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Transition of biological wastewater treatment from flocculent activated sludge to granular sludge systems towards circular economy

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ABSTRACT

The public health concern of wastewater treatment spans many centuries and civilizations. The most significant advancement and evolution of wastewater treatment in the last century is the advent of conventional activated sludge (CAS) process to bacterial and/or algal-bacterial aerobic granular sludge (AGS) systems. This research is aimed to overview the importance of wastewater treatment, the development of biological wastewater treatment plants (WWTPs), and the changing focus in the last century by comparing the CAS and bacterial/algal-bacterial AGS systems. In addition, this review has introduced the circular economy concept and multiple sustainable development goals (SDGs) together with a systematic cross-sectional analysis of academic literature and government publications. The dynamic needs of society are the dominant factors in wastewater treatment transition and innovations, reflecting that bacterial and algal-bacterial AGS would be the main process for the future modern WWTPs with prospects to enhance a circular economy transition.

1. Introduction

Wastewater streams are products of developing societies' daily water use. The solvency of water contributes to wastewater richness in material resources that enhances microorganism growth processes. If improperly treated, wastewater poses water-related public health risks, alongside producing offensive odors that impact air quality, particularly in restricted urban spaces. Progressively, wastewater generation has kept pace with population growth, urbanization, industrial development, increasing agricultural production from the late 18th and early 19th centuries to the 21st century.

Advanced wastewater treatment technologies are imperative for public health safety, owing to population growth, high pollutant load discharge, and rising wastewater generation. Thus, the modern technological design of centralized sewage treatment plants began in the 1900s. Meanwhile, diverse environmental, public health, and social challenges have characterized the shifting focus and bioengineering advances in wastewater treatment. However, bioengineering innovations such as the flocculent conventional activated sludge (CAS) and aerobic granular sludge (AGS) processes have significantly influenced the outlook of wastewater treatment within the last 100 years.

With the emerging biotechnologies, wastewater's material resource recovery potential to transform society and foster sustainable development has gained much attention from the beginning of the 21st century. This appeals to a circular economy transition in wastewater treatment systems, adopting specific indicators for resource recovery monitoring (Preisner et al., 2022). Previous researchers on circular economy in wastewater treatment plants (WWTPs) focused on descriptive perspectives for energy recovery (Gherghel et al., 2019; Kundu et al., 2022; Zarei, 2020) and environmental impact from life cycle assessment (LCA) and greenhouse gases (GHGs) emission viewpoints (Pahunang et al., 2021; Rufí-Salís et al., 2022). Meanwhile, AGS rather than flocculent CAS process systems possess high potentials of dominating the next century's environmentally friendly biotechnologies for wastewater treatment in the context of circular economy.

This research mainly adopted peer-reviewed articles and analysis from Web of Science core collections in addition to the relevant research materials searched from ScienceDirect. This review is divided into three parts: (1) a brief account of wastewater generation through the century, underpinning the need for wastewater treatment with population growth; (2) the modern development of wastewater biotechnologies in the century, focusing on the flocculent CAS and AGS systems; and (3) the new outlook for wastewater treatment advanced by AGS systems, towards a circular economy transition, sustainable development, and

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environmental safety in the millennia.

2. Population growth and wastewater generation in the past 100 years

Human population increase in the last 100 years is the most significant as recorded in human history. In retrospect, on the global scale, the human population tripled from 2.5 billion in 1950 to 7.9 billion in 2021 (UNDESA_PD, 2021). This enlightens the increased water abstraction, uses, and subsequent wastewater generation from rapid urbanization, industrial growth, and agricultural production activities. Meanwhile, the critical value of water for human sustenance in direct and indirect applications inevitably produces wastewater, transporting high organic matters, nutrients, and varying hazardous material concentrations that threaten public health and life forms in water resources and the environment. Hence, proper wastewater treatment is prerequisite and crucial for a sustainable society.

2.1. A brief outlook on wastewater generation in the century: underpinning need for wastewater treatment

Increased wastewater generation, low collection and treatment ratio, and poor disposal are precursors to environmental pollution that have entrenched societies' view of wastewater as a nuisance. Characteristically, unsanitary conditions and contaminated water resources use have preceded repeated disease outbreaks. The first cholera epidemic of 1823 was in St. Petersburg (Barabanova, 2014), and the most recent one was in Yemen (Ng et al., 2020). Economic prosperity and advances in public health delivery on life expectancy triggered the stable population of the 1920s and rapid population growth from the 1950s to the 1970s. Moreover, intensified agricultural production, industrialization, and urbanization exposed surface water resources to high nutrient loads from agricultural runoffs, domestic sewage, and industrial effluents (Strokal et al., 2014).

High nitrogen (N) or phosphorus (P) nutrient loads discharged from wastewater into water resources influence nutrient cycling, aquatic life, and water use. The relatively shorter residence time for natural water purification through biogeochemical processes can induce massive eutrophication. Meanwhile, excess nitrate concentration in water resources causes "blue baby syndrome" in infants. Bat et al. (2018) reported that rapid population growth and rural-urban migration prompted the direct dumping of municipal and industrial effluents into the Black Sea, which subsequently influenced its fast eutrophication and degradation. Meanwhile, eutrophication caused the loss of almost 60 million tonnes of living marine resources and 5 million tons of fish in the Black Sea between 1973 and 1990 (Strokal et al., 2014). Similarly, several reports on coinciding eutrophication of vital lakes in various regions during the 1970s, including Lake Erie and Lake Kasumigaura (Mizunoya et al., 2021; Scavia et al., 2014), are found in the literature.

Primarily, wastewater treatment aims to protect and promote human health and the environment from spreading water resource contamination and associated diseases (Capodaglio et al., 2017). Thus, wastewater treatment until 1970 predominantly involved the removals of colloidal/suspended solids and floatable materials. However, more stringent measures for maximal removal of biological oxygen demand (BOD) and total suspended solids (TSS), and elimination of pathogenic microorganisms were implemented in the 1970s (Tchobanoglous et al., 2014). Additionally, N and P removal began (Kehrein et al., 2020a).

While originally designed to lower BOD by heterotrophic microorganisms, modifications for N and P removal by the flocculent CAS in different redox environments necessitated the introduction of multiple process units and recirculation flows (Nancharaiah et al., 2019). Moreover, the flocculent CAS process only became the dominant biological treatment process in urban environments from the beginning of the 1970s (Wanner, 2021), although it was discovered in 1910s (Ardern and Lockett, 1914).

In recent decades, the increasing complexity of wastewater streams has entailed more stringent effluent discharge standards prospecting innovative WWTPs' design a continuum. Therefore, the conventional focus, design, and upgrading of WWTPs to meet increasingly strict discharge standards (Fernández-Arévalo et al., 2017) characterize the innovation and the engineering of emerging second-generation biotechnologies. Moreover, the needs for energy use efficiency, economic viability, technical feasibility, and social acceptance have become relevant and critical features for WWTPs' sustainability evaluation, considering the indispensable requirements for environmental sanitation and protection.

2.2. Early development of wastewater treatment technologies

The pressure of increasing discharge of high pollutant load effluents in huge volumes and frequencies shortens the residence time and reduces the self-purification capacity of water resources. Before the industrialization and increased urbanization in the 18th and 19th centuries, the need for technological advancement in wastewater treatment had not drawn much attention. Most probably, sufficient pollutant load reduction could be achieved from a high dilution factor and natural water body's self-purification for safe withdrawal during that period.

