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1	A critical review on Life Cycle Assessment and plant-wide models towards emission
2	control strategies for greenhouse gas from wastewater treatment plants
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28 Abstract

For decades, there has been a strong interest in mitigating greenhouse gas (GHG) emissions 29 from wastewater treatment plants (WWTPs). Numerous models were developed to measure 30 the emissions and propose the quantification. Existing studies looked at the relationship 31 between GHG emissions and operational cost (OCI), which is one of the most important 32 indicators for decision-makers. Other parameters that can influence the control strategies 33 include the effluent quality (EQI) and total environmental impacts. Plant-wide models are 34 reliable methods to examine the OCI, EQI and GHG emissions while Life cycle assessment 35 (LCA) works to assess the potential environmental impacts. A combined LCA and plant-wide 36 model proved to be a valuable tool evaluating and comparing strategies for the best 37 performance of WWTPs. For this study involving a WWTP, the benchmark model is used 38 39 while LCA is the decision tool to find the most suitable treatment strategy. LCA adds extra criteria that complement the existing criteria provided by such models. Complementing the 40 cost/performance criteria is proposed for plant-wide models, including environmental 41 evaluation, based on LCA, which provides an overall better assessment of WWTPs. It can 42 capture both the dynamic effects and potential environmental impacts. This study provides an 43 44 overview of the integration between plant-wide models and LCA.

Keywords: Life cycle assessment, greenhouse gas, emission control, wastewater treatment
plant, plant-wide model

Wastewater treatment plants are important systems in the water treatment sector because they 48 ensure the quality of the aquatic environment. However, under various treatment processes, 49 given their usage of chemicals and energy, greenhouse gas are produced and emitted from 50 wastewater treatment plants. The quantity of these emissions is increasing and been reported 51 52 in the Global Atlas of the three major types of greenhouse gas emissions for the period 1970-2012 (Janssens-Maenhout et al., 2017). Therefore, wastewater treatment plants do contribute 53 to global warming. GHG emissions become the key factor when evaluating the overall 54 performance of a WWTP. The inclusion of GHG emissions is an additional criterion when 55 evaluating control strategies in a WWTP, offering a better idea about their overall sustainability 56 (Flores-Alsina et al., 2014). In recent years, various methods of quantification and 57 58 measurement have been proposed in order to increase the available data and literature on GHG emissions. It is also pointed out that our incomplete knowledge of GHG production influences 59 the output results. The direct measuring methods have certain uncertainties and limitations due 60 to the variability of the influent, complexity of treatment process, operational time and different 61 standard evaluation criteria. 62

There is a need for developing new tools to estimate and evaluate GHG emissions from 63 different processes that prevent or mitigate their generation in WWTPs (Flores-Alsina et al., 64 2011). Mathematical models of GHGs are useful tools to assess the quantity of emissions and 65 examine different mitigation approaches. Emissions under various operating conditions and 66 from different units could be estimated with great accuracy by using these models. Control 67 strategies could be developed for these models to include GHG emissions during design, 68 operation and optimisation of WWTPs (Mannina et al., 2016). As the results of using different 69 models in different plant configurations and a variety of influent wastewater characteristics, all 70 treatment systems and other related activities should be evaluated. Flores- Alsina et al. (2011) 71

found influent, effluent and operational variables at each simulation step; hence, the control strategies might influence the conditions of GHG generation and emissions. Therefore, multiple evaluation criteria, which include effluent quality, economic cost and GHG emissions, should be involved in the operational strategy. Plant-wide models were built for the design and testing of control strategies in WWTPs. Existing studies focus on either implementation of the control on these models or adapt the benchmark framework for real-life plants (Barbu et al., 2017).

Evaluating the control strategies for WWTP via models has several uncertainties regarding using different mathematical models, different plant configuration, a variety of influent characteristic, and numerous environmental indicators. Life Cycle Assessment (LCA) was suggested as something that can complete the performance assessment. Of the different methodologies available in estimating the environmental impacts of WWTPs, LCA as an environmental management tool can investigate a product or a service throughout its whole life cycle, from raw material to production, use and disposal (Nausad, 2018).

Use of LCA ensures that all environmental impacts are analysed within the LCA framework and this helps to avoid shifting problems from one place to another. In the wastewater treatment field, LCA was applied in the 1990s and is considered an effective tool to evaluate the environmental effects of WWTPs in both design and operation (Corominas et al., 2013). During the last two decades, LCA has answered several concerns within the wastewater treatment industry. Numerous research has been conducted and reviewed in recent years demonstrating different LCA application methods (Sabeen et al., 2018).

