systems were collected from a review of relevant literature. Peer-reviewed journal articles were the primary sources, while some web search engines were also searched using keywords such as "sewage treatment", "wastewater treatment", "energy consumption", "energy recovery", "ANAMMOX", and so on. From these sources, a dataset detailing the energy consumed in the treatment process was developed, covering two major types of nitrogen removal systems: conventional N/DN and ANAMMOX-based processes (**Fig. 2**). A large number of process configurations, including membrane integrated bio-treatment processes, have been reported and clustered under different categories of membrane bioreactor (MBR); biofilm or bio-granular based systems grouped (BG); activated sludge-based continuous flow reactors (CF); or sequential batch reactors (SBR), depending on their operating modes. A total of 119 domestic wastewater treatment systems are listed for an evaluation of their energy consumption. When the specific energy consumption data were not available in the sources, the KPI of nitrogen or COD removal was recalculated (**Eqs. 1** and **2**), based on the removal rates.

$$KPI.COD\ (kWh/kg) = \frac{\text{Electric energy consumption (kWh/m3)}}{COD\ removal\ rate\ (\%) \times influent{-}COD\ (kg/m3)} \qquad \textbf{(Eq. 1)}$$

$$KPI.N\ (kWh/kg) = \frac{\text{Electric energy consumption (kWh/m3)}}{TN\ removal\ rate\ (\%) \times influent{-}TN(kg/m3)} \qquad \textbf{(Eq. 2)}$$

It should be noted that the influent concentrations of COD and nitrogen were defined in terms of kg per cubic meter (kg/m^3), the electric energy consumption was in kilowatt-hours (kWhs), the hydraulic retention time (HRT) was in the unit of days, and wastewater flowrate was defined as cubic meters per day (m^3/day). In addition, for those studies that only reported the values of KPIs, the COD or nitrogen KPIs were calculated according to **Eqs. 1** and **2**, respectively.

Energy Balance Analysis. The energy potential contained in domestic wastewater is mainly recovered through two means: (i) the biogas generated through the anaerobic treatment (AT) of wastewater or sludge anaerobic digestion (AD), and (ii) the incineration of dewatered sludge digestate in both N/DN and ANAMMOX–based systems (**Fig. 2**). Instead of collecting data on the energy recovered from domestic wastewater, the empirical values related to the characteristics of domestic wastewater,[11] bio-mass (sludge) generation, and energy-electricity conversion processes were utilized in this study. Regarding the electricity output of the biogas produced from sludge, a conversion coefficient of COD to energy was used (see **Table S2** for supporting information (SI)). The calculation (**Eq. 3**) is as follows.

$$\text{Biogas.P.E (kWh/kg)} = \frac{\text{biogas production efficiency(\%)} \times 13.9\text{MJ/kgCOD} \times \text{CHP efficienty(\%)}}{3.6\ (MJ/kWh)} \qquad \textbf{(Eq. 3)}$$

Incineration is commonly practiced to minimize solid waste and recover energy, and has widely been applied to treat sewage sludge in WWTPs.[12] Key parameters related to incineration fuels, such as heating values and organic portions, are listed in **Table S2**. Regarding the incineration of digestate (**Fig. 2**), the amount of the electricity output (Inc.E) was calculated according to **Eq. 4**.

$$\text{Inci.E (kWh/kg)} = \frac{\left(13.9\text{MJ/kgCOD} - \frac{2.67(MJ/kgH2O) \times \text{WC\%}}{SC\% \times OP\%}\right) \times \text{digestate.COD(kg)} \times \text{CHP efficienty(\%)}}{3.6\ (MJ/kWh)} \qquad \textbf{(Eq. 4)}$$

Here, the parameters of WC, SC, and OP, representing water content (70%), solid content (30%), and the organic portion of sludge digestate (50%), were empirical values

(**Table S2**), respectively; and the values of 13.9 MJ/kg-COD and 2.67 MJ/kg-H_2O indicate the energy potential of raw municipal wastewater and the energy lost due to water evaporating from the drying of the sludge, respectively. The CHP efficiency was defined as 35%. The energy balance analysis comprised two sections, namely energy consumption and energy recovery. To estimate the energy consumption, the mean values of the reviewed COD and nitrogen removal KPIs, and the characteristics typical of sewage were used (**Table 1**). All three proposed systems were selected to treat the same domestic wastewater (**Fig. 2**). It should be noted that the mean KPI.COD value was used in the N/DN system to calculate energy consumption (**Fig. 2**), which was applied to calculate the TN concentrations in the effluent (**Eq. 3**). By contrast, the mean value of KPI.N was used in the ANAMMOX-based systems to calculate the COD concentrations in effluents. If the COD or TN concentration in treated wastewater did not comply with the wastewater discharge guidance/regulations (European Communities Council 1991; Environment Protection Agency of USA 1994), a post-treatment stage had to be added to the proposed process (**Eq. 1 and Eq. 2**). The sum of the energy consumption was defined as the electricity input (E.input). As such, the energy balance analysis was conducted according to **Eq. 5.**

Extra.E = Inci.E + Biogas.S.E. + Biogas.P.E. – E.Input (**Eq. 5**)

If the Extra.E < 0, the selected system was considered to be energy negative and extra electricity input was needed. Otherwise, the system was considered to be either energy balanced (Extra.E = 0) or energy positive (Extra.E > 0). Additional information related to the calculation is provided in the supporting information (SI).

