



Nature-based solutions coupled with advanced technologies: An opportunity for decentralized water reuse in cities

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ABSTRACT

Decentralized water reuse in cities is a prominent alternative to mainstream top-down models for urban water treatment, which are based on centralized, linear dynamics of resource management. In this sense, Nature-based Solutions (“green” technologies) coupled with advanced technologies (“grey” technologies) constitute a promising approach for fomenting onsite water treatment and reuse in cities, while also providing multiple co-benefits. This article puts forward a conceptual advancement by providing a better understanding of coupled “green-grey”/“grey-green” technologies (CGGT). To do this, we critically discuss the main reasons for pairing these technologies instead of using them separately, as well as their treatment performance and constraints regarding data reporting issues. Moreover, the article discloses the most common treatment configurations, water quality parameters being evaluated, potential reuse schemes, costs, and energy requirements. A systematic selection and analysis of scientific articles was carried out to this end. Of 395 pre-selected articles, only 17 addressed coupled (green-grey/grey-green) technologies in the treatment of urban wastewaters for further reuse or safe discharge onsite. Despite the relatively low number of articles, 80% were published in the past five years, showing the increased interest in this novel topic. The selected articles were analysed and here we present the resulting comprehensive Excel database (343 datasets) containing detailed information about the design, operation, and performance of such systems. Green-grey technologies were found to be predominant, the configuration constructed wetlands followed by advanced oxidation process and electrochemical process being the most studied. Grey technologies are normally applied at a second stage to remove pathogens in compliance with reuse standards (normally when green technologies alone cannot deliver the standards). Meanwhile, green technologies are commonly used at a second stage to break down slowly biodegradable substances that have not been completely removed by grey technologies (normally as a polishing step following grey technology). The design parameters for combining these technologies have not yet been fully optimized, since they were mainly designed as sole technologies and forcibly put together as a coupled treatment. Hence, further studies should focus on variables and parameters influencing the functioning of coupled technologies as a whole. Finally, due to the novelty and relevance of the topic, transparency and consistency in data reporting is essential to support the optimization and competitiveness of coupled green-grey/grey-green technologies against existing decentralized/centralized approaches.

; EC, electrochemical process.

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Abbreviations			
NbS	Nature-based Solutions	TSS	total suspended solids
CGGT	coupled green-grey/grey-green technologies	TB	turbidity
CW	constructed wetlands	COD	chemical oxygen demand
HSSF	horizontal subsurface flow constructed wetland	BOD	biochemical oxygen demand
VSSF	vertical subsurface flow constructed wetland	TOC	total organic carbon
SVF	saturated vertical flow constructed wetland	TN	total nitrogen
FL	floating treatment wetland	TKN	total kjeldahl nitrogen
BSF	baffled constructed wetland	TP	total phosphorus
GW	green walls	DWS	decentralized water systems
PL	ponds and lagoons	TA	total area
GR	green roofs	HL	hydraulic load rate
MB	membrane filtration	OLR	surface organic loading rate stage
AOP&EC	advanced oxidation process and electrochemical process	HRT	hydraulic retention time
DF	disinfection methods	NR	not reported
UV	ultraviolet radiation	NC	not clear
ADS	adsorption techniques	NA	not applicable
		LMH	operating flux

1. Introduction

By 2050, 68% of the global population is projected to be urban and water demand is expected to increase between 20% and 30%, mainly due to population growth, socio-economic development, changes in consumption habits and higher demands from industrial and domestic sectors (OECD, 2015; United Nations, 2019). These pressures on the availability of water in cities raise concerns over the current linear models of urban water management, which do not consider onsite reuse. Decentralized water systems (DWS) are emerging as a systematic approach to water management in cities, these being key to water sustainability (Lu et al., 2019). DWS promote a change in the current conventional paradigms from linear treatment and disposal to a circular model that prioritizes treatment of low amounts of water as near as possible to the original source, promoting local water reuse schemes for non-drinking purposes and thus reducing pressure on the potable water supply (Novotny and Brown, 2007; Nelson, 2008; Capodaglio et al., 2017). Moreover, in contrast to centralized systems, which involve long distances for wastewater transportation, huge piping networks and multiple lifting pump stations (Oliveira et al., 2021), most DWS require lower structural costs and lower amounts of energy, mainly due to their usually smaller collection and distribution networks taking advantage of gravity flow (Capodaglio et al., 2017).

Most wastewater treatment technologies currently being applied under centralized water schemes can be implemented in decentralized settings (Capodaglio et al., 2017). Several studies have demonstrated the feasibility of implementing advanced technologies such as membrane filtration (MB) (Kümmerer et al., 2016), advanced oxidation and electrochemical processes (AOP&EC) (Otter et al., 2020), adsorption techniques (ADS) (Schumann et al., 2020) and disinfection methods (DF) (Alfiya et al., 2017) in decentralized schemes. These advanced “grey” technologies are highly efficient in removing a wide range of contaminants and leave a smaller footprint, facilitating their implementation in urban contexts where space is limited (Andersson et al., 2017). However, these technologies usually imply a high energy demand and a considerable cost for their implementation and maintenance (Garrido-Cardenas et al., 2020). Moreover, toxic disinfection by-products (e.g., nitrosamines, bromate), catalyst residues (e.g., nano TiO₂) and brine wastes can be formed depending on the technology selected, diminishing the sustainability of such technologies (Jahan et al., 2021) (Funke et al., 2021) (Keyikoglu et al., 2021).

In this regard, Nature-based Solutions (NbS) such as constructed wetlands (CW) (Capodaglio et al., 2017; Kobayashi et al., 2020), ponds and lagoons (PL) (Rizzo et al., 2020; Fiorentino et al., 2016), green walls

(GW) (Svete, 2012; Masi et al., 2016a; Fowdar et al., 2017) and green roofs (GR) (Zehndorf et al., 2019) have emerged as alternative “green” technologies to facilitate water reuse in cities. Besides ensuring a set of environmental and socio-economic co-benefits (for example, enhancing biodiversity, improving air quality and promoting wellbeing), these “green” technologies are also recognized for being environmentally friendly and cost-effective technologies for water treatment, mainly due to their low energy demand and low implementation and maintenance costs (Fowdar et al., 2017; Zehndorf et al., 2019; Kobayashi et al., 2020).

Nevertheless, the implementation of CW and PL can be severely limited due to the larger area footprint, especially in cities with a high population density and low land availability (Cheng et al., 2018; Fiorentino et al., 2016). As an alternative, GR and GW bring the advantage of using empty vertical façades and rooftops in cities. However, due to the novelty of this field, most studies are at the pilot stage, and the relationship between the required surface area and hydraulic/contaminant load rates is often under-reported. On top of that, not all available areas (vertical or rooftop) might be suitable for implementing GR and GW. For example, they could compete with other functional structures (e.g., windows, balconies, solar panels, air conditioning structures), be restricted due to structural conditions (e.g., lack of proper slope, reinforcement or required water connections) or even face legal restrictions concerning the change of architectural aspects of façades. All in all, the larger land footprint of CW and PL and uncertainties over the area requirements for implementing GR and GW are inhibiting the mainstreaming of these technologies for urban water reuse schemes. Moreover, the sole use of green technologies depends on climatic conditions and entails a limitation regarding potential water reuses because the treated water does not always achieve the required reuse standards in terms of microbiological pollutants, microcontaminants and emergent contaminants (Arden and Ma, 2018).

With regard to the above, coupled green-grey/grey-green technologies (CGGT) may represent suitable alternatives for on-site water treatment to overcome the above-mentioned limitations of using grey or green technologies separately in urban areas. “Green” technologies can contribute to reducing costs/energy and the formation of toxic by-products. As an example of this, Talekar et al. (2018) showed that the use of CW prior to electrochemical (EC) treatment reduced energy expenditure from 27 kWh/m³ (only EC) to 16 kWh/m³ (CW + EC), meaning a 40% decrease in energy investment in the long term. Moreover, the associated operation and maintenance costs are lower, due to the reduced organic and mineral load going through the EC reducing calcium precipitation between the electrodes and membrane (Talekar

et al., 2018). Additionally, the combination of compact grey technologies (e.g., DF, AOP&EC) with cost-efficient green technologies (e.g., CW or green roofs) may prove especially suitable in situations in which land availability is limited and overall high efficiency in terms of pathogen removal (bacteria and viruses) is needed to ensure reuse standards for diverse water reclamation purposes, including potable reuses (Chen et al., 2011; Alves et al., 2012; Kümmerer et al., 2016; Pei et al., 2019). In respect of this, Arden and Ma (2018) and Wagner et al. (2020) demonstrated that combining CW with advanced technologies such as ultraviolet radiation, reverse osmosis, electrodialysis or capacitive ionization may be a suitable strategy for treating greywater while reliably meeting water reuse standards.