The processes for domestic wastewater separation and collection through connected sewer networks for treatment began in the 1900s. Meanwhile, the first modern technological designs of centralized sewage and municipal WWTPs were mainly to reduce pollutant loads. This combined physical, biological, and chemical processes first in the United Kingdom and the United States in the late and early 19th - 20th centuries (Ambulkar and Nathanson, 2022). Moreover, introducing tertiary treatment with chlorine disinfection as a public health strategy in 1915 reduced cholera and typhoid mortalities.

2.3. Discovery of the flocculent CAS process

Ardern and Lockett commenced sewage aeration studies in 1912 at the Manchester Sewage Works based on Dr. Fowler's observations at the Lawrence Experimental Station in Massachusetts, New York. By retaining the sludge to accumulate in the reactor after decantation from each 6-h cycle from preliminary experiments for five weeks, they achieved complete nitrification within 6 h with clear oxidized effluent. Thus, exploiting the 19th century theory of natural selection, the retained solid matter from prolonged aeration of sewage termed "activated sludge" intensifies the oxidation process under suitable aeration (Ardern and Lockett, 1914). Sludge accumulated from several cycles could successfully purify sewage to acceptable standards in a shorter period, radically revolutionizing the dynamic advance of biological wastewater treatment and sanitation. The loosely settled microbial structures or activated sludge particles at the end of the aeration cycles are irregularly shaped flocs in the range of 100 μm in average (Nancharaiah et al., 2019). The first full-scale continuous-flow treatment system was installed at Worcester in 1916, rapidly spreading to developed countries with sewer systems.

Based on the logical concept of organic carbon (C) conversion into CO_2 and sludge to produce "clean water" after sedimentation, the flocculent CAS process has evolved over decades to incorporate nitrification/denitrification for N removal and enhanced biological P removal (Verstraete and Vlaeminck, 2011). Hence the robust, broad adaptability and environmentally friendly features of biological methods, easy operation, and low chemical use have been essential to their distinctive preference in the past century.

3. Sustainability of biological wastewater treatment systems in the 21st century

A growing preference for biological wastewater treatment technologies is becoming more apparent internationally in effluent discharge

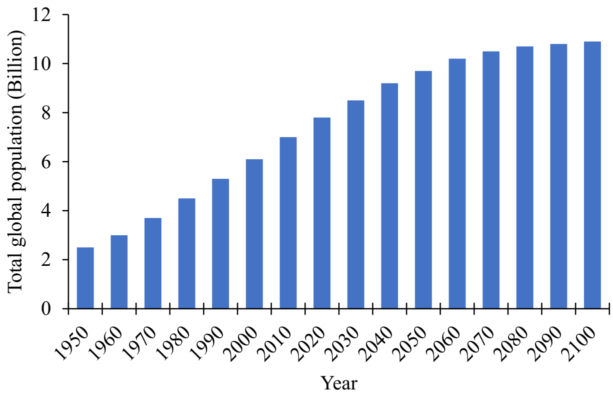

Fig. 1. Total global population growth since 1950, and projections to the end of the millennia.
(Data source: UNDESA_PD, 2021, 2022).

policy planning. For example, tertiary treatment requirement is recommended in EU countries only when biological treatment processes do not meet discharge limits. However, high aeration energy cost accounting for over 50 % of energy use in wastewater treatment remains a challenge for biological processes. Meanwhile, stricter treatment standards for wastewater by 2040 may contribute to over 50 % increase in energy use (IEA, 2016) and substantial GHGs emission from high fossil fuel-derived energy consumption.

The high energy demand in flocculent CAS processes mainly includes biological aeration, suspended solids mixing and recycling, solid waste dewatering, and pumping (Sid et al., 2017). Moreover, the relatively ineffective toxic substances treatment, including contaminants of emerging concern (CEC) and salinity tolerance, are critical challenges to the flocculent CAS process. Besides, waste activated sludge (WAS) disposal remains a significant drawback in urban environments with limited avenues for sludge landfills development, since sludge disposal in developed urban environments is becoming increasingly expensive and complicated (Han et al., 2021).

Innovative and competitive wastewater treatment options with comparably lower energy demand, better effluent quality, and sludge handling are quickly becoming the preferred alternatives to flocculent CAS processes. According to Abinandan et al. (2018) and Al-Jabri et al. (2020), inefficient treatment and high energy costs in conventional wastewater treatment systems are increasingly claimed for alternative treatment options, such as microalgae technologies, to maximize treatment efficiency, biomass production, and resource recovery. Moreover, aerobic granular sludge (AGS) systems with efficient sludge separation, compact infrastructure, and high energy efficiency are promising, which can become the standard for future WWTPs engineering (Nancharaiah et al., 2019; Nancharaiah and Sarvajith, 2019). These meet current requirements for optimizing WWTPs sustainability in energy use by incorporating climate-smart thinking, which promotes public health and social development.

The prospective integration of resource recovery into wastewater treatment is innovative in transforming the outlook, successfully realizing a circular economy model transition in the water sector. Meanwhile, a growing consensus on WWTPs' energy recovery capacity to transform them into energy neutral or net positive facilities from renewable energy production is gaining much attraction. Furthermore, there is a growing need for WWTPs to contribute to carbon neutrality in the future (Bae and Kim, 2021). Huang et al. (2022) proposed energy neutrality as primary, with the three main pathways for decarbonization: energy reduction, resource recovery, and renewable energy generation.

3.1. *Resource recovery: innovative and strategic solution for the 21st century challenges*

Global material resource use almost tripled in less than five decades from 27 Gt in 1970 to 89 Gt in 2017 (OECD, 2019). Meanwhile, the UN projects a rise in the worldwide population of 7.8 billion in 2020 to 8.5, 9.7, and 10.9 billion by 2030, 2050, and the end of the millennium (2100), respectively (UNDESA_PD, 2021). Fig. 1 shows the population growth trend and future projections. This impending rise would increase resource demand and use, building up more waste resources with

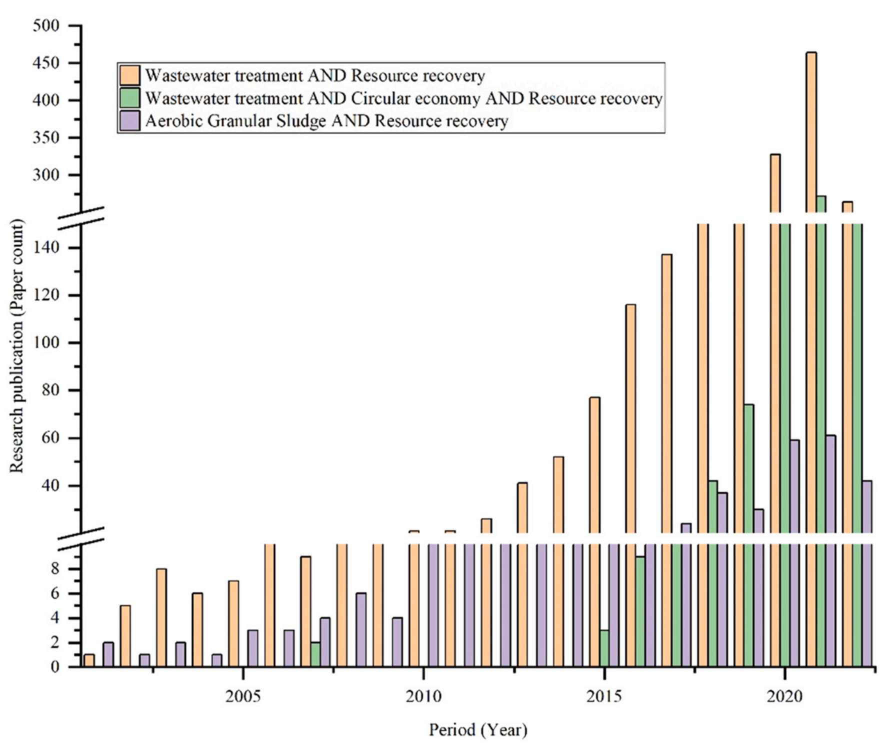

Fig. 2. The publications on wastewater treatment and resource recovery, aerobic granular sludge resource recovery, and circular economy application in the past two decades (Web of Science database from 2001-01-01 to 2022-09-09). Search terms used: Wastewater treatment/ resource recovery; Wastewater treatment AND Resource recovery; Wastewater treatment/Circular economy/ Resource recovery; Wastewater treatment AND Circular economy OR Circular bioeconomy AND Resource recovery; Aerobic Granular Sludge/resource recovery; Aerobic granular sludge AND Bacterial aerobic granular sludge OR Algal-bacterial granular sludge AND Microalgal-bacterial granular sludge AND Resource recovery.