93 Several reviews on LCA studies dealing with WWTPs have been published. Corominas et al.
94 (2013a) conducted a review of 45 articles. According to their paper, previous studies focused
95 on identifying methods and how they communicated the results. All of the reviewed studies

defined their respective goals and scopes. One-third of them provided inventory data. Eightytwo per cent addressed the impact assessment, while thirty-eight per cent did not indicate the
methodology. Only thirty-three per cent provided the assessment procedure and limitations of
the approach and/or determining the parameters that influence the LCA outcome (Corominas,
Foley et al., 2013). The purpose of this paper is to determine the need for guidelines for LCA
studies when applied to WWTPs and the upcoming challenges in the water treatment field.

Friedrich et al. (2007) reviewed the use of LCA in the water treatment sector and its local applications. Their paper reflected the limitations of the LCA tool. Aspects of these limitations related to the tool and framework in general while the other problems emerged from the specific locations. The general shortcomings of LCA are listed as follows: the energy required, raw materials, toxicological impacts which are included without an analysis of the gap. The consumption of water and land, and habitat alterations are mostly not considered. Finally, the environmental impacts are underestimated due to insufficient data (Friedrich et al., 2007).

A review of 22 studies using LCA in wastewater treatment focused on the impact of toxicity, 109 was carried out by Larsen et al. (2007). Their findings hightlighted the importance of different 110 stages in LCA. The "use stage" is the most important process due to energy consumption, the 111 level of GHG emissions, effluent, and sludge production (Larsen et al., 2007). The 112 environmental impacts, which include an energy-related category and chemical-related 113 component, play an important role in an LCA study of wastewater treatment technology. The 114 objective of this research was developing a methodology that could analyse impacts of micro-115 pollutants and pathogens in the life cycle impact assessment. 116

117 Zang et al. (2015) reviewed 53 LCA studies on biological WWTPs and indicated the source of 118 each impact category that may influence the results. In their analysis, the development and 119 application of the model are reviewed to improve accuracy. The authors showed the current 120 use of LCA in WWTPs. According to their study, LCA is normally employed to compare between wastewater treatment systems based on their environmental impacts. Another benefit 121 of LCA is to evaluate the influence of one or more WWTPs to identify the limitations of the 122 process. The third purpose for using LCA is to integrate WWTPs with the whole water use and 123 treatment cycle, which consists of all processes from water supply to wastewater treatment. 124 Finally, LCA can be applied when some impact categories need more focus or further 125 development (Zang et al., 2015). This paper also indicated the influence of GHG on global 126 warming and the surrounding technological systems of the WWTPs affecting the LCA results. 127 128 For example, in the methane recovery process, when converting biogas into energy, the material for energy conversion would contribute a significant amount of GHG emissions. 129

Sabeen et al. (2018) reviewed numerous papers published from 1990 to 2016 that have used LCA to evaluate the domestic wastewater treatment processes. This article reviewed how LCA was applied to cover the objectives, boundaries, functional units (FUs) and life cycle impact assessment (LCIA). According to Sabeen et al., using LCA helps to improve some aspects of municipal wastewater treatment. However, choosing different FUs, system boundaries or LCIA methods affect the total results. Therefore, in future studies more attention should be paid when selecting FU.

The most recent review on the application of LCA to wastewater treatment in developing countries was conducted by Gallego-Schmid et al. (2019). This paper assessed 43 articles and indicated that the sources and technical parameters could influence the quality of LCA when considering GHG emissions. Technical parameters include influent wastewater quality, efficiency in removing pollutants, treatment size, and treatment technologies (Gallego-Schmid et al., 2019).

To the best of the our knowledge, no review has yet investigated the GHG emissions estimation 143 methods when using LCA to evaluate the total environmental impacts. There are numerous 144 strategies applied to quantify GHG emissions. However, the assessment of different control 145 strategies is qualitative, not quantitative (Barbu et al., 2017). To capture both the plant's 146 performance and environmental impact evaluation, including GHG emissions, LCA and plant-147 wide models were combined in some prior studies (Meneses et al., 2015, Arnell et al., 2017). 148 The integration of LCA and other benchmarking tools to develop successful mitigation 149 strategies for GHG emissions from WWTPs has never been reviewed previously. This study 150 aims to provide an overview of the LCA application in evaluating the total environmental 151 impact, including plant-wide models to quantify GHG emissions from WWTPs. The scope of 152 the review includes available journals articles, which consider and provide results for GHG 153 154 emissions. This paper also proposes an advanced integration of plant-wide models within the LCA procedural context. 155

- 156 2. Method and materials
- 157 2.1.Method

This paper is structured as can be seen in Figure 1: first, the application of LCA and plant-wide models are presented to describe the gap of the existing methods; then the reviewed studies are analysed in detail. The LCA reviewed papers are examined follow four phases: goal and scope, inventory, impact assessment, and interpretation in order to investigate the common elements, the distinguishing conditions and the shortage performance. Numbers of articles that conduct plant-wide models to measure GHG emissions were inspected to explore the pros and cons. The opportunities for future reseach are based on limitations of different applied methods.



