



Perspective

Carbon neutrality of wastewater treatment - A systematic concept beyond the plant boundary



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ABSTRACT

Recently, every industry has been working to achieve carbon neutrality, and the wastewater sector is no exception. However, little research focuses on the carbon accounting of wastewater treatment and the roadmap to carbon neutrality. Here, to systematically perform accounting, we provide a sketch that describes three boundaries of the wastewater system and propose that the carbon neutrality of the wastewater system is far beyond the plant boundary. Moreover, we identify the direct and indirect carbon emissions of wastewater treatment. In addition to direct emissions of CH₄ and N₂O, direct fossil CO₂ emissions from wastewater treatment should be included in accounting to set accurate guidelines. Next, the technologies that assist in achieving carbon-neutral wastewater treatment both within-the-fence of wastewater treatment plants and beyond the plant boundary are summarized. All measurements of energy recovery, resource recovery, and water reuse contribute to reaching this goal. The concepts of energy neutrality and carbon neutrality are identified. Successful wastewater treatment cases in energy self-sufficiency may not achieve carbon neutrality. Meanwhile, resource recovery methods are encouraged, especially to produce carbon-based materials. Ultimately, the trend of preference for the decentralized sewage treatment system is pinpointed, and systematic thinking to set the urban infrastructure layout as a whole is advocated.

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1. Introduction

In 2015, the Paris Climate Agreement was signed to limit the temperature rise to within the 2-degree threshold. To reach this objective, many countries have announced their carbon neutrality targets. Again, in 2021, leaders from 200 countries attended the COP26 to review and update their Nationally Determined Contributions regarding climate change [1]. As an essential part of urban sanitation, wastewater treatment can contribute approximately 1–2% of the total greenhouse gas (GHG) emissions in the world [2,3].

Currently, decreasing carbon emissions has become an urgent objective in the wastewater industry. Many effective measures such as sludge anaerobic digestion are practiced to pursue

environmental sustainability [4]. Meanwhile, emerging wastewater treatment technologies such as anaerobic ammonia oxidation processes and microbial fuel cells are moving out of laboratories and into the field [5,6]. The technical effect and economic profit of these methods have been evaluated, and optimistic results have been obtained [7]. Although several scholars have also assessed their multi-environmental impacts [8], few studies have been mainly conducted on their carbon emissions or the standardization of accounting.

Thus, here, we systematically analyze the scope and inventories of carbon emissions of wastewater system and carbon credit of environmentally friendly measures. First, a sketch that describes the boundaries for carbon accounting of wastewater system is provided. Then, GHG emission sources of the wastewater treatment are identified. On these bases, we point out the roadmap to carbon-neutral wastewater system both within-the-fence of wastewater treatment plants (WWTPs) and beyond the plant boundary. Since CO₂, CH₄ and N₂O are the three main GHGs to consider in

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wastewater treatment, in this study, carbon neutrality refers to the neutrality of these three GHG emissions, which is the net zero sum of CO₂, CH₄ and N₂O emissions (in CO₂ equivalent).

2. Multiple boundaries for carbon accounting of the wastewater system

The implementation of WWTPs' basic function, removing pollutants and providing a good sanitation environment, cannot be done independently and require cooperation with other municipal facilities such as sewer system, sludge treatment and disposal sites. Currently, wastewater treatment facilities are undergoing a transition from traditional functions into those that include energy recovery and resource recovery. Based on these two considerations, the scope of carbon accounting in the field of wastewater treatment can be gradually expanded from wastewater treatment processes to the entire human society and ecological system (Fig. 1). In the methodology, a "system expansion" approach is adopted here to solve the multifunctionality of WWTPs. The products reclaimed from wastewater may replace similar products on the market, which implies avoided production and a negative contribution to the carbon emissions of the system.