Green and grey technologies have been widely implemented, both individually or combined (CGGT), as supportive units under centralized/sectorial wastewater treatment schemes either for polishing effluent in wastewater treatment plants before its disposal (Ahmed et al., 2008; Rizzo et al., 2020) or supporting peri-urban or rural areas not covered by the sewage network (Li et al., 2021). Nevertheless, the implementation of CGGT in decentralized schemes to foster reuse or safe discharge onsite has also begun to garner more attention in recent years. According to Bakheet et al. (2020), their research (from 2020) on GW coupled with electrochemical treatment is the first study to explore such a combination offering a reliable and an environmentally-friendly method for greywater reuse onsite. Other examples include horizontal subsurface flow constructed wetlands (HSSF) and ozonation for the removal of trace pharmaceuticals in domestic wastewaters (Lancheros et al., 2019), and mobile vertical flow CW coupled with MB for treating black and greywater for potable reuse (Lakho et al., 2020b).

However, to the best of our knowledge, no publication has yet summarized design features and performances of CGGT treating urban wastewater under decentralized schemes for water reuse or onsite discharge (in buildings, restaurants, houses, universities etc.). The consolidation of existing knowledge regarding such a promising coupling of technologies constitutes an important step in providing guidance on their potential as an alternative to current centralized water management in cities. The main goal of this article is therefore to critically discuss CGGT design and performance, as well as their advantages and constraints. In view of this, our method included a comprehensive literature review, followed by data collection and analysis. Based on the results, we provide a Microsoft® Excel database with a detailed dataset containing more than 300 data entries regarding the design and performance of CGGT, which is designed to evolve together with advancements in the field. Moreover, we put forward a conceptual advancement in this field by providing a critical discussion of the following topics: most common treatment configurations, reasons for applying CGGT to decentralized reuse schemes, water quality parameters evaluated, water treatment performance and potential reuses, energy requirements, and CGGT as an alternative for the removal of emergent contaminants. Next, we present several insights concerning difficulties faced during the data collection process and translate them into a set of recommendations regarding the type of data to be reported, units and operational features. Finally, we draw some conclusions and discuss aspects of further research.

2. Materials and methods

In this section, we detail the systematic protocol followed to carry out this review. It included three main stages: literature collection on CGGT treating urban wastewaters (Section 2.1), systematic selection of suitable articles (Section 2.2) and data collection - procedures and analysis (Section 2.3).

2.1. Literature collection

A search was performed of the Web of Science and Scopus on February 25th, 2021. Search terms referring to NbS/green technologies,

advanced/grey technologies and urban wastewaters (Table A1 and Fig. 1) were applied to the title, abstract and keywords. Search terms referring to NbS/green technologies were limited to four types (based on Castellar et al., 2021): **constructed wetlands (CW)**, **green roofs (GR)**, **green walls (GW)** and **ponds and lagoons (PL)**. Search terms referring to advanced/grey technologies were also limited to four types (based on Wang and Xu, 2012; Kümmerer et al., 2016; Saqib Ishaq et al., 2019; Hube et al., 2020): **membrane filtration (MB)**, which includes ions or cation exchange, ultra-filtration, reverse osmosis and membrane bioreactors; **advanced oxidation processes and electrochemical process (AOP&EC)**, which include photocatalysis (e.g., TiO₂, TiO₂/-Al₂O₃, TiO₂O₃), photo-electrochemical oxidation, cathode and anodic oxidation, solar-driven anodic oxidation, photo-Fenton, fluidized-bed Fenton and ozonation; **disinfection methods (DF)**, which include chlorine/chlorination and ultraviolet radiation; and **adsorption techniques (ADS)**, which include zeolite filtration and activated carbon filter. The search terms for urban wastewaters were limited to the following six types adapted from Council of the European Economic Community, (1991) – European council directive, concerning urban wastewater treatment (91/271/EEC): domestic wastewater, greywater, black water, sewage, industrial wastewater and agricultural wastewater. The search was limited to full articles (reviews, proceeding papers and book chapters were excluded), in English, and published in scientific journals between 2012 and February of 2021. From these full articles, authors, title, abstract and DOI were exported and organized in a Microsoft® Excel sheet.

2.2. Systematic selection of suitable articles

The authors reviewed the abstracts and full text (if needed) of pre-selected articles (Section 2.1). A control procedure was implemented, whereby 10% of the articles were randomly read and annotated by two of the authors to ensure consistent choices by the team. The selection process was based on the following eligibility criteria: i) Articles must contain experimental primary data on design and performance of CGGT for urban wastewater treatment; ii) Articles must deal with CGGT as a self-standing technology for treating urban wastewater for reclamation or safe discharge under a decentralized scheme (e.g., in buildings or single households, restaurants, university campus facilities, industries and temporary use facilities). Therefore, articles dealing with CGGT as a polishing treatment for supporting wastewater treatment plants (WWTP, whether centralized or sectorial) were excluded. iii) Full text should be available.

2.3. Data collection: procedures and analysis

Data concerning design and performance were collected from the selected articles (Section 2.2) and organized in a Microsoft® Excel database (supplementary material). Data collection was performed in two steps: i) each author reviewed a certain number of articles and entered all the information required in the Excel datasheet; each line of the Excel database constitutes one data entry. Each data entry refers to the performance of a certain CGGT with regard to removing a specific pollutant under a certain experimental condition; ii) the information entered during the first step was double-checked by a different author to ensure a robust and reliable database.

The design data included CGGT type of sequence (green-grey, grey-green), treatment configuration (e.g. CW- AOP&EC, AOP&EC – CW), type urban wastewater origin (e.g., residential, industrial, university, etc.), experiment scale (lab, pilot or full scale), design/operation features (e.g., hydraulic load rate, surface area, filtration material, plants), proposed reuse application (e.g., irrigation of edible crops, toilet flush, recharge aquifers), and where possible, energy demand and costs. Performance data were collected from tables, manuscript text or supplementary data. If the information was not available from the previous sources, it was estimated from figures. All data regarding influent or

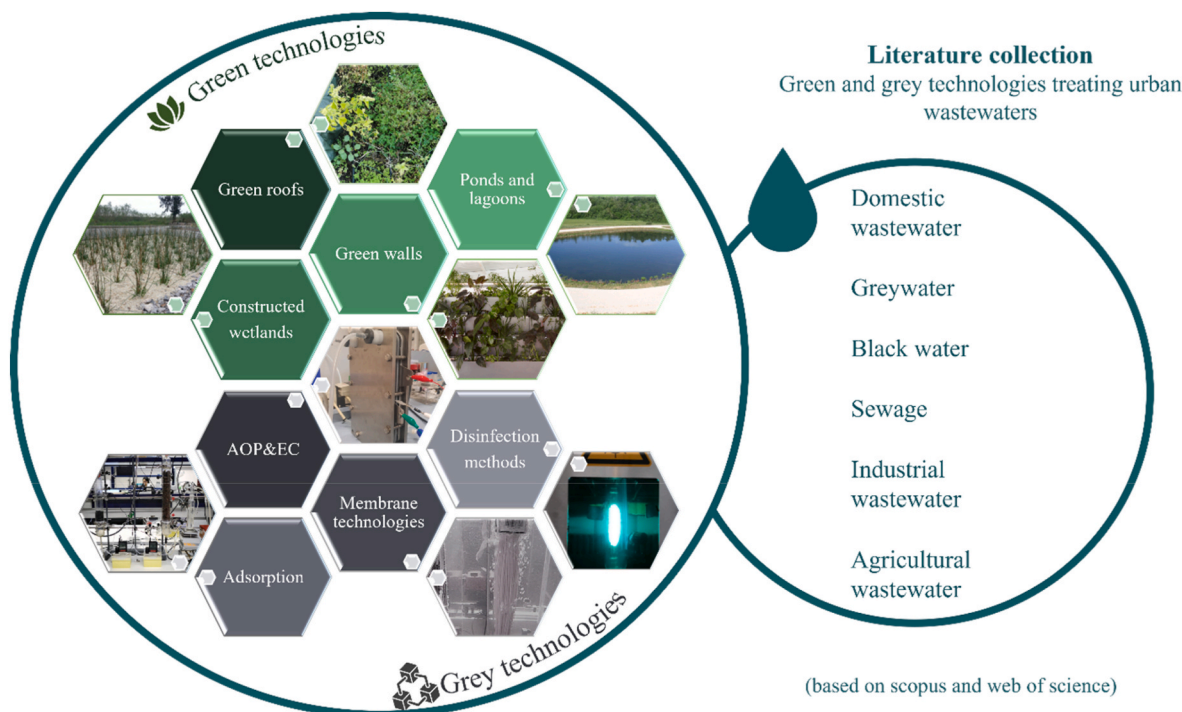


Fig. 1. Main technologies and types of urban wastewater included in the literature collection on coupled green-grey/grey-green technologies (CGGT) for treating urban wastewaters. Source: pictures from author’s personal archive, including constructed wetlands (CW), ponds and lagoons (PL), green roofs (GR), AOP&EC (advanced oxidation process and electrochemical process), disinfection methods (DF), adsorption techniques (ADS) and membrane filtration (MB). The green wall (GW) picture represents the vertECO technology from Alchemia-Nova. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

effluent concentration was expressed as mg/L, except for the data relating to pathogens, which was normalized to log CFU. When data on removal performance were not available, performance was calculated according to equations (1)–(3).