166 *Figure 1. Framework for the evaluation process proposed in this study*

167 2.2. Materials

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168 2.2.1. LCA

LCA completes the whole picture as much as is possible, and all the environmental impacts are taken into account (Guinée, 2002). Use of LCA ensures that all environmental impacts are analysed within the LCA framework. There are three types of LCA models, namely the process-based LCA, input-output LCA, and hybrid LCA (Chen et al., 2012). The first type focuses on energy and materials flow in a manufacturing process, while input-output LCA emphasises the environmental data (Awad et al., 2019). The application of LCA to WWTP is illustrated in Figure 2.



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177 Figure 2. The advantages of using LCA in WWTPs

Using LCA has several benefits as listed below. LCA can determine what technique and management tools provide the best environmental performance (Niero et al., 2014). LCA aims to determine the environmental impact indicators by analysing the emissions from all relevant processes (Yoshida et al., 2014). LCA can identify improvement alternatives for a single plant and to compare different technologies. LCA could be applied to evaluate different types of WWTPs, to compare the environmental impacts (Delre et al., 2019), and to select the best treatment unit process operating scenario (Tangsubkul et al., 2006).

Despite the advantages, LCA has some limitations as follow. Firstly, the results and influence 185 indicators do vary among papers. The reason for this may due to different assessment methods, 186 simulated inventory data, and integration of different models to quantify the environmental 187 impacts (Zang, Li et al., 2015). Secondly, although the environmental impacts of WWTPs are 188 assessed in detail by using LCA, the economic variables are excluded from LCA because they 189 might affect the control strategies. Thirdly and finally, the data availability and data quantity 190 191 are limited in the life cycle inventory. In some cases, the researcher used secondary data to model effluent emissions (Niero, Pizzol et al., 2014). In other cases, the impact categories were 192 site-dependent (Corominas et al., 2013). 193

Existing studies have limited scope, either in terms of alternative processes, size of facility or exclusion of significant aspects of the WWTP system (Foley et al., 2010). A limited number of studies have examined the relative environmental impacts of different treatment standards. As research has developed over the years, the objective of LCA research has changed from protecting human health, minimising the consumption of finite resources, reducing the amount of energy required, and reusing pollutants so that they can help in the nutrients recycling process.

201 2.2.2. Plant-wide models for GHG emissions calculation

WWTPs include many different processes and these comprise biological, transports and 202 hydraulic phenomena. These factors make it difficult to propose control and operation 203 alternatives. For a better understanding of GHG emissions that originate from wastewater 204 205 treatment plants, models are used as effective and low-cost tools to examine the new technologies and control strategies in GHG management. The application of models to estimate 206 and mitigate GHG estimation has been demonstrated for many years (Bani Shahabadi et al., 207 2009). Furthermore, improving measurement techniques to reduce uncertainty related to GHG 208 emissions, means that models can describe the GHG production from each process in the 209 WWTPs (Corominas et al., 2012). Therefore, the modelling of GHG emissions from WWTPs 210 was proposed in many publications to provide an accurate estimate of how much GHG was 211 being emitted from wastewater treatment plants (Barbu, Vilanova et al., 2017). 212

According to previous research, these models can be divided into three main types. The first group, which has a high level of uncertainty and variability, consists of the empirical models based on data for emission factors at treatment units (Pagilla et al., 2009). The second group includes simple comprehensive process-based models at treatment units (Corominas, Flores-Alsina et al., 2012). The third group consists of dynamic mechanistic models at treatment units or plant scale (Mannina, Ekama et al., 2016). For a quick evaluation of GHG emissions, the
second group is more popular than the third one (Mannina et al., 2019). The process models
combine with instrumentation, control and automation (ICA) to create the benchmarking tool
for assessment.

