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Wastewater treatment for carbon capture and utilization

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A paradigm shift is underway in wastewater treatment as the industry heads toward ~3% of global electricity consumption and contributes ~1.6% of greenhouse gas emissions. Although incremental improvements to energy efficiency and renewable energy recovery are underway, studies considering wastewater for carbon capture and utilization are few. This Review summarizes alternative wastewater treatment pathways capable of simultaneous CO_2 capture and utilization, and demonstrates the environmental and economic benefits of microbial electrochemical and phototrophic processes. Preliminary estimates demonstrate that re-envisioning wastewater treatment may entirely offset the industry's greenhouse gas footprint and make it a globally significant contributor of negative carbon emissions.

The Paris Agreement on Climate Change aims to hold the global average temperature increase below 2 °C, and special efforts are made to limit the increase to 1.5 °C by 2100 relative to pre-industrial levels. The United Nations Environment Programme (UNEP) recently analysed the major emission gap to achieve this goal, but it found current state pledges cover no more than one third of the needed emission reductions[1]. To meet the 2 °C target, many studies suggest it is necessary to balance emission reductions with CO_2 removal and the deployment of negative emission methods, because most models indicate it is impossible to reach 1.5 °C with a 50% chance without significant negative emissions[2–4]. Popular negative-emission approaches include bioenergy combined with carbon capture and storage (BECCS), alternative and adjusted agricultural practices such as biochar production and utilization, ambient air capture and utilization, and accelerated natural mineral weathering via electrogeochemical methods coupled with H_2 generation[1,5].

Although many infrastructure sectors such as energy, industry, transportation and building systems have been extensively studied with regards to emission reduction and sustainability, studies in the wastewater industry have been sparse. Because wastewaters are generated everywhere there is human activity, this study strives to discuss the substantial opportunities for carbon capture and utilization (CCU) in the context of the wastewater–carbon nexus. To this end, we first provide a summary and critical review of wastewater treatment pathways with CCU capabilities and then quantitatively compare an illustrative example of a CCU-enabled treatment process against conventional activated sludge with regards to their carbon balance, energy balance and economic implications. The findings from this Review demonstrate the significant potential of the wastewater industry to contribute to negative emission practices while simultaneously protecting the aquatic environment and public health.

Every year, nearly 1,000 km^3 of wastewater is generated around the world, which includes more than 300 km^3 municipal wastewater and more than 600 km^3 industrial wastewater[6–8]. Approximately 70% of these waste streams are treated in developed countries, while the treatment percentage in low-income countries is only around 8%[6,9]. For many cities and towns, wastewater treatment plants (WWTPs) are the largest energy consumers and wastewater treatment could account for up to 3% of global electricity if treated with conventional technologies[10,11]. In addition, WWTPs have been recognized as one of the largest of greenhouse gas (GHG) emitters. Direct GHG emissions at WWTPs stem from biological carbon, nitrogen and phosphate removal, sludge management and off-gas from wastewater collection systems. In addition, indirect GHG emissions are incurred from imported electrical and thermal energy, the production and transportation of chemicals and fuels, as well as waste disposal[12,13]. It was estimated the degradation of organics during wastewater treatment contributed 0.77 Gt CO_2-equivalent GHG emissions in 2010, equivalent to nearly 1.57% of global GHG emissions[14] (49 Gt CO_2e). In particular, wastewater treatment is a major contributor of non-CO_2 GHG (for example, CH_4 and N_2O) emissions, accounting for 0.56–0.71 Gt CO_2e per year between 2005 and 2030, which is equivalent to roughly 4.6–5.2% of the global total non-CO_2 GHG emissions[15]. Although the direct release of CO_2 from the degradation of wastewater organics is largely considered a carbon-neutral process in GHG accounting (despite evidence of some carbon being of fossil origin[16]), the non-CO_2 GHG emissions are of significant concern as they are 25–298 times stronger (100-year time horizon) in greenhouse effect than CO_2 (ref. [17]).

Great progress has been made to increase energy efficiency and recover renewable energy from wastewater using technologies such as anaerobic digestion, anaerobic membrane bioreactors and microbial electrochemical systems, because the chemical energy embedded in wastewater is estimated to be more than nine times that required to treat the wastewater (17.8–28.7 kJ g^{-1} chemical oxygen demand, COD)[18,19]. However, these methods only reduce fossil fuel consumption and its associated carbon emissions, whereas few have looked at the additional possibility of using wastewater treatment for active and direct CO_2 capture and utilization. Considering

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the sizeable amount of wastewater generated each year and its positive correlation with population and industrial activities, wastewater's potential contribution to meet the goals of the Paris Climate Agreement is significant. Additionally, because CCU can occur within existing wastewater infrastructure during treatment, no additional land or transportation may be needed for such operations. Therefore, integrating CCU with wastewater treatment may transform energy-intensive, carbon-emitting WWTPs into integrated water resource recovery facilities (WRRFs) that recover energy, nutrients, water and other valuable carbon products with economic, environmental and social benefits. In this context, the wastewater industry may become a major player in the global carbon market[20,21].

