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Increasing resource circularity in wastewater treatment: Environmental implications of technological upgrades



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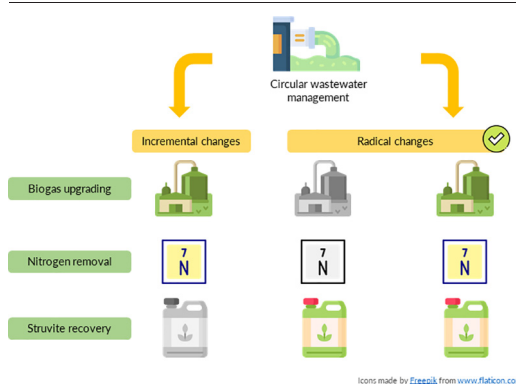
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HIGHLIGHTS

- Rethinking wastewater treatment is key to recover energy and resources in cities.
- Increased biogas production, nitrogen removal and struvite recovery are studied.
- Radical upgrades through nutrient recovery decrease the plant's environmental impacts.

GRAPHICAL ABSTRACT



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ABSTRACT

A paradigm shift is needed in wastewater treatment plants (WWTPs) to progress from traditional pollutant removal to resource recovery. However, whether this transformation produces overall environmental benefits will depend on the efficient and sustainable use of resources by emerging technologies. Given that many of these technologies are still being tested at the pilot scale, there is a lack of environmental assessments quantifying their impacts and benefits. In particular, an integrated approach to energy and nutrient recovery can elucidate the potential configurations for WWTPs. In this study, we conduct a life cycle assessment (LCA) of emergent wastewater treatment technologies aimed at increasing resource circularity in WWTPs. We focus on increasing energy self-sufficiency through biogas upgrades and a more radical circular approach aimed at nutrient recovery. Based on a case-study WWTP, we compare its current configuration with (1) implementing autotrophic nitrogen removal in the mainstream and deriving most of the organic matter for biogas production, which increases the quality and quantity of biogas available for energy production; (2) implementing struvite recovery through enhanced biological phosphorus removal (EBPR) as a radical approach to phosphorus management, offering an alternative to mineral fertilizer; and (3) a combination of both approaches. The results show that incremental changes in biogas production are insufficient for compensating for

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the environmental investment in infrastructure, although autotrophic nitrogen removal is beneficial for increasing the quality of the effluent. Combined phosphorus and energy recovery reduce the environmental impacts from the avoided use of fertilizers and phosphorus and the nitrogen release into water bodies. An integrated approach to resource management in WWTPs is thus desirable and creates new opportunities toward the implementation of circular strategies with low environmental impact in cities.

1. Introduction

The circular economy is becoming increasingly popular in cities to approach the sustainable management of natural resources. A circular urban metabolism can help “keep products, components, and materials at their highest utility and value at all times” (Bocken et al., 2017) by taking advantage of the local resources available in cities. Urban circular strategies target a large variety of resource flows and urban sectors, from infrastructure to consumers. (Petit-Boix and Leipold, 2018). The conception and implementation of strategies requires, however, a paradigm shift in different urban sectors to increase the availability of local resources while reducing the environmental impacts of urban infrastructure. Wastewater treatment plants (WWTPs) are an essential component of urban infrastructure; this paradigm shift has been discussed but has yet to become a reality.

In recent decades, water research has called for a transition from “what must be removed” from wastewater to comply with water quality standards to “what can be recovered” from wastewater (Guest et al., 2009). The environmental consequences of focusing on removal technologies stress this need. For example, heterotrophic biological nitrogen removal (BNR) would be the classical choice when implementing nitrogen removal technologies in WWTPs, but it would entail a large energy consumption due to increased aeration needs to convert ammonia nitrogen to nitrite and to convert its subsequent oxidation to nitrate (ENERWATER, 2015). Embracing the “circular water economy” has the potential to expand the functions of urban wastewater systems, as becomes a valued service (e.g., clean water from sanitation), energy source (e.g., biogas) and carrier of resources (e.g., nitrogen and phosphorus) (Brears, 2020). Circular wastewater management aims to reuse, recycle, recover and reclaim the resources embedded in water flows, which is the result of rethinking the current system (Smol et al., 2020). Examples include water reuse in agriculture e.g., Almanaseer et al., 2020; Chen et al., 2021) or nutrient recovery in the form of struvite (e.g., Mavhungu et al., 2020). Earlier in the circularity debate, energy recovery and efficiency dominated the wastewater industry's efforts, (Guest et al., 2009), whereas current political and scientific debates encourage nutrient recovery. For instance, the European Union's updated Circular Economy Action Plan mentions nutrient recovery and wastewater among the planned measures (European Commission, 2020a). Both energy and nutrient recovery offer opportunities for resource self-sufficiency; the question that remains is what kind of circularity approach holds the largest potential to minimize the environmental impacts of wastewater treatment.

In the case of energy self-sufficiency, process optimization and onsite energy generation through biogas have become widespread circular strategies (Jenicek et al., 2012). To date, most self-sufficient WWTPs take advantage of biogas to heat their digesters and to generate electricity (Gu et al., 2017), and the literature has long suggested that replacing conventional energy sources can reduce a WWTP's environmental impacts (e.g., Foley et al., 2010; Pasqualino et al., 2009; Stokes and Horvath, 2010). However, the system often depends on external waste inputs, as many WWTPs apply codigestion of their sludge with urban organic waste to enhance biogas production (Mata-Alvarez et al., 2014). Considering that current environmental targets (e.g., CO₂ emissions) demand a more stringent implementation of energy self-sufficiency measures, emergent treatment technologies might be more beneficial than classical alternatives. An example is the implementation of autotrophic (i.e., without organic matter) BNR in the mainstream WWTPs, i.e., implementation of partial nitrification and anammox for removing nitrogen species from wastewater (Jetten et al., 1997; Kartal et al., 2010; Siegrist et al., 2008), instead of the classical heterotrophic

BNR. This new configuration of the mainstream line of an urban WWTP (refer to Section 2.1 for more details) reduces the energy consumption for aeration and uses most of the organic matter contained in the wastewater for biogas production, which enhances the energy recovery potential and reduces the overall operational costs of the facility.

This type of technology produces incremental changes in energy recovery based on the existing infrastructure, meaning, in this case, that a larger amount of biogas is available. Pilot-plant studies of new technologies, however, tend to focus on process efficiency, whereas the environmental impacts associated with their implementation are often disregarded. Upgrading WWTPs to enhance circularity requires an investment in natural resources that needs offsetting. We need to ensure that the environmental benefits outbalance the impacts, which might not be accomplished with a single technological upgrade. Here, nutrient recovery becomes an asset. Notably, the literature highlights the interest in examining the combined benefits of resource recovery approaches, such as onsite energy generation, nutrient recovery and water reuse (Mo and Zhang, 2013; Robles et al., 2020; Villarín and Merel, 2020; Wang et al., 2018). Additionally, Life Cycle Assessments (LCAs) are becoming abundant to capture the environmental effects of emergent technologies targeting nitrogen (N) and phosphorus (P) recovery in WWTPs (Roldán et al., 2020; Ruff-Salís et al., 2020; Sena and Hicks, 2018; van Zelm et al., 2020). Nutrient recovery is generally presented as an environmentally friendly strategy due to reduced chemical use and the substitution of mineral fertilizer (Lam et al., 2020).