Within-the-fence of WWTPs. Generally, when accounting for the carbon emission inventories of the wastewater system, the most basic and indispensable part of the statistics is the water line, which starts from sewage flowing in and ends with the effluent of physical, chemical, and biological treatment processes. Due to different regional regulations and use purposes of the effluent, wastewater treatment processes are optional. For instance, the effluent from secondary biological treatment can be directly discharged to natural waters or used for irrigation [9]. However, if the effluent is to be reused for industrial production or as reclaimed water, a tertiary treatment process such as a membrane reactor must be added [10].

Sludge is an inevitable byproduct of wastewater treatment, and pollutants may transfer from wastewater to sludge. Thus, sludge

treatment and disposal are integral parts of wastewater treatment, which should definitely be included in carbon emission accounting [11]. After thickening, there are several routes for sludge treatment and disposal [12]. Conditioning and dewatering are routine processes of sludge treatment to reduce volume. The processes of anaerobic digestion (AD), composting, and pyrolysis are possible methods to recover energy and resources from sludge. After stabilization, the sludge is transported to landfill sites, incineration furnaces, or farmlands for disposal. The carbon emissions of sludge treatment within WWTPs are usually included here in the first circle. The emissions of sludge treatment and disposal out of WWTPs can be counted in the second circle.

In addition to water line and sludge line, other energy conservation and exploitation measures to pursue energy self-sufficiency within the fence of WWTPs, such as water source heat pumps, wind power, solar energy, etc., are considered in the first circle.

Expanding to urban infrastructure related to WWTPs. Before flowing into WWTPs, wastewater is collected and transported along the sewer system. WWTPs and sewer system are physically connected and form a fairly complete urban wastewater system. Moreover, because sewage treatment is being given more functions, systematic management with other urban municipal facilities has become more frequent. For example, municipal waste, especially food waste, can be transported to WWTPs and co-digested with sludge to improve the biogas production of the AD process while reducing the sludge amount for incineration or landfill. In addition to energy recovery, some WWTPs with tertiary treatment processes can be considered reclaimed water plants, which are alternative sources for municipal water or industrial water supply. Thus, the carbon neutrality of wastewater treatment is far beyond the boundary of the treatment plant; it is relevant to the entire water/wastewater system and even the entire urban infrastructure.

Further expanding to human society and ecological system. Natural water, soil-vegetation, and human society together form the third circle. Water bodies serve as water sources for human activities and as ultimate receiving sites for treated wastewater. The