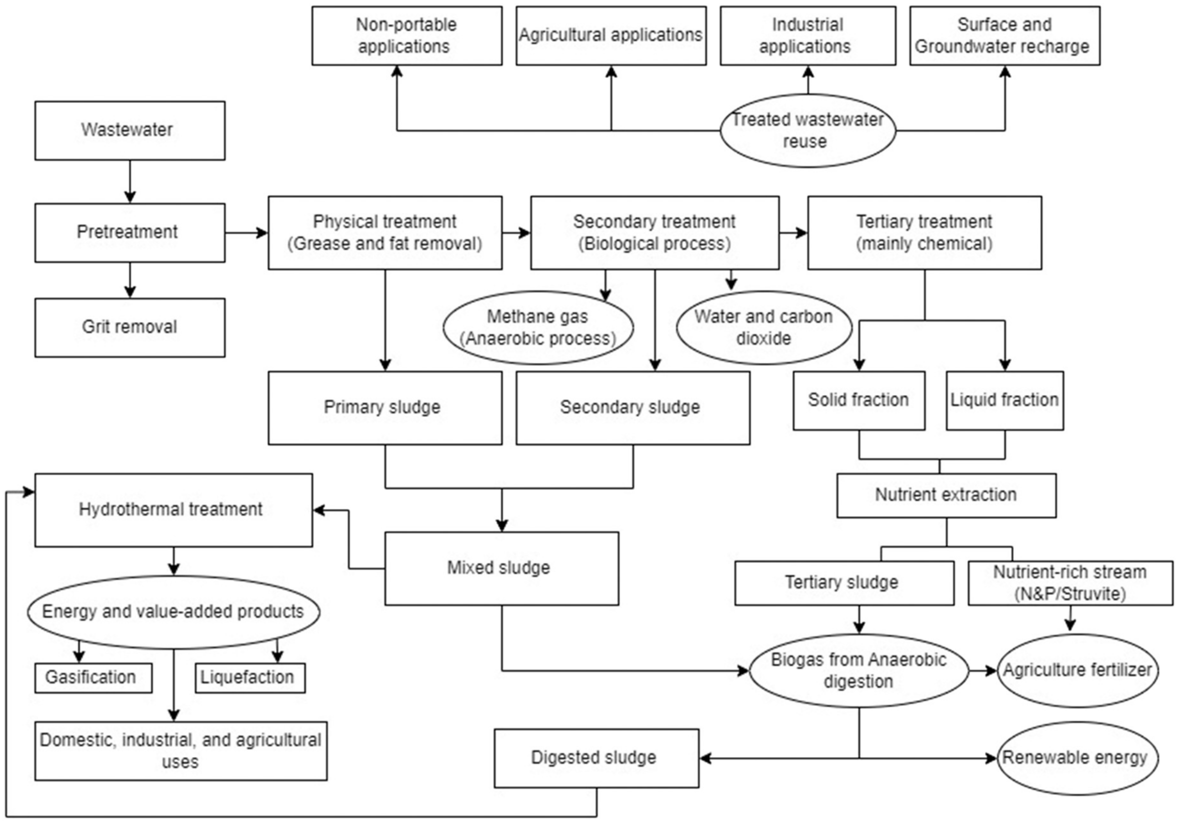

Fig. 3. Key steps and potential resource product recovery from the conventional wastewater treatment process and their uses. (Modified from Djandja et al. (2021) and Mbavarira and Grimm (2021)).

potentially negative consequences. Considering the projected rise in global material use from 2017 to 167 Gt by 2060, 123 Gt would be contributed from non-metallic minerals and biomass (OECD, 2019).

The dynamic challenges of the 21st century and the rise of a global economy present new opportunities, notably to harness material resources availability in wastewater. This underscores the recent additional focus on resource recovery from WWTPs. Besides the fundamental functions in nutrient removal and promoting environmental safety, WWTPs are transforming into water resource recovery factories (WRRFs) (Kehrein et al., 2020b). Simultaneously, this advances a wastewater circular economy transition that increases prospects for reclaimed water reuse and other pathways for recovery of multiple value-added products. The wastewater new outlook as a resource transforms observations of over 80 % of global wastewater discharged untreated and over 95 % among some least developed countries (UNEP, 2017) into a sustainable resource value to exploit. This potentially incentivizes more significant wastewater treatment planning and value creation in developing countries.

Predictably, resource recovery from wastewater treatment can stabilize price volatility from disparities in global natural resources availability and distribution, such as phosphates (Reijnders, 2014). Moreover, wastewater treatment and resource recovery applications are relevant to attaining multiple sustainable development goals (SDGs) and a circular economy globally, such as SDGs 6, 7, 9, 11, 12, 13, and 14. Substantially, this will limit natural resource exploitation and pollution effects. Thus, as the circular economy model of conscious design and efficient resource use for as long as possible in the production chain provides a suitable opportunity for maximal material use, which also facilitates a safe environment. The biological material cycles, on the other hand, are critical to creating a sustained renewable resource opportunity that keeps resource demands and uses in balance for economic development by exploring renewable energy, nutrients, metal ions, bioplastics, biofuels, and other value-added products from wastewater treatment.

3.2. Advancement in resource recovery and circular economy through biological treatment systems

The main drivers for developing the wastewater industry are global nutrient needs and water/energy recovery from wastewater (Neczaj and Grosser, 2018). However, the potentially high economic benefits from circular economy model application in wastewater treatment systems incentivize the increasingly growing research and industry interests. In the past decade, more circular economy development has been observed in the snowball of research publications in wastewater treatment and the same trend has been observed in AGS systems (Fig. 2). Meanwhile, different resource recovery and potential reuse routes are relevant for a circular economy transition between the existing conventional wastewater treatment processes and emerging alternatives in water, energy, and value-added products (Fig. 3). Moreover, the better effectiveness, economic value, and energy production potential of sludge from biological treatment systems underline their considerable preference over mechanical and chemical treatment processes (Ali et al., 2020). Utilization and recycling of sewage sludge can vary by treatment methods and toxic metal concentrations (Ghahdarijani et al., 2022).