Nutrient recovery thus constitutes a more radical change in wastewater treatment, as it shifts from the traditional removal paradigm and provides valuable resources that can be utilized by other urban sectors, such as local food production (Ruff-Salís et al., 2020). In parallel, enhancing energy recovery generates a partial increase in a WWTP's existing energy self-sufficiency. The question that remains is what kind of recovery strategy (i.e., incremental, radical or a combination of both) can increase the availability of local resources with the lowest environmental cost. Addressing this question can help redesign WWTPs and prioritize circular strategies from an environmental standpoint. However, while academics call for LCAs integrating both energy and nutrient recovery (e.g., Mo and Zhang, 2013), an increasing number of recovery solutions are being developed at the pilot level and still lack an evaluation of their impacts and benefits (Bohra et al., 2022).

To fill this gap, we assessed the environmental impacts and benefits of upgrading WWTPs for energy and nutrient recovery based on two emergent technologies. Additionally, we analyze the extent to which increased energy and nutrient recovery compensate for the environmental investment in infrastructure. Based on a case-study WWTP, we perform an LCA to analyze scenarios with technological upgrades, including i) implementing autotrophic BNR allowing an increase in biogas production, ii) recovering nutrients in the form of struvite through enhanced biological phosphorus removal (EBPR) and iii) implementing EBPR and autotrophic BNR allowing the recovery of nutrients in the form of struvite and simultaneously increasing biogas production.

2. Methods

2.1. Emergent resource recovery technologies

The exemplary technologies selected in this study are autotrophic BNR and EBPR. Autotrophic BNR can be implemented in the water line (mainstream) of an urban WWTP with several configurations, but the most promising setup includes two stages (Pérez et al., 2015). The first biological stage

aerobically removes organic matter through a high-rate activated sludge (HRAS) system operated at high organic loading rates (higher than 1 kg of chemical oxygen demand (COD) $\text{m}^{-3} \text{d}^{-1}$), low dissolved oxygen concentrations (lower than $1 \text{ g O}_2 \text{ m}^{-3}$) and low sludge retention times (lower than 3 days) (Carrera et al., 2022; Jimenez et al., 2015). Therefore, the secondary sludge produced in this stage would have higher anaerobic biodegradability than the secondary sludge produced in the conventional activated sludge (CAS) system that is available in most urban WWTPs. The effluent of this HRAS stage is wastewater with a low COD/N ratio, but the system does not remove ammonia nitrogen. HRAS can be implemented as the sole stage for organic matter removal before the autotrophic BNR process, or it can be implemented after a primary sedimentation and just before the autotrophic BNR process, as demonstrated like in the study system. In the selected configuration, organic matter removal is carried out in the primary sedimentation and HRAS processes. In the first configuration, which is only an HRAS process, the percentage of COD removal is approximately 40–60% (Güven et al., 2019). In the second configuration, which comprises the primary sedimentation plus HRAS process, the percentage of COD removal can be increased up to 65–80% (Carrera et al., 2022). The second stage is the autotrophic BNR process, which consists of two biological steps: partial oxidation of ammonium to nitrite (partial nitrification) and anaerobic ammonium oxidation (anammox) to di-nitrogen gas. Autotrophic BNR in the mainstream can be implemented through a single reactor (Laureni et al., 2016; Lotti et al., 2014) or through two reactors (Isanta et al., 2015; Pérez et al., 2015; Reino et al., 2018; Juan-Díaz et al., 2022).

From a nutrient recovery perspective, EBPR has been extensively utilized in recent years, as it is able to remove P without chemical precipitants (Oehmen et al., 2007). Instead, EBPR uses bacteria that accumulate P and transfer it to the sludge line. EBPR is implemented in current WWTPs with CAS systems by adding an anaerobic reactor to produce volatile fatty acids for P-accumulating bacteria (Izadi et al., 2020). If EBPR is implemented with autotrophic BNR in the mainstream, the HRAS stage should also be modified to include an anaerobic reactor. This technology has been successfully demonstrated at the lab scale by operating the EBPR-HRAS system at sludge retention times (SRTs) as low as 3 days (Roots et al., 2020; Zhang et al., 2021). Regardless of how the EBPR is applied (i.e., in a CAS or HRAS configuration), P is recovered in the sludge line after anaerobic sludge digestion. In the digester, P accumulates in the supernatant or digestate (Pastor et al., 2008), which is the main target of P recovery technologies. In this case, we selected Ostara® technology (Ostara, 2019) to recover struvite, which is a promising new P fertilizer with slow-release characteristics (Latifian et al., 2012; Rahman et al., 2014). Ostara® was selected because of the environmental performance of this technology when compared with other alternatives (Amann et al., 2018; Ruff-Salís et al., 2020). Moreover, Ostara® is one of the few struvite recovery technologies that operates worldwide and produces a commercial struvite fertilizer.

2.2. Upgrade scenarios in a case-study WWTP

The environmental effects of these technological upgrades are exemplified through an existing urban WWTP selected for our case study. The WWTP is located in the Metropolitan Area of Barcelona and serves 135,000 population equivalents (p.e.). This WWTP is representative of large facilities (urban agglomerations $>100,000$ p.e.), which represent 52% of the EU's wastewater load (European Commission, 2020b). The analysis compares four scenarios that result in various modifications of the original WWTP configuration (Fig. 1). These modifications lead to incremental changes in the quantity and quality of biogas production and/or radical changes in nutrient management, i.e., from removal to recovery (Table 1).

The baseline scenario (BS) (Fig. 1) corresponds to the current configuration of the WWTP. The facility removes organic matter through a CAS system operated at an SRT of 14 days, while P is chemically precipitated by adding iron chloride. This WWTP does not remove nitrogen species, being representative of 34% of the WWTPs in the European Union that

do not comply with more stringent treatment, according to article 5 of the Urban Wastewater Treatment Directive (European Commission, 1991). In fact, the nitrification process does not take place at SRT of 14 days because the plant was limited by the available volume of the reactors of the biological treatment. Due to this limitation, the WWTP had an exemption of the Public Administration regarding the nitrogen removal. Moreover, the WWTP was operated at a very low dissolved oxygen concentration to save energy consumption and to completely stop the nitrification process at any season of the year. In terms of energy self-sufficiency, this WWTP includes a biogas cogeneration unit for producing electricity and heat. The biogas produced by anaerobic digestion of a mixture of primary sludge and secondary sludge has 57% methane (CH_4) content based on the WWTP records. The selected technologies were assessed in three different scenarios.

The SAVING-E scenario (SE) (Fig. 1) considers the implementation of the autotrophic BNR in the mainstream through an HRAS system for removing organic matter and two separated reactors (partial nitrification and anammox) for removing nitrogen species. SAVING-E technology (www.saving-e.eu) has been tested at the pilot scale for three years in the same full-scale WWTP that is considered the baseline scenario. With this configuration, biogas production would be increased by 61%, and the CH_4 content would reach 75%. In this case, nitrogen species are removed through autotrophic BNR, whereas P is still precipitated through the addition of iron chloride.

The struvite recovery scenario (SR) considers a theoretical CAS-EBPR to transfer P to the sludge line (Izadi et al., 2020), where it can be recovered from the digestate in the form of struvite through an Ostara® process. From a technical point of view, struvite precipitation also contributes to N recovery since struvite is composed by a mol of potassium, a mol of ammonium and a mol of phosphate. However, if we make a mass balance, only around 5–6% of the total inflow N of the WWTP can be recovered in form of struvite. Considering the number and difficulty of the calculations needed in this study, we decided not to consider that small N-recovery amount. The total resource recovery scenario (TRR) combines autotrophic BNR and HRAS-EBPR by adding an anaerobic reactor to the HRAS system (Roots et al., 2020; Zhang et al., 2021), thus simultaneously enhancing the production of biogas and P recovery.