Approaches for CCU-enabled wastewater treatment

The traditional objective of wastewater treatment is to remove carbon, nitrogen, phosphorus and other contaminants (for example, pathogens and suspended solids) so the effluent meets environmental quality regulations. In this context, CCU must be realized without compromising treatment efficacy given that protection of the local aquatic environment and public health will remain paramount for the wastewater industry. If strategically integrated with existing wastewater infrastructure and treatment constraints, CCU may go beyond global environmental benefits and have direct local benefits through product valorization and added alkalinity for better water chemistry and treatment[22]. Here we discuss emerging technologies and processes that may advance CCU from wastewaters and demonstrate the environmental and economic potential of this integration through a quantitative analysis of one particular scenario with integrated operations of treatment and CCU (Fig.1).

Microbial electrolytic carbon capture. This method uses wastewater as the electrolyte for microbially assisted water electrolysis. Microorganisms, especially electroactive bacteria (EAB) in the anode chamber, oxidize biodegradable substances in wastewater to produce electrons, protons and CO_2. Electrons are accepted by the anode and transferred through an external circuit to the cathode, where they reduce water to produce H_2 and OH^- (ref. [23]). The H^+-rich anolyte can liberate metal ions (Ca^{2+}, Mg^{2+} and so on) from abundant silicate minerals (for example, wollastonite $CaSiO_3$) or waste materials (for example, coal fly ash). When the metal ions migrate to the OH^- catholyte, they form metal hydroxide, whose subsequent reaction with CO_2 leads to spontaneous CO_2 capture and transformation into stable carbonate or bicarbonate[22,24–27]:

$$\mathrm{CH_3COOH(aq) + 2H_2O + 6CaSiO_3(s) + 4CO_2(g) \xrightarrow{V_{d.c.}} 6CaCO_3(s) + 6SiO_2(s) + 4H_2(g)}$$

Such materials can be collected for cement production or provide additional alkalinity in water effluent. Compared with abiotic electrolysis, the electrons are from microbial organic degradation, so microbial electrolytic carbon capture (MECC) does not generate toxic chlorine-containing compounds and it produces large quantities of high-purity H_2, which can make the process energy-positive[19].

MECC can be realized in situ in one reactor or ex situ involving separated acid and alkali production. For in situ operation, one study demonstrated that by using both artificial and actual industrial wastewater, almost all CO_2 derived from organic oxidation as well as additional exogenous CO_2 (80–93% of the total CO_2) could be captured[24]. In addition, 56–100% of organics (as COD) were removed depending on the wastewater, and a net energy gain of ~2 kJ mol^{-1} of CO_2 captured was achieved via high-rate H_2 production (>1 m^3 m^{-2} d^{-1})[24]. Similar approaches were not only used in concentrated CO_2 sources (5–15%) but also for ambient CO_2 capture in combination with an ion exchange resin[28]. MECC carries advantages in wastewater treatment as well, because similar microbial electrochemical processes have been operated in both low-strength municipal wastewater (COD = 200–600 mg l^{-1}) and high-strength waste streams such as industrial wastewater and sludge (COD> 5,000 mg l^{-1})[29]. Unlike anaerobic digestion, it can also be operated in low temperatures (~4 °C)[30], avoiding the need for heating and enabling energy efficiencies above 80%. Because MECC is an anaerobic process and can become energy-positive, it is much more energy efficient than traditional activated sludge systems, for which aeration alone can account for 40–60% of an entire treatment plant's electricity consumption. Additionally, the microbial electrochemical process produces ~80% less sludge and therefore reduces the burden of sludge management[23,31]. Because microbial electrolysis is endothermic, it does require a small applied voltage (0.6–0.8 V) to operate[32], though such voltage can be provided by a renewable power source such as a microbial fuel cell, reverse-electrodialysis cell, photocatalysis cell or by a fuel cell leveraging produced H_2 (refs. [26,33,34]). All reported MECC studies to date are still at bench scale, so more research and development is needed to reduce cost, simplify the configuration and increase reaction kinetics[24,35,36]. Additionally, systems need to be tested under actual wastewater conditions and appropriate silicate sources need to be identified based on specific scenarios to make sure the costs are justified and no secondary contamination is introduced. Beyond carbon removal, considerations should also be given to remove nutrients and other contaminants to fulfil the mission of wastewater treatment.

Microbial electrosynthesis. In anode reactions this method shares a similar principle with MECC, in which electroactive bacteria oxidize organics present in wastewater and generate current on the anode[37]. Therefore, microbial electrosynthesis (MES) carries similar benefits in wastewater treatment, such as low energy operation, low sludge production and adaptability to organic loading and changes in environmental conditions[34]. However, instead of using alkalinity change for carbon mineralization, the cathode reaction in MES relies on autotrophic bacteria to capture and convert CO_2 into value-added organic compounds[37]. Though MES is still bench scale, compared with low efficiencies in solar-to-product conversion by plants and photosynthetic microorganisms (<1–3%)[38], the cathode microorganisms could be orders of magnitude higher in energy efficiency to convert CO_2 to organic molecules (>10%) and therefore demonstrate good potential in CO_2 reduction using renewable electrons[39].