Equation (1) - Pollutant removal by stage (PR_x)

$$PR_x(\%) = \frac{(CFS_{in} - CFS_{ef}) \cdot 100}{CFS_{in}} \frac{(IN_x - EF_x) \cdot 100}{IN_x} \quad \text{Equation 1}$$

Where IN and EF stand for pollutant concentration in the influent and effluent, respectively, and x represents the treatment stage (which can be 1st, 2nd, 3rd, and so on).

Equation (2) - Representativeness of the removal taking place in the second stage in relation to the total removal ($RPR_{2^{stage}}$)

$$RPR_{2ND}(\%) = \frac{(IN_{1st\ stage} - EF_{2nd\ stage}) \cdot 100}{IN_{1st\ stage}} \quad \text{Equation 2}$$

Where $IN_{1st\ stage}$ and $EF_{2nd\ stage}$, respectively, stand for pollutant concentration in the influent of first treatment stage and effluent of second treatment stage.

Equation (3) - Total removal (TR)

$$TR(\%) = \frac{(CFS_{in} - CFS_{ef}) \cdot 100}{CFS_{in}} \frac{(IN_{1st\ stage} - EF_{last\ stage}) \cdot 100}{IN_{1st\ stage}} \quad \text{Equation 3}$$

Where $EF_{last\ stage}$ stand, respectively, for pollutant concentration in the effluent of last treatment stage. $\frac{(CFS_{in} - CSS_{ef}) \cdot 100}{CFS_{in}}$

Additionally, we calculated the averaged removal of each contaminant for CGGT in general (not distinguishing by type or configuration), per type of CGGT (green-grey and grey-green) and per configuration of CGGT (e.g., CW – AOP&EC, AOP&EC-CW). These averages were only calculated for pollutants with at least three data entries in the database and appearing in more than one article. Standard deviation was calculated using data entries taken either from different articles or from the

same article but under different experimental conditions.

3. Results and discussions

In this section, we present and discuss results for the systematic selection of articles (Section 3.1), design and performance of CGGT (Section 3.2) and difficulties faced during data collection (Section 3.3).

3.1. Systematic selection of articles

Of the 395 articles pre-selected during the literature collection (Section 2.1), 17 were selected for further analysis (Section 2.2). Most of the excluded articles (352, 89%) were found to be outside the scope, due to the following reasons: a) articles comparing the technologies separately (e.g., MB and CW performance comparison; Andleeb and Hashmi, 2018); b) articles dealing only with NbS, or only advanced technologies, or neither; c) articles not dealing with wastewater treatment performance (e.g., ultrasound technology applied as extraction method of endocrine-disrupting chemicals, Azzouz and Ballesteros, 2016); d) articles dealing with hybrid technologies in which NbS and advanced technologies are not coupled in sequence but merged into one single technology. In this case, the majority of excluded articles addressed CW using adsorptive materials as filter media (e.g., activated carbon, zeolite, nanofillers) and not as an independent advanced ADS (Du et al., 2020; Fu et al., 2020). Other examples of hybrid technologies that were excluded are electrolysis and photocatalysis wetlands and integrated bioelectrochemical wetland systems, which include such processes inside the CW and not as an independent advanced technology (Ju et al., 2014; Li et al., 2020; Zhong et al., 2020); e) articles dealing with chemical or other removal processes occurring inside the NbS and not as a coupled advanced technology (e.g., PL in which natural photolysis occurs, Maiga et al., 2018).

Of the remaining 43 articles, 24 addressed CGGT applied in central

or sectorial WWTP. These articles were excluded since they do not meet the decentralized scope proposed in this article. During data collection, of the 19 pre-selected articles, two were excluded due to lacking data (Alves et al., 2012; Sklarz et al., 2013). In fact, in these two cases there was no sampling point between previous treatments (e.g., sedimentation tank, Sklarz et al., 2013; up-flow anaerobic sludge blanket digestion, Alves et al. 2012 and CGGT, hence the performance of the CGGT could not be evaluated without interference of the previous treatment.

Therefore, 17 articles were finally selected as having met all eligible criteria. The selected articles were from 11 countries and three different continents. Of these 17 selected articles, seven (41%) were from Asia/Oceania, India being the country with the highest number of publications (3). Of the remaining articles, six (35%) were from European countries (three articles from Belgium, and Greece, France and Portugal with one article each) and 24% from Latin American countries (Brazil and Colombia with two articles each) (Fig. 2A). Finally, most of the selected articles were published within the last five years (up to 80%, Fig. 2B) and the number of publications tripled when comparing 2019 with 2020. Therefore, despite the relatively low number of articles, there is an increasing interest in this novel research topic.

3.2. Design and performance of CGGT

3.2.1. Main treatment configurations and motivations for coupling green and grey-technologies

After an exhaustive revision of the selected articles, a general set of advantages were identified, concerning the coupling of these technologies in contrast to stand-alone green or grey technologies: i) **Reduced land footprint**. This feature of CGGT can facilitate urban decentralized schemes for water reuse or safe discharge, especially, in high densified cities (Alves et al., 2012) or areas not covered by centralized sewage collection (Talekar et al., 2018). ii) **Reduced costs and low maintenance needs**. For example, green technologies can decrease the organic load in the influent of AOP&EC, and thus increase efficiency while decreasing the maintenance costs of grey technology (Talekar and Mutnuri, 2020; Jaén-Gil et al., 2021). iii) **Enabling water reclamation or reuse, normally not achieved by green technologies alone**. For example, grey technologies can help polish the effluent of green technologies by efficiently removing pathogens (Álvarez et al., 2017; Talekar et al., 2018; Colares et al., 2019; Bakheet et al., 2020; Gomes et al., 2020). Some examples include systems designed to meet water reuse standards and even potable water needs in remote areas (Lakho et al.,

2020a) or in seasonal events (Lakho et al., 2020b). This feature of CGGT may represent an excellent option for low and middle-income countries, where traditional facilities often fail due to frequent energy supply failures and the lack of technical manpower (Álvarez et al., 2017). In these conditions, the use of green technologies such as CW was already considered adequate, given the low costs and low maintenance needs. Nevertheless, grey technologies can bring the added value of producing higher quality reclaimed water (Mishra et al., 2018). iv) **Enhancing performance in removing emerging contaminants and toxic or bio-recalcitrant compounds** (Chow et al., 2017; Casierra-Martinez et al., 2020; Gomes et al., 2020). CGGT can favour the breakdown of persistent organic compounds into more biodegradable substrates (Jaén-Gil et al., 2021). For example, a CW coupled with ozonation was statistically better at removing pharmaceuticals than a CW alone (Lancheros et al., 2019). Moreover, constructed wetland-solar photo-fenton coupled systems represent an alternative for reducing the discharge of pharmaceuticals in the environment by achieving high removal efficiency at low cost (Casierra-Martinez et al., 2020)

A greater focus on the green-grey rather than grey-green sequence was observed when targeting urban wastewater reuse onsite under decentralized schemes. Of all of the selected articles, 14 addressed green-grey technologies, four were about grey-green technologies and only one studied both green-grey and grey-green (Fig. 3). One of the reasons for this current “preference” for green-grey technologies is the added value brought by green technologies, which provide a more “environmentally friendly” appeal to operation of the system, while preserving the functional efficiency of grey technologies. In respect of this, green technologies are used as a pre-treatment to ensure a low content of suspended solids and organic matter in the influent of the grey technology and thus avoid increasing the dose, potential shielding from pathogens and the formation of toxic compounds (García-Segura et al., 2020). Another reason for the higher number of studies on green-grey technologies is their clear appeal in terms of ensuring diverse reuse schemes. In this regard, grey technologies are used to polish the effluent of green technologies, mainly through the removal of pathogens, and thus ensure a wider range of reuse schemes. In contrast to green-grey technologies, the focus of grey-green technologies in the selected articles was not water reuse. A clear example of this is that none of the articles analysed pathogens (e.g., *Escherichia coli* (*E. coli*) or total coliforms), which are key parameters in safe water reuse. Rather, the reported grey-green technologies focused on improving the efficiency of organic contaminant removal (Ma et al., 2018), emerging pollutants and

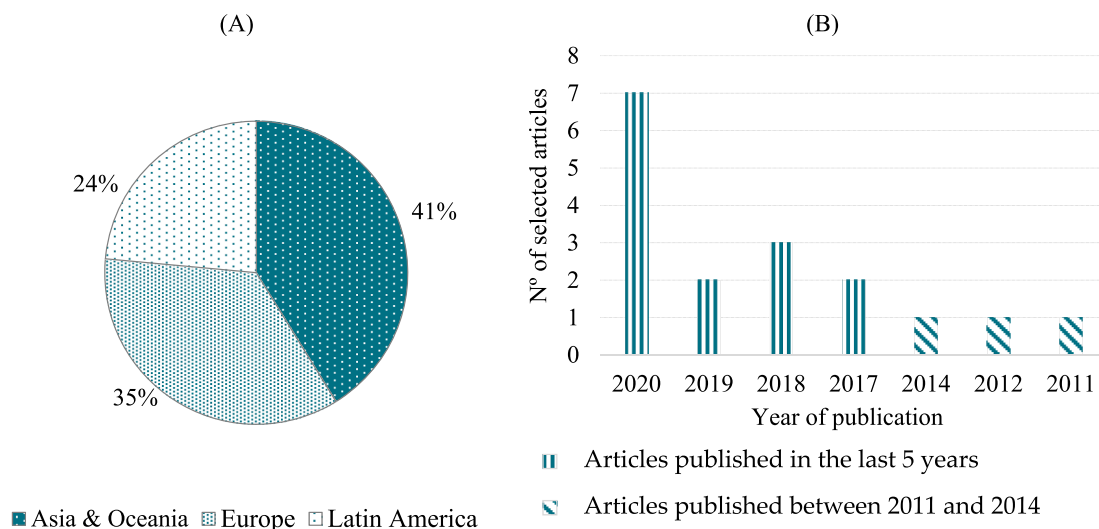


Fig. 2. Graphical representation of the main results of the systematic selection of articles: (A) selected articles per continent of origin; (B) selected articles per year of publication.