2.3. Life cycle assessment methodology

This section includes the main considerations and data for three of the phases of an LCA, namely, goal and scope definition, life cycle inventory analysis, and life cycle impact assessment (ISO, 2006).

2.3.1. Goal and scope definition

To assess the environmental impacts of the four scenarios, a functional unit needs to be defined to enable a scenario comparison. In this case, the impacts refer to 1 m^3 of treated wastewater, which is one of the most common functional units in wastewater-related LCAs (Corominas et al., 2013). We used the WWTP's average treated effluent for the period 2014–2016, which was $7,342,338 \text{ m}^3/\text{year}$. A lifespan of 30 years was assumed for the WWTP based on previous estimations (Morera et al., 2017; Risch et al., 2015). The infrastructure associated with the upgrades has an expected lifespan of 20 years according to the producer.

The system boundaries are shown in Fig. 2. For the purpose of our analysis, the operation stage will elucidate the short-term variations in the environmental impacts of WWTPs resulting from technological upgrades. The construction of the whole WWTP was excluded from the analysis, as it is assumed to remain constant before and after the upgrades (Güven et al., 2018; Hadjimichael et al., 2016). However, we did consider the additional impacts related to the construction of new infrastructure required for the technological upgrades. The end of life of the new infrastructure was not modeled as it is still uncertain whether the tanks could be reused or recycled for similar purposes. Sludge final disposal and solid waste management were also excluded. In our case, sludge final disposal undergoes thermal drying, which is an energy-intensive process (Bennamoun et al., 2013)

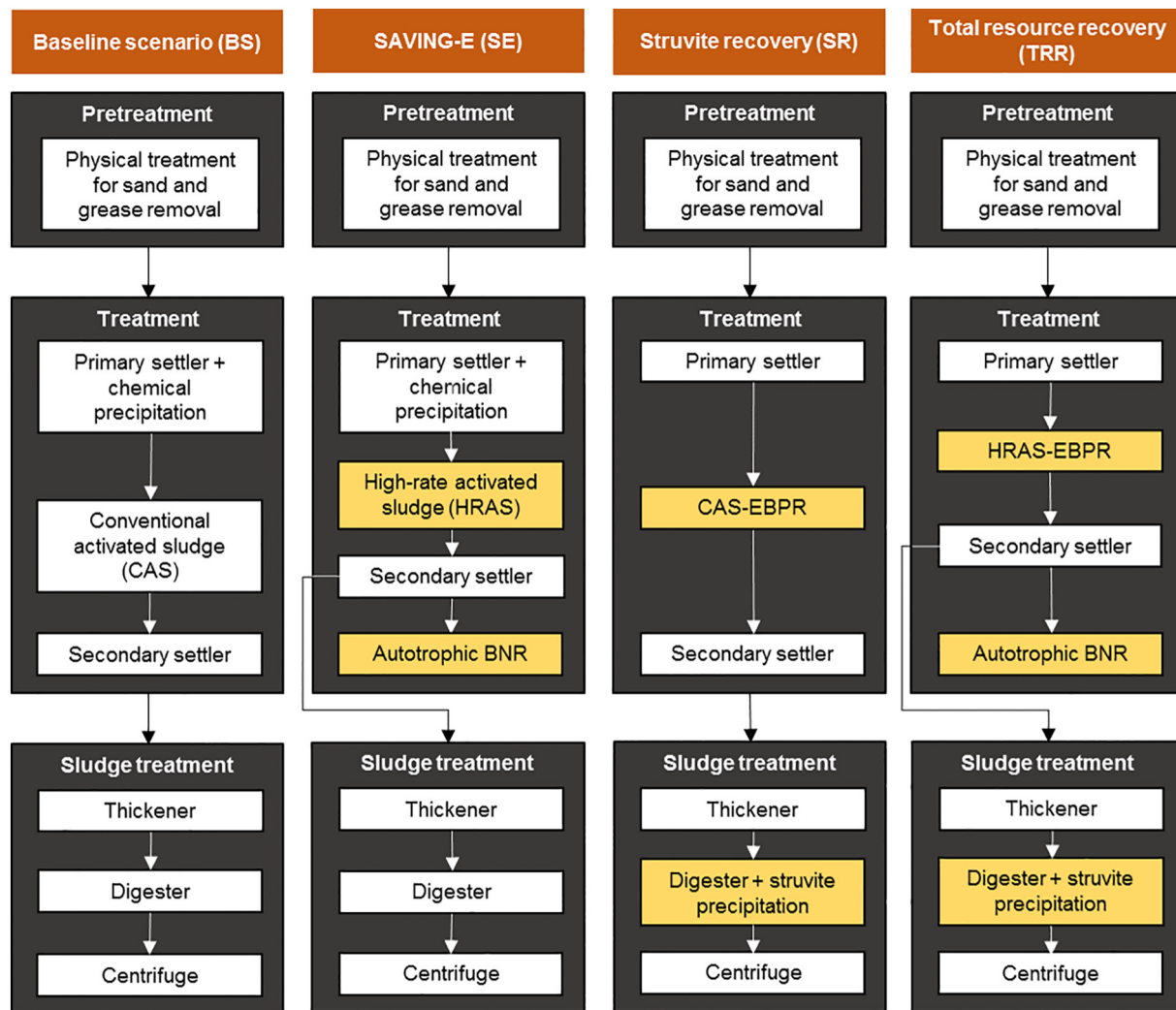


Fig. 1. Original configuration of the wastewater treatment plant (left column) and modifications to the treatment units in the selected scenarios (SE, SR and TRR). Autotrophic BNR: autotrophic biological nitrogen removal; EBPR: enhanced biological phosphorus removal.

that might overshadow the actual operational energy use of the WWTP. Discussing the best sludge treatment processes is under debate (Hospido et al., 2005) and is beyond the scope of this analysis.

2.3.2. Life cycle inventory

Table 2 shows the life cycle inventory for each scenario. Facility managers provided detailed flow charts that helped us to identify the main flows involved in the operation of the baseline scenario, which are electricity, chemicals, and direct pollutant emissions to air and water. Each flow was quantified for the main treatment blocks depicted in Fig. 1, i.e., wastewater pretreatment and treatment and sludge treatment. As

explained in Section 2.3.1, waste management was excluded. The consumption of electricity and chemicals was also available in the WWTP reports and databases provided by facility managers. To address the changes in biogas production, we applied the average electricity and chemical values for the period 2014–2016. In the case of electricity, the WWTP is connected to the low voltage power grid, which we modeled using the Spanish electricity mix (Red Eléctrica de España, 2014). The WWTP's monitoring system already provided data on the amount of cogenerated and purchased electricity, and no assumptions were needed. The chemicals comprise iron chloride and polyelectrolyte, which are utilized in the primary settler and sludge management, respectively. Background inventory

Table 1 Features of the scenarios applied to study technological upgrades in the WWTP.