There have been excellent reviews discussing microbial electron transfer, logical data representation and economic feasibility of MES for carbon valorization, with wastewater having been considered as one source of renewable energy as well as solar and wind[37,40,41]. Because methanogens and acetogens have been found to be dominant in MES reactors, methane and acetate are the primary products from CO_2 reduction. When normalized to the projected cathode area, the CH_4 production rates have ranged from 0.005–9.78 l m^{-2} d^{-1}, while the rates of acetic acid ranged from 0.02–685 g m^{-2} d^{-1} (ref. [42]). The Wood–Ljungdahl pathway is known as the primary metabolism in electroactive bacteria, with acetyl-coenzyme A (acetyl-CoA) serving as a key intermediate to produce various organic compounds from CO_2, including formic acid, propionic acid, butyric acid, 2-oxobutyrate, ethanol, isopropanol, butanol and isobutanol[43–46]. The production of these compounds has generally been accompanied by acetic acid production and the conversion rates from CO_2 have been much lower than acetate. The electrode serves as the source of reducing power for CO_2 reduction. The electron transfer mechanisms can be direct or mediated by H_2 or other redox agents and are largely influenced by the cathode working potential[47,48]. Despite the promise of the MES process, major breakthroughs are needed to produce higher titre

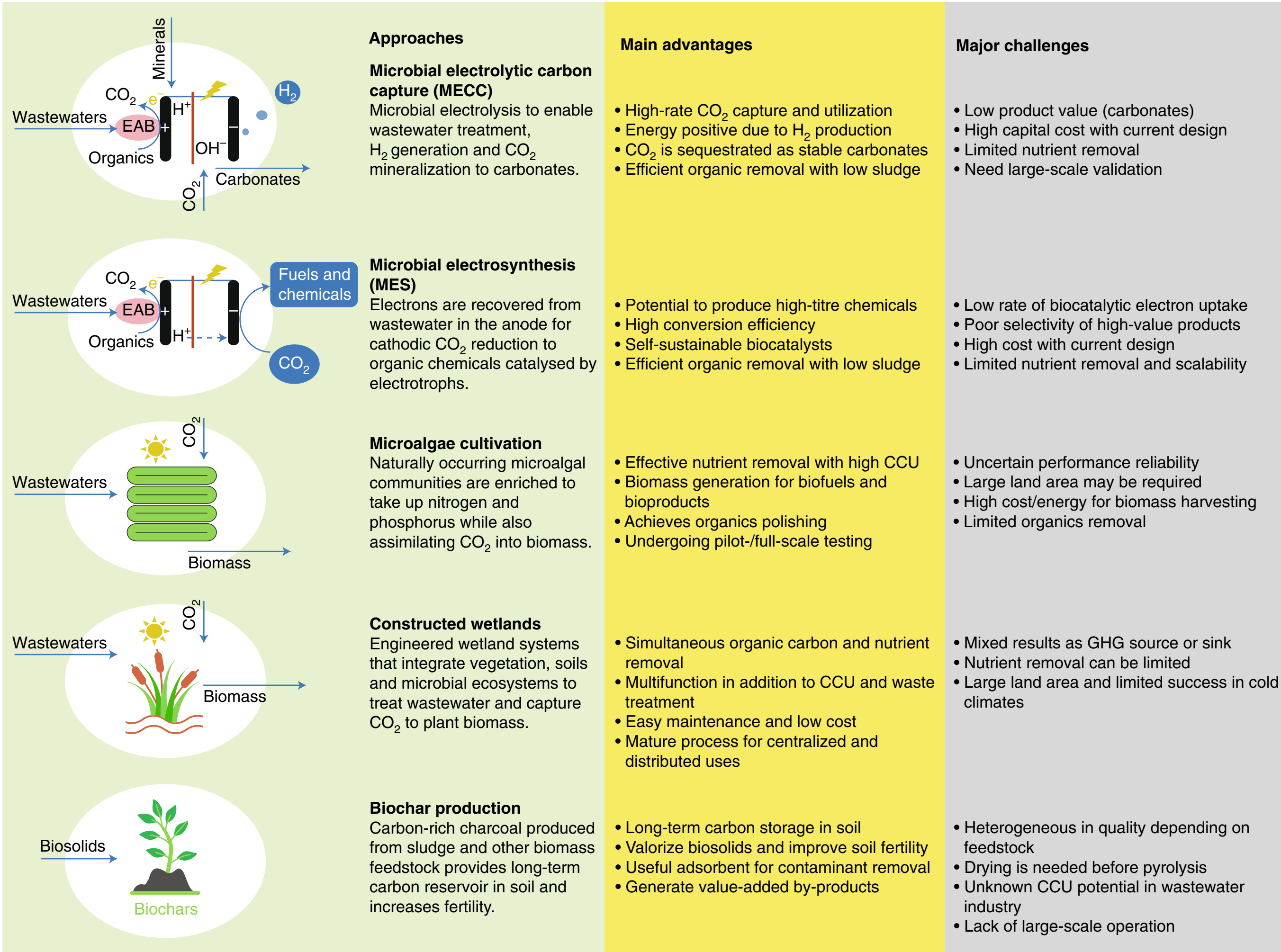

Fig. 1 | The different CCU processes that can be integrated with wastewater treatment. The major advantages and current limitations of these processes are evaluated based on data and observations from the literature.

products, develop high-performing reactors, reveal electron transfer mechanisms and reduce cost. A recent study showed that only a few MES products, such as formic acid and ethanol, are economically competitive in the market when compared with conventional processes[41]. Synthetic biology may be critical in this endeavour for engineered microorganisms to convert CO_2 into targeted chemicals and engineering innovation is needed for more-efficient CO_2 conversion and product separation[41,49].