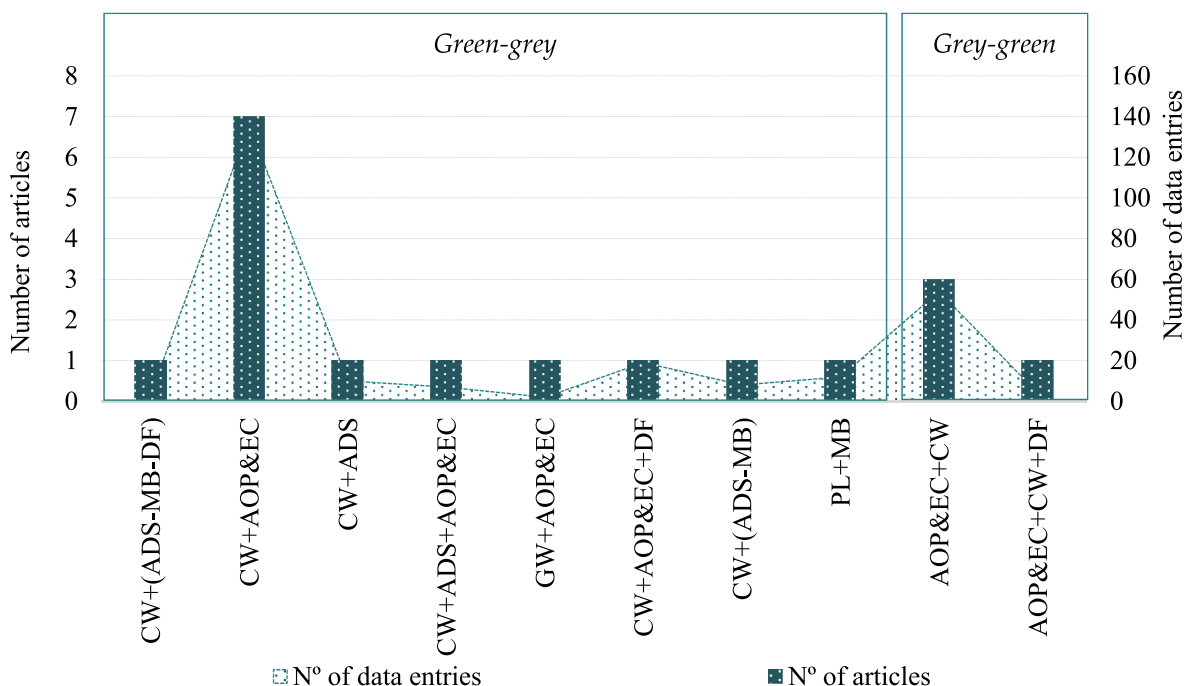


Fig. 3. Number of articles and data entries for each CGGT configuration (CW: Constructed wetlands; GW: Green walls; PL: Ponds and lagoons; AOP&EC: Advanced oxidation process and electrochemical process; ADS: Adsorption techniques; MB: Membrane filtration; DF: Disinfection methods). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

complex organic compounds (Chow et al., 2017; Gomes et al., 2020) not completely removed by grey technologies, by taking advantage of the biodegradation processes occurring in green technologies. This is achieved via the breakdown of pollutants in grey technology (e.g., advanced oxidation), and further removal via the biological treatment provided by green technology (Gomes et al., 2020).

Most of the articles dealt with CW implemented either before (12/17 articles) or after (4/17) grey technologies, representing approx. 90% of the total data entries in the Excel database. With regard to CW as a first stage, 11 of the 17 articles dealt with subsurface flow filters, of which six used horizontal sub surface flow (HSSF), five vertical sub surface flow (VSSF) and one a hybrid VSSF-HSSF. AOP&EC was the most common second stage, even though green-grey technologies sometimes included multiple steps, such as ADS-MB-DF, AOP&EC + DF, ADS + AOP&EC. HSSF were the only type of CW used in grey-green technologies as second stage after ozonation, cathode and anoxic oxidation and photocatalysis. This may be due to the greater potential of HSSF in comparison with other types of CW, since they provide further biodegradation of organic pollutants (not completely removed in the first “grey” stage) and by-products formed during the first stage.

AOP&EC was found to be the most studied grey technology (13/17) and the only one in the grey-green sequence. In contrast, the most common grey technologies when used as a tertiary treatment of secondary conventional technologies were MB and DF. Cathode and anoxic oxidation (4/17) was the most common AOP&EC, closely followed by ozonation (3/17) and photocatalysis (3/17), and then photo-Fenton (1/17). The second most prominent grey technology was ADS (4/17), zeolite and activated carbon being the most widely used reactive materials. MB was only found in three of the 17 articles, and mostly included as one step of potable water systems (that include diverse advanced technologies) applied right after green technologies. DF was only used in the final stage (3/17), using either chlorine or UV light.

Finally, only two of the articles addressed green technologies such as GW and PL. Although Pradhan et al. (2019) analysed the potential of greywater treatment with green roofs, no studies on wastewater treatment using GR coupled with advanced technologies could be found in the literature. The most likely reason for this is that, nowadays, GR are

mostly used for stormwater control (Razzaghmanesh and Beecham, 2014).

3.2.2. Design and operational features

For green-grey technologies, the most commonly applied pre-treatments were anaerobic, up-flow anaerobic sludge blanket reactor, septic tank, secondary treatment and settling tanks. The only grey-green technology that provided details on pre-treatment used a sedimentation tank. Therefore, due to the limited number of grey-green studies, no conclusions can be drawn regarding the different uses or aims of the pre-treatment stage. The most widely reported design parameters for green technologies in general were treatment area, depth, type of plant (genus or specie) and, for subsurface flow CW, substrate type and size. As Table 1 shows, treatment areas for CW varied greatly, mainly due to differences in implementation scale (full real scale, pilot plants, lab scale). The reported depths of CW were the usual ones, around 1 m for VSSF, around 0.6 m for HSSF and shallow depths for PL (Torrens et al., 2020; Vymazal et al., 2021). The plants used were the most common ones for CW: Phragmites, Typha, Cyperus (Vymazal, 2013), except for some specific native plants and Geranium. The standard filtration materials were also reported (gravel, sand), and using the normal sizes (sand or fine materials for VSSF, and coarse materials or gravels for HSSF) (Wang et al., 2020).

With regard to operational parameters, VSSF were fed intermittently and HSSF were mainly fed continuously. Overall, CW did not include intensification or mechanical devices, with some exceptions for VSSF (two filters with recirculation and one with aeration). HL presented typical values for all the green technologies (Boano et al., 2020; Torrens et al., 2021). For VSSF, the HL ranged from 0.02 to 0.2 m/day. Only one pilot plant (Lakho et al., 2020b) tested HL up to 2 m/day, which is extremely high for CWs. OLR for BOD₅ and COD for VSSF were within the usual ranges for vertical systems (Molle, 2014; Torrens et al., 2021). The only exception was the experience documented by Lakho et al. (2020a), with 710 gBOD₅/m².d and 830 gCOD/m².d due to very high HL. OLR for first stage HSSF were normal, ranging from 1 to 21 gBOD₅/m².d. HRT presented usual values for HSSF, the hybrid CW and the pond system (2–7 days). HRT was not reported for VSSF, since tracer

Table 1

Overview of main design and operation parameter for green technologies used in CCGT. Horizontal subsurface flow constructed wetland (HSSF), vertical subsurface flow constructed wetland (VSSF), saturated vertical flow constructed wetland (SVF), floating treatment wetland (FL), baffled constructed wetland (BSF), green walls (GW), ponds and lagoons (PL).