High effluent quality for water reuse is an essential and primary function of resource recovery. Hence, future evaluation of wastewater treatment systems based on water reuse, other resource recovery potential, and energy use efficiency is imminent. This could entail prospective self-sustainability (economic sustainability), energy use efficiency, waste reduction, and pollution control (ecological sustainability), and technical feasibility. Meanwhile, WWTP's economic affordability and improvement of the local environment are vital to socio-cultural acceptance, which may differ by region (Muga and

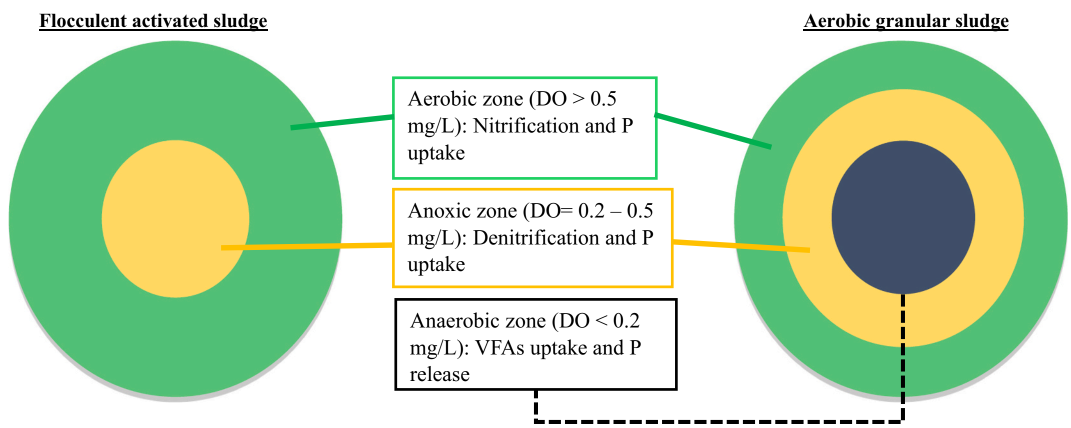

Fig. 4. Schematic representation and comparison of flocculent conventional activated sludge and aerobic granular sludge structures modified from Gogina and Gulshin (2016) and Nereda (2022). DO, dissolved oxygen; VFAs, volatile fatty acids.

Mihelcic, 2008).

Two cardinal requirements for effective wastewater treatment are contaminant removal and biomass separation from effluent. However, three-dimensional sustainability of WWTPs with enhanced profitability, environmental protection, and social relevance is critical to meeting future treatment demands and requirements. The key components include energy and resource recovery (Neczaj and Grosser, 2018; Zarei, 2020), environmental friendliness, ease of operation, capacity to withstand toxicity, low capital investment, and operation & management costs (Ali et al., 2020) in addition to minimized footprint (Nielsen, 2017). Besides, wastewater treatment systems are essential to the society, demanding more research on innovative strategies that adapt to climate change mitigation from anthropogenic GHGs emission.

4. Granular sludge systems: a new paradigm for sustainable wastewater treatment

4.1. Bacterial AGS

The recent discovery of bacterial AGS systems advances the prospects of addressing the society's dynamic and evolving needs in sustainable sanitation development, waste reduction and resource recovery, and recycling to a circular economy shift. Advancing the conventional focus, design, and upgrading of WWTPs is promising to meet the stringent discharge standards (Fernández-Arévalo et al., 2017).

Bacterial AGS biotechnology is considered the second generation of wastewater treatment, revolutionizing the outlook for sanitation in sustainable cities and integrating concepts of climate-smart thinking. Bacterial AGS has been over three decades since its first report (Mishima and Nakamura, 1991), in which granules of 2–8 mm in diameter were formed from aerobic activated sludge by self-immobilization in an upflow sludge blanket reactor and exhibited more excellent settleability compared to the CAS flocs. Thereafter, the novel bacterial AGS gained much research interest and subsequently global industrial application under the tradename Nereda® (Pronk et al., 2015; Robertson et al., 2016), with over 90 full-scale treatment plants in 20 countries and a cumulative 158,152,813 kWh savings in electricity (HaskoningDHV, 2022).

Bacterial AGS's compact structure, excellent settleability, lower energy requirement, 50–75 % reduction in land footprint (Bengtsson et al., 2019; Robertson et al., 2016), simultaneous nitrification and denitrification capacity, and ability to withstand toxicity and shock loadings

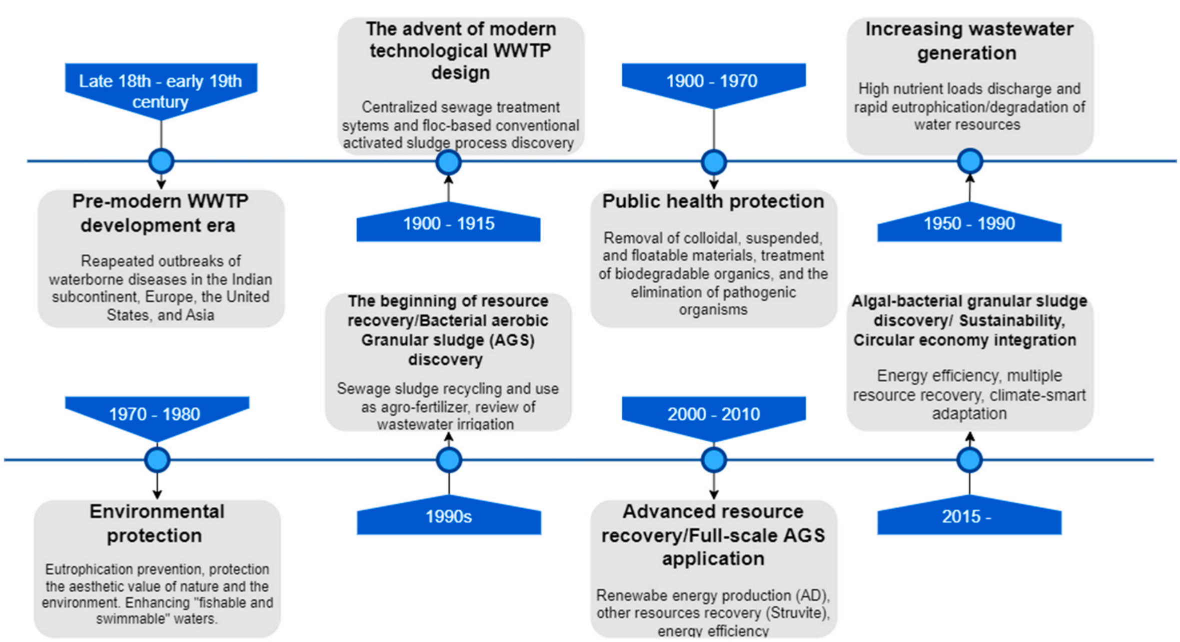

Fig. 5. A timeline of sanitation and environmental challenges shaping the advance of biological wastewater treatment from 19th century to 21st century.

Table 1
Effluent quality from full- and pilot-scale bacterial aerobic granular sludge systems.