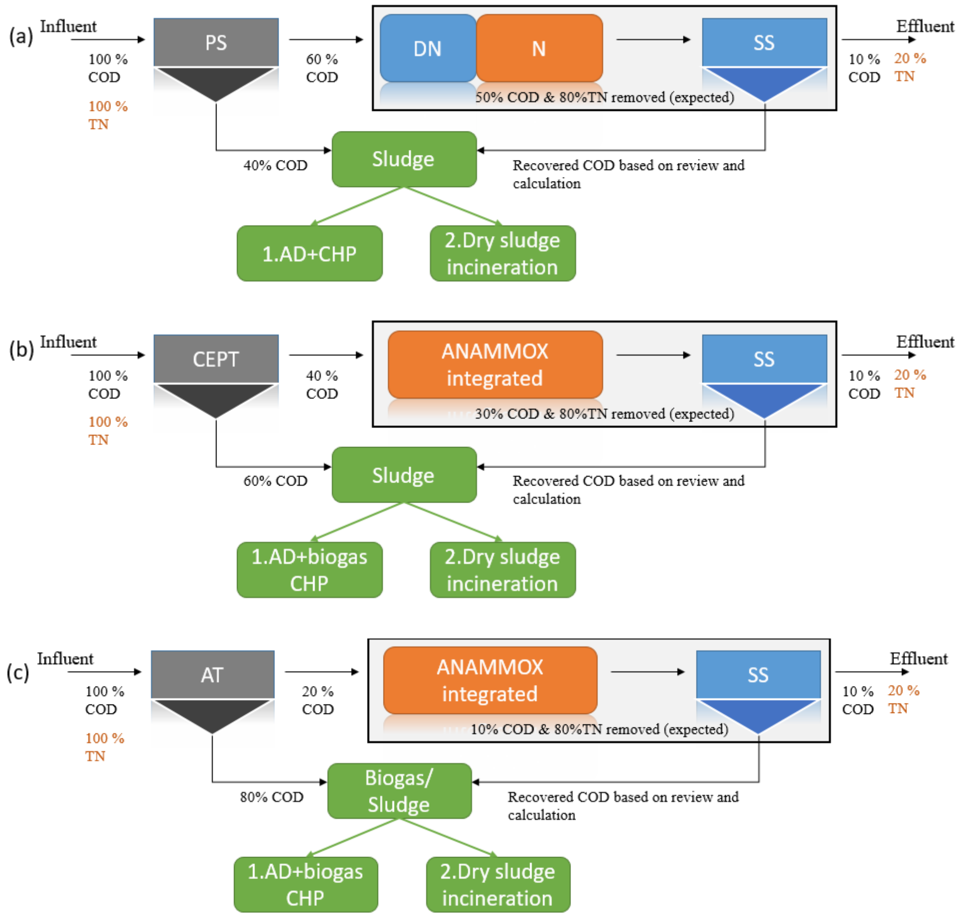


Fig. 2 Illustration of the proposed three treatment processes including the PS + N/DN process (a), the CEPT + ANAMMOX integrated process (b), and the AT + ANAMMOX integrated process (c). In these processes, the same removal efficiencies of COD and TN were expected to be 90% and 80%, respectively, and the sludge solids and biogas generated were utilized to recover energy.

Statistics. Before conducting the statistical analysis for this study, the data were log-transformed or scaled to improve sample normality or fitness to specific methods. The descriptive analyses of the collected data were performed on Excel 2010 (Microsoft Corp., USA). Statistical significance was defined as a 95% confidence interval, with a

P-value of < 0.05 (two-tailed). The statistical analyses were processed using SPSS (IBM, USA).

RESULTS AND DISCUSSION

Comparison of Energy Consumption Performance. Fig. 3 shows the processed data based on the overall energy consumption of the studied WWTPs. To elucidate the energy performance of wastewater treatment systems, the analysis was carried out using electrical energy (kWh) per kilogram removed COD or TN as the KPI. An indication of the specific type of studied reactor involved (i.e., whether MBR, SBR, BG, or CF) is given below each data point.