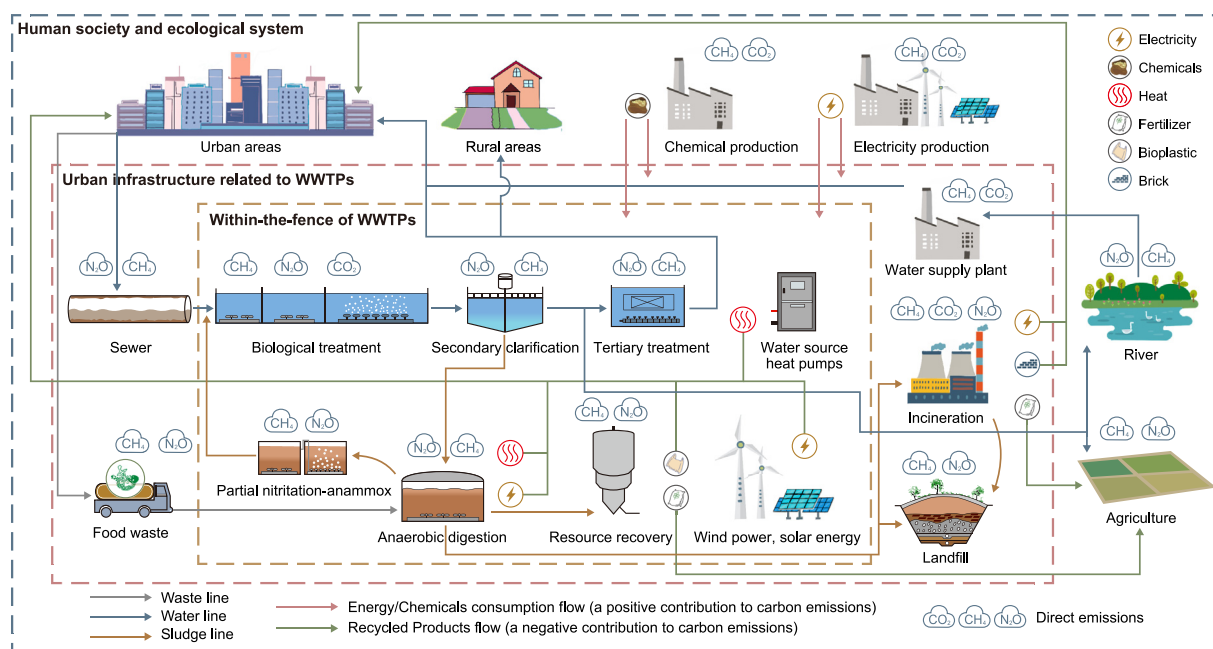


Fig. 1. A sketch of multiple boundaries for carbon accounting of wastewater treatment. Three boundaries: Within-the-fence of WWTPs (yellow dashed line), urban infrastructure related to WWTPs (pink dashed line), human society and ecological system (blue dashed line). Since the operation of most facilities in boundaries 1 and 2 requires energy and chemicals, their inputs are simplified as flows pointing into the boundary. Direct carbon emissions are presented by specific symbols. Indirect carbon emissions are implied in the flows of energy/chemicals consumption and recycled products, which represent positive and negative contributions to carbon emissions, respectively.

Table 1
Quantities and global warming impacts of CH₄ and N₂O emissions in WWTPs.

Reference	Inventory	Quantity ^a (g CO ₂ m ⁻³)	Global Warming Impact ^b (g CO ₂ m ⁻³)	Data Source
[16]	CH ₄	11.36	284	Measurement
	N ₂ O	1.83	546	
[18]	CH ₄	2.40	60	Simulation
	N ₂ O	0.81	240	
[20]	CH ₄	2.384	59.6	Simulation
	N ₂ O	0.479	142.6	

^a The quantity values refer to the wastewater flow rate.

^b Global warming potential: 25 kg CO₂ per kg CH₄ for CH₄ and 298 kg CO₂ per kg N₂O for N₂O.

resources recovered from wastewater treatment processes are distributed to human society and agricultural system, which can reduce the demand for corresponding products manufactured by other industrial processes. Moreover, the layout in urban or rural areas may shift the scale of sewage treatment. Generally, the centralized sewage treatment system is mostly applied in densely populated urban areas, and the decentralized sewage treatment system is more appropriate for rural areas. Although both systems have their strengths, there is a trend to make decentralization a preferred system where applicable in considering resource recovery [13].

3. Inventory for direct and indirect carbon emissions within-the-fence of WWTPs

Generally, the GHGs generated by wastewater treatment can be divided into two parts: direct and indirect carbon emissions. Direct carbon emissions usually refer to onsite CH₄ and N₂O emissions, which are usually generated by microbial metabolic activities throughout the processes of wastewater treatment and sludge treatment/disposal. Indirect carbon emissions mainly stem from the consumption of energy and resources. From a life cycle perspective, they can be traced to the production of electricity, production of chemicals and even transportation of chemicals, which are considered background processes for wastewater treatment. Any type of incomplete carbon inventory, regardless of whether direct or indirect emissions sources are ignored, will result in an underestimation.