Parameters (unit)	Constructed wetlands			GW	PL
	VSSF	HSSF	FL - BSF-SVF		
Total area (m ²)	3–960	0.05–1000	1.38	0.12	73
Substrate (mm)	Sand (0–2), Gravel (2–10), Lava rock (8–16)	Gravel (10–32), crushed stone, LECA	Gravel	Perlite, coco coir	NA
Depth (m)	0.8–1.3	0.45–0.6	0.4–0.45–0.4	0.45	0.3
Plants (genus)	Phragmites, Typha, Cyperus, Géranium	Phragmites, Cyperus, Hymenache, Comelina	Hymenachne, Pistia, Lemna	Carex	NA
HL (m/d)	0.02–2	0.01–0.1	0.013	0.03	0.05
OLR (gBOD/m ² .d)	6–710	0.006–0.1	NR	4.6	NR
OLR (gCOD/m ² .d)	15–830	5–26	NR	10.7	16
HRT (d)	NR or NC	2.5–7	7–7–7	NR	6
Reference/ stage	A, B, C, D, E, F	G, H, I, E, J, K, L, M, N, O	P	Q	R

TA: total area, HL: hydraulic load, OLR: surface organic loading rate stage 1, HRT: hydraulic retention time, NR: not reported, NA: not applicable. Reference/ stage. A: Lakho et al. (2020b)/1; B: Talekar et al. (2018)/1 C: Gikas and Tsihrintzis (2012)/1; D: Talekar and Mutnuri (2020)/1; E: Álvarez et al. (2017)/1; F: Lakho et al. (2020a)/1; G: Horn et al. (2014)/1; H: Lancheros et al. (2019)/1; I: Mishra et al. (2018)/1; J: Casierra-Martinez et al. (2020)/1; K: Gomes et al., (2020)/1; L: Chow et al. (2017)/2; M: Ma et al. (2018)/2; N: Gomes et al. (2020)/2; O: Chen et al. (2011)/2; P: Colares et al. (2019)/1; Q: Bakheet et al. (2020)/1; R: Robles et al. (2020)/1.

tests are needed.

For the AOP&EC, the most widely reported design parameters were flow rate, HRT and type of dosing substance or light (see Table 2). There was a large variation in flow rate values depending on the scale used (lab scale, pilot or full-scale systems), while HRT values varied greatly according to the type of technology, from a few minutes for ozonation to a few days for photocatalysis. The HRT values for ozonation were of the

Table 2

Overview of the main design and operational parameters for grey technologies used in CCGT. Adsorption techniques (ADS), membrane filtration (MB), disinfection methods (DF), ultraviolet radiation (UV), advanced oxidation process and electrochemical process (AOP&EC).

Parameters (unit)	ADS	MB	DF	AOP&EC				
	Activ. carbon/ Zeolite	Ultra filtration	Reverse Osmosis	UV	Cathode/Anode oxidation	Photo catalysis	Ozone	Photo-Fenton
Substance/light (dose or ranges)	NA	NA	NA	NR	NaCl (50 mg/L), Iron (0.5–0.8 m ² /g)	TiO ₂ Light (103 μmol/m ² .s), UVA-V (4 × 10W)	O ₃ (10–347 mg/L)	Fe ²⁺ /H ₂ O ₂ (Ratio 0.1)
Reactor Volume (L)	2.34–27	NR	NR	NR	50000		1.4	NR
Flow (L/h)	NR	NR	NR	25	0.036–30	NR	0.31–50	NR
HRT (h)	24	NA	NA	NA	0.37–6	12–72	0.01–0.1	NR
Operating flux (LMH)	NA	3.3–28	11.5	NA	NA	NA	NA	NA
Energy used (kWh/m ³)	NR	0.29	NR	NR	0.73–27	NR	NR	NR
Pressure (bar)	NA	1	4	NA	NA	NA	NA	NA
Current (A)	NA	NA	NA	NA	4.4–30A	NR	NA	NA
Reference/ stage	A, B, C	D, E, F	E, F	G	H, I, J, K	L, M, N	O, P, Q, R	S

HRT: hydraulic retention time, LMH: operating flux (units of litres per square metre hour), NR: not reported, NA: not applicable. Reference/ stage (1 = first stage and 2 = second stage). A: Gikas and Tsihrintzis (2012)/2; B: Mishra et al. (2018)/2; C: Álvarez et al. (2017)/2; D: Lakho et al. (2020b)/2; E: Robles et al. (2020)/2; F: Lakho et al. (2020a)/2; G: Lakho et al. (2020b)/2; H: Talekar et al. (2018)/2; I: Bakheet et al. (2020)/2; J: Talekar and Mutnuri (2020)/2; K: Ma et al. (2018)/1; L: Horn et al. (2014)/2; M Chow et al. (2017)/1; N: Chen et al. (2011)/1; O: Lancheros et al. (2019)/2; P: Colares et al. (2019)/2; Q: Gomes et al. (2020)/2; R: Gomes et al. (2020)/1; S: Casierra-Martinez et al. (2020)/2.

same order of magnitude as when applied as tertiary treatments in conventional WWTPs, where green technologies are not present (Helmreich and Metzger, 2017). For photocatalytic technologies, two studies reported very high HRT (12–72h) when used as a first stage.

Once again with respect to operational parameters, the dosing of ozone ranged between 10 and 347 mg/L, which is rather high in the upper range when compared to ozonation in tertiary treatments (Helmreich and Metzger, 2017; Rizzo et al., 2019). The upper range doses corresponded to Gomes et al. (2020), where ozonation was tested both as first and second-stage treatments in combination with CW. The current density for EC was of the same order of magnitude as that used in electrochemical oxidations as stand-alone water treatment technologies to remove contaminants of emerging concern - CECs (García-Segura et al., 2020; Stirling et al., 2020). These densities provided a range of energy consumption from 0.7 to 27 kWh/m³, even though there was little information on real water matrices. Very little information has been reported for the design parameters of MB and ADS. Finally, operating flux (LMH) and pressure were reported for MB, with usual values ranging from 11.5 to 28 LMH and 1–4 bar, respectively, except for one case where the flux was very low (3.33). Specific aeration demand was also provided for one reference, with tested values ranging from 0.1 to 1.2 m³/m².h.

Overall, designers did not apply specific design and operation methods or values when linking green and green techs. The parameters were the usual ones when dealing with individual green technologies (e. g., organic load rate, hydraulic load rate) or grey technologies (e.g., electrochemical processes, optimum batch reactor time, ozone doses or ozone to COD ratios and Fe²⁺/H₂O₂ ratios). Therefore, it seems that design/operational/experimental parameters have been not optimized to take advantage of the pairing. Consequently, the area does not seem to be reduced when compared to single green technologies.

Given the above, in order to optimize CCGT design, further studies should focus on variables or parameters aimed at enhancing the functioning of CCGT as a whole and not as two separate technologies joined together. For example, in the case of green-grey technologies, investigating the role of influent characteristics required for the proper functioning of advanced grey technologies, and thus avoid “over designing” either the grey or the green technology. In respect of this, it is well known that photo-based AOPs and ECs require influents with a very low total number of suspended solids, respectively, to reduce risk of blocking UV radiation, specifically when disinfection is desired (Pirnie et al., 2006) and to avoid clogging (Radjenovic and Sedlak, 2015). Moreover, AOPs require influent with a low content of organic matter (up to 10–20

mg/L of total organic carbon), while ECs can tolerate higher contents. However, in both cases, the organic matter competes for radicals with the target microcontaminants and for photon absorption with H_2O_2 (Kwon et al., 2019), thus reducing the effectiveness of the treatment and increasing costs, for instance. Hence, design projects and experiments dealing with green technology as a first stage should consider achieving the minimum (no more, no less) removal of total suspended solids (TSS) and organic matter content, in order that the grey technology can work properly. Another aspect to take into consideration are the variables affecting the potential formation of negative by-products when chloride is present, especially for EC (Bagastyo et al., 2012). Finally, it would also be interesting to ascertain the minimum area required for CW to achieve the target removal rates for the whole treatment as well as a good functioning of grey technology. Therefore, the land footprint of CGGT can be reduced as far as possible, fact which might play an important role on the mainstreaming of this technologies in urban settlement.

3.2.3. Water quality parameters

A total of 58 water quality parameters were analysed in the 17 selected articles for this review. However, there were enough data (at least three data entries) to calculate the average performance of CGGT for only approximately 50% of all analysed parameters. The number of analysed parameters varied across the different articles. For CGGT in general, 19 parameters were analysed in three or more articles, while this number is even lower for green-grey and grey-green technology, with 15 and two parameters, respectively. The less common parameters were the pharmaceutical compounds such as ibuprofen, naproxen, diclofenac and carbamazepine (fewer than four articles), organic acids, and other pharmaceuticals such as metoprolol, propranolol, sulfamethoxazole, trimethoprim and clarithromycin (in just one article). In contrast, the most common parameters for CGGT were related to organic matter measurements. All papers except one (Chow et al., 2017) analysed at least one form of organic matter, with the most common parameters chosen being COD and BOD₅, followed by TOC and DOC. Of all the parameters, only COD, BOD, TSS and DOC were reported for both green-grey and grey-green configurations.