Modeling scenarios	Organic matter removal	Nitrogen management	Phosphorus management	Biogas features
Baseline scenario (BS)	CAS	Not implemented	Removal: Chemical precipitation	57% of CH ₄ content
SAVING-E (SE)	HRAS	Removal: Autotrophic BNR	Removal: Chemical precipitation	61% production increase with respect to BS scenario 75% of CH ₄ content
Struvite recovery (SR)	CAS-EBPR	Not implemented	Recovery: Struvite precipitation	57% of CH ₄ content, no increment in the biogas production compared to BS scenario
Total resource recovery (TRR)	HRAS-EBPR	Removal: Autotrophic BNR	Recovery: Struvite precipitation	61% production increase with respect to BS scenario 75% of CH ₄ content

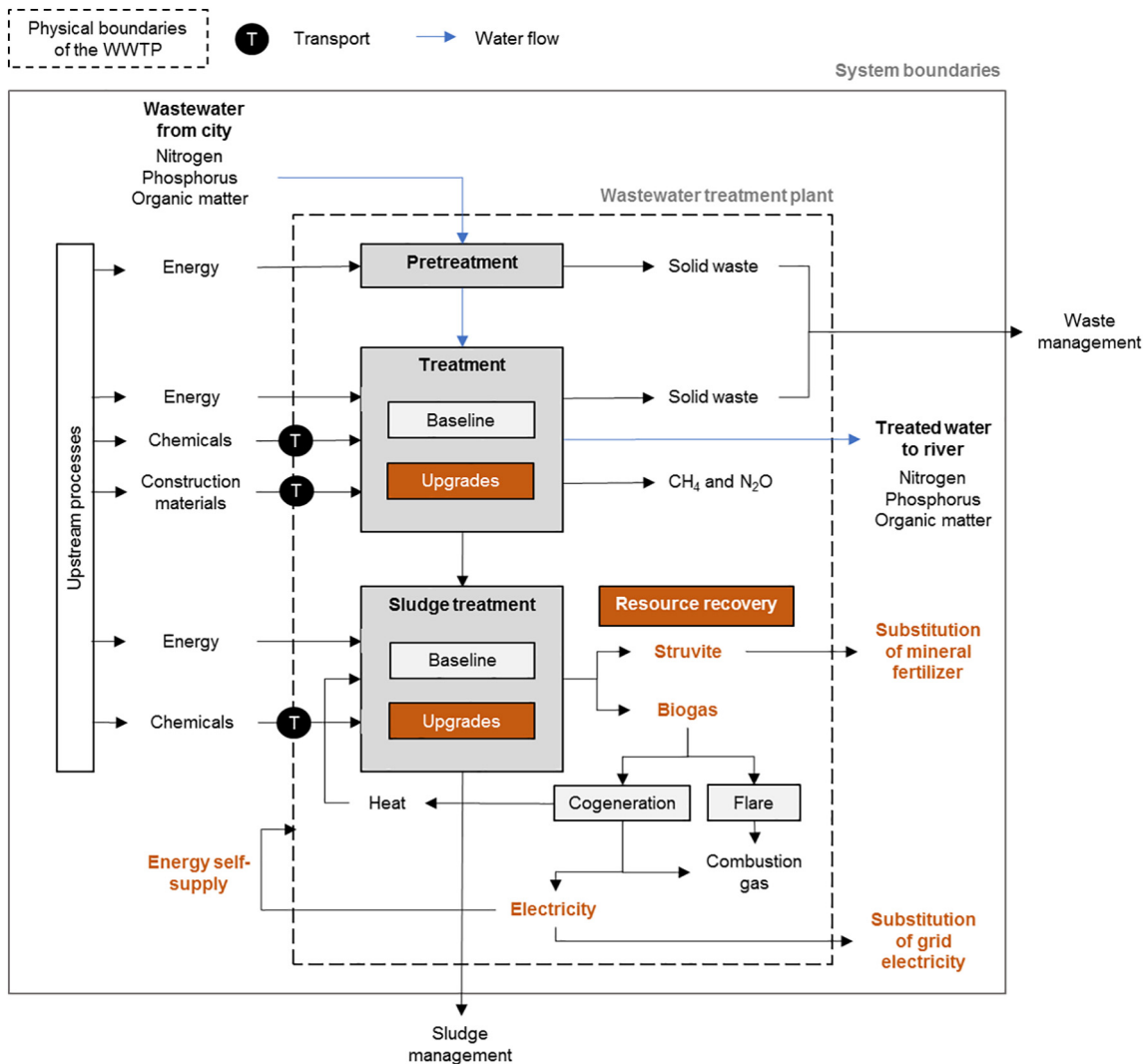


Fig. 2. System boundaries of the WWTP under study, including the flows and processes involved in its current configuration and with the application of technological upgrades.

data were retrieved from Ecoinvent v3 (Wernet et al., 2016) (refer to Table 1 in the Supplementary Material).

The new infrastructure required for the emergent technologies (i.e., autotrophic BNR and EBPR) was included in the assessment (lifespan of 20 years). We assumed that the current treatment capacity would be covered with these additional or substitutive technologies, per the technical assessment of the pilot plant (Section 2.2). We did not account for space limitations in the installation of the new reactors to show the full potential of these upgrades. As a result, several airlift reactors (ALR) for the partial nitrification process (Isanta et al., 2015) and Upflow Anammox Sludge Blanket (UAnSB) for the anammox process (Reino et al., 2018) were designed using steel for the SE scenario, and steel and concrete reactors were considered for the HRAS-EBPR unit. Regarding the operation, the SE scenario demands less energy due to decreased oxygen consumption and a slight increase in polyelectrolyte consumption. These data were obtained from the SAVING-E pilot plant. In terms of P recovery, we accounted for the electricity and chemicals (magnesium chloride and sodium hydroxide) required in the Ostara® process as detailed by (Amann et al., 2018), considering a P recovery efficiency of 25% with respect to the P content in the influent. Iron chloride is thus not needed. We included the transport of chemicals from the actual suppliers to the WWTP.

As resources, both in terms of energy and nutrients, are recovered from WWTPs, the system becomes multifunctional. First, we have a function of treating wastewater that aligns with the functional unit described in

Section 2.3.1. Second, we have a function of energy provision from biogas cogeneration, either to increase the energy self-sufficiency of WWTPs or to provide burden-free energy to the electricity market. Last, we have a function of phosphorus fertilizer provision to the regional agricultural market. To account for the main function of treating wastewater and the benefits of nutrient recovery through struvite and energy production via biogas cogeneration, we conducted a system expansion and considered the substitution of mineral fertilizer, considering stoichiometric ratios and the market for electricity considering direct substitution, and accounted for this product displacement as environmental credits. Diammonium phosphate (DAP) was chosen as the baseline fertilizer, as it is the most prevalent P fertilizer worldwide (IPNI, 2007). To model the effects of electricity cogeneration through biogas, we accounted for the credits to energy production, i.e., avoided consumption of energy from the grid. We made a theoretical assumption in scenarios with enhanced biogas production (SE and TRR). Ideally, increased biogas generation implies that all the gas is consumed to boost energy production. For this reason, we assumed that the capacity of the cogeneration turbines would be adapted, but the additional infrastructure required in these cogeneration units was not modeled due to a lack of data. The substitution process for each scenario is provided in Eq. (1) for the BS and SE scenarios and in Eq. (2) for the SR and TRR scenarios.

$$F_{TW} = F_{TW+EP} - F_{EM} \tag{1}$$

Table 2

Average life cycle inventory data of the WWTP in its current configuration (baseline scenario) and with technological upgrades. Values per 1 m³ of treated wastewater. km for the flows of chemicals refers to the distance between the factory and the WWTP under study. These values were converted to tons per km (tkm) to perform the LCIA. The lifespan of "Construction" flows is 20 years. TSP: total suspended particles.