Direct carbon emissions. CH₄ is mainly produced by anoxic and anaerobic degradation of organic matter along both water line and sludge line. Based on whether the generated CH₄ is collected, it can be divided into two situations. If effectively utilized, it can be considered an energy source and indirectly diminish CO₂ emissions. However, if released into the atmosphere through leakage or incomplete combustion, it will cause a positive greenhouse effect. N₂O is mainly generated by a series of nitrifying and denitrifying microorganisms. Based on the production mechanisms, CH₄ and N₂O are mainly produced in oxygen-limited conditions, anaerobic processes for CH₄ and anoxic processes for N₂O [14,15]. However, in the anaerobic/anoxic/oxic (A²/O) process, a plug-flow system, the aerobic tank has been identified as one major emission source of CH₄ and N₂O [16,17]. This phenomenon shows that the place of GHGs generated may not necessarily coincide with the place of GHGs released, which can be explained by the specific solubility of CH₄ and N₂O. In other words, only the dissolved GHGs generated in the pre-anaerobic tank or sedimentation tank are stripped off and released by aeration.

The estimated direct emissions of CH₄ and N₂O from the wastewater sector were responsible for over 5% of global non-carbon dioxide GHG emissions in 2005 [18]. Meanwhile, an increase of 22% was predicted by 2030 [18]. Moreover, from the point of carbon footprint within one WWTP, the contribution of direct gaseous emissions indeed plays an important role, especially for

those with high influent concentrations of pollutants [19]. Although the quantity of N₂O emissions is less than that of CH₄ emissions (Table 1), N₂O emissions usually have much higher impact than CH₄ emissions [16,18,20]. This is mainly because that N₂O is a potent GHG with a global warming potential (GWP100) 298 times larger than that of CO₂ and approximately 10 times larger than that of CH₄ [21].

The accounting of direct CO₂ emissions lacks consistency. It was excluded by the Intergovernmental Panel on Climate Change (IPCC), given the reason that they are generally derived from biogenic organic matter in human excreta or food waste. However, this assumption ignores the presence of fossil carbon, such as pharmaceuticals and personal care products, which may actually add the amount of direct carbon emissions. An estimation conducted in four Australian WWTPs demonstrates an underestimation of 2–12% of direct carbon emissions if fossil CO₂ emissions are ignored [22]. In comparison, this inaccuracy may vary according to wastewater compositions. Some industrial wastewater, especially from petrochemical plants, inevitably emits more non-biogenic CO₂ [23]. Since fossil CO₂ emissions from wastewater are gaining more attention, the IPCC (2019) amended its 2006 guidelines with a discussion of it. In short, including fossil CO₂ in GHG accounting is necessary for setting accurate guidelines.

Indirect carbon emissions. To run a WWTP, the inputs of electricity and chemicals are indispensable. It is estimated that approximately 3–4% of the total global electricity consumption is attributable to wastewater treatment [3,24]. Specifically, within WWTPs, the aeration system, lifting pumps, and sludge dewatering units contribute the main share of the total electricity consumption [25]. Chemicals are mainly dosed to enhance the phosphorus removal and sludge dewatering or utilized as carbon sources for denitrification. Recently, for WWTPs subjected to stringent discharge standards, additional chemicals have also been in large demand, such as coagulation agents for advanced treatment and cleaning agents for membrane filters.

Based on the calculation of the carbon footprint within one WWTP, electricity consumption is an essential source of GHG emissions, and the indirect carbon emissions of chemical use are limited [19]. However, it is worth noting that, influenced by the IPCC accounting framework of excluding CO₂ emissions from bioprocess, the in situ non-biogenic carbon emissions of the added methanol or acetate to enhance denitrification were rarely counted, which might underestimate the carbon contribution of chemicals [26]. Thus, direct fossil CO₂ emissions are again emphasized to be included in GHG accounting.

4. Roadmap to carbon-neutral wastewater treatment: within-the-fence of WWTPs

Energy saving and recovering. The cost of electricity is high to run a WWTP. To reduce economic cost, applying energy conservation technologies is the first and most effective solution. Among

many types of wastewater treatment processes, the oxidation pond, which is a natural sewage treatment system, has the highest energy efficiency [27]. However, its shortcomings such as occupying large areas and releasing odors may seriously limit its application. For an existing WWTP, the most practical method is to upgrade obsolete equipment and apply real-time controllers, since the quantity and quality of wastewater always fluctuate, and the steady operation of equipment is not appropriate [28]. Moreover, applying automatic control within WWTPs has been demonstrated to achieve multiple benefits in a case study, which can save energy and reduce GHG emissions by up to 9.6% [29].