As Fig. 4 shows, other relevant water quality parameters when dealing with green-grey technologies were nutrients (NH_4-N , TN, PO_4^- , TP) and microbiology indicators (total coliforms). Surprisingly, *E. coli* and TSS were only analysed in six articles addressing green-grey technologies, even though both parameters are widely required to comply

with reuse standards. Only articles dealing with green-grey technologies analysed *E. coli* and TSS, which were measured more often in the effluent of the first-stage green technology (20 data entries) than in the effluent of the second-stage grey technology (13 data entries). The latter indicates that even though one of the main reasons for coupling such systems is to achieve reuse standards, the role of grey technology as the final step for removing microorganisms is not being prioritized, which is conflictive considering that the performance of green technologies is usually limited in this regard. Organic pollutants, pharmaceutical micropollutants and heavy metals were only addressed in three or fewer articles, although in many cases at least three data entries were obtained.

3.2.4. Water quality performance

The lack of information regarding several treatment configurations and, to a greater extent, differences between the designs applied hinder a comprehensive interpretation of the performance of specific configurations and further comparisons between them. Therefore, in this section we focus on providing an overview of the overall performance of CGGT and the role of green and grey stages for pollutant removal.

In terms of overall CGGT performance, average organic matter removal was higher than 80% for COD, BOD. Regarding nutrient removal, the average removals of NH_4-N , TN, TP were above 70% (Fig. 5, A). In contrast, negative removals were obtained for NH_3 and TKN in two systems (although not the same systems). Indeed, most of the systems analysing NO_3-N and NO_2-N also revealed negative removals (see Supplementary material), all resulting from CW. However, average results for nitrogen species and corresponding removal efficiency need to be analysed with caution, since the number of articles differs for each parameter, and the variability of TKN is much higher than for TN. Nevertheless, these results reflect the natural contribution that CW can introduce in some nitrogen species and reflect known issues with good nitrification but limited denitrification in VSSF, or limited nitrification in HSSF. Thus, the need to ensure that full nitrification-denitrification conditions are achieved, especially when water reuse is intended, leaves space for optimizing through adequate CGGT conjugation. Also, total coliforms and *E. coli* were removed at a high rate (4.8 log and 4.3, respectively), with very low standard deviation, regardless of the type, configuration or operational differences across the data. This indicates that CGGT are consistent when it comes to removing such contaminants.

When comparing green-grey and grey-green technologies, COD



Fig. 4. An overview of parameters analysed across CGGT. Note that the graphs show only parameters appearing in more than one article and with at least three data entries in the database.

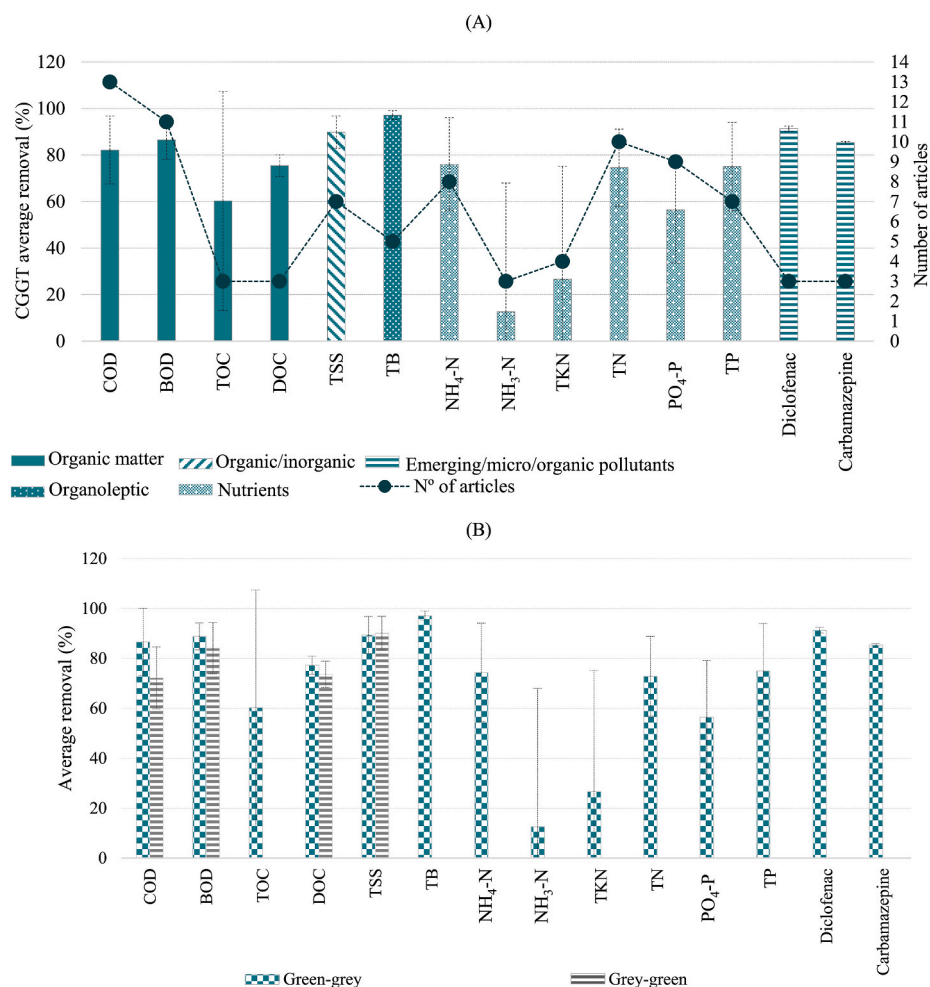


Fig. 5. Overview of the performance of CGGT at removing pollutants from urban wastewater. A: CGGT overall performance. Note that the graph shows averages taking into consideration all data for CGGT, and thus does not discriminate by types of configurations. B: CGGT performance (green-grey and grey-green configurations). Detailed data can be seen in Supplementary data. Nitrates are not included in the chart, being largely negative. E. coli and TSS are not included due to differences in units. Detailed data regarding other parameters can be seen in Supplementary data. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

removal tended to be higher in the published green-grey technologies (Fig. 5, B). Overall, BOD and DOC removal tended to be similar in green-grey and grey-green systems. TSS removal appeared to be similar in both types of CGGT. For all other studied pollutants, the lack of data for grey-green systems limited the opportunity for comparison.

There is a substantial lack of data regarding the performance of each stage of CGGT (see Supplementary data, Excel database). The majority of articles only provided data concerning the total removal of pollutants, and thus, it was difficult to clarify the role of green and grey stages for respective total removal. Despite this, removals in the 1st and 2nd stages were compared wherever possible. Overall, most of the removal was achieved in the 1st stage (for all parameters for which it was possible to calculate removal in both stages), except for total coliforms, where the higher removal rate was achieved in the 2nd stage (especially for green-grey technologies). It was observed that the removal of COD was mostly achieved in the 1st stage. Although the 2nd stage achieved COD removals >30% relative to its inflow concentration, its contribution to the overall removal tended to be on the low side (5–20%, in some cases negative, in a specific case >40%). This suggests that in green-grey systems, the 2nd stage deals mostly with recalcitrant material. For BOD₅, the trend was similar to that of COD. Most of the removal was found to occur in the 1st stage, while the contribution of the 2nd stage was limited (<40%). The removal of Turbidity (TB) and TSS in the first stage of green-grey technologies was above 80%. The latter is especially important for 2nd stage (grey technologies such as UV) to be able to work properly, and thus neither the green nor grey tech is overdesigned.

Generally speaking, negative contributions of the second stage were observed for NH₄-N, NH₃-N, and TKN, although a comparison across

different nitrogen forms was not possible, since not all studies analysed the different N parameters at the same time. NH₄-N removal was mostly in the 1st stage, but in some systems the 2nd stage may be relevant if nitrification is not favoured by the design of the 1st stage. Some grey technologies may result in the release of NH₄-N, probably due to the oxidation of recalcitrant organic matter, but the limited data did not allow us to further explore this. NO₃-N was mostly generated in the first stage, due to nitrification, but several different grey technologies could remove the nitrate in the 2nd stage.

3.2.5. An emerging alternative for the removal of emerging contaminants

One promising application of CGGT is the removal of emerging contaminants (organic micropollutants, microplastics, antibiotic resistance genes, etc.). With regard to organic micropollutants, it is well documented that the disappearance of the original form of micropollutants (= parent compound) may be achieved via a variety of processes, including green or grey technologies. Whatever the cause, in order to fully understand the fate of these pollutants, it is vital to also consider transformation products (TPs), micropollutant metabolites and conjugates (Castaño-Trias et al., 2021). TPs and parent compounds can behave differently. TPs can be less biodegradable, more toxic and inhibitory compared to the parent compound. Although a decrease in parent compound effluent concentrations compared to the influent is the most frequent scenario, it is not the only one. A higher mass load in the effluent than in the influent can be detected if the TP is more recalcitrant than the parent compound (e.g., metoprolol acid vs. metoprolol (Carballa et al., 2017)). The importance of this issue is reflected by the fact that national and/or international research strategies have been

developed to assess health risks associated with exposure to trace concentrations of multiple TP (Evgenidou et al., 2015).