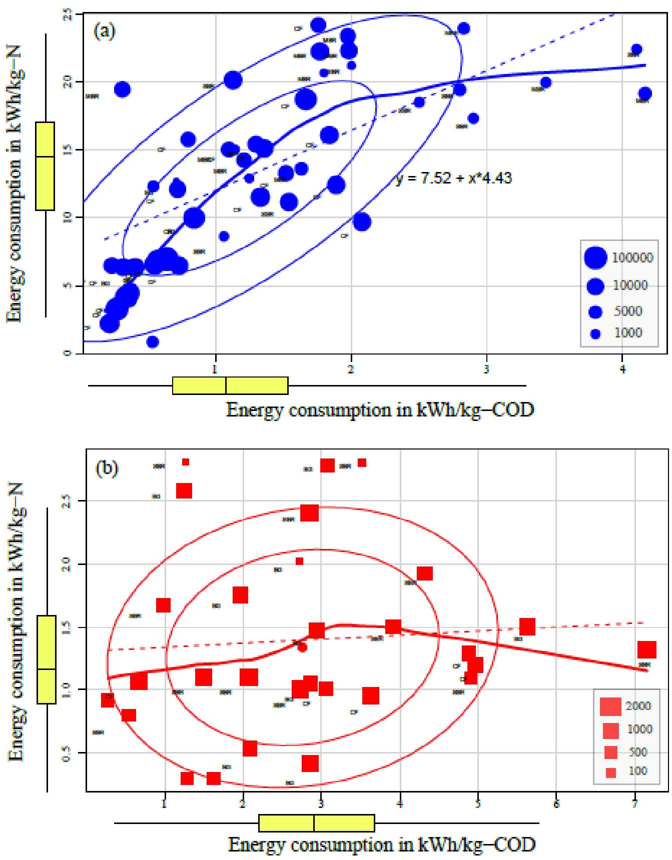


Fig. 3 Specific energy consumption per type of treatment (n = 119). The inner and outer circles encompass 50% and 85% of the plotted data in the nitrification-denitrification (N/DN; a) and ANAMMOX (b) groups, respectively; the linear regression between KPI-COD and KPI-N was plotted using dashed lines (Pearson, $P < 0.05$), and the solid line shows the general variation in the collected data (Loess, span = 0.4). The boxplot bars paralleling the x and y-axis represent the averaged energy KPI (kWh/kg) of samples in two different groups. The size of the legend symbols is proportional to the treatment capacity of the selected treatment system (m^3/d). The characters below each data point indicate the configuration of the collected wastewater treatment bioreactors.

Energy performance of the N/ND-based nitrogen removal process. Of interest is the fact that the N/DN systems displayed a wide range of KPI values for the removal of COD (KIP.COD; **Fig. 3**) of from 0.3 to 4 kWh/kg-COD. This could have been the result of the influence of large differences in treatment capacity, reactor type, operation control, regional energy policy, and so on.[13]

Table 1 Target treatment techniques and sewage characteristics

Treatment process			Sludge types	Energy recovery approaches	
Primary settling plus conventional N/DN $^{\#}$(0.58 kWh/m^3)			Primary and secondary	Biogas(AD-CHP)	Digestate (Incineration-CHP)
CEPT plus ANAMMOX $^{\#}$(0.25 kWh/m^3)			CEPT and ANAMMOX	Biogas(AD-CHP)	Digestate (Incineration-CHP)
AT plus ANAMMOX $^{\#}$(0.17 kWh/m^3)			AT and ANAMMOX	Biogas(AT+AD-CHP)	Digestate (Incineration-CHP)
COD (mg/L)		**Dis[b]**	**Nitrogen (mg/L)**		**Dis[c]**
Total	500	50	Total	50	10
Soluble	245		TKN/NH_4-N	40/25	n.a./8
Suspended	255		Others-N	10	

[a] Chemically enhanced primary treatment
[b,c] Discharge standards of Urban Waste Water Treatment (England and Wales) Regulations 1994

(complying to EU Council Directive 91/271/EEC)
#Calculations were provided in SI.

Regarding the cases that were reviewed in this study, the larger treatment systems such as the CF (plug-flow A/O system), had an average capacity of > 9,000 m^3/day and cost ~ only 0.5 kWh/kg-COD (**Fig. 3a**), due to the relatively stable characteristics of the influents.[14] This was seldom the case for small WWTPs,[15] the KPI.COD of which generally ranged from 2.5 to 4.5 kWh/kg-COD (< 5,000m^3/d; **Fig. 3a**). The SBR system under study was a variation of the conventional CF process,[16] the latter being more flexible regarding the requirements of both the land footprint and treatment capacities. The SBR is generally constructed in plants of below 1,000 m^3/d in size,[17] a figure which is consistent with the observations made in this study (**Fig. 3a**). Their KPI.COD values substantially increased, thus becoming comparable to the MBR's, i.e., ranging from 2 to 3 kWh/kg-COD. This was because they were using larger and more efficient equipment, such as pumps and compressors, and at a large scale, allowing these WWTPs to achieve a per capita reduction in electricity consumption.[3]

Fig. 3a also shows the relevance of the configurations of the reactors to energy consumption. Active aeration is considered the process that consumes the greatest amount of energy, accounting for 50% to 80% of the total electricity budget of WWTPs.[18] The intensive aeration in MBR systems enables organic degradation to take place and anti-fouling processes to be managed. Such systems exhibit an average KPI.COD of 2.13 ± 0.7 kWh/kg-COD (**Fig. 3a**). The capita energy consumption of the