Literature statistics show that the electricity used to run one WWTP is usually 0.3–0.6 kWh m⁻³ [30]. The thermal energy of combustion of the organic compounds in wastewater is approximately 9–10 times greater than this value [31], so recovering the chemical energy contained in sewage is economically profitable. The most feasible method is using biogas produced by AD to generate power and heat, which has been widely practiced. Generally, this technology can provide a quarter to half of the energy needs in one WWTP with conventional aerobic treatment [24,25]. To further increase energy recovery, the co-design of wastewater processes is beneficial. For example, a two-stage activated sludge process (A/B process) is usually implemented to sequester more organics from wastewater to sludge [32]. To boost the methane production process and reinforce process stability, co-substrates such as organic fractions of municipal solid waste may be added, and/or sludge pretreatment methodologies such as thermal hydrolysis processes can be applied [4]. One relatively successful case in the WWTP of Strass (Austria) shows that the recovered chemical energy can compensate for up to 80% of the annual total energy consumption in 2003 [5]. With other modifications, including organic waste addition, the energy autarky in the Strass WWTP was achieved at 158–178% in 2012 [5].

Technical innovations for energy saving and recovering.

Applying anaerobic wastewater treatment such as upflow anaerobic sludge beds (UASBs) and expanded granular sludge beds (EGSBs) is another promising option to recover energy. Based on the theoretical calculation, anaerobic wastewater treatment can double the CH₄ production over sludge digestion in conventional treatment for a WWTP with a chemical oxygen demand (COD) concentration of 500 mg L⁻¹ [24]. However, the limited organic matter content in some domestic wastewater may restrict the application of well-established anaerobic reactors [33]. Recently, anaerobic membrane bioreactors (AnMBRs) have been developed. Coupling membranes in the anaerobic process can retain the suspended solids instead of letting them flow away [34]. By prolonging the materials' degradation time, AnMBR may provide possibilities for low-strength municipal wastewater [35]. However, membrane fouling becomes the greatest challenge that prevents this technology from scaling [24].

Bioelectrochemical systems (BESs) can directly convert organic energy into electricity or valuable products such as methane or hydrogen [36]. Although this is hoped to achieve more efficient conversion, restricted to the low reaction rate, huge efforts are required to transform it into a practical technique [6]. For example, one pilot MFC operated in Harbin, China performed poorly and only converted 7% of the embodied energy in organic substances to electricity [35].

Among these emerging technologies, the anammox process has been successfully applied in practice and shows significant advantages compared with the conventional nitrogen removal process. For example, based on theoretical calculations, the need for external carbon sources by applying the anammox process decreases by 100%, aeration demand by approximately 60%, and sludge production by approximately 90% [37]. However, this

process is mainly used as sidestream treatment, and turning it into mainstream processes remains challenging.

Energy self-sufficiency is a narrow sense of carbon neutrality.

As introduced in the inventory section, energy is heavily consumed in both water line and sludge line for running equipment. Reducing the external input of energy and achieving energy self-sufficiency have become important goals for many WWTPs. By applying energy conservation and exploitation technologies, there are opportunities for WWTPs to achieve energy neutrality. However, it should be noted that energy neutrality and carbon neutrality are two different terms. In the definition of energy neutrality, "breaking even" is checked within the plant boundary and easy to meter with online facilities. However, carbon neutrality traces all life-cycle GHGs within and beyond the plant boundary. Since gases are fugitive, even in-plant emissions, especially from open top tanks, are difficult to measure. Unlike energy neutrality, which is powerfully driven by economic benefits, research and practice on carbon-neutral WWTPs from long-term environmental benefits is just getting started.