The above being said, the effectiveness of combined treatments will always depend on the type of water effluent to be treated. The use of green technology has been widely suggested as an eco-friendlier solution for the removal of organic pollutants from complex wastewater matrices (Jaén-Gil et al., 2021). However, incomplete elimination of non-biodegradable pollutants and bio-recalcitrant intermediates generated has also been reported. For the remediation of low biodegradable effluents, highly reactive and non-selective grey treatment has been widely suggested as a suitable post-treatment solution. On the other hand, the application of grey treatment as a pre-treatment step to biological treatments has been suggested to convert the contaminants into more readily biodegradable intermediates, and thus reduce the total cost of the treatment process (Cesaro et al., 2013).

Of the articles reviewed here, only two reported on the performance of green-grey systems. Lancheros et al. (2019) studied ibuprofen and naproxen, with the combined systems achieving 95–97% and 96–97% removal, respectively. The 1st stage by CW ensured the highest removal (92% and 80%, respectively), while the 2nd stage (AOP) ensured the remaining fraction. Ibuprofen and naproxen are known to be easily biodegraded, so in this case the grey technology ensured the removal of a minor recalcitrant fraction. Casierra-Martinez et al. (2020) studied diclofenac and carbamazepine, two recalcitrant compounds in biological processes. In this case, from a total removal of 90–92% and 85–86%, respectively, the 1st stage by CW only provided 40% removal, while solar photo-fenton advanced oxidation complemented the treatment. No details were provided in either study regarding intermediates or transformation products. On the other hand, data from grey-green systems applied under centralized conditions revealed the limited capacity of post-treatments to complement the removal achieved by ozonation for pharmaceutical metabolites (Kharel et al., 2021). Therefore, while the reviewed papers reported the expected performance for organic micropollutants based on single technologies alone, they also revealed that the field is lacking further studies to fully potentiate the synergies of CGGT in addressing emerging contaminants. No studies were found on microplastics and CGGT in the literature.

3.2.6. Water reuse

Legislation can be found around the world that regulates parameters for water reuse at national or supranational level, with different demands, also based on their final application. In some cases, such as emerging pollutants, limits are not usually provided. Some binding values have not yet been reported on the EU Watch list decision (e.g. 2020/1161) and the EU regulation for water reuse (European parliament and the council of the european union 2020. Regulation, 2020/741) states that additional requirements may be considered for contaminants of emerging concern, depending on the outcome of risk assessment. On the other hand, standard parameters, including TSS, organic and microbiological parameters, are regulated in many cases (e.g., European parliament and the council of the european union 2020. Regulation, 2020/741). That being said, not enough data were provided in the studies for some of the regulated parameters in the evaluated CGGT configurations, or at least not consistently (e.g., in the case of nitrate, and in general of nitrogen forms, the results were not provided in a homogeneous way). Nitrate may well be an issue in some of the CGGT configurations and needs to be carefully considered to ensure safe reuse schemes. On the other hand, the reclamation of nutrients coming from wastewater treatments, for example in agricultural areas, could be fostered as an alternative to replace mineral fertilizers (Masi et al., 2018b; Muys et al., 2021).

For some of the remaining parameters, EU minimum requirements for water reuse (EU 2020/741) were checked after the 1st stage (when possible), and at the end of the treatment line for both green-grey and grey-green configurations (Table 3). For green-grey and TSS concentration, in those articles reporting the parameters it was possible to

observe that TSS already complied with the EU limits after the first green stage (<10 mg/L) in four of the articles, whereas they did not yet comply in another one. In the latter case, the application of the grey treatment was required to reduce TSS concentration to 14.9 mg/L, sufficient for classes B, C and D of EU 2020/741. Similarly, for BOD, class B quality was obtained after the first green stage as a minimum in five of the reviewed articles, but not obtained in three others. Another article complied with class B after the first grey stage. After the second grey stage, class B was obtained in terms of BOD concentration (<25 mg/L) in all cases. Finally, very few articles reported *E. coli* removal in all treatment stages. Nevertheless, it was observed that green treatment, even though largely reducing *E. coli* in some cases, was not always sufficient in itself. As expected, in fact, a grey treatment was usually required for disinfection. In all of the reported cases, following a CGGT the effluent complied with *E. coli* values even for the most restrictive A class. On the other hand, where other classes were considered (e.g., class D, limit 10,000 number/*E. coli*), it was apparent that only a single stage might be sufficient.

3.2.7. Energy requirements and costs

None of the reviewed articles provided information regarding the energy requirements and costs of combining grey-green technologies. Few articles on green-grey sequence even reported any of these data (only four articles addressed energy demands and only two costs), and no comparison was provided with sole technologies or other CGGT. On top of that, limited data were reported on the contribution made by each treatment stage. Usually, the data reported concerned either the performance of CGGT as a whole (Robles et al., 2020) or the performance of the grey technology alone (Talekar et al., 2018; Bakheet et al., 2020; Talekar and Mutnuri, 2020). Very little information was provided concerning the contribution of green technologies to overall CGGT energy demand, which may be related to the fact that these technologies are recognized as passive energy tech (consuming very little or no energy at all). As a consequence, it was difficult to estimate the role played by each treatment stage in energy expenditure or compare the energy performance among different types or configurations of CGGT and between CGGT and sole technologies.

Broadly speaking, the energy requirements of grey technologies and costs were found to vary between 0.73 and 27 kWh/m³ and between 0.04 and 0.4 €/m³, respectively. The configurations with the lowest energy requirements were GW + AOP&EC and PL + MB, with 0.29–0.73 kWh/m³. Such values are promising if we consider that centralized treatment uses around 1 kWh/m³, including transportation (Capodaglio and Olsson, 2020) and the sole use of membrane bioreactor uses around 1–2 kWh/m³ (Gabarrón et al., 2014). In addition, CW + AOP&EC were found to require approximately 25 times more energy than GW + AOP&EC. Such a large variation may be related to different implementation scales (pilots, lab and full scale), type of wastewater (and thus different pollutants loads), hydraulic load rate (and thus different amount of water to be pumped through the system) and other specific requirements of each treatment configuration (e.g., light for the photoelectrochemical process). Therefore, such aspects should be considered when comparing CGGT in terms of energy requirements. Overall, due to its novel nature, more research is needed on the topic of energy and costs to overcome existing “path dependency” in the field of water management and thus promote the implementation of CGGT at real scale as an alternative to conventional and centralized approaches.

3.3. Difficulties during data collection and recommendations

In some cases, the results were only reported in plots or figures, which hindered the collection of accurate data regarding their performance and consequently restricted the reliability of data comparison. In respect of this, we recommend that further CGGT studies include more user-friendly data interpretation formats, either in the manuscript or as supplementary data. Moreover, there was a clear lack of consistency on

Table 3

Influent and effluent concentrations in both the 1st and 2nd stage of CGGT configuration and verification of compliance with EU 2020/741* for water reuse.

Pollutant (unit)	CGGT configuration	References	Influent	1st stage		2nd stage	
				Effluent	Comply EU 2020/741	Effluent	Comply EU 2020/741
TSS (mg/L)	CW + AOP&EC + DF	Álvarez et al. (2017)	41.0	6.7	YES	6.4	YES
	GW + AOP&EC	Bakheet et al. (2020)	90.7	1.4	YES	N.R	N.R
	CW+(ADS-MB-DF)	Lakho et al. (2020b)	136.7	6.7	YES	N.R	N.R
	CW + ADS	Gikas and Tsihrantzis (2012)	124.1	77.9	NO	14.9	NO ^a
	CW + AOP&EC	Lakho et al. (2020a)	71	5.6	YES	N.R	N.R
BOD (mg/L)	CW + DS + AOP&EC	Mishra et al. (2018)	48	26	NO	18	NO ^a
	CW + AOP&EC + DF	Álvarez et al. (2017)	48	35	NO	N.R	N.R
	CW + AOP&EC + DF	Álvarez et al. (2017)	64	14	NO ^a	4.5	YES
	GW + AOP&EC	Bakheet et al. (2020)	140	2	YES	N.R	N.R
	CW+(ADS-MB-DF)	Lakho et al. (2020b)	355	16.3	NO ^a	N.R	N.R
	AOP&EC + CW + DF	Ma et al. (2018)	46	22	NO ^a	4.3	YES
	CW + AOP&EC	Horn et al. (2014)	224	20.2	NO ^a	N.R	N.R
	CW + ADS	Gikas and Tsihrantzis (2012)	211	155	NO	20	NO ^a
	CW + ADS	Gikas and Tsihrantzis (2012)	211	110	NO	N.R	N.R
	CW + AOP&EC	Lakho et al. (2020a)	119	4	YES	N.R	N.R
E. coli (number/100 mL)	GW + AOP&EC	Bakheet et al. (2020)	29100	3200	NO ^b	0	YES
	CW + AOP&EC	Colares et al. (2019)	43	0	YES	N.R	N.R
	CW + AOP&EC	Horn et al. (2014)	8.2·10 ⁵	10 ⁵	NO	0	YES
	CW + AOP&EC	Horn et al. (2014)	8.2·10 ⁵	0	YES	0	YES

*Class A: All food crops consumed raw where the edible part is in direct contact with reclaimed water and root crops consumed raw; all irrigation methods. Class B: Food crops consumed raw with edible part above ground and not in direct contact with reclaimed water, processed food crops and non-food crops; all irrigation methods. Class C: Food crops consumed raw with edible part above ground and not in direct contact with reclaimed water, processed food crops and non-food crops; drip irrigation or other irrigation method that avoids direct contact with the edible part of the crop. Class D: Industrial, energy and seeded crops; all irrigation methods.