The benchmark is a simulation environment consisting of a plant layout, a simulation model, 222 influent loads, treatment procedure and a set of evaluation criteria. Benchmark models are 223 effective tools for the design and testing of the control strategies of WWTPs, and their function 224 is to overcome the difficulties in engineering techniques (Barbu, Vilanova et al., 2017). The 225 first version, named Benchmark Simulation Model no. 1 (BSM1), was proposed in 2002 to 226 develop efficient control strategies for WWTPs (Copp, 2001) and then followed by the BSM2 227 in 2007. Both the BSM1 and BSM2 include simulation for all treatment units, influent loads, 228 229 test procedures and evaluation criteria. However, the BSM1 does not allow for evaluation of the interaction between processes, only local strategies can be evaluated. The BSM2 is 230 available for different simulation platforms so it can easily to compare the results of different 231 control strategies of different platforms (Henze et al., 2000). The BSM2 consists of existing 232 models that can describe processes in the WWTP. It includes all the units within the WWTP, 233 234 and makes it possible to fully evaluate the plant's performance. This model consists of the biological reactions, liquid-gas interactions and GHG production as well. BSM2 calculates the 235 GHG through the following stages: biotreatment, sludge treatment, sludge reuse, chemical 236 237 usage, power consumption and biogas usage. Many studies use benchmarks on applying control strategies by simulation or on building control frameworks in real plants (Zhou et al., 238 2011, Santín et al., 2015, Santín et al., 2017). The limitation of BSM2 is that the reduction of 239 240 nitrate to nitrogen is considered as a one-step process that leads to N₂O production, which 241 cannot be accurately determined (Sweetapple et al., 2013).

An extended version of the BSM2, BSM2G, was proposed later on. BMS2G includes GHG emissions within the model and considers all the units in which the emissions may occur (Flores-Alsina, Corominas et al., 2011, Corominas, Flores-Alsina et al., 2012). This model allows the dynamic evaluation of the GHG emissions in the biological treatment units. BSM2G was employed in some case studies to investigate the influence of some control actions and operational strategies on GHG emissions (Flores-Alsina, Corominas et al., 2011, Sweetapple et al., 2015).

The diffusive emissions estimation model (DEEM) was developed to focus on CO₂ and N₂O 249 emissions originating from biological processes (Rodriguez-Garcia et al., 2012). The biological 250 model was divided into four main categories: oxidation of organic matter, nitrification, 251 denitrification and hydrolysis. DEEM takes into account the CO₂ emissions associated with 252 endogenous decay and microbial growth of autotrophs. N₂O can be captured in the nitrification 253 and denitrification processes where the possibility of AOB reducing nitrite and the possibility 254 of N₂O increasing due to NO inhibition are considered. DEEM can be applied only to the water 255 line, but nonetheless it presents the benefit of simplicity and is suitable for LCA. 256

A new plant-wide model was developed by Mannina et al. (2019), which can quantify both direct and indirect GHG emissions from the biological and physical processes of a WWTP. This model considers both the contribution of the water line and the sludge line. The model is based on COD, TSS mass balance. The novel features of the model include the following massbalance-based model regarding nitrogen; a two-step nitrification process; and the ability to quantify N₂O generation both in dissolved and gas forms (Mannina, Rebouças et al., 2019).

Numerous of existing models were developed but none of them consists of multi-criteria
evaluation combining GHG with effluent quality and operational cost. Moreover, each models

has limitations in measuring GHG emissions due to complex condition. Therefore, more effortshould be paid on improving the accuracy of quantifying GHG emissions.

267 3. The current use of LCA applications to WWTP

While numerous LCA studies investigate the total environmental impacts and potential 268 outcomes from a whole wastewater treatment system cycle, few examined the relevant 269 environmental influences, especially with reference to GHG emissions from wastewater 270 treatment plants. When considering GHG emissions from WWTPs, the majority of LCA 271 studies concentrate on: firstly, carbon dioxide emissions originating from energy consumption; 272 and secondly, methane emissions from sludge treatment. To the best of our knowledge, only 273 24 studies quantify GHG emissions when using LCA to evaluate the WWTPs performance as 274 can be seen in Table 1. The papers which quantified the volume of GHG emissions from 275 276 WWTPs while doing life cycle assessment were selected for review in this study.

277 Table 1. Articles included in the review and main characteristics

CUC	Mathad	Caal	Decoard
GHG	Method	Goal	Process
CO_2	Estimation	Evaluate LCA	All treatment
		framework and	processes,
			1
		alternative	energy
		process options	consumption
		process options	consumption
CO_{2} CH, NoO	ASM1	Assess the	Enorgy
$CO_2, CI14, IN_2O$	ASIMI	Assess ule	Ellergy
		environmental	consumption
			1 1 1
		outcomes in a	and sludge
		WWTP under	treatment
		three scenarios	
	GHG CO ₂ CO ₂ , CH ₄ , N ₂ O	GHGMethodCO2EstimationCO2, CH4, N2OASM1	GHGMethodGoalCO2EstimationEvaluateLCAframeworkandalternativeprocess optionsCO2, CH4, N2OASM1AssesstheenvironmentaloutcomesinaWWTPunderthree scenarios