In some cases, the endeavor on energy neutrality may lift the other end of the carbon seesaw. For example, advanced nitrogen removal technologies such as the successful implementation of the sidestream partial nitrification-anammox process in the Strass WWTP hold considerable advantages in external carbon sources saving, meanwhile, illustrated a reduction of electricity consumption by 36–44% [38]. Nevertheless, partial nitrification-anammox, simultaneous nitrification-denitrification and nitrification-denitrification processes may emit more N₂O than traditional nitrification/denitrification processes due to NO₂⁻ accumulation, limited oxygen and other operating conditions [14]. For another example, the energy recovery technologies from wastewater, such as methane recovered by AD, may come at the cost of GHG emissions. In the biogas collection and purification process, methane leakage inevitably occurs. In this situation, methane should be counted as GHG instead of an energy source. As mentioned above, gas production is closely related to the organic content of sludge. In some poorly operated sewage plants, energy is still consumed to maintain temperature, and chemicals are dosed to break down the cell wall to enhance the biodegradability of sludge, which implies indirect carbon emissions. From a life cycle perspective, the carbon emissions of the AD process may not necessarily be net zero [39,40].

Engineering measures for energy production beyond the water and sludge lines.

Due to the temperature differences, the thermal energy contained in wastewater offers another source to indirectly offset the energy demand for wastewater treatment [41]. The calculated amount of thermal energy that can be recovered is 6–8 times larger than the amount of chemical energy recovered by AD [41]. Thus, the water source heat pump technology has great potential to facilitate WWTPs approaching the carbon-neutral goal, if the thermal energy recovered can be associated with an effective municipal heat grid and be sufficiently utilized [25]. Thus, it is a technology with low technical difficulty but strict application scenarios where the distance of heat transport is limited. In Northern European countries, for example, wastewater source heat pumps are widely applied, and it has been reported that 3% of all buildings in Switzerland and Germany can be heated or cooled in this manner [42].

In addition to utilizing the chemical energy and thermal energy contained in wastewater, installing facilities of new energy sources such as solar energy, has been practiced within WWTPs. Due to the need for technological procedures, most WWTPs have large architectures such as biological reaction tanks and secondary sedimentation tanks, which usually have large installation spaces for photovoltaic systems. For example, the Aquaviva WWTP in Cannes,

France, which has achieved carbon neutrality, installed 4000 m² of photovoltaic panels to produce electricity [25]. Another example in Pennsylvania was a 3-MW solar project completed in a WWTP, which is expected to produce more than 3 million kWh annually and is sufficient to avoid 3515 tons of CO₂ emissions [43].

Resource recovery within the plant. Since wastewater contains large amounts of organics and nutrients, emerging treatment processes have been developed to capture and transform these valuable resources into value-added products [44–46] such as struvite, vivianite, biodiesel, bioplastics, biochar, and protein. Moreover, it has been demonstrated that resource recovery within WWTPs plays an important part in achieving carbon neutrality. For example, the reduction effects of the struvite precipitation process on global warming were simulated to vary from 3% to 38% [19].

It is worth noting that a competitive relation exists between energy recovery and resource recovery. For instance, bioplastic polymers are produced as carbon storage materials by various microorganisms, which may compete for organics with the sludge anaerobic digestion process [47]. Thus, there is a trade-off between these energy and resource processes. On one hand, with the advent of the energy technology revolution, producing cleaner electricity may become more common. Consequently, recovering energy from wastewater may be less attractive. On the other hand, the production of carbon-based materials from wastewater treatment can promote CO₂ sequestration and decrease GHG emissions [3]. Thus, it is supposed that WWTPs will achieve more obvious benefits in resource recovery in the future [48].

5. Roadmap to carbon-neutral wastewater treatment: beyond the plant boundary

Upstream of the WWTPs. In the current sanitation system, municipal wastewater is mainly collected by sewer pipes. The space of sewer pipes is relatively closed with poor air ventilation. Because it is easy to form an oxygen-deficient environment, organic matter in sewage is prone to anaerobic fermentation to produce CH₄. The manholes of rising mains and the outlet of the sewage conveyance system are hotspots of CH₄ emissions [49]. Although the IPCC considers closed sewers as negligible sources of carbon emissions, one field measurement in Australia shows that significant quantities of CH₄ are produced in sewer systems. According to rough estimates, the sewer system generated an additional GHG contribution of 12–100% for methane compared to the WWTP itself [50].