^a Sufficient for class B, C, and D.

^b Sufficient for class D. CW: Constructed wetlands; GW: Green walls; PL: Ponds and lagoons; AOP&EC: Advanced oxidation process & electrochemical process; ADS: Adsorption techniques; MB: Membrane filtration; DF: Disinfection methods N.R: Not reported.

the reporting of CGGT performance concerning units. We found more than 11 different units referring to CGGT performance at removing pollutants (e.g., mg/L, µg/L, g/m³·d, g/L·d, % of removal, g/m²·d, nmol/L, NTU, CFU, MPN/100 ml). We consider it very important to standardize units when referring to the same type of pollutants or, at least, to provide enough information so the data can be standardized. Indeed, as CGGT includes green technologies, we strongly recommend reporting mass balance or water flow amount across treatment stages, since concentrations can be overestimated. Furthermore, detection limits were not usually reported in the reviewed articles, making it difficult to estimate some parameters.

In addition to the above, data reporting on the performance of each treatment stage (first, second, third, etc.) was very limited. It was observed that overall removal was reported more often than removal performance in each treatment stage. Moreover, when overall removal was not reported, the performance of the entire CGGT could not be estimated for some parameters, due to missing data regarding the performance in one of the treatment stages. For example, Robles et al. (2020) evaluated the performance of a membrane-coupled high-rate algal pond when removing COD, NH₄⁺-N, PO₄³⁻-P, and TP. However, only the overall removal of COD, NH₄⁺-N, PO₄³⁻-P was reported (or could be calculated) and no information was provided regarding each treatment stage. Lakho et al. (2020a) evaluated 35 parameters. However, the performance of 1st stage, 2nd stage, and overall performance was only reported (or could be calculated) for 22, 5 and 8 parameters, respectively. Of the 19 parameters evaluated by Lakho et al. (2020b) in the effluent of the 1st stage (BOD, COD, TSS, EC, turbidity, metals and micropollutants, mainly pharmaceuticals), only three were evaluated in the effluent of the 2nd stage.

Furthermore, a disparity was found in the type of parameters evaluated in each treatment section. Our results indicate that the selection of parameters was mostly based on the target contaminants to be addressed by each treatment stage or according to research objectives or priorities. For example, Bakheet et al. (2020) evaluated the performance of GW coupled with EC at removing 13 different parameters (including organoleptic, organic matter, biological, nutrients and heavy metals). The authors reported data concerning the performance of the 1st stage

(GW) for all parameters. This comprehensive evaluation may be related to the novelty of this technology, which is reflected in a current need to validate their potential to remove a wide range of contaminants. Meanwhile, data concerning the 2nd stage (grey technology) was focused on microbiological parameters, such as *E. coli* and coliforms, which is in line with the main motivation to couple these technologies: meeting water reuse standards. Lakho et al. (2020b) also proposed a similar technology that couples a mobile vertical flow CW with a drinking water production system to achieve potable water quality standards. However, only nitrates, nitrites and ammonium were measured in the effluent of the 2nd stage (grey technology), while microbiological parameters such as *E. coli* and total coliforms, which are important for various water reuse standards, were not measured at all. Lakho et al. (2020a) proposed a VSSF coupled with MB-based potable water system, but only the overall removals of *E. coli* and total coliforms were reported, and thus no information was provided regarding the performance of each treatment stage.

Whether certain contaminants are evaluated as a whole and/or in different stages of the treatment, we believe that establishing a key set of parameters to be evaluated across all treatment stages of CGGT is an important step towards a comprehensive understanding of the functioning of coupled technologies. This will allow both for their optimization and a comparison either across CGGT or with sole technologies. Moreover, in order to facilitate the quantification of additional benefits of coupled systems, studies comparing CGGT with sole technologies are needed (only one of the 17 articles compared the performance of the combined system to stand-alone grey, namely, Talekar et al., 2018).

Although characterization of the parameters varied across different articles, we found that green technologies, and particularly CW, tend to be the most consistent, offering largely plant species, material, surface area, hydraulic load rate and organic load. In contrast, information regarding the operation and design of grey technologies was frequently insufficient to calculate operational conditions (e.g., hydraulic retention time, volumes, dosage). Moreover, data concerning operational features (e.g., HLR, fouling rate, surface area) also varied across articles and were sometimes not available, making it difficult to correlate CGGT performance and design features. On top of that, only a few articles reported

data concerning other relevant topics beyond treatment performance, such as, for example, potential oxidation by-products (Chen et al., 2011), energy requirements (Talekar et al., 2018; Bakheet et al., 2020; Robles et al., 2020; Talekar and Mutnuri, 2020) and costs (Bakheet et al., 2020; Robles et al., 2020). Finally, due to the novelty of the topic, no article dealt with simulations/modelling using secondary data. Therefore, modelling and simulations could be an interesting research field to pursue in the coming years. All in all, such recommendations can play an important role in terms of facilitating knowledge transfer and comparison, especially considering the novelty of the field.

4. Conclusions

CGGT have a great potential for facilitating decentralized treatment schemes in cities by enabling water reclamation or reuse, enhancing the treatment of emergent contaminants (higher removal of bio-recalcitrant compounds) and eliminating toxic products or by-products. Moreover, CGGT represent a promising alternative for reducing the land footprint (the main limitation to implementing green technologies in urban settlements), as well as the energy and costs usually associated with standalone grey technologies by increasing pre-treatment and reducing the need for maintenance. However, due to the novelty of the topic, the performance of CGGT in comparison with sole technologies needs to be explored further and better documented, including data on the reduction of costs, energy demand and required land footprint.

All in all, the field is still at an early stage of development, as reflected by the low number of publications available for each specific technology configuration. However, the main reasons for coupling such technologies seem to be consistent across different articles. Grey technologies are usually included as a first stage to remove recalcitrant materials and/or microbiological parameters, or to break down slowly biodegradable substances so that they can be further removed in green systems. In turn, grey technologies are normally applied at a second stage to remove pathogens that had not been eliminated via green technologies and thus meet safe reuse standards. Green-grey configuration was the predominant coupled type across all articles, and the configuration CW + AOP&EC the most widely studied. In respect of this, further studies should take a more in-depth look at existing configurations and explore new ones (especially grey-green technologies).

Generally speaking, designers do not apply specific design and operation methods or values when linking green and green technologies. Both are mainly designed as sole technologies and later forcibly put together as a coupled technology. There is therefore room for optimization and the field would benefit from more research focused on this integration and the comparison with sole technologies. Further studies should focus on variables and parameters influencing the functioning of the CGGT as a whole, such as, for example, the minimum influent quality required for a proper functioning of advanced grey technologies (in the case of green-grey) and minimum area required for CW to maintain target removal rates.

Despite the large number of data entries collected (343), the average performance of CGGT could only be calculated for approximately half (setting a minimum of three data entries), since several studies did not share many of the parameters. The most common parameters across the 17 articles were COD, BOD, NH₄-N, TN, PO₄⁻, TP and Total coliforms. Further research should be more consistent on data reporting, mainly concerning water quality parameters (units and type of parameters), treatment performance across treatment stages, design, operation details (e.g., loads, flow, volume, area), energy demand and costs (maintenance and implementation). All are crucial for a better understanding of the functioning of CGGT and thus essential to support their optimization and competitiveness, as well as their comparison across different studies.

The above being said, it is important to highlight that, in addition to technological development, the transition from centralized urban water management towards a decentralized model will also require a set of changes in the governance and institutional framework of water management in cities. In respect of this, it is crucial that the water community (scientists, urban planners, decision makers) explore strategies to overcome current barriers to implementing coupled technologies, such as the following: costs for implementing water source separation in existing buildings/houses, limited social acceptance due to the “yuck factor”, bureaucratic burden (e.g. permits for implementation and water reuse onsite), discrepancies concerning local/regional/national/european normatives for water reuse (lack of a common baseline for water quality standards), path dependency affecting decision making and policies development, required adaptations of central wastewater treatment plants due to reduced water flow and much more concentrated wastewater.

Author statement

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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.

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Appendix A. Supplementary data

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