Flows			Units	Scenarios			
				Baseline (BS)	SAVING-E (SE)	Struvite recovery (SR)	Total resource recovery (TRR)
Inputs	Construction	Steel	kg	–	4.53E-03	2.61E-05	4.55E-03
		Concrete	kg	–	–	1.36E-03	1.36E-03
		Transport	km	–	5.00E+01	5.00E+01	5.00E+01
	Energy	Wastewater pretreatment	kWh	6.00E-02	6.00E-02	6.00E-02	6.00E-02
		Wastewater treatment	kWh	1.80E-01	1.20E-01	1.80E-01	1.20E-01
		Sludge treatment	kWh	4.60E-02	4.60E-02	4.60E-02	4.60E-02
		Struvite recovery	kWh	–	–	9.63E-03	9.63E-03
	Chemicals (wastewater treatment)	FeCl ₃	kg	6.50E-02	6.50E-02	–	–
			km	3.09E+01	3.09E+01	–	–
		Polyelectrolyte	kg	2.25E-03	2.48E-03	2.25E-03	2.48E-03
			km	1.88E+01	1.88E+01	1.88E+01	1.88E+01
	Chemicals (struvite recovery)	MgCl ₂	kg	–	–	1.36E-02	1.36E-02
			km	–	–	1.88E+01	1.88E+01
		Na(OH)	kg	–	–	3.52E-04	3.52E-04
			km	–	–	1.88E+01	1.88E+01
Outputs	Emissions to water	WWTP effluent	kg N	6.30E-02	1.00E-02	6.30E-02	1.00E-02
			kg P	1.00E-03	1.00E-03	3.35E-04	3.35E-04
	Emissions to air	WW degradation	kg CH ₄	5.95E-06	5.95E-06	5.95E-06	5.95E-06
			kg N ₂ O	1.26E-03	1.26E-03	1.26E-03	1.26E-03
		Biogas cogeneration	kg CH ₄	7.93E-08	1.38E-07	7.93E-08	1.38E-07
			kg CO	1.74E-07	3.02E-07	1.74E-07	3.02E-07
			kg N ₂ O	8.92E-10	1.55E-09	8.92E-10	1.55E-09
			kg NO _x	9.42E-10	1.64E-09	9.42E-10	1.64E-09
			kg PM ₁₀	3.92E-10	6.80E-10	3.92E-10	6.80E-10
			kg PM _{2.5}	3.92E-10	6.80E-10	3.92E-10	6.80E-10
		kg TSP	3.92E-10	6.80E-10	3.92E-10	6.80E-10	
	Energy and nutrient recovery	Energy production	kWh	1.75E-01	2.31E-01	1.75E-01	2.31E-01
		Energy self-sufficiency	%	61%	102%	59%	98%
		Struvite recovery	g	–	–	1.33E-02	1.33E-02
		Avoided diammonium phosphate	kg	–	–	7.14E-03	7.14E-03

$$F_{TW} = F_{TW+EP+SP} - F_{EM} - F_{FM} \quad (2)$$

where F is a specific function, TW is treating wastewater, EP is energy provision (from cogeneration), EM is energy from the market, SP is struvite provision and FM is fertilizer from the market (diammonium phosphate). This substitution process was performed in the current attributional LCA to avoid allocation that otherwise had to be made to a service (treating wastewater), a mass-based product (struvite) and an energy-based product (energy from biogas). Additionally, we opted for a simple substitution (without accounting for specific market characteristics, contrary to consequential LCA) over just a system expansion to allow the comparability of our study with published literature (mainly on resource-intensive WWTPs) and future research (potentially focused on converting WWTPs to resource-recovery facilities).

Additionally, we estimated the direct pollutant emissions to air and water, which are associated with three different processes, i.e., wastewater treatment, treated water discharge, and biogas cogeneration. First, wastewater degradation is a known source of significant CH₄ and N₂O emissions at different stages of the treatment process (Corominas et al., 2013; Eijo-Río et al., 2015; Lorenzo-Toja et al., 2016a). CH₄ emissions were calculated based on the emission factors reported by Lorenzo-Toja et al. (2016a), who conducted experimental studies in a similar area. N₂O emissions were calculated based on emission factors reported by Reino et al. (2017) for partial nitrification under mainstream conditions. Second, water quality tests provided information about the amount of N and P discharged into the river, which are the main drivers of eutrophication. Last, the use of biogas for energy cogeneration produces direct air emissions. Biogas cogeneration emissions were calculated using the methods suggested for WWTPs by the Catalan Office for Climate Change (2015). This method uses data from Spanish air emission inventories (Ministry of Agriculture Food and Environment, 2014).

2.3.3. Life cycle impact assessment

We conducted the mandatory classification and characterization stages of the impact assessment using the ReCiPe (H) method (Huijbregts et al., 2016) and SimaPro 9 (PRé Consultants, 2017). As this paper addresses energy self-sufficiency, the results focus on the global warming indicator (in kg CO₂ eq) and cumulative energy demand (in MJ). Global warming is an easily recognizable indicator in the context of decision-making, whereas the cumulative energy demand gives an indication of the energy consumption throughout the life cycle of the WWTP. Moreover, because the changes applied in the wastewater treatment may affect the quality of the effluent, we also included the categories freshwater eutrophication (in kg P eq) and marine eutrophication (in kg N eq). Additionally, due to the toxicity potential associated with steel, we also included the ecotoxicity impact category (in kg 1,4-DB eq), which consists of the product of the sum of terrestrial, freshwater and marine ecotoxicity.

2.4. Impact payback times and eutrophication net environmental impact

Impact payback times (IPTs) indicate the amount of time a module or a technology must produce environmental credits to compensate for the environmental impacts generated to produce the module or technology. For upgrades in WWTPs, the IPT for every impact category is estimated based on Eq. (3), which is an adaptation to wastewater management of existing proposals in renewable energy literature (Alsema and Phylipsen, 1995; Sumper et al., 2011). The investment in natural resources and environmental impacts is depicted in the nominator, where I_p and I_T are the impacts generated by the production and transport, respectively, of the infrastructure required to implement technology i . In our case, i represents the scenarios, i.e., SE, SR and a combination of both (TRR). The denominator includes the avoided impacts (AI) resulting from technology i in terms of electricity from the grid (E) and mineral fertilizer (F). Since many WWTPs already include a biogas cogeneration unit to produce electricity, avoided impacts might

already occur in the baseline scenario. To account for the actual benefits of the incremental or radical changes to resource recovery, the initial avoided impacts (if any) need to be subtracted ($AI_{E,BS}$).