Downstream of the WWTPs. After being processed, the treated effluent is typically discharged to a receiving water environment (e.g., river, lake, estuary). Direct carbon emissions may still be generated in the receiving water bodies, although the quantities may be limited compared with those produced within WWTPs [51]. In addition to being directly discharged, treated effluent can be reused to water trees or clean streets. This is a typical behavior crossing multiple boundaries; since the reclaimed water production process is conducted in the first circle, the recycled water is applied in the third circle and accomplished through the infrastructure in the second circle. Certainly, a negative carbon effect is expected, since the enclosed reclaimed water system can replace the water works. However, whether carbon emissions from tertiary treatment are offset by carbon emissions from reused waters remains in doubt. For example, the estimation of the WWTP in Taragona, Spain, shows a slight increase in carbon emissions if wastewater is reused for the industrial process [52].

After the initial treatment in WWTPs, the sludge is transported outside for final disposal, which can be generally distributed to landfills, incineration, land application, etc. Among the three common approaches, landfilling is the most widely applied due to its relatively low cost and simplicity. The carbon emissions during

the sludge disposal process are closely related to the technical routes. Generally, energy and resource recovery measures are essential to reduce environmental and economic burdens. To this end, methods on resourceful utilization of sludge have been gradually developed and play a much larger role. For example, heat can be recovered through sludge incineration, and the ash produced after incineration can be utilized as building materials or transformed into fertilizers [39]. When technology constantly evolves, uncertainties remain regarding whether carbon emissions are reduced [39,40].

Resetting and reassessment of the water system. Over the past few decades, in most urban areas, the centralized sewage treatment system has been mainly adopted. Sewers collect municipal wastewater from homes, industries, and businesses and deliver it by gravity and pumping to WWTPs. Since WWTPs are usually located in a low-lying place, the reclaimed wastewater must be lifted back to the areas of users. Thus, a large amount of energy is demanded for pumping, which adds indirect carbon emissions [24]. In other words, although the management of a centralized sewage treatment system is mature, it has fewer advantages in the sustainable development of wastewater treatment [53].

To solve these problems, an alternative approach is to apply a decentralized wastewater treatment system, i.e., a paradigm that can close the water loop [54]. A decentralized system outcompetes the centralized system in less energy input and more efficient resource recovery that is not limited to water reuse. In a decentralized wastewater treatment system, short-distance conveyance may eliminate anaerobic environmental factors, methane emissions may be alleviated, and the energy consumption for pumping may be partly saved [55,56]. Moreover, keeping the higher temperature of raw wastewater, decentralized heat recovery from warm water sources at the household level holds a higher potential to extract useful thermal energy [48]. With the development of source separation technology, nutrient resources can also be more efficiently recycled [57].

Systematic thinking is required to achieve carbon neutrality in wastewater treatment. Since the effective recovery of energy depends on the high organic content of the influent wastewater in WWTPs [19,31], the sewer collection system can be coordinated and changed to achieve this goal. Constructing rain and sewage diversion system to avoid dilution of pollutant concentrations, deprecating urban septic tanks to reduce unnecessary consumption of organic matter, and maintaining the drainage system to prevent groundwater from backlogging are recommended as best management practices [58]. Meanwhile, low impact development (LID) technologies such as green roofs and bioretention facilities mainly focus on the source control of stormwater management and are designed to prevent the waterlogging and overloaded operation of WWTPs [59]. In China, under the concept of sponge city construction, based on the technology route of source reduction, process control and system governance, rearranging the infrastructure layout can systematically achieve the sustainability of wastewater system, with carbon neutrality as one of the goals.

Declaration of interests

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

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