(Houillon et al.,	CO ₂ , CH ₄ , N ₂ O	Literature	The	Sludge
2005)		review	contribution of	treatment
			energy and	process
			emissions to	
			global warming	
(Hospido et al.,	CH ₄ , N ₂ O	Literature	Evaluate the	Sludge
2007)		review	most common	treatment
			technical	
			options to	
			remove organic	
			matter	
(Foley, de Haas	CO ₂ , CH ₄ , N ₂ O	BioWin	Using LCA	All treatment
et al., 2010)		simulator	framework to	processes
			analyse	
			alternative	
			process options	
(Wu et al., 2010)	CO ₂ , CH ₄	Not specificed	Assess a WWTP	All treatment
			based on energy	processes
			and material	
			flows	
(Pan et al.,	CO ₂ , CH ₄ , N ₂ O	IPCC	Estimate GHG	All treatment
2011)			from a vertical	processes,
			subsurface flow	energy
			constructed	consumption
			wetland; and	

					compare with a	
					group of five	
					WWTPs by	
					using LCA	
(Rodrigu	lez-		N ₂ O	IPCC	Assess the	Sludge
Garcia	et	al.,			environmental	treatment
2011)					performance of	
					24 WWTPs	
					using LCA with	
					Eutrophication	
					Potential, GWP	
					and operation	
					costs	
(Godin	et	al.,	CH ₄ , N ₂ O	IPCC	Propose a new	All treatment
2012)					methodology to	processes
					perform LCA on	
					WWTPs	
(Wang	et	al.,	CO ₂ , CH ₄ , N ₂ O	Literature	Employ LCA to	Biological
2012)				review	construct and	treatment
					evaluate 6 AAO	process and
					wastewater	energy
					treatment	consumption
					systems to meet	
					standards	

(Wang	et	al.,	CO_2	BioWin	To choose the	Biological
2012)			CH4	simulator	wastewater	treatment
			N ₂ O		treatment	process and
					processes that	energy
					mitigate the	consumption
					environmental	
					impacts and	
					promote	
					bioenergy and	
					nutrient	
					recovery	
(Corom	inas,	,	CO ₂ , CH ₄ , N ₂ O	Neptune	Using LCA to	Biological
Larsen	et	al.,		Simulation	evaluate the	treatment
2013)				Benchmark	environmental	process and
				(NSB)	impacts of	energy
					enhancing	consumption
					strategies	
					applied to	
					wastewater	
					nutrient removal	
(Zhu	et	al.,	CO ₂ , CH ₄ , N ₂ O	Literature	Present a	Biological
2013)				review	methodology to	treatment
					evaluate the	process and
					environmental	energy
					impacts of	consumption

						perfor	manc	e		
(Cao	et	al.,	CO ₂ equivalent	Literature		LCA	was	used	Energ	SY.
2013)				review		to eva	aluate	e and	consu	mption
						compa	are	the	and	sludge
						energ	у	and	treatm	nent
						GHG	emis	sions		
						implic	cation	s of		
						two	se	wage		
						sludge	e-to-			
						energ	y syst	ems		
(Rodrig	guez-		CO ₂ , CH ₄ , N ₂ O	Literature		The	pot	ential	All	treatment
Garcia	et	al.,		review		enviro	onmei	ntal	proce	sses
2014)						impac	ts o	of 3		
						differe	ent	side-		
						strean	1			
						treatm	nent			
						techno	ologie	es		
						were	ass	essed		
						by LC	ĊA			
(Yoshi	da,		CO ₂ , CH ₄ , N ₂ O	IPCC,	direct	The ir	nfluer	nce of	All	treatment
Clavre	ul et	al.,		measurem	ent	data	inve	ntory	proce	sses
2014)				and literat	ure	to		LCA		
						outco	mes			

process

(Lorenzo-Toja	CH ₄ , N ₂ O	Direct	The role of	All treatment
et al., 2016)		measurement	direct GHG	processes
			emissions in the	
			GWP of two	
			WWTPs	
(Piao et al.,	CH ₄ , N ₂ O	Assumed to be	Evaluate several	All treatment
2016)		based on	WWTP based	processes,
		literature and	on LCA and	energy
		IPCC	economic	consumption
			efficiency	
			analysis	
(Lu et al., 2017)	CO ₂ , CH ₄ , N ₂ O	Not specified	Complement the	Biological
			economic	treatment
			criteria and	process
			environmental	
			implications	
			with suggested	
			management	
			policies	
(Casas Ledón et	CO ₂ , CH ₄ , N ₂ O	Not specified	LCA was used	All treatment
al., 2017)			to assess GHG	processes,
			emissions,	energy
			environmental	consumption
			remediation	
			costs and the	

		specific	
		environmental	
		remediation	
		costs	
(Garfi et al., CO ₂ , CH ₄ , N ₂ O	Literature	Three	All treatment
2017)	review	wastewater	processes
		treatment	
		systems were	
		assessed to	
		compare the	
		total	
		environmental	
		impacts	
(Polruang et al., CO ₂ , CH ₄ , N ₂ O	IPCC	The	All treatment
2018)		environmental	processes,
		impacts of seven	energy
		WWTPs were	consumption
		investigated	
(Awad, Gar N ₂ O, CH ₄ , CO ₂	Literature	LCA was	Biological
Alalm et al.,	review	applied to four	treatment
2019)		scenarios to	Sludge
		study the	treatment
		environmental	
		impacts of a	
		WWTP	