MBR unit was generally maintained at 0.7 kWh/m^3 (2.0 ~ 2.8 kWh/kg-COD).[19] Only the membrane separation module of an MBR system consumed electricity at a rate of 0.6 – 1 kWh/m^3 – permeate.[20] However, the extremely high value of > 2.5 kWh/m^3 (> 5 kWh/kg-COD) was caused by the fouling of the membrane and the failure to equalize influent loads; such inefficient management could cause the whole MBR system to underperform by 50% of the nominal design.[21] Therefore, the aeration rate, covering the oxygen demand for organic degradation and system operation, was indexed as the "total oxygen demand" for the performance of wastewater treatment systems.[22] For example, optimizing MBR systems to reduce the operational aeration demand could make the related consumption of energy comparable to that of the CF system. The Ulu Pandan WWTP (Singapore) optimized the energy cost of its MBR unit to ~ 0.5 kWh/m^3 by balancing the biomass retention and membrane fouling processes.[22] However, the biofilm-granular systems (categorized into the BG) that utilize passive aeration techniques fell to a low range of KPI-COD,[23, 24] varying from 0.3 to 0.5 kWh/kg-COD, equivalent to ~ 0.1 kWh/m^3, in the studied cases (0.25 kg-COD/m^3), while the EU had a mean energy consumption rate of 0.3 kWh/kg-COD. Similar cases were also observed in the US, where the unit electricity requirement of trickling filters was 0.25 kWh/m^3.

Notably, the review of the energy required for COD removal also covered the use of nitrogen stripping for a standard-complying discharge (TN < 10 mg/L; **Table 1**). As shown in **Fig. 3a**, the lowest mainstream treatment KPI value of nitrogen removal

(KPI.N) via a conventional N/DN system (CF; > 5 kWh/kg-N) was substantially higher than that of systems for side-stream treatment (C/N < 1.5; NH_4-N > 500 mg/L), which reportedly consumed approximately 4.0 kWh/kg-N.[25] This finding suggests that an unnecessary amount of energy was inputted in the conventional nitrogen removal processes (14 kWh/kg-N; **Fig. 3a**). Of particular interest, from the perspective of COD removal, is that the mean energy consumption of the N/DN treatment systems was 0.61 kWh/kg-COD, or only 70% of the energy usage of a conventional activated sludge (CAS, without denitrification) system.[26] This implies that N/DN is a process that offsets the external electricity inputted for removing organic carbon.[27] It could be concluded that reducing the energy consumption of mainstream wastewater treatment processes still hinges on manipulating the association between carbon and nitrogen streams.

Energy Performance of ANAMMOX-based Systems. ANAMMOX is an energy-efficient process for removing nitrogen, but not for removing COD. In most cases, most ANAMMOX systems are independently applied for side-stream (low C/N ratio) nitrogen removal.[25] For application to mainstream sewage treatment, the system should include two parts:[28] Part A mainly involves the removal of COD to decrease the C/N ratio, while Part B involves the removal of N via an ANAMMOX process.[10] Regarding the proposed pretreatment process of chemically enhanced primary treatment (CEPT) or anaerobic treatment (AT) at the Part A stage, 60% to 80% of total COD can be removed from raw wastewater, which then reduces COD (biodegradable)/TN ratios to

3.0 or below. This ratio may apply to an ANAMMOX process (**Fig. S1** of the supporting information).

As shown in **Fig. 3**, the KPI.N values of the selected ANAMMOX systems were significantly lower than those of the N/DN group (One-way ANOVA, F = 108.4, $P < 0.001$). The average KPI.N values of the ANAMMOX and N/DN groups were 1.3 ± 0.7 and 14.6 ± 9.1 kWh/kg-N (**Fig. 3**), respectively. The lower energy input in nitrogen removal is majorly ascribed to a half nitration process.[29] In addition, the discrepancies in the KPIs, across all reviewed ANAMMOX systems, appeared to be less associated with the reactor's overall configurations (**Fig. 3b**). Take the SBR-ANAMMOX system, for example (**Fig. 3b**). Its KPI.COD and KPI.N values ranged widely from 0.3 to 7.0 kWh/kg-COD and 0.5 to 2.5 kWh/kg-N, respectively. One possible explanation for this large variation was that the treatment capacity of the study cases was below 5,000 m^3/d. In comparison with large-scale treatment plants, smaller-scale treatment plants could be more energy intensive.[30]

In addition, the linear regressions between KPI.COD and KPI.N further demonstrate that, unlike the N/DN systems (Pearson, R = 0.67, $P < 0.01$), the energy inputted for the removal of nitrogen and carbon by the ANAMMOX systems appears to be completely dissociated (Pearson, $P > 0.05$). A decrease in COD usually results from bio-absorption by sludge or co-occurs with early-stage nitration.[31] As such, full-scale applications of ANAMMOX are often used to treat low C/N ratio wastewater.[32]

However, a lower C/N ratio does not indicate that ANAMMOX-based systems are more efficient. As validated by this study, the optimal C/N ratio in wastewater-fed ANAMMOX treatment systems was observed to be within a range of 1 - 1.5 (**Fig. S1**). A massive aggregation of ANAMMOX bacteria (Candidatus *Brocadia caroliniensis*) was observed in a full-scale glycerol external COD-fed denitrification tank,[33] which suggested the possible co-existence of heterotrophic denitrification and ANAMMOX.[34] It should be noted that ANAMMOX-based (full-scale) systems are constructed for energy efficient N-removal;[25] and the co-occurrence of reduction in the contents of organic carbon can take place when wastewater has high levels (> 500 mg/L) of ammonia nitrogen.[25] Therefore, when using ANAMMOX technology for removing nitrogen from mainstream wastewater that is low in nitrogen but has a high C/N ratio, it is necessary to separate the removal of COD from that of N using a two-stage process.