WWTP (country)	Wastewater composition	Volumetric flowrate ($m^3 d^{-1}$)	Operation conditions	Influent concentration ($kg m^{-3} d^{-1}$)	Effluent quality (mg/L)	Reference
Yancang WWTP (China)	Municipal wastewater (30 % domestic and 70 % industrial)	50,000	Filling: 40 min; Aeration: 240 min; Settling: 40 min Discharging: 30 min; (Settling & discharge: 70–80 min) Idling: 0 min Operation duration: 155 days	COD: 0.56 NH_4^+-N: 0.022	COD: 85[c] NH_4^+-N: 95.8[c] TN: 59.6[c]	Li et al. (2014)
Garmerwolde WWTP (The Netherlands)	Municipal wastewater	28,600	Sludge loading: 0.10 kg TSS^{-1} d^{-1} HRT: 17 h SRT: 20–38 days Max. recycle ratio: 0.3 DO: 1.8–2.5 mg/L Temp.: 20 °C SRT: 20 days HRT: 0.7 days	COD: 0.506 BOD: 0.224 TP: 0.0067 NH_4^+-N: 0.039 PO_4^{3-}-P: 0.0044 TN: 0.0494 SS: 0.236	COD: 64 BOD: 9.7 TP: 0.9 PO_4^{3-}-P: 0.4 TN: 6.9 SS: 20	Pronk et al. (2015)
Garmerwolde (The Netherlands)	Municipal wastewater	28,600	60 min: Anaerobic feeding/simultaneous effluent withdrawal 240 min: aeration 60 min: Settling 15: Excess sludge discharge	COD: 0.528 BOD_5: 0.232 TP: 0.0072 TN: 0.053 SS: 0.247	COD: 57 BOD: 9.3 TP: 0.7 TN: 7.4 SS: 8.9	Guo et al. (2020)
Garmerwolde (The Netherlands)	Municipal wastewater	20,355	SRT: >30 days HRT: 10–12 h Temp: 8–8.6 °C	n.d.	Meets EU Water Directive (91/271/EEC) standards	Barrios-Hernández et al. (2020)
Österröd WWTP (Sweden)	Municipal wastewater	1800[a] 7980[b]	5.2 h cycle VER: 50 % SRT: >30 days HRT: 11.4 ± 0.7 h (average of 2 SBRs) Temp.: 13 °C	COD: 0.230 BOD_7: 0.081 NH_4^+-N: 0.019 TP: 0.0021 PO_4^{3-}-P: 0.0074 TN: 0.0177 SS: 0.102	COD:42–44 BOD_7: 6 NH_4^+-N: 0.75 TP: 0.06 PO_4^{3-}-P: 0.03 TN: 5.3–5.4 SS: 11–12	Burzio et al. (2022)
Frielas WWTP (Portugal)	Municipal wastewater (domestic, storm, and industrial)	55,000–60,000	n.d.	COD: 0. 310–0.5625 BOD: 0.1367–0.2825 SS: 0.1667–0.290	COD: 31–64 BOD: 6–10 SS: 6–21	Oliveira et al. (2020)
Vroomshoop (The Netherlands)	Municipal wastewater	1541	SRT: >21 d HRT: 11–24 h Temp.: 8.5–17.8 °C	n.d.	Meets EU Water Directive (91/271/EEC) standards	Barrios-Hernández et al. (2020)
Adelaide (South Australia) Pilot scale	Saline Municipal wastewater	63.9 L	100 % anaerobic condition 60 min: Anaerobic feeding 120 min: Aeration 8 min: Settling 2 min: Decant Operation duration: 113 days	COD loading: 1.55 COD: 534.9 mg/L NH_4^+-N: 35.1 mg/L TN: 55.8 mg/L Sulphate: 668.6 mg/L TSS: 535 mg/L Salinity: 5.8–7 g/L	NH_4^+-N: 77.8–99.7 % TN: 16.0–97.5 % PO_4^{3-}-P: 5.4–49.7 %	van den Akker et al. (2015)
Adelaide (South Australia) Pilot scale	Saline Municipal wastewater	63.9 L	33 % anaerobic condition 20 min: Anaerobic feeding 40 min: Aerobic feeding 80 min: Aeration 15 min: Settling 10 min: Decanting Operation duration: 95 days	COD loading: 0.98	PO_4^{3-}-P: 4.3–17.3 % NH_4^+-N: 96.1–99.8 % TN: 27.5–94.2 %	van den Akker et al. (2015)
Adelaide (South Australia) Full scale	Saline Municipal wastewater	n.d.	100 % aerobic 54 min: Aerobic feeding 108 min: Aeration 54 min: Settling 54 min: Decant	COD loading: 0.80	NH_4^+-N: 70.8–99.6 % TN: 75.7–92.9 % PO_4^{3-}-P: n.d.	van den Akker et al. (2015)
Lubawa WWTP (Poland)	Low strength (30–40 % from dairy industry) wastewater	3200	216 min: Aeration 20 min: Settling 40 min: Feeding/Discharge DO: 2 mg/L VER: about 25 %. Superficial gas velocity: 0.18 cm/s SRT: about 30 days; HRT:	COD: 1319.5 mg/L BOD_5: 1120 mg/L TP: 19.5 mg/L TN: 90.5 mg/L NH_4^+-N: 64.3 mg/L	COD: 39.1 BOD_5: 20.0 TP: 0.9 TN: 11.8 NH_4^+-N: 0.4	Świątczak and Cydzik-Kwiatkowska (2018)

(*continued on next page*)

Table 1 (*continued*)

WWTP (country)	Wastewater composition	Volumetric flowrate ($m^3 d^{-1}$)	Operation conditions	Influent concentration ($kg m^{-3} d^{-1}$)	Effluent quality (mg/L)	Reference
			1 day, Minimum settling velocity: about 1.6 m/h.			
Dinxperlo WWTP (Aalten, the Netherlands)	Domestic wastewater	3100	n.d.	COD: 531 mg/L BOD: 202 mg/L NH_4^+-N: 54 mg/L P: 6.4 mg/L	COD: 28 BOD: 2 NH_4^+-N: 6 P: 1.1	van Dijk et al. (2021)
Pilot scale test (Hangzhou, China)	Medium strength wastewater	Total working volume of 3 m^3 1 m^3 (A)	70: Feeding 120 min: Stirring 480 min: Aeration 5 min: Settling 5 min: Discharge 10 min: Idling VER: 33 % Air flow rate: 3.6 m^3/h Temp: 25 ± 5 °C	COD: 447.9 mg/L TN: 111.9 mg/L NH_4^+-N: 95.5 mg/L TP: 9.3 mg/L iron shavings: 10 kg with filling rate of 3.3 g/L	COD: 34.0 TP: 0.12 NH_4^+-N: 8.4 TN: 30.1 TFe: 0.30	Pan et al. (2022)
Pilot scale test (Hangzhou, China)	Medium strength wastewater	1.5 m^3/cycle (B)	110 min: Stirring 400 min: Aeration: 40: Settling 40: Discharge 5 min: Idling 45 min: Feeding VER: 50 % Air flow rate: 3.6 m^3/h Temp: 25 ± 5 °C	COD: 447.9 mg/L TN: 111.9 mg/L NH_4^+-N: 95.5 mg/L TP: 9.3 mg/L Iron shavings: 10 kg with filling rate of 3.3 g/L	COD: 21.5 TP: 0.07 NH_4^+-N: 3.7 TN: 19.1 TFe: 0.23	Pan et al. (2022)

BOD, biochemical oxygen demand; COD, chemical oxygen demand; HRT, hydraulic retention time; SRT, solids retention time; SS, suspended solids; TN, total nitrogen; TP, total phosphorus; TSS, total suspended solids; VER, volumetric exchange ration. n.d., no data, TFe; total iron.

[a] Dry weather.

[b] Rainy weather.

[c] Removal rate (%).

(Jiang et al., 2020, 2021; Wu et al., 2020) are superior advantages over the century-old CAS process. Besides the sludge density enhancement of fast settling, bacterial AGS's distinctive aerobic, anoxic, and anaerobic zones advance higher effluent quality over the flocculent CAS systems (Nereda, 2022). Fig. 4 illustrates the characteristics of activated sludge flocs and aerobic granules in addition to their nutrient removal zones.