(Delre,		ten	CO ₂ , CH ₄ , N ₂ O	IPCC	LCA was u	sed	The	whole
Hoeve	et	al.,			to calculate	the	plants	
2019)					site-specific			
					carbon			
					footprints	for		
					WWTPs			

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The level of coverage is illustrated in four main steps for the LCA in Figure 3 and then discussed in the following sections.



281

282 Figure 3. Level of LCA coverage in 25 reviewed papers

283 3.1. Goal and scope definition

Goal and scope definition is the first step to understand the main purpose of the assessment. It is necessary to describe in detail the function of the systems and to ensure they operate exactly as determined in the event where different WWTPs are being compared. The functional unit (FU) should be clarified. During this step, reference flow is verified to measure product components and materials. All data used in the LCA must be calculated or scaled by the reference flow. Futhermore, all the reviewed papers have identified the FU and described the system boundary.

The FU provides a reference from which inputs and outputs of the process can be standardised. 291 The potential impacts of a WWTP can be calculated and referred to the FU. When defining the 292 FU of a WWTP, different choices were selected. The most common FU is the quantity of 293 treated water at a certain time (m³d⁻¹), named FU1; it is based on the realistic data as used in 294 86% of reviewed articles. Some studies chose FU based on treated wastewater associated with 295 a population equivalent (PE) to minimise the difference between the influent composition and 296 flow. In addition to this, FU considers the removal of nutrients and organic matter and this is 297 known as FU2 (kg PO4³⁻ eq) (Zhu, Liu et al., 2013). FU1 has the ability to indicate the 298 differences between facilities with reference to influent characteristics, while FU2 focuses on 299 300 the differences between environmental and economic costs of mitigating the potential eutrophication of the effluent (Rodriguez-Garcia, Molinos-Senante et al., 2011). When 301 302 investigating the pollution removal capacities of the WWTPs, Delre et al. (2019) used the FUs 303 in term of 1 kg of carbon removed, 1 kg of total nitrogen removed, and 1 kg of phosphorus removed. The reason for choosing these FUs was to separate the pollution removal abilities 304 according to the relevant pollutant. The life-span of the plants has improved from fifteen years 305 306 (Emmerson, Morse et al., 1995) to twenty years (Foley, de Haas et al., 2010) and now their maximum life-span is fifty years (Piao, Kim et al., 2016). 307

308 System boundary definition plays an important role in describing which processes will be included or excluded from the system, so the system boundary should be consistent with the 309 objectives of the study. Choosing the system boundary is one of the challenges when 310 implementing LCA. Various system boundaries lead to different results because choosing the 311 system boundary defines which flows of information, energy or material transfer from one 312 system to another (Pan et al., 2018). With reference to GHG emissions, only 9% of the papers 313 could be defined as the "Cradle to Grave" approach, and 22% represented the "Cradle to Gate" 314 research. Most papers focus on the "Gate to Gate" analysis when only the operational phases 315 316 are being considered. Among the reviewed articles, half of these considered all treatment processes in the water line and sludge line contexts. Three of these focused on sludge treatment 317 only. It was concluded that the construction phase contributed to a negligible impact compared 318 319 with the operational phase. The effectiveness of these boundary influences on the LCI is discussed in the following section. 320

321 3.2. LCI – inventory analysis

LCI is a list of all the material and energy inputs and outputs. This step includes the collection 322 and definition of inputs and outputs of a system throughout its life cycle. LCI is defined as the 323 procedure of data collection and data calculation. For example, inputs comprise raw materials, 324 energy, products or semi-finished products, which are outputs from other processes. 325 Conversely, outputs are emissions, products, and semi-finished products, energy, which are 326 emitted to the environment or used in another process. The input and output data are based on 327 the boundary selection in step 1. The construction and demolition processes are normally 328 excluded from WWTP studies because the operation stage contributes approximately 80% of 329 environmental impacts (Polruang, Sirivithayapakorn et al., 2018). Yoshida et al. (2014) 330 conducted a research study to determine the influence of the inventory data on the outcomes of 331 an LCA study. 332