Energy Flowchart in Primary Settling Plus N/DN-based Systems. Nitrogen removal is the most energy intensive process in mainstream wastewater treatment (**Fig. 3**). Therefore, in this study, the electricity consumed in the process of removing nitrogen from typical wastewater was calculated (SI), and this amount of electricity was used to estimate the concentrations of COD in wastewater effluents, to assess whether the effluents can meet discharge standards (**Table 1**). In addition, the extra energy used in the administration sections and in the sludge treatment process (e.g., for lighting buildings and dewatering sludge) accounts for ~ 10% (0.06 kWh/m^3) of the total energy costs of a conventional activated sludge WWTP.[35] Thus, this common value was added to calculate the energy consumption of each of the selected systems. **Table 1** shows that the energy cost of the conventional N/DN system was calculated to be 0.58 kWh/m^3. The COD content in its effluent was below 50 mg/L. This simulated result is consistent with the situation in on-site operations, in which external carbon sources are usually added for denitrification.[36] As shown in **Fig. 4**, the energy harvested from the conventional treatment system was only 0.63 MJ/m^3 (0.17 kWh/m^3), insufficient to achieve an energy-balanced situation, in which it was expected that 1.78 MJ/m^3 (2.41-0.63 MJ/m^3) of energy would be invested in one cubic meter of treated wastewater. Hence, an extra 0.47 kWh/m^3 (1.78 MJ/m^3) of electricity was needed for the on-site WWTP, even though the energy potential in raw wastewater was as high as 6.9 MJ/m^3. The sludge generated from primary and secondary treatment accumulated ~60% of the potential energy of raw wastewater, and around 2.0 MJ/m^3 of the organic matter was removed via a form of carbon dioxide. The assumption is that the latter was induced by the intensive aeration released in the conventional N/DN system. The collected data further revealed that the biogas energy recovered from sludge digestion provided 2.08 MJ/m^3 of heat for drying the dewatered digestate before the incineration process. Additionally, an energy loss of 1.16 MJ/m^3 was expected to take place when the biogas was converted to electricity (**Fig. 4**). Hence, apart from the energy lost due to the nature of the CHP-system (electricity conversion efficiency), the drying of dewatered sludge was another process involving the major consumption of energy (water evaporation). This implies that a reduction in the amount of sludge/digestate that is generated is key to achieving energy balance in this type of WWTP.

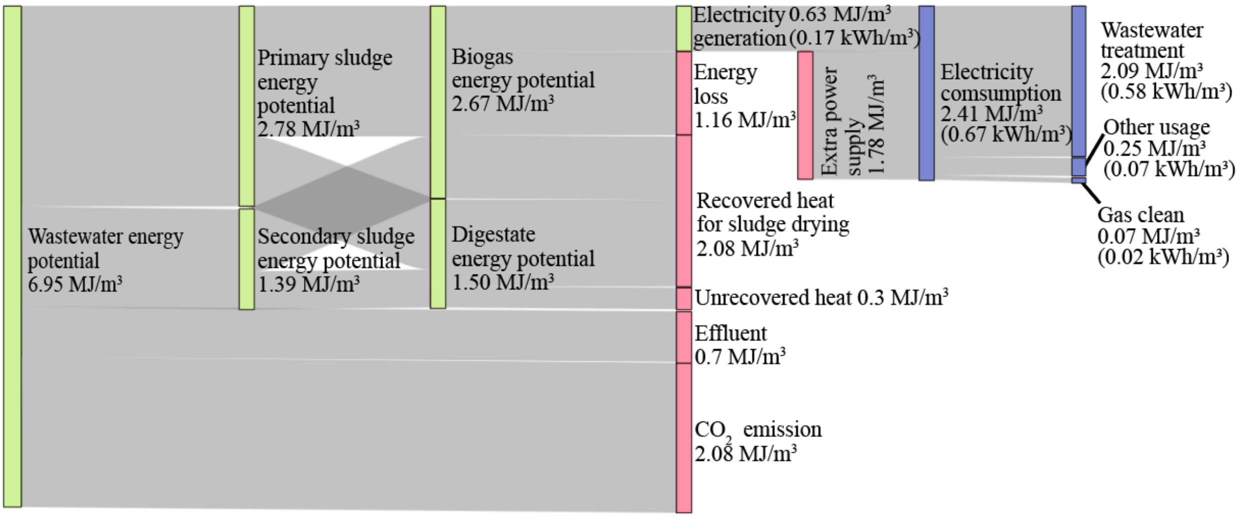


Fig. 4 Estimated capita energy consumption and recovery of targeted treatment systems (kWh/m^3) of a conventional nitrification/denitrification (N/DN) process. The grey strips represent energy flows (from left to right). The green, pink, and blue nodes indicate the remaining energy potential, energy loss, and energy consumption of the treatment systems, respectively. The specific amount and usage of energy are labeled on the right side of each node.