$$IPT_i = \frac{I_{P,i} + I_{T,i}}{AI_{E,i} + AI_{F,i} - AI_{E,BS}} \quad (3)$$

Additionally, we calculated the Eutrophication Net Environmental Impact (ENEI). The ENEI is an equation proposed by Lorenzo-Toja et al. (2016b) adapted from the originally proposed Net Environmental Benefit by Godin et al. (2012). With the goal of complying with legislation removal standards, this indicator accounts for the net improvement in water quality from a removal/recovery technology plus the thresholds imposed by current legislation (Eq. (4)). Current emission limits in Spain are 1 and 10 mg/L of P and N for WWTPs with more than 100,000 p.e. (Royal Decree 509/1996.OE, 1996). To adapt to the ENEI, which is based on the Eutrophication Potential impact category from the CML-IA impact method (Guinée, 2002) in Lorenzo-Toja et al. (2016b), we quantified the Freshwater ENEI (FENEI) and Marine ENEI (MENEI) replacing the Eutrophication Potential from CML-IA by Freshwater Eutrophication and Marine Eutrophication impact categories from the ReCiPe (H) impact method (Huijbregts et al., 2016). In terms of interpretation, values equal to 1 mean that N or P are removed according to legislation. Values greater than 1 mean that the WWTP is performing better than what the legislation imposes, whereas values less than 1 mean that the WWTP is underperforming.

$$ENEI = \frac{EP_{influent} + EP_{process (indirect)} + EP_{effluent(real)}}{EP_{influent} + EP_{effluent (limit by - law)}} \quad (4)$$

3. Results

3.1. Illustrating the potential environmental impacts of technological upgrades

Our impact assessment results highlight the environmental benefits of struvite recovery (SR scenario) and a combination of energy and nutrient recovery technologies (TRR scenario) (Fig. 3; refer to Table 2 in the Supplementary Material for detailed results). In particular, the TRR scenario generates lower environmental impacts than the baseline in all impact categories except for global warming, even without accounting for the avoided impacts. The benefits of the emergent strategies are particularly

apparent in marine eutrophication. Scenarios SE and TRR score six times better than the baseline and SR scenarios due to the removal of N species in the autotrophic BNR process, which is present in both scenarios.

Struvite precipitation offers the possibility of obtaining a valuable resource at a low environmental cost (Ruff-Salís et al., 2020), as the reconfiguration of the WWTP has several associated benefits. First, the lower impacts of SR and TRR compared with BS and SE are related to the EBPR process (either operated as CAS or HRAS), which is capable of removing up to 99% of the influent P under optimal conditions (Izadi et al., 2020; Roots et al., 2020; Zhang et al., 2021). Furthermore, given that the technology recovering struvite precipitates P from the digestate, the quantity of P in the effluent in the SR scenario is equivalent to that in the TRR scenario (ca. 0.34 mg/L), as are the emissions to water in freshwater eutrophication ($3.4 \cdot 10^{-4}$ kg P/m³).

Second, the TRR scenario generates lower impacts on ecotoxicity and cumulative energy demand than the BS scenario due to the avoided use of iron chloride in the HRAS-EBPR. In contrast, the BS and SE scenarios are able to remove P through chemical precipitation, leaving an effluent concentration of ca. 1 mg/L. Third, struvite is a substitute fertilizer with proven feasibility and environmental benefits at different scales (Gell et al., 2011; Latifian et al., 2012; Liu et al., 2011; Ruff-Salís et al., 2020). In our case, the avoided impacts due to fertilizer substitution in the SR and TRR scenarios are approximately 3 times higher than the impacts of struvite recovery on global warming and cumulative energy demand.

Enhanced energy recovery entails increased environmental burdens for certain indicators, although the net balance is positive for other indicators. The construction of the autotrophic BNR infrastructure in the SE and TRR scenarios represents a major impact on ecotoxicity (45%) due to the ecotoxicity potential of steel. Moreover, because of this new infrastructure, the SE scenario generates slightly larger impacts on freshwater eutrophication than the BS scenario. Nonetheless, if the avoided impacts of increasing biogas cogeneration are subtracted from the total impacts (Fig. 4), the final performance of SE is slightly better than BS in this impact category because the avoided impacts obtained through biogas cogeneration are 16% higher in SE and TRR than in BS.

The aggregated credits obtained in the TRR scenario could help mitigate 24%, 67% and 102% of the plant's impacts on global warming, ecotoxicity and cumulative energy demand, respectively; these values are much higher than those quantified for the BS scenario in the same categories (15%, 16% and 37%, respectively). As reported by Lorenzo-Toja et al. (2016a), direct emissions to air comprise the main contributor to global warming in

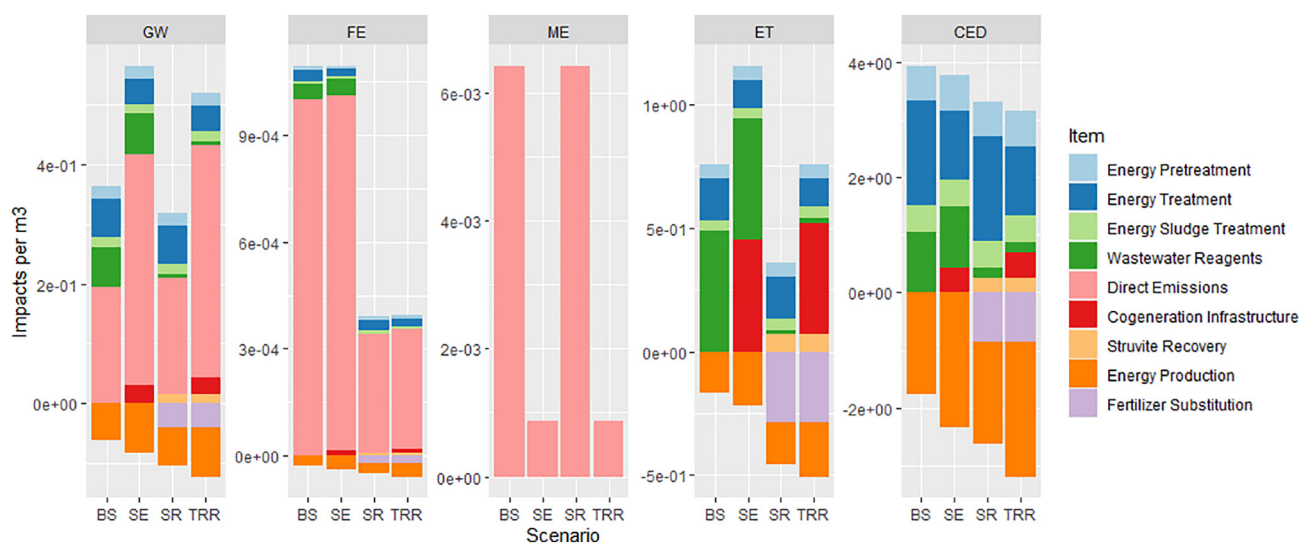


Fig. 3. Impacts of the baseline (BS) and upgrade (SE, SR and TRR) scenarios in the assessed impact categories. Data per m³ of wastewater. The “struvite recovery” component includes the chemicals, energy and infrastructure required to recover struvite. “Cogeneration infrastructure” includes the infrastructure (concrete and steel) required to implement the improvement in the cogeneration process as described in Sections 2.1 and 2.2. GW – global warming (kg CO₂ eq); FE – freshwater eutrophication (kg P eq); ME – marine eutrophication (kg N eq); ET – ecotoxicity (kg 1,4-DB eq); CED – cumulative energy demand (MJ).