One of the most critical issues is establishing reliable inventory data (Yoshida, Clavreul et al., 334 2014). Data collection is time- and labor-consuming because establishing the on-site data 335 monitoring process is expensive or even impossible in some cases. Due to the lack of data, 336 some processes were excluded from the studies and this meant that the output from the whole 337 cycle and its impact on the environment was underestimated. The input data was based on the 338 WWTP design standards, environmental reports, plant operator communications or on-site 339 measurements. The output data was obtained from on-site measurements, modelling or 340 simulations. Of the 20 papers that investigated on the treatment processes only, 9 of them 341 covered all the treatment stages whilst the number of papers reviewed biological and sludge 342 treatment were 6 and 5, respectively. Regarding GHG emissions, 68% of the articles quantified 343 344 total GHG emissions in terms of carbon dioxide, methane and nitrous oxide. The remaining papers covered only one or two gases in their research. 345

346 - Data quality

The number of reviewed papers indicating the method to quantify GHG emissions based on 347 the literature review and IPCC guidelines, accounted for 63.6%. Using IPCC has some 348 limitations, such as the exclusion of direct carbon dioxide emissions, uncertainties with default 349 emissions factors, and the accuracy of default data. There are two types of emission factors 350 used to report the quality and quantity of the data. Most of the emission factors are based on 351 the IPCC guidelines or have been calculated on-site. To ensure the quality and reduce the 352 uncertainties, the data need to be updated and less than three years old. Approximately 20% of 353 354 the studies excluded the evaluation method, which was similar to the proportion of articles assessing GHG emissions through the use of models. Only one study was conducted with an 355 on-site measurement technique (Lorenzo-Toja, Alfonsín et al., 2016). The uncertainties of this 356

357 method might relate to technical problems, the number of sampling points, the position of 358 sampling points, and the operator's experience.

359 3.3. LCIA

The impact assessment step aims to study the potential effects on human health, the availability of resources, and the natural environment. This step makes the results of an LCA easier to interpret. The energy use and emissions generated are classified and characterised into impact categories and impact potentials, including global warming, acidification and eutrophication (UNEP, 2003, ISO, 2006). Because of the limitation of the boundaries, the reviewed papers mostly considered the environmental impacts associated with the operation phase, which included primary, secondary, tertiary treatments, and effluent discharge.

Total GHG emissions from WWTP, including methane, carbon dioxide and nitrous oxide, are the sum of direct and indirect emissions. The direct emissions are GHG emitted from each treatment processes within the system boundary while the indirect emissions are related to energy consumption. For the treatment processes, the energy inputs are strongly correlated to the potential outcomes such as global warming, and contributing up to 70% of several impact categories (Sabeen, Noor et al., 2018). However, half of the reviewed papers excluded energy utilisation.

374 3.4. Improvement analysis and interpretation

In the interpretation phase, LCA users aim to identify the most important aspects of the inventory analysis and the impact assessment. Furthermore, they evaluate the study's outcomes, do a completeness check, undertake a sensitivity analysis and uncertainty analysis, check for consistency, make conclusions, and recommendations (UNEP, 2003, VanDuinen et al., 2009). The reason for interpreting data are to: firstly, determine the contribution of each component to each environmental outcome; and secondly, evaluate the level of confidence in 381 the results. Sensitivity analysis was used to determine the effect of each environmental indicator when the inventory data varied $\pm 10\%$. The uncertainty analysis confirms the absence 382 of any outcome caused by sensitive factors on the findings. The recommendations of the 383 384 proposed strategies were made in term of the objectives of this research. These included several improvements that would mitigate GHG emissions from the WWTPs. The result was that the 385 power produced from the total GHG emissions exceeded 59% (Zhu, Liu et al., 2013). There is 386 an opportunity to reduce life cycle impacts relating to energy utilisation (Emmerson, Morse et 387 al., 1995, Wu, Meng et al., 2010). The largest proportion of the energy consumption was used 388 389 in the aeration unit, which accounted for a maximum of 58.8% (Wang, Liu et al., 2012). In contrast low aeration reduces the amount of carbon dioxide emissions but increases the 390 emissions of nitrous oxide (Nguyen et al., 2019). It is necessary to maintain a suitable level of 391 392 aeration to balance operational efficiency and energy requirements. However, these reviewed 393 studies were limited in the outcomes of the weighting step (Corominas, Larsen et al., 2013). Most of the LCA studies involved the overall assessment of large wastewater treatment 394

systems. However, regarding GHG emissions, the performance of each treatment unit may lead
to different outcomes. Therefore, what is essential here is to propose the best system for the
WWTP in term of how each treatment unit operates.