4.2. Algal-bacterial AGS

More recently, algal-bacterial AGS developed from the bacterial AGS concept is gaining much research attention as a promising granular sludge option (Lee and Lei, 2019, 2022). The prospective application for high effluent quality through the algal-bacterial symbiosis process has been the basis for water purification in natural resources. Comparatively, algal-bacterial AGS exhibits a more stable granular structure and great potential for fast biomass growth, and its dense biomass per unit area can reduce 76 % of footprint (Wang et al., 2022). Additionally, algal-bacterial AGS has the potential to reduce aeration energy costs from microalgae respiration, resulting in 58 % decrease in energy consumption (Liu et al., 2020; Wang et al., 2022; Zhao et al., 2019). Moreover, the high nutrient accumulation and bioavailability (Wang et al., 2020; Zhao et al., 2019) in algal-bacterial AGS biomass is an innovative solution to alternate P recovery from the biological wastewater treatment process. Furthermore, it provides an alternative strategy for industrial-scale, low-cost cultivation, and a less cumbersome harvesting route for microalgae production (Wang et al., 2022) and high value-added products recovery (Meng et al., 2019a, 2019b, 2019c) for industrial applications.

Given the need for resource recovery (water reuse, nutrient recycling, energy recovery, and value-added product development), AGS systems, including bacterial and algal-bacterial AGS, are the future of sustainable WWTPs. Although its operation is still laboratory-based, algal-bacterial AGS has excellent potential for future application due to its excellent effluent quality, low energy use, high biomass production and productivity for multiple value-added resource recovery, excellently competing with the flocculent CAS process (Zhang et al., 2021) and bacterial AGS biotechnology (Guo et al., 2021; Semaha et al., 2020). Furthermore, algal-bacterial AGS system shows great potential for excellent wastewater treatment, concurrently pioneering low-cost microalgae cultivation and harvesting compared to suspended microalgae systems (Wang et al., 2022).

Fig. 5 summarizes the timeline of the advancements of biological wastewater treatment, highlighting the transition of WWTPs from sanitation and environmental protection to resource/energy recovery from wastewater, especially by bacterial/algal-bacterial AGS systems.

5. Treatment performance, potential use, and resource recovery via AGS systems

5.1. Treatment performance and potential use

Multiple use of treated wastewater is enhanced by effluent quality and can significantly contribute to increased reclaimed water use in water-stressed regions or for varied non-potable services. Agricultural reclaimed water usage in the water-scarce areas is critical to efficient freshwater abstraction for portable use. According to Liao et al. (2021), between 2015 and 2019, 398 million people were affected by drought in Asia. Meanwhile, nutrient recycling from WWTPs as fertilizer positively impacts the environment by reducing the demand and production of conventional fossil-based fertilizers, consequently reducing water and energy consumption (Mo and Zhang, 2013; Neczaj and Grosser, 2018).

AGS systems' excellent treatment performance in full and laboratory scales are promising to realize safe effluent discharge without tertiary treatment, potentially reducing cost and chemical use. Meanwhile, this also provides an avenue for agricultural irrigation use. Bacterial and algal-bacterial AGS demonstrate excellent nutrients removal that meets international discharge and effluent reuse standards (Barrios-Hernández et al., 2020; Guo et al., 2021; Ji et al., 2020). Tables 1 and 2 respectively summarize the effluent quality from full-scale bacterial AGS and lab-scale algal-bacterial AGS systems under the specific operation

Table 2
Effluent quality from lab-scale algal-bacterial granular sludge systems.

Reactor volume and type	Operation conditions	Influent concentration (mg/L)	Effluent quality (mg/L) and removal efficiency (%)	Reference
1.4 L Sequencing Batch reactor (SBR)	Temp.: 25 ± 2 °C Cycle: 4 h Influent filling: 2 min Non-aeration: 28 min Aeration: 185–200 min Settling: 5–20 min Effluent discharge: 5 min VER: 50 % HRT: 8 h Airflow rate: 2.0 cm/s DO: 7–9 mg/L Natural sunlight Operation duration: 100 days	COD: 600 PO_4^{3-}-P: 10 NH_4^+-N: 100 Ca^{2+}: 10 Mg^{2+}:5 mg Fe^{2+}: 5 mg	COD: (< 30[a]) 95.2 TP: (<1[a]) 44 TN: 43.1	Huang et al. (2015)
1 L Continuous flow reactors (CFR)	Temp.: 25 ± 2 °C Seed sludge: Mature bacterial & algal-bacterial AGS (1:1 w/w) Alternative aeration (60 min) and no-aeration (30 min) regime HRT: 6 h Airflow rate: 0.5 cm/s Aver. DO: 7–8 mg/L (aeration); 2–5 mg/L (no aeration) Operation duration: 120 days Illumination: ~ 900–1100 lx (room light; no light control)	COD: 300–600 PO_4^{3-}-P: 10–20 NH_4^+-N: 100–200 COD/N/P = 30:10:1	COD: 43–50 [a] DOC: 96–95 NH_4^+-N: >99 TN: 29–80 TP: 44–50	Ahmad et al. (2019)
0.25 L Shaking glass flasks	Temp.: 25 ± 2 °C Cycle: 12 h Filling: 1 min Shaking: 715 min Settling: 2 min Effluent discharge: 2 min Shaking: 150 rpm VER: 50 % HRT: 24 h SRT: ~30 days Operation duration: 25 days Seed sludge: Mature algal-bacterial AGS Light on/off period: 12 h/12 h Light intensity: 88–122 μmol m^{-2} s^{-1}	COD: 400 PO_4^{3-}-P: 10 NH_4^+-N: 50	DOC: (<14[a]) 94.4–94.8 TP: 55 TN: 71 NH_4^+-N: >99	Zhao et al. (2019)
SBRs	Temp.: 23 ± 2 °C Cycle: 4 h Feeding: 2 min No aeration: 28 min of Aeration: 200 min Settling: 5 min Decanting: 3 min Idling: 2 min VER: 50 % HRT: 8 h SRT: 40–50 days Airflow rate: 3 L/min DO: 7 mg/L Light on/off: 12 h/12 h Light intensity: 180 μmol m^{-2} s^{-1} Seed sludge: Sewage sludge	COD: 600 PO_4^{3-}-P: 10 NH_4^+-N: 50	TOC: 97.5 PO_4^{3-}-P: 57–63 NH_4^+-N: > 99 TIN: 69.8–71.3	Meng et al. (2019b)
0.92 L SBR	Temp.: 20 ± 2 °C Cycle: 4 h Feeding: 2 min No aeration: 60 min Aeration: 172 min Settling: 3 min Decanting: 2 min Idling: 1 min pH: 7.4 VER: 50 % HRT: 8 h SRT: 23 days Airflow rate: 0.8 L/min Light on/off: 12 h/12 h Light intensity: 835 μmol$m^{-2}s^{-1}$[a] Seed sludge: Mature AB-AGS	COD: 500 PO_4^{3-}-P: 10 NH_4^+-N: 50	COD: > 98 PO_4^{3-}-P: 71 NH_4^+-N: > 99 TN: 78	Wang et al. (2021)
2 L SBRs	Temp.: 23 ± 2 °C Cycle: 4 h		COD: 95 PO_4^{3-}-P: 31–42	Meng et al. (2019c)

(continued on next page)

Table 2 (*continued*)