Microalgae cultivation. The cultivation of microalgae is complementary to carbon-based treatment processes like MECC and MES, as it can fix CO_2 during autotrophic growth while assimilating nutrients (N and P) in wastewater. Microalgae have been widely studied for CO_2 capture and utilization, including extensive research on their use in large-scale (>5,000 acres) cultivation systems to produce feedstocks for biofuels and bioproducts[50]. Microalgae have higher areal productivities than terrestrial plants, with production rates generally in the range of 15–30 g m^{-2} d^{-1} (ref. [51]) and research initiatives targeting 40–60 g m^{-2} d^{-1} (ref. [50]). The potential of microalgae to simultaneously achieve wastewater treatment in conjunction with CO_2 capture is now attracting more attention, as the recovery of nutrients from wastewater not only reduces financial burdens on algae feedstock production but also increases the recoverable chemical energy in municipal wastewater by three-to-four times[52]. When growing autotrophically, microalgae fix 1.8–2.4 kg of CO_2 per kg of biomass grown[53,54] and yield roughly 9.4–116 grams of CO_2 fixed per gram of nitrogen assimilated (C/N ratio = 2.6–32)[53–55], with higher values for both ratios stemming from the storage of lipids and carbohydrates. There are numerous stressors that can trigger biopolymer (lipid and carbohydrate) storage (for example, intense light and salinity stress), but the most common approach in process engineering is to subject cells to conditions with limited light, nitrogen or phosphorus[50]. Nitrogen to phosphorus ratios in microalgae are dictated by both microorganism ecology and physiology and are typically in the range 2.3–23 (although ratios of >40 have also been observed in phototrophic microorganisms)[55,56]. Thus, given that most municipal wastewaters have inorganic C/N ratios less than 3 and N/P ratios on the order of 6–9 (ref. [57]), process engineering may enable the complete use of nutrients present in wastewaters[58,59] for carbon recovery and capture of exogenous CO_2. Once grown and harvested, downstream uses being explored include use as a carbon- and nutrient-rich soil amendment[50], animal feed[60] or as a feedstock for bioenergy (for example, ethanol, diesel and methane)[61,62] or bioplastic[62] production.

The state of the art for small-footprint, high-productivity microalgal processes (that is, high areal productivity) is the use of suspended, enriched cultures that achieve reliable treatment performance by creating selective environments through process engineering[63,64]. Mixed communities of microalgae (and bacteria) have demonstrated high levels (>99%) of nutrient recovery from wastewaters[52] and are likely to increase productivity and reliability relative to pure cultures[65]. To maximize the conversion of light energy into chemical energy via photosynthesis, the low organic concentration in the influent are preferred to limit light absorption by organics and heterotrophic microorganisms. Algal systems could, for example, be used in series after technologies that capitalize on influent organic carbon (for example, MECC and MES technologies), with the algal process providing the added benefit of COD polishing to meet discharge standards. The general concept of integrating phototrophic and chemotrophic processes dates back more than half a century[66], with recent efforts exploring the integration of microalgae and microbial fuel cells (MFCs) for simultaneous organic carbon removal, nutrient removal and electricity generation[67]. Given the potential of MECC technologies to valorize organic carbon and achieve greater CO_2 capture, a more effective approach for CCU would be a combined MECC–microalgae process (Fig. 2b). Future research on microalgal technologies should focus on process engineering to leverage local microbial communities and achieve reliable performance that meets effluent nitrogen, phosphorus and biochemical oxygen demand (BOD) permit requirements in small-footprint, intensive reactor systems with efficient solid–liquid separation technologies.

Constructed wetlands. These are engineered systems that utilize and enhance natural processes involving vegetation, soils and the associated microbial ecosystem for wastewater treatment[68,69]. In addition, constructed wetlands (CW) often provide other functions such as flood control, biomass production, biodiversity and recreational and educational services. Organic carbon removal is high in all types of CW as the microbial degradation processes are mostly aerobic, but nutrient removal can also be accomplished when CWs are designed to achieve nitrification–denitrification. Different classes of CW can have a wide range of emission and carbon capture profiles depending on the level of treatment, seasonal variation and system variation. For example, vegetated CWs can capture a substantial amount of CO_2 into biomass, but they also emit large quantities of CH_4 and N_2O, complicating GHG accounting of CW net benefits[70]. A recent review summarized 158 CW studies and found that free water surface CWs (351 mg m^{-2} h^{-1}) had ~30% lower CO_2 emissions than subsurface flow CWs (502 mg m^{-2} h^{-1}), and vertical subsurface flow CWs showed lower CH_4 emissions than horizontal subsurface flow CWs. However, they also concluded that the absolute value of CH_4 emission from CWs can be much lower than that found in conventional wastewater treatment plants[71].