Energy Flowchart in the CEPT Plus ANAMMOX-based System. The overall performances of the various ANAMMOX techniques, which are aimed at capturing more organic carbon, were initially assessed with the addition of CEPT pretreatment (**Fig. 2**). In comparison with the primary settling, more COD were captured at the CEPT stage (**Table S1**). With regard to the ANAMMOX, the KPIs were collected from the side-stream wastewater treatment systems (**Fig. 3**). The latter does not include the energy usage of a CEPT process. Thus, an energy consumption rate of 0.07 kWh/m^3, which includes the cost of pumping influents and that of the coagulant mixing electrical process,[37] was used (**Table 1**). The energy inputted in the ANAMMOX was estimated to be 0.25 kWh/m^3. Compared to the N/DN systems, the decreased energy consumption of the ANAMMOX integrated techniques were ascribed to their comparatively low KPI-N values (**Fig. 3b**). Notably, in this scenario, however, the estimated COD concentration in the final effluent was 132 mg/L, thus failing to meet the designated discharge standard of 50 mg/L. According to previous studies,[38] carbon invested in the metabolism of microbes accounted for only 4% to 6% of the total COD fed into the ANAMMOX bio-systems. Hence, the implication is that more efficient carbon removal techniques, as a necessary post-treatment unit, should be installed following the introduction of an ANAMMOX function system.

Given that the COD contents of the effluents from the CEPT + ANAMMOX system were around 100 mg/L, equivalent to medium-strength domestic wastewater,[39] an aeration tank was selected as the post-treatment unit for the further removal of COD. Its energy consumption rate was assigned an empirical value of 0.85 kWh/kgCOD.[26] To comply with a discharge standard of 50 mg-COD/L, extra energy consumption of 0.09 kW/m^3 was expected from the energy cost of a post aeration tank. In summary, the CEPT + ANAMMOX + post-aeration system consumed 0.34 kWh/m^3 of energy (**Table 1**). This was 0.24 kW/m^3 lower than that for the N/DN system. It should be noted that the C/N ratio of the influent to the ANAMMOX treatment unit was 3:1 (COD = 150 mg/L; **Table S1**), and that ~60% of the COD of CEPT effluent was detected as soluble COD in previous full-scale and pilot-scale trials.[40] This relatively high portion of soluble COD may be better for the growth of heterotrophic denitrifying bacteria than of ANAMMOX bacteria.[41] However, so far, a limited number of existing large-scale mainstream ANAMMOX treatment systems, such as that used in the operations of the Changi Water Reclamation Plant[42], appear to have been unaffected by the transfer of soluble COD to the ANAMMOX system. The COD residual was recirculated from the ANAMMOX zone into an anoxic tank (minimum aeration), to facilitate denitrification and the uptake of phosphorus.[42] However, and notably, the per capita electricity consumption rate of the whole treatment system has yet to be reported from the on-site data.