Reactor volume and type	Operation conditions	Influent concentration (mg/L)	Effluent quality (mg/L) and removal efficiency (%)	Reference
	Feeding: 2 min No aeration: 28 min Aeration: 190–200 min Settling: 5–15 min Discharge: 5 min VER: 50 % HRT: 8 h Airflow rate: 3 L/min DO: 7–9 mg/L Operation duration: 120 days Light on/off: 12 h/12 h Light intensity: 45–225 μmol m^{-2} s^{-1} Seed sludge: Mature algal-bacterial AGS	COD: 600 PO_4^{3-}-P: 10 NH_4^+-N: 50	NH_4^+-N: > 99 TN: 61–80	
0.84 L Sealed glass reactor	October–November weather in Wuhan city, China (Open terrace) Temp.: 13–19 °C Operation duration: 30 days Light on/off: 12-h day cycles Light intensity: 60–400 μmol m^{-2} s^{-1} CO_2: 52 mL (99.9 % purity) /148 mL air Seed sludge: Mature bacterial AGS	COD: Glucose: 250 Peptone: 80 Urea: 15 Meat extract: 55 PO_4^{3-}-P: 3.7 NH_4^+-N: 19.2	COD: 78.3 TP: 95 PO_4^{3-}-P: 31–42 NH_4^+-N: 85.4 TN: 84.5	Sun et al. (2022)
4 L Photo SBR	4-h cycle: Feeding: 30 min No aeration: 90 min Aeration: 190–204 min Settling: 15–1 min Discharge: 5 min VER: 50 % HRT: 8 h SRT: 10 days Airflow rate: 2 L/min DO: 3–4 mg/L Light on/off: 12 h/12 h Light intensity: 3000 μmol m^{-2} s^{-1} Operation duration: 100 days Static magnetic field: 5 mT Seed sludge: Sewage sludge	COD: 400 PO_4^{3-}-P: 12 NH_4^+-N: 70	COD: 91 TP: 95 PO_4^{3-}-P: 71.5–83.3 NH_4^+-N: 96.6 TN: 49.3	Zhang et al. (2022b)
0.06 L SBR	8 cycles (3 of 8 h and 5 of 6 h, respectively). Light intensity: 200 μmol m^{-2} s^{-1} Seed sludge: Mature bacterial AGS	COD: 552.8 PO_4^{3-}-P: 13.2 NH_4^+-N: 99.4	COD: 92.69 TP: 87.16 PO_4^{3-}-P: 71.5–83.3 NH_4^+-N: 96.84 TN: 84.10	Ji et al. (2020)
6.0 L SBR	Temp.: 22–28 °C Cycle: 8 h Feeding: 3 min Anaerobic phase: 120 min Oxidation phase: 210 min Anoxic phase: 114–142 Precipitation: 2–30 min Settling: 2 min Discharge phase: 3 min DO: 4–5 mg/L Operation duration: 60 days VER: 50 % SRT: 30 days Light intensity: 4000 lx Seed sludge: Mature bacterial AGS	COD: 320 PO_4^{3-}-P: 9 NH_4^+-N: 35	COD: (13.0ᵅ) TP: (0.93ᵅ) 97 PO_4^{3-}-P: 71.5–83.3 NH_4^+-N: 15.9 TN: 0.38	Guo et al. (2021)
0.5 L SBR	6-h cycle Feeding: 3 min No aeration: 90 min Aeration: 262 min Settling: 2 min of settling Discharge: 3 min VER: 50 % pH: 7.5 LED light: 5500 lx Uplift air flow velocity: 0.86–0.87 cm/s Seed sludge: Matura algal-bacterial AGS Operation duration: 25 days	DOC: 150 NH_4^+-N: 50 PO_4^{3-}-P: 10	DOC: 90 NH_4^+-N > 99 TN: 75 NO_2^--N: (0.18^{α}) TP: 64	Zhang et al. (2020b)
0.5 L SBR	4-h cycle Feeding: 6 min No aeration: 60 min Aeration: 161 min 2 min of Settling: 2 min Discharge: 11 min	COD: 300 NH_4^+-N: 30 PO_4^{3-}-P: 5 Ca^{2+}: 10 Mg^{2+}: 5 Fe^{2+}: 5	DOC: 96.6 NH_4^+-N: 99.9 TN: 65 TP: 70	Dong et al. (2021)

(continued on next page)

Table 2 (*continued*)

Reactor volume and type	Operation conditions	Influent concentration (mg/L)	Effluent quality (mg/L) and removal efficiency (%)	Reference
	VER: 50 % SRT: 30 days Aeration: 0.87 cm/s Illumination: 3600 lx			
0.5 L SBR	4-h cycle Feeding: 6 min No aeration: 60 min Aeration: 161 min 2 min of Settling: 2 min Discharge: 11 min VER: 50 % SRT: 30 days Aeration: 0.87 cm/s Illumination: 3600 lx Light duration: 12 h/day Temp: 25 °C	COD: 300 NH_4^+-N: 30 PO_4^{3-}-P: 5 Ca^{2+}: 10 Mg^{2+}: 5 Fe^{2+}: 5 Salinity: 1–3 g/L	DOC: 92–94 NH_4^+-N: 99.9 TN: 63–16 TP: 33–38	Dong et al. (2021)
0.05 L	LED light: 200 μmol/m^2/s Light: 12 h light/12 h dark VER: 70 % Temp.: 25 °C via water bath No aeration/stirring Seed sludge: Bacterial AGS	COD: 281 NH_4^+-N: 11 PO_4^{3-}-P: 3 Ca^{2+}: 20 Mg^{2+}: 50 Fe^{2+}: 40	COD: > 80 NH_4^+-N: 99 PO_4^{3-}-P: 92.3	Hu et al. (2022)
0.04 L SBR	8-h cycle Temp: 30 °C Biomass concentration: 5.7 ± 0.1 g VSS/L LED light: 200 μ mol/m^2/s. Operation duration: 36 continuous cycles No mixing/aeration Seed sludge: Bacterial AGS	COD: 280.91 NH_4^+-N: 11.44 NO_2^--N: 9.86 NO_3^--N: 16.61 PO_4^{3-}-P: 2.83 Ca^{2+}: 20 Mg^{2+}: 50	COD: 64.8 NH_4^+-N: 84.9 NO_2^--N: 70.8 NO_3^--N: 50 PO_4^{3-}-P: 84.2	Fan et al. (2021a)
0.05 L	Batch reactors HRT: 8 h LED light: 70, 140, 210 μmol/m^2/s No mixing or aeration VSS/SS: 0.86 pH: 7.0. Temp.: 26 °C	COD: 400 NH_4^+-N:50 PO_4^{3-}-P: 5 Ca^{2+}: 20 Mg^{2+}:50 Fe^{2+}: 40	COD: 52.1–70.5 NH_4^+-N: 64.0–80.7 PO_4^{3-}-P: 73.9	Fan et al. (2021b)

BOD, biochemical oxygen demand; COD, chemical oxygen demand; DO, dissolved oxygen; DOC, dissolved organic carbon; HRT, hydraulic retention time; LED, light-emitting diode; TN, total nitrogen; TP, total phosphorus; SS, suspended solids; SRT, solids retention time; VER, volumetric exchange ration; mT, Millitesla; [α] effluent concentration (mg/L).

Table 3
WWTP effluent discharge and reuse standards in different countries.