Most GHG-related studies on CWs focus on understanding the carbon and nitrogen fluxes because they fluctuate dramatically across both space and time. There have been limited studies explicitly investigating the potential CCU benefits of CWs during wastewater treatment. Overall, CO_2e emissions of anaerobic CWs include those from soil, root, rhizome respiration and organic decomposition (3.2–8.7 kgCO_2 ha^{-1} yr^{-1}), but the emissions were orders of magnitude higher in aerated CWs with low water table (up to 14,000–27,000 kgCO_2 ha^{-1} yr^{-1})[72]. On the other hand, CO_2 is assimilated by plants, and in optimal conditions horizontal subsurface flow CWs may capture 57,000–76,000 kgCO_2 ha^{-1} yr^{-1} (ref. [71]). This carbon is not stable and can easily be mineralized when the water table drops or the CW filter is saturated, but the benefits of CCU can be extended if the biomass is harvested for BECCS or biochar production. Specifically, a study focusing on carbon cycle in wetlands dominated by *Phragmites australis* concluded that CWs are sources of GHGs on a timescale of decades, but they can become carbon sinks on longer timescales[73]. A recent study converted CH_4 and N_2O fluxes to CO_2 equivalents in a CW and found that it is most likely a sink of CO_2 with an annual net capture of 2,700–24,000 kgCO_2 ha^{-1} yr^{-1}, which represents 12–67% of the CO_2 fixed in the biomass[74]. To reduce CH_4 and N_2O emissions, CWs can be integrated with plant microbial fuel cells to minimize anaerobic decomposition of rhizodeposits[75].

Biochar production. Biochar is a carbon-rich charcoal produced through pyrolysis under anaerobic conditions[76]. Biochar production is typically self-sufficient in terms of energy requirements and can produce surplus energy as heat or as other by-products such as bio-oil or syngas[76]. Although terrestrial plant residues have been the primary feedstock for biochar production, wastewater sludge, microalgae and wetland plants have also been used to produce biochar. When used as a soil amendment, biochar enhances soil fertility and improves soil's water and nutrient retention, which can lead to higher crop yields[76]. Biochar links the short-term photosynthesis-mediated carbon cycle to a long-term carbon reservoir in the soil and it was found suppress soil CH_4 and N_2O emissions, even though the levels of benefit may vary significantly depending on the properties of biochar and soil, type of crop planted as well as the local climate and so on[21,77–80]. The CO_2 removal potential through biochar globally across industries is estimated[81] between 1.8–3.3 GtCO_2e yr^{-1}.

Compared with the current disposal practices of landfilling and direct land application, carbonizing the sludge into biochar may provide higher environmental and economic benefits. Sludge biochar eliminates pathogens, improves soil structure and increases agricultural output[82]. Moreover, studies have shown that such biochar can adsorb pesticides, reduce heavy metal leaching and increase soil fertility. For example, a study found that when sludge biochar was applied at 10 t ha^{-1}, the production of cherry tomatoes increased by 64% over the control soil due to increased nutrient availability (N and P) and improved soil conditions[83]. For a wastewater treatment plant that generates 100 dry tons of sewage sludge per day, an estimated 65 tons of biochar can be generated assuming a median yield of 65% and ~21 ktCO_2e may be captured per year based on a GHG emission of 0.9 kgCO_2e per kg biochar[77,84]. Though limited literature is available on biochar production from wastewater microalgae or constructed wetland plants, similar benefits can be obtained and the biochar can be utilized on-site for the removal of toxic contaminants from wastewaters[85]. Compared with woody biomass, the high moisture content in these wastewater treatment associated feedstocks can make the conversion more energy intensive and require additional treatment steps, so more studies are needed in product quality control, life-cycle assessment and technoeconomic analysis.

Illustration of simultaneous CCU and C/N/P removal

The world's two largest GHG emitters are the USA and China, which respectively produce an estimated 6.51 and 2.97 million dry tonnes of sewage sludge and 60.4 and 38.0 km^3 municipal wastewater per year[8]. Of the produced wastewaters, it is estimated that roughly 40.9 (USA) and 26.6 (China) km^3 are actually treated[8]. If treated with activated sludge (the most common engineered process for large-scale, centralized facilities), direct fugitive GHG emissions from activated sludge processes can be around 14 and 8.9 MtCO_2e per year in the US and China, respectively[17]. Additionally, indirect emissions stemming from electricity consumption — which are sensitive to the carbon intensity of the local electrical grid — further contribute to the system's carbon footprint (Supplementary Table 2)[10,86]. On the other hand, biomass growth due to organic removal can lead to biogenic carbon capture in a form of biosolid or biochar, which could be applied for land applications. Such a method of adding carbon as a soil amendment has been shown to increase soil organic carbon reserves and plant activity. In total, the US and China currently net release roughly 13.5 (median; 5th–95th percentile range 0.2–26.8) and 14.9 (5.3–24.7) MtCO_2e during conventional wastewater treatment[8], respectively. More than half of the emission can be attributed to electricity consumption, where China has a higher CO_2 intensity of electricity (Fig. 2c).