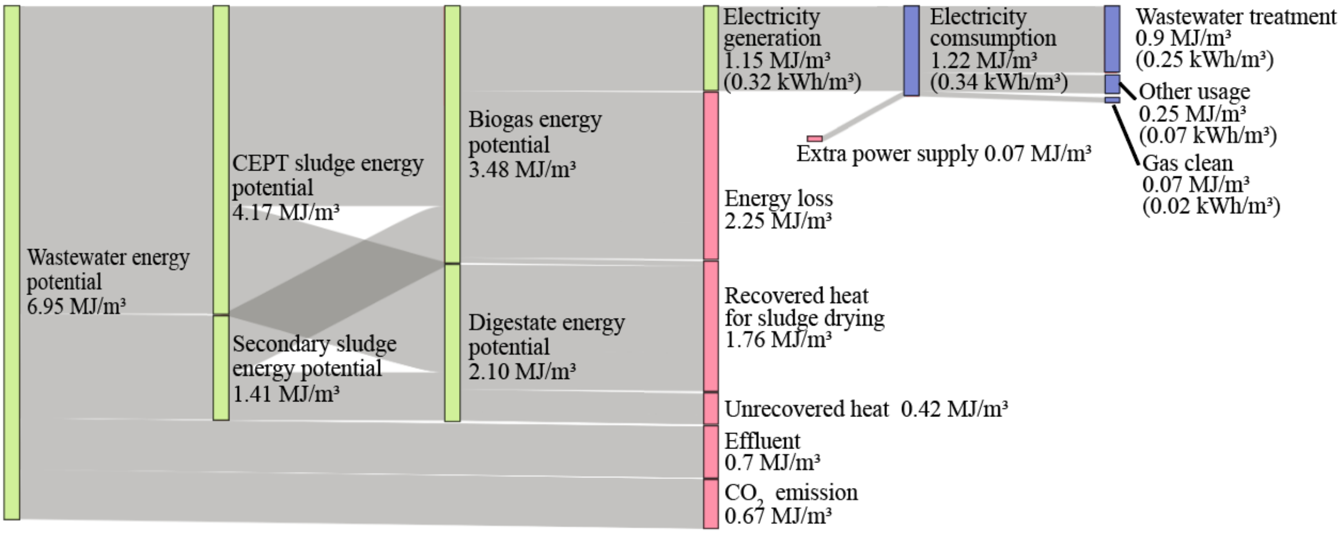


Fig. 5 Estimated capita energy consumption and recovery of targeted treatment systems (kWh/m^3) of the CEPT + ANAMMOX treatment process. The grey strips represent energy flows (from left to right). The green, pink, and blue nodes indicate the remaining energy potential, energy loss, and energy consumption of the treatment systems,

respectively. The specific amount and usage of energy are labeled on the right side of each node.

In terms of energy recovery, compared with primary settling sludge, CEPT can capture more organic content from raw sewage with a higher volume of sludge. In comparison with the primary settling plus N/DN based system, 4.17 MJ/m^3 of energy, capsuled in raw wastewater, was expected to be harvested in the primary sludge via the CEPT (**Fig. 5**). It is notable that, although the portion of organic content in CEPT sludge is comparatively lower than that in normal primary settling sludge due to chemical dosing, the application of co-digestion with food or husbandry wastes can enhance the production of biogas.[43] However, due to the ANAMMOX process, the whole system appears to need less energy (~ 0.7 MJ/m^3) due to the release from the CO_2 emission. Although the CEPT sludge has a lower portion of organic content than that of the primary settling sludge, in terms of a mixture of fluctuation additives, it captures more COD substance than does conventional primary settling sludge (**Table S1**), Hence, 36% of the total amount of COD can be converted to biogas (**Fig. 5**). The increase in the recovery of heat from biogas, together with the decrease in the demand for energy from the drying of sludge, produced (0.32 kWh/m^3) of electricity, which was twice that of the primary settling plus N/DN based system (**Fig. 4**). More importantly, the wastewater treatment system consumed only 0.25 kWh/m^3 of electricity. This relates to i) the use of ANAMMOX techniques, which require substantially less electricity for aeration; and ii) a reduction in the burden of removing organic carbon after CEPT. As a result, in this scenario, the energy harvested from wastewater can, in theory, almost drive the CEPT plus the ANAMMOX based energy-autarkic system (**Fig. 5**).

Energy Flowchart in the AT plus ANAMMOX-based Systems. **Table S1** shows that 80% of the total COD can be removed from raw domestic wastewater through the AT process. At this early stage, however, carbon removal treatment had rarely been conducted in the cases reviewed in this study (**Table 1**). An AT tank's energy cost of 0.12 kWh/m^3 was adopted from a previous study.[44] In total, the summarized energy consumption rate of the AT + ANAMMOX system was 0.17 kWh/m^3, of which the AT consumed energy of ~ 0.1 kWh/m^3, which was slightly higher than that consumed by the ANAMMOX system. The direct anaerobic treatment of sewage has recently drawn much attention due to the much lower energy input involved in the removal of COD compared to that of conventional systems.[11] The application of AT plus ANAMMOX to domestic wastewater treatment, in comparison with the approach in two other mainstream systems, has been conceptualized as the most energy-efficient process (**Table 1**). Featured among the three systems as having the highest portion of biogas harvested at 65% of COD (**Table S1**), the AT unit converts ~ 4.6 MJ/m^3 of energy directly from wastewater to biogas (**Fig. 6**). Simultaneously, it substantially reduces the amount of sludge, the energy potential of which is merely equivalent to approximately 1.5 MJ/m^3, thereby cutting the energy demands for drying sludge to less than 1.3 MJ/m^3 (**Fig. 6**). Hence, rather than being simply drained from the "sludge sink," more heat can be utilized to generate electricity. As shown in **Fig. 6**, although with an energy loss of 2.97 MJ/m^3, 1.57 MJ/m^3 of the energy flow was generated as electricity (0.44 kWh/m^3). The enhanced generation of electricity not only covers that needed for the holistic treatment of wastewater/sludge, but also leads to an output of surplus electricity at a predictable value of 0.17 kWh/m^3.