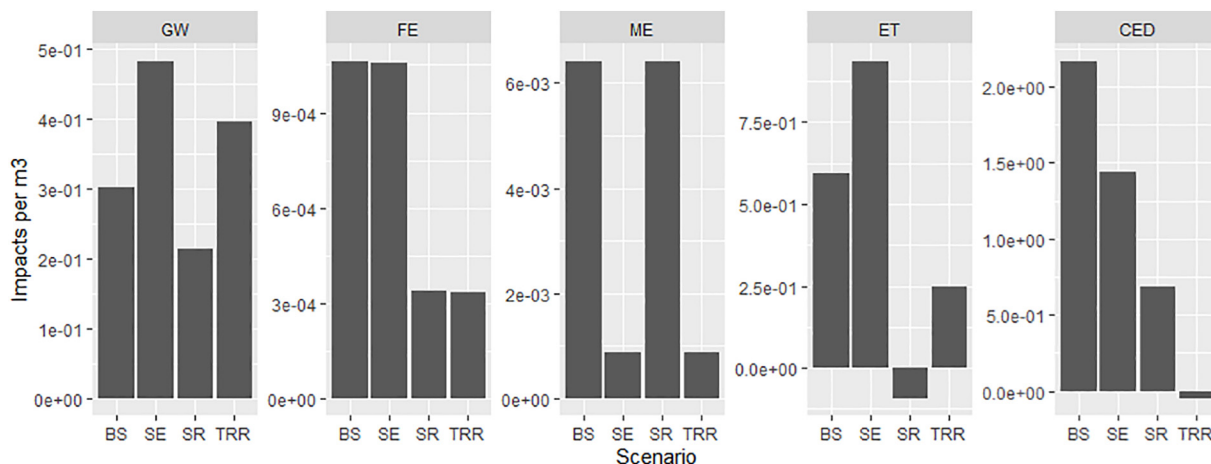


Fig. 4. Net environmental impacts of the BS and upgrade (SE, SR and TRR) scenarios in the assessed impact categories after subtracting the avoided burdens. Data per m³ of wastewater. GW – global warming (kg CO₂ eq); FE – freshwater eutrophication (kg P eq); ME – marine eutrophication (kg N eq); ET – ecotoxicity (kg 1,4-DB eq); CED – cumulative energy demand (MJ).

WWTPs. For this reason, the percent contribution of the credits is lower in the global warming indicator, as the direct emissions to air contribute 0–2–0.4 kg CO₂ eq/m³, representing 48% (BS), 64% (SE), 61% (SR) and 75% (TRR) of the impact in the analyzed scenarios. The greatest impact observed in global warming in the SE and TRR scenarios is attributed to the presence of the BNR process. The nitrogen removal achieved by these technologies is key to complying with nutrient discharge legislation but increases the amount of N₂O emitted, which radically increases the WWTP's impact on global warming due to the high radiative force of this greenhouse gas (Vasilaki et al., 2019).

3.2. Quantifying the infrastructure payback time

The avoided impacts of struvite recovery (SR scenario) or a combination of nutrient and energy recovery (TRR scenario) set all payback times below the lifespan of the new required infrastructure (20 years), except for ecotoxicity (Table 3). However, the benefits of the SE scenario alone would be insufficient for compensating for the environmental investment in the short term. For example, we would need an extra 12% of energy produced by cogeneration to obtain a carbon payback time lower than the infrastructure's lifespan. Enhanced biogas recovery alone represents an ecotoxicity payback time that is 8.5 times higher than the infrastructure's lifespan, mainly due to the production of steel for the reactors. This effect is mitigated in the TRR due to the benefits of fertilizer substitution. On the other hand, SR presents the lowest payback times because the benefits related to fertilizer substitution substantially outperform the additional impacts generated by the required infrastructure. This finding is attributed to not only to the need for less steel infrastructure compared with SE but also the lack of P recovery technologies in the WWTP under study in contrast to

biogas cogeneration, which greatly benefits the payback times of the scenarios that consider fertilizer substitution (SR and TRR).

In terms of freshwater eutrophication payback time, Table 3 shows the expected results as SR and TRR considered that the P recovered in the form of struvite will be used to displace fertilizer from the market. However, we cannot appreciate a distinct benefit of applying BNR processes in SE and TRR in marine eutrophication payback time. Since nitrogen in BNR is removed but not recovered, thus not displacing another product from the market, the benefits of cleaning wastewater cannot be observed with payback time quantification. Based on this limitation, we quantified the FENEI and MENEI (Table 3), which aim to quantify the degree of improvement of a technology aiming to remove nutrients from wastewater considering the thresholds imposed by national legislation (Lorenzo-Toja et al., 2016b).

Since the SE scenario only considers an improvement in terms of nitrogen removal and increased biogas production, its FENEI value is equal to 1.00, which means that the P removal in this technology strictly complies with the legislation. This removal is achieved by the same means employed in the baseline scenario: application of iron chloride to chemically precipitate P. On the other hand, SR and TRR have a FENEI value of 1.12, meaning that these technologies are outperforming compared with the legislation. This behavior is related to the implementation of EBPR, which increases P removal compared with chemical precipitation processes.

The methodological potential of the ENEI is observed when analyzing the MENEI values. For this indicator, the SE and TRR scenarios have a value of 1.17 (Table 3), meaning that they remove nitrogen above the value imposed by the legislation. This increased removal in SE and TRR compared with SR is attributed to the addition of BNR processes.

4. Discussion

4.1. Incremental and radical changes in wastewater management

Shifting from a removal approach to a recovery approach in WWTPs unveils the environmental effects of focusing on individual strategies toward a circular water economy. The baseline scenario already pursued energy recovery through biogas, but it focused on a removal approach when addressing nutrients such as P. Indeed, the WWTP had a 61% self-sufficiency potential and already avoided 37% of the cumulative energy demand through energy self-supply. Upgrading the plant through the implementation of autotrophic BNR in the mainstream allowed for approximately 30% additional self-supply, thus reaching energy self-sufficiency under optimal conditions. Nevertheless, the environmental investment in infrastructure outbalances the benefits when accounting for payback

Table 3
Impact payback times and ENEI of each technological upgrade.

Indicator	Impact category	SE	SR	TRR
Payback time (years)	Global warming (GW)	29.5	7.2	10.9
	Freshwater eutrophication (FE)	27.7	6.2	8.7
	Marine eutrophication (ME)	30.3	7.5	9.2
	Ecotoxicity (ET)	169.4	4.8	26.8
	Cumulative energy demand (CED)	15.0	6.1	7.3
Eutrophication Net Environmental Impact	Freshwater eutrophication (FENEI)	1.00	1.12	1.12
	Marine eutrophication (MENEI)	1.17	1.07	1.17

times. The environmental impacts of producing more energy for self-supply are not paid off within the expected lifetime of the infrastructure (i.e., approximately 20 years). This limitation is related to two specific reasons.

First, the assumed benefits of upgrading biogas production depend on the initial status of the WWTP. This is a recurrent issue when conducting LCAs, as the results are case-specific and highly determined by the baseline scenario (Corominas et al., 2013). In this sense, the implementation of autotrophic BNR in the mainstream might score better when the baseline scenario does not include a cogeneration unit or when planning a new WWTP from scratch. To show an example, the WWTP used as a case study was affected by a national policy on renewable energy self-supply (Ministerio de Industria Energía y Turismo, 2015) and stopped the production of electricity for several months. If we compare the benefits of upgraded biogas production with a scenario without electricity generation, the environmental investment in the implementation of autotrophic BNR in the mainstream is compensated for in a shorter period (7 years). Nevertheless, biogas production is a common practice in moderate- to medium-sized WWTPs (Jenicek et al., 2012), and thus, this scenario only seems realistic in the construction of new WWTPs in small municipalities aiming to increase energy self-sufficiency.