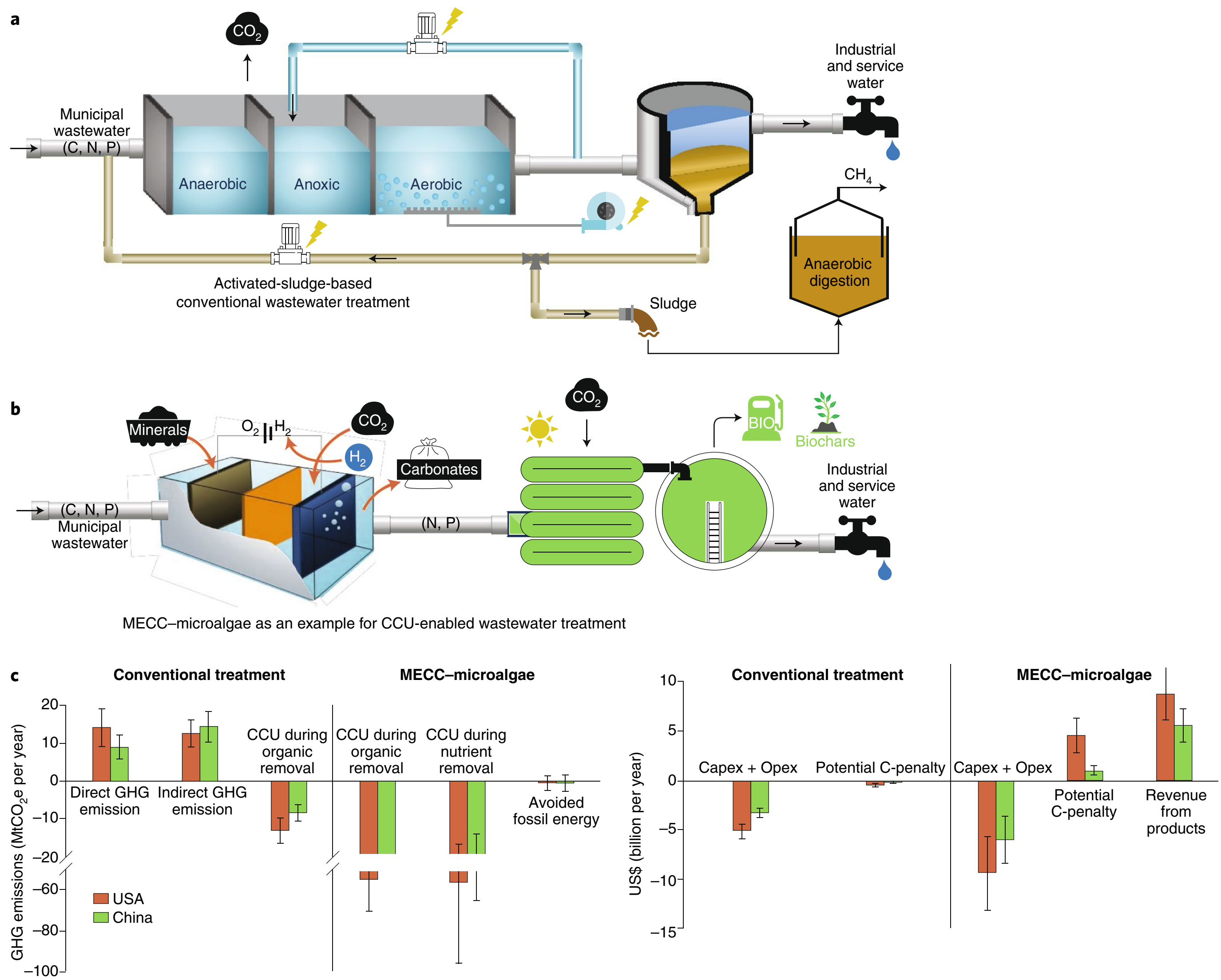

Fig. 2 | Preliminary estimates of CCU benefits from an example integrated MECC–microalgae process compared with a conventional activated-sludge process. a, Conventional anaerobic/anoxic/aerobic activated sludge process for simultaneous carbon and nutrient removal. **b**, Integrated MECC and microalgae cultivation for carbon and nutrient removal with resource recovery and CCU. **c**, The preliminary estimates of CCU potential and economic impacts of a conventional process compared with the MECC–microalgae process. Direct and indirect GHG emissions are associated with pollutant degradation and fossil energy expenditure. CCU during the activated-sludge process is due to biomass growth (sludge) for organic removal. Monte Carlo simulation with Latin hypercube sampling (100,000 simulations) was used to characterize the likely ranges of performance of each technology. (The probability density functions can be found in Supplementary Table 2.) Varied values include volumes of municipal wastewater treated per year, COD and nitrogen concentration, pollutant removal rate, Coulombic efficiency, electricity consumption, CO_2 intensity of electricity and MECC; data are from the literature with detailed calculations provided in the Supplementary Information, including energy balance, material uses and economic analysis, potential carbon tax, price of products and electricity, direct fugitive emissions in conventional WWT, carbon capture in biomass and capital expenditures and operating expenses. Capex, capital expenditure; Opex, operating expense.