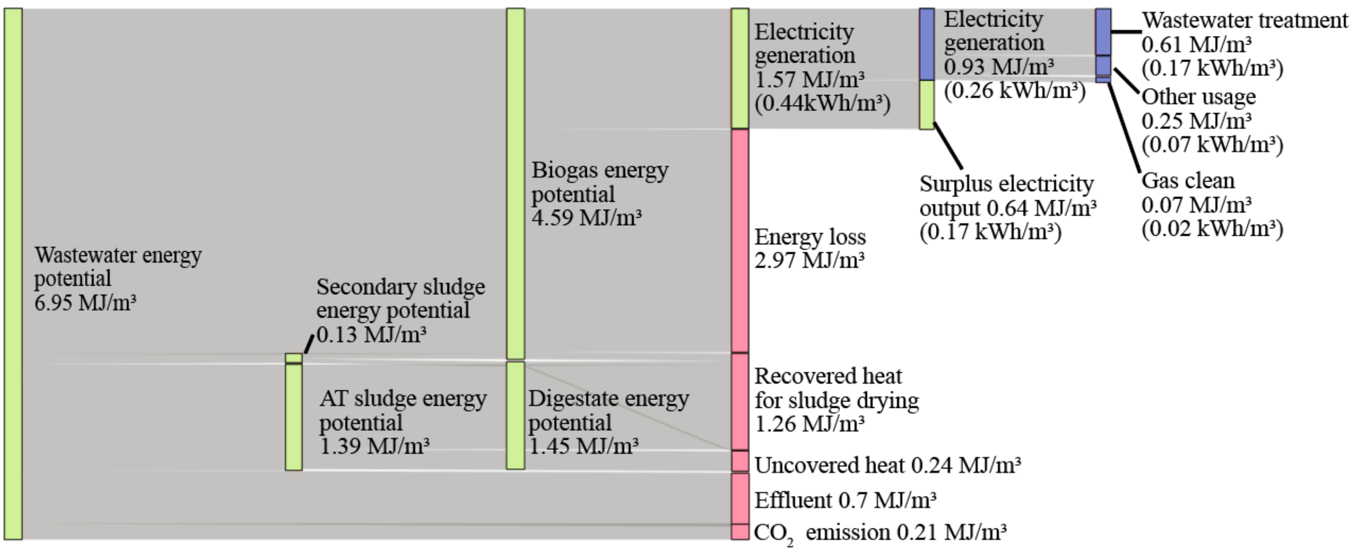


Fig. 6 Estimated capita energy consumption and recovery of targeted treatment systems (kWh/m^3) of the AD + ANAMMOX process. The grey strips represent energy flows (from left to right). The green, pink, and blue nodes indicate the remaining energy potential, energy loss, and energy consumption of the treatment systems, respectively. The specific amount and usage of energy are labeled on the right side of each node.

ENVIRONMENTAL IMPLICATIONS

By evaluating energy consumption and generation potential from more than 100 cases and analyzing the energy flowcharts in three conceptual processes (**Fig. 2**), it is concluded that 1) the conventional N/DN process cannot recover sufficient energy from raw wastewater to support the energy that it consumes, thereby necessitating inputs of energy from external sources; 2) as a better option with the potential to lead towards energy neutrality, the CEPT + Anammox + Post-treatment process could recover more energy to cover almost all of its energy usage; and 3) it is expected that the AT+ANAMMOX will use the least amount of energy in meeting discharge standards, as well as recover the highest amount of energy. A further result would be the possible output of surplus electricity, which would the most feasible option for designing and operating an energy sufficient WWTP.

The technical feasibility of treating wastewater and the associated energy recovery approaches are the major issues. Indeed, the ultimate goal in designing and establishing an energy-autarkic municipal WWTP is to reduce its dependence on inputs of energy, rather than to simply enable the purification of water. In the proposed energy-balance cases, further consideration should be given to the CEPT + ANAMMOX with regard to the extra chemical dosing costs involved and the decreases in the digestibility of sludge.[45, 46] The additional operation costs, associated with coagulation (e.g., coagulants, chemicals for pH adjustment) ranged from 0.02 to 0.05 US$/m^3.[47] When the CEPT process was adopted for the pretreatment of domestic wastewater, a maximum of 0.4 kWh/m^3 of energy could be generated. This is equivalent to 0.03 US$/m^3 in the USA, 0.06 US$/m^3 in the EU, and 0.05 US$/m^3 in China. In terms of energy-electricity cost, harvesting electricity from the CEPT + ANAMMOX system may be an economical choice, but a detailed cost-benefit analysis is needed before implementation.

The space requirements needed for designing energy recovery facilities for a WWTP would vary substantially, due to the different types of land sources. An incineration plant with a high sludge processing capacity, which requires a smaller land footprint, could be an option for areas with limited suitable land, such as Hong Kong and Singapore. Future efforts should be put into researching how to scale up from pilot-scale to full-scale AT techniques for treating mainstream domestic wastewater, such as enhancing carbon removal efficiency and biogas generation rates, and decreasing hydraulic retention time. Consideration might be given in future studies to expanding the energy calculations from the treatment scope of the WWTPs to the upstream processes, such as chemical production, land footprints, and transportation.[48, 49] Evaluating all the categories utilizing life circle analysis (LCA) would provide further comprehensive technical background information. Such information could then be used to conduct holistic comparisons of WWTPs using different configurations, and thereby further contribute to the enablement of energy neutrality in mainstream wastewater treatment systems.

ASSOCIATED CONTENT

Supporting Information

The supporting materials include raw data lists and details of calculations.

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X.D.L. and X.Z.L. designed this study; D.W. and X.Z.L. collected and analyzed the data; and D.W., X.Z.L., and X.D.L wrote the paper. All of the authors read and approved the final manuscript.

Notes

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