Parameters	COD (mg/L)	BOD (mg/L)	NH_4^+-N (mg/L)	TN (mg/L)	TP (mg/L)	pH	TSS (mg/L)	Reference
The EU (Agricultural irrigation)	125	≤10	NA	10	1	NA	35	EPC (2020)
The UK	125	25	NA	10	1	NA	35	Oleszkiewicz et al. (2015)
The Netherlands	125	20	NA	7	1	NA	30	Pronk et al. (2015)
The United States (urban/irrigation use)	25–30	10–30	NA	NA	NA	6–9	≤ 30	USEPA (2012); Sauder (2018)
China (Class I-A)	50	10	5(8)[a]	15	0.5	6–9	10	GB18918-2002[b]
China (Class II)	100	30	25(30)[a]	NA	3	6–9	20	GB18918-2002[b]
Northern Territory and Victoria (Australia)	NA	10–20	NA	NA	NA	NA	10–30	NTG (2020)
State of Victoria (Australia) (Class A)		< 10	NA	NA	NA	6–9	< 5	SVEPA (2021)
State of Victoria (Australia) (Class B)		< 20	NA	NA	NA	6–9	< 30	SVEPA (2021)
Canada (Manitoba)	25	25	1.25	15	1	NA	25	Oleszkiewicz et al. (2015); CWN (2018)
India NGT 2019	50	10	NA	10	1	5.5–9	20	Schellenberg et al. (2020)
Egypt (Agricultural irrigation) Indirect reuse	50–80	30–60	NA	5–15	1–3	6–9	30–50	Elbana et al. (2017)

COD, chemical oxygen demand; BOD, biochemical oxygen demand; USEPA, United States Environmental Protection Agency; EU, European Union; NA, not available; TN, total nitrogen; TP, total phosphorus; TSS, total suspended solids; UK, United Kingdom; EPA, Environment Protection Authority.

[a] Data outside the brackets are concentrations at water temperature > 12 °C; those inside the brackets are concentrations at water temperature ≤ 12 °C.

[b] https://www.mee.gov.cn/ywgz/fgbz/bz/bzwb/shjbh/swrwpfbz/200307/W020061027518964575034.pdf.

conditions, and the WWTP effluent discharge and reuse standards in different countries are listed in Table 3.

Besides ethical considerations, the standards for effluent discharge and reclaimed water use differ by region and local conditions. As shown in Table 3, secondary biological treatment is common for meeting acceptable discharge standards and non-direct consumable agriculture use in most developed economies. AGS systems with lower effluent nutrient and organics concentrations are excellent for more reclaimed water use at lower cost. Considering high treatment cost influences reuse (Liao et al., 2021), effluent quality standards are mostly unattainable by

Table 4
Resource recovery from bacterial and algal-bacterial AGS systems.

Wastewater source	Operation scale and volume	Effluent quality (mg/L)	Recoverable resources		Reference
			Bacterial AGS	Algal-bacterial AGS	
Low strength wastewater	110 L SBR	NH_4^+-N: 11.48 TP: 4.8	ALE: 236 ± 27 mg/g VSS		Schambeck et al. (2020a)
Synthetic (acetate/propionate based) wastewater	Lab scale	NA	ALE: 261 ± 33 mg/g VSS		Schambeck et al. (2020b)
Synthetic saline wastewater	Lab-scale SBR 2 L	TOC: 5–10 NH_4^+-N: ~0 PO_4^{3-}-P: 0.1	ALE: 26.8–49.8 mg/g VSS		Meng et al. (2019b)
Synthetic wastewater	5 SBRs 7.8 L	COD: 24–80 NH_4^+-N: 1.4–6.3 PO_4^{3-}-P: 3.6–8.2	ALE: 180–418.7 mg/g VSS Tryptophan: 0.9–4.1 mg/g VSS		Ferreira dos Santos et al. (2022)
Low strength municipal (raw and settled) wastewater	Lab-scale SBR 28 L	COD: 55–130 NH_4^+-N: 25–72 PO_4^{3-}-P: 0	PHA: 10.8 and 9.3 %		Karakas et al. (2020)
Synthetic wastewater	Lab-scale SBR 0.9 L	DOC: <8.97	P: 0.29 kg/day	P: 0.56 kg/day ALE: 13.37 mg/g VSS	Chen et al. (2022)
Synthetic wastewater	Lab-scale SBRs 16 L	NA	TP: 33.43 ± 0.69 mg/g-SS P: 25.10 ± 1.85 mg/g-SS	ALE: 8.81 mg/g-VSS TP: 27.54 ± 0.23 mg/g-SS P bioavailability: 97 %	Chen et al. (2021)
Synthetic wastewater	Lab scale 2 L	COD: 32.6	Lipids: 34.6 mg/g-SS	Lipids: 57.4 mg/g-SS	Zhang et al. (2020a)
Synthetic wastewater	Lab scale 2 L	NH_4^+-N removal: >99 %	Lipids: 33.4 mg/g-SS	Lipids: 68.7 mg/g-SS	Huang et al. (2020)

ALE, alginate-like exopolymers; COD, chemical oxygen demand; DOC, dissolved organic carbon; NA, not available; P, phosphorus; PHA, polyhydroxyalkanoates; SS, suspended solids; TOC, total organic carbon; TP, total phosphorus; VSS, volatile suspended solids.

conventional treatment processes. Meanwhile, reclaimed water's economic value is an entry point for resource recovery advancement.

5.2. Resource recovery and potential applications

The last decade has seen an increasing focus on resource recovery from AGS biotechnology in research publications (Fig. 2), mostly in the past five years. Meanwhile, bacterial AGS is observed to have an increasing focus on alginate-like exopolymers (ALE), polyhydroxyalkanoates (PHA), tryptophan, and P recovery (Amorim de Carvalho et al., 2021). AGS-based PHA production can maximize waste for bioplastic development, reducing overall operational cost. Chen et al. (2021, 2022) recently reported that algal-bacterial AGS biomass has a higher potential for simultaneous P and ALE recovery than bacterial AGS. Meanwhile, ALE has a commercial value of US$ 80–140/kg (Ferreira dos Santos et al., 2022), and could generate € 1000–2000/t if processing cost being excluded (Tavares Ferreira et al., 2021). Moreover, Meng et al. (2019b) reported an enhanced ALE yield under moderately saline conditions for bacterial AGS. This is promising for industrial wastewater treatment and can be researched in the more saline adaptable algal-bacterial AGS (Dong et al., 2021; Semaha et al., 2020). Furthermore, the high P bioavailability in AGS (Zhao et al., 2019) is promising for phosphate biofertilizer production.

Meng et al. (2019a, 2019c) reported that algal-bacterial AGS has high lipids content and productivity for biodiesel production. Additionally, algal-bacterial AGS can accumulate higher crude protein (313.28 ± 26.67 mg/g-VSS) for animal feed production compared to suspended microalgae cells (174.10 ± 11.47 mg/g-VSS) (Wang et al., 2022). Although considerable financial investment is required to address bottlenecks and realize algal-bacterial AGS in future full-scale applications (Zhang et al., 2022a), its prospects are worthwhile. Besides, a circular economy transition will require a change of mindsets and the commitment of governments and the private sector to attain (Guerra-Rodríguez et al., 2020). This could potentially increase the economic benefits of modern wastewater treatment from varied value-chain opportunities into the end of the millennia from AGS biotechnology. Table 4 summarizes the results of resource recovery from AGS in recent publications.

6. Conclusions

The century-long dynamic challenges of society have dictated technical innovations and the development of WWTPs. Moreover, the environmental suitability and resource recovery potentials of biological treatment processes have established them as preferred options to realize environmental safety and sustainable development transition for the circular economy. Although the CAS process remains in sanitation realization for urban environments, the prominent AGS systems are most adaptable to the needs of society. Bacterial/algal-bacterial AGS systems can drive the modern wastewater treatment from merely pollutants removal and sludge stabilization to focusing on material recycling, water reuse, energy/nutrients recovery, and value-added biomaterials production with climate-smart thinking.

CRediT authorship contribution statement

Philip Semaha: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Zhongfang Lei:** Formal analysis, Funding acquisition, Project administration, Supervision, Writing – review & editing. **Tian Yuan:** Writing – review & editing. **Zhenya Zhang:** Writing – review & editing. **Kazuya Shimizu:** Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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