Current wastewater treatment focuses on removing different contaminants, such as organic carbon (BOD), nitrogen, phosphorus and solids to comply with environmental regulations to protect public health and the aquatic environment. Although technologies and locality-specific wastewater treatment needs vary widely, it is reasonable to expect roughly 5.3 (median; 5th–95th percentile range of 4.8–5.8; USA) and 3.3 (3.0–3.7; China) billion US dollars in expenditure per year for wastewater treatment systems operation. If a carbon penalty (US\$35 and US\$10 per metric tonne of CO_2e for US and China, respectively) were applied in the future, the costs could increase to a total of 5.7 (5.1–6.4; USA) and 3.5 (3.1–3.9; China) billion dollars per year. If the sludge feedstocks from these countries are converted to biochar, an estimated 4.2 (U.S.A.) and 1.9 (China) Mt of biochar can be produced. Accordingly, 3.8 (USA) and 1.7 (China) $MtCO_2e$ may be captured per year based on a median biochar yield of 65% and a GHG emission of –0.9 $kgCO_2e$ per kg of biochar[77,84]. These carbon offsets would equate to roughly 0.13–0.20 (USA) and 0.02–0.03 (China) billion US dollars in value. Sludge management on its own, however, does not address the need for adequate wastewater treatment (Fig. 2c).

In order to achieve concurrent removal of C, N and P, activated sludge processes with sequential anaerobic/anoxic/aerobic zones can be used (Fig. 2a)[10,11,87]. These processes, however, consume a significant amount of energy (0.6–1.0 kWh m^{-3} wastewater) for aeration and pumping[10] and generally require the addition of an external carbon source (for example, methanol) to support denitrification. To investigate the potential of using CCU-enabled processes for wastewater treatment, we further explore the implications of an integrated MECC and microalgal process. This scheme is used as an example because the processes are complementary in treating wastewater: MECC specializes on organic carbon removal while microalgae are effective in nutrient removal. Moreover, compared with other emerging treatment processes, they both demonstrated excellent CCU capability during treatment and generated value-added products. Many other processes such as anaerobic digestion, anaerobic membrane reactors (AnMBR), anammox and adsorption/bio-oxidation also reduce energy consumption and its associated GHG emissions and/or recover energy, but they have not been used for direct CO_2 capture and utilization[88–90]. The dissolved methane in AnMBR effluent also remains a challenge that contributes to GHG emission[88].

If conventional processes were replaced with MECC–algae systems, instead of being a net emitter of GHGs, up to 112 (median; 5th–95th percentile range of 84–145; USA) and 75 (57–97; China) net $MtCO_2e$ can be captured and converted to valued-added products. Among these negative emissions, approximately 41–56% and 47–58% are attributed to CCU during organic and nutrient removal, respectively; and −2 to 2% is credited to avoided consumption of fossil energy during CCU (avoided aeration and so on; the negative value stems from uncertainty analysis). In terms of economic benefits, while the proposed system is likely to have even greater capital expenditure and operating expense costs than conventional processes, the recovered mineral and biofuel products may create 8.7 (6.9–10.9) and 5.6 (4.4–6.9) billion dollars in value per year for the US and China, respectively. Furthermore, carbon credits could also be leveraged as an additional source of revenue to finance infrastructure investment. China recently launched the world's largest carbon market, with an average price on carbon ranging from US$3–10 per ton (ref. [91]). The US government recently passed a new US$35 per ton tax incentive for CCU for up to 12 years, and it greatly expanded such credits from fossil power plants to industrial and air capture facilities using biologically based CCU systems[92]. Altogether, the carbon capture credits in the two countries could have significant economic benefits for such operation, mobilizing 4.5 (3.3–6.2) and 1.0 (0.7–1.5) billion dollars for US and China wastewater industries, correspondingly. These estimates demonstrate that wastewater industry can become a significant contributor of negative carbon emissions. However, we do acknowledge that the MECC–algae combination only represents one example of carbon-negative wastewater treatment and because these technologies have not yet been demonstrated in full scale, the real impacts remain to be quantified. Similar methods can be used to assess the potentials of other technology combinations as well.