Second, the benefits of increasing the quality of the effluent are not reflected in the payback times if the implemented technology is aimed at removing instead of recovering nutrients. Since the removal of nitrogen does not substitute for any other marketable product (such as biogas cogeneration for primary energy or struvite for chemical fertilizers), credits cannot be given to this improvement, and thus, the implementation of autotrophic BNR increases the impact on global warming and cumulative energy demand. To exploit the potential of implementing this process, we adapted the ENEI proposed by Lorenzo-Toja et al. (2016b), although other authors suggested adding complementary water quality-related functional units (Rodríguez-García et al., 2011).

In this case, a removal approach seems to be more attractive when assessing the benefits of autotrophic BNR, as the SE and TRR scenarios integrate a new N removal strategy. Although this nutrient was managed from a removal standpoint, it succeeded in dramatically reducing the plant's impacts on marine eutrophication because the baseline scenario includes an urban WWTP without N removal. This is not the case in urban WWTPs, where N removal is implemented through classical heterotrophic BNR systems. Previous LCAs confirm the benefits of implementing autotrophic BNR processes to the environmental performance of WWTPs, although tradeoffs may arise in terms of larger air emissions and electricity use (Hauck et al., 2016). This was the case in our study, in which direct N_2O emissions substantially contribute to the final value of global warming. However, most studies have focused on the implementation of autotrophic BNR in the sidestream, i.e., for treating the reject water generated from the dewatering of the digested sludge. Only a few studies have explored the implementation of autotrophic BNR in the mainstream, highlighting their benefits compared with heterotrophic BNR removal (Arias et al., 2020; Cho et al., 2019). However, this paper clearly shows the synergistic effect of modifying existing WWTPs by implementing autotrophic BNR in the mainstream and P recovery as struvite. This study also opens a research avenue to understand whether N recovery is environmentally friendlier than N removal.

From a resource recovery standpoint, our study shows that the incremental benefits of enhanced biogas production might become more attractive when combining this technology with additional technological upgrades. Existing LCA studies point out the environmental benefits of nutrient recovery and reuse, as they avoid the consumption of mineral fertilizer and reduce the WWTP's demand for chemicals (Lam et al., 2020). Indeed, the payback time of combined energy and nutrient recovery is much shorter and within the lifetime of the infrastructure. Additionally, it also scores better than the baseline in most categories. For this reason, combining an incremental change in an existing strategy with a "radical" change in an unexplored flow not

only increases the circularity of the system but also improves its environmental performance. These findings can help elucidate the potential pathways of resource recovery that reduce the environmental impacts of large WWTPs (> 100.000 p.e.) such as the WWTP examined in this analysis.

The selected technologies are meant to be exemplary, and therefore, other configurations are possible, which is a limitation of this analysis. Furthermore, their economic feasibility needs to be assessed in future studies to ensure that strategies are prioritized from a holistic standpoint. A cost-benefit analysis will determine the investment costs, externalities and profit associated with the selected technologies, which is needed to complement the literature on the economic evaluation of wastewater treatment (Lin et al., 2016). Such analyses need to account for the externalities of N removal, which can be estimated through shadow prices (Molinos-Senante et al., 2010), as well as the uncertainties of a slowly developing struvite market.

4.2. Perspectives and prospects: toward a circular water economy in cities

The combination of upgrades can constitute a radical change to wastewater management by taking stock of the circular perspective of water as a service, energy source and nutrient carrier (Brears, 2020). The benefits of this integration span beyond the boundaries of WWTPs, which aligns with the interconnected nature of urban systems and the effects of circular strategies at different scales (Kirchherr et al., 2017; Nika et al., 2020). Upscaling the effects of these incremental and radical changes at the urban or regional level will provide further insights into the environmental effects of resource recovery. Notably, in addition to the energy and nutrient values associated with water flows, the integration of carbon capture technologies into WWTPs could turn the water industry into a global source of negative carbon emissions (Lu et al., 2018).

Given the local value of WWTPs, the interconnections between sanitation systems and the urban environment need to be further explored to understand the extent to which a circular water economy can be beneficial for other urban sectors. These localized innovations might trigger changes at larger scales due to the interconnections among food, energy, water, materials and land (Bleischwitz et al., 2018). The relationships between these small-scale upgrades and the metabolism of a city are worthy of future research, especially given the influence of economic incentives and social awareness on the implementation of circular strategies (Robles et al., 2020). For instance, there seems to be a direct relationship between wastewater and local food production (Mehta et al., 2015; Massey et al., 2007; Ruff-Salís et al., 2020), whereas excess energy could be shared with other industries to generate a symbiosis. In this case, we did not model the effects of distributing struvite as a fertilizer to local farmers and its spreading on agricultural soil, but it could also enhance urban food self-sufficiency and reduce the impacts of local food production (Ruff-Salís et al., 2020). However, "nexus thinking" seems to remain a conceptual approach that does not translate to actual strategies, which means that a transformation into "nexus doing" is necessary to take stock of radical actions and transform them into real implementation projects (Simpson and Jewitt, 2019). In parallel, it is imperative that "nexus doing" in a circular water economy not only aligns with environmental and economic goals but also ensures that "exporting" recovered resources from wastewater to other sectors does not generate environmental injustice toward certain urban sectors and social groups (Cox, 2020).

5. Conclusions

Our analysis shows that incremental changes in biogas production are insufficient for compensating for the environmental investment in infrastructure, although autotrophic nitrogen removal is beneficial for increasing the quality of the effluent. In contrast, combined phosphorus and energy recovery reduces the environmental impacts from the avoided use of fertilizers and phosphorus and nitrogen release into water bodies. An integrated approach to resource management in

WWTPs (i.e., upgrading existing facilities with a set of emergent technologies) is thus desirable and creates new research avenues toward the implementation of circular strategies. The payback times support our recommendations. Investing in incremental energy recovery has a carbon payback time of 30 years, whereas combining energy and nutrient recovery barely exceeds the 10-year mark, which is lower than the lifespan of the infrastructure. We suggest that future wastewater management strategies and respective policies consider these new insights. Our analysis sets the stage to further analyze circular strategies at the urban scale, highlighting potential tradeoffs between traditional waste management and innovative resource recovery strategies. The lessons learned from wastewater infrastructure are an asset for identifying the best possible combinations of strategies in future circular cities to minimize their impacts on the environment.

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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CRedit authorship contribution statement

All authors were responsible for the conception and design of the study. M.E. Suárez-Ojeda and A. Petit-Boix conceived the original idea of the article. E. Moliné provided the necessary data, which were then analyzed by M. Ruff-Salís and A. Petit-Boix with the help of E. Moliné and M.E. Suárez-Ojeda. M. E. Suárez-Ojeda and J. Carrera provided data on HRAS experimental results and calculated biogas features in Table 1. A. Petit-Boix and M. Ruff-Salís took the lead in writing the manuscript. M.E. Suárez-Ojeda, J. Carrera and S. Leopold wrote individual sections of the manuscript, and with J. Rieradevall, X. Gabarrell and G. Villalba, critically revised the draft for important intellectual content. All authors gave their final approval of the manuscript.

Data availability statement

The data that support the findings of this study are available in the supporting information of this article.

Appendix A. Supplementary data

Detailed impact assessment results of the WWTP under analysis with and without technological upgrades. Supplementary data to this article can be found online at doi:10.1016/j.scitotenv.2022.156422

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