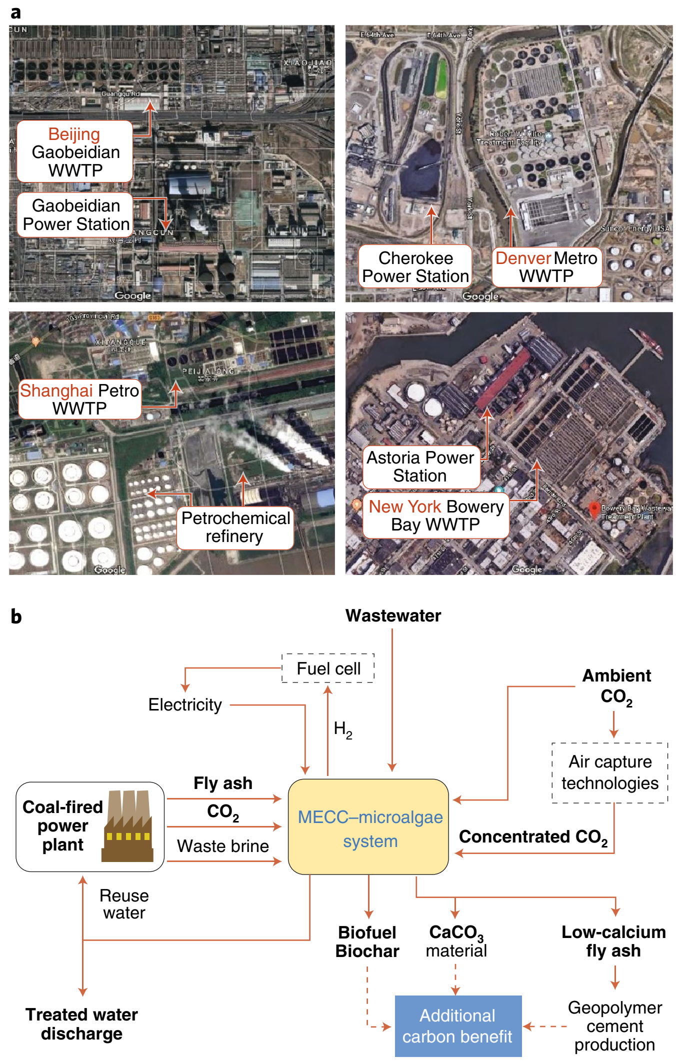

Fig. 3 | The low-hanging fruit on co-location of emission-point-source and water-resource recovery plants. a, Example co-locations of CO_2 point sources and wastewater treatment plants that enable complementary CCU in Beijing, Shanghai, New York City and Denver. **b**, Schematic of the mutual benefits between a point source (coal-fired power plant) and the proposed CCU-enabled wastewater treatment process integrating microbial electrochemistry with microalgae. Credit: CNES/Airbus and DigitalGlobe (Beijing map); CNES/Airbus, TerraMetrics (Shanghai map); 2018 Google (US maps); ref. [25], Elsevier (schematic in **b**)

Co-locations and ambient CO_2 capture

Enabling wastewater treatment with CCU not only transforms the societal contributions and design of WRRFs but also brings tremendous opportunities for major CO_2-emitting industries, such as power plants, cement works and refineries (Fig. 3a). Our previous study identified that many of these facilities are co-located with or near major WRRFs[24], which could result in mutual benefits, for instance, between a coal-fired power plant and a nearby WRRF equipped with MECC and algae treatment processes (Fig. 3b). The WRRF takes the CO_2 from the point source to produce value-added products and generate carbon credits, which is likely to be more cost-effective than standalone CCU implementation by the power plant[93]. The calcium/magnesium-abundant fly ash generated by the power plant may be used as a silicate supplement to facilitate CO_2 mineralization[25]. The WRRF-treated wastewater could be reused by the power plant and the WRRF-generated renewable energy could be integrated into the power grid. The generated products — including biofuels, biochar, $CaCO_3$ and purified fly ash — would increase the profitability of the process for both industries[25]. Even for WRRFs without a nearby concentrated CO_2 source, anaerobic digesters could also serve as a source of CO_2. Plus, the feasibility of air capture has been demonstrated using both MECC and microalgae. Commercially available ion-exchange resin with dry–wet cycle can concentrate ambient CO_2 by>200 times for MECC to generate concentrated H_2 and carbonate and similar practice can be used to improve microalgae yield and biofuel production[28,94].

In addition to municipal WRRFs, similar CCU-enabled wastewater treatment practice can be implemented in many industries for which wastewater treatment is needed. For example, nearly 3.3

billion cubic metres of wastewater are generated from the oil and gas industry every year in the US alone[24]. The use of such waste stream for in situ CCU of GHGs from oil and gas could have significant benefits for the industry, not the least of which would stem from environmental and economic benefits of water reuse relative to deep well injection. Both MECC- and algal-based wastewater treatment have also been demonstrated in the alcohol and beverage industry, which generates 1.1 billion cubic metres of wastewater in the US and 21 billion cubic metres of wastewater globally[95]. High operational costs to manage their concentrated wastewaters and large CO_2 emissions make such industry an ideal entry market for new technologies. For WRRFs located on the coast that discharge billions of cubic metres of treated effluent to the ocean, carbonate-enriched effluent generated after MECC treatment can facilitate more ambient CO_2 capture and storage in the ocean, which can be a cheaper and simpler alternative to an industrial air capture process. Such effects can be easily implemented with cities like Hong Kong, which uses>270 million cubic metres of seawater every year for toilet flushing[5,24].

Conclusion

While this prospective review demonstrates the great potential of using wastewater treatment for CCU, this concept is still in its infancy. It is challenging to meet and balance multiple objectives to fulfil different treatment needs and capture and utilize CO_2 for resource recovery. Therefore, technological development and implementation as well as more-detailed technoeconomic, lifecycle and socioeconomic analyses will be required to understand the potential of such an approach for different industries. We have listed several promising wastewater treatment processes that are known to have CCU capability, but we acknowledge that each process has its niche application areas. We used MECC and algae as an example to quantify the preliminary environmental and economic benefits in large-scale municipal water resource recovery facilities, but constructed wetlands may be more appropriate for distributed systems. Similarly, biochar derived from wetland plants or wastewater sludge can provide long-term negative emission benefits, but its production and application are associated with current mainstream wastewater treatment process. While this Review strives to bring attention to the important role that the wastewater industry can play in global carbon management, different stakeholders including regulators, technology providers, utilities and investors must also recognize that carbon is an integral part of waste management process. When managed properly, CCU can be realized without compromising current waste treatment efficacy, rather it can instead bring tremendous value to the wastewater industry, CO_2-generating industries and to society as a whole.

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Author contributions
Z.J.R. and L.L. designed the study. L.L, J.S.G. and Z.J.R. performed data analysis. L.L., J.S.G., C.A.P., X.Z., G.H.R. and Z.J.R. wrote the manuscript.

Competing interests
The authors declare no competing interests.

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