398 4. Developing strategies based on results of plant-wide models

GHG emissions can be influenced by influent and effluent characteristics and operational conditions (Flores-Alsina, Corominas et al., 2011); it is vital to understand and predict their power generation procedure. Flores-Alsina et al. (2011) demonstrated that GHGs could be quantified during the evaluation of control strategies. Models are useful tools to quantify GHG production and their emissions as well as to evaluate the systems' performance before implementing them in real-life plants, thus the models serve as an effective solution for control development. Flores-Alsina et al. (2011) suggested using empirical equations and mechanistic 406 models to estimate GHG emissions during the evaluation of WWTP control strategies. The differences between strategies were the quantity of GHG emitted from secondary emission and 407 power consumption sources. This kind of study provides a better picture of overall WWTP 408 409 performance with a new dimension dealing with emissions. Using simulation in the evaluation process can offer better guidance on the sustainability of different treatment options. Various 410 studies were conducted on the Benchmark Simulation Models (BSM) to explore the influences 411 412 of some control strategies on several indicators, specifically with reference to their economic and operational aspects. 413

Some early research demonstrated the quantification of GHG emissions during the evaluation 414 of the control strategies conducted on BSM2 (Corominas et al., 2010) and BSM2G (Flores-415 Alsina, Corominas et al., 2011). The authors analysed the influence of some operational 416 417 parameters and identified the strategies to control the emissions. The purpose of these studies were to: (1) consider the change in the influent characteristic and operating conditions; and (2) 418 quantify multi-criteria including effluent quality (EQI), operational costs (OCI), and GHG 419 emissions procedure when comparing the strategies. The results showed that the plant under 420 control could reduce GHG emissions by up to 9.6% (Flores-Alsina, Corominas et al., 2011) 421 422 and 12% (Corominas, Flores-Alsina et al., 2010). However, when considering other indicators, 423 the scenario with the lowest GHG emissions had the worst EQI due to limited biodegradable 424 organic carbon preventing denitrification. The scenario with the best EQI produced the highest 425 GHGs and proved to be expensive because of the increase in aeration energy.

In fairy recent research, Flores-Alsina et al. (2014) emphasised the importance of multi-criteria evaluation, including GHG emissions when analysing the WWTPs control strategies. The plant under study was simulated by BSM2G. The results showed that the DO set point has a major influence on the plant's total GHG emissions, the effluent quality and operating cost. Low DO set points resulted in less CO₂ being generated and lower operational costs due to less energy 431 consumption. Also, incomplete nitrification increased the quantity of N₂O emissions, and subsequently the overall GHG emissions increased. This trend was also found in research 432 conducted by Corominas et al. (2010). However, there was a big difference between studies 433 434 that examined the high DO set point. According to Corominas et al. (2010), at a high DO set point, high energy consumption and incomplete denitrification leads to more emissions of CO₂ 435 and N₂O, respectively. Meanwhile, Guo et al. (2012) indicated that a different DO distribution 436 437 resulted in different N₂O emissions even at the same energy consumption level. Hence, the influence of DO on GHG emissions should be investigated further. 438

BSM-e, a modified version of BSM2, was developed to assess control strategies with multiple 439 objectives. The research found a large range of options for mitigating GHG emissions without 440 increasing the operational cost, and also maintaining an acceptable effluent quality (Sweetapple 441 442 et al., 2014). It is pointed out that to simplify the comparison between strategies, using a single index to present effluent quality is more effective than a focus on specific pollutants. BSM-e 443 includes the modelling of dynamic GHG emissions, but it is unsuitable for achieving multiple 444 objectives due to the high simulation time and the many simulations required (Sweetapple, Fu 445 et al., 2014). 446

When considering a multi-criteria evaluation, including EQI, OCI and GHG emissions, Barbu 447 et al. (2017) presented a method to enhance the control strategies by adding more control 448 actions. Twelve control strategies were examined from which to choose the best option for 449 these three elements. The best one is the one that controls DO in the fourth tank, nitrate 450 concentration in the last anoxic tank, and ammonium concentration, which refers to the total 451 suspended solids in the last aerobic tank. Santin et al. (2017) researched similar objectives 452 using the same method. Their results showed that the solution for mitigating GHG emissions 453 is to control the dissolved nitrous oxide concentration, ammonium, and ammonia nitrogen 454 concentrations (Santín, Barbu et al., 2017). 455

456 Another application of model is to describe the GHG generation mechanism and investigate the effects of operational conditions on the emissions (Boiocchi et al., 2017). Different dynamic 457 simulations were conducted to evaluate the ability to reduce N₂O emissions, which were the 458 459 smallest in quantity but with the highest global warming potential of all GHG emissions from the WWTP. It is indicated that controlling N₂O emissions will benefit the reduction of total 460 GHG emissions. The model used was the BSM for Nitrous oxide (BSM2N). However, this 461 application could only satisfy one specific aim of N₂O emissions mitigation (Boiocchi et al., 462 2017). 463

The plant-wide models make it possible to evaluate the performance of the systems before 464 implementing them in real-life scenarios, thus using the benchmark models are an effective 465 solution for control development. Moreover, plant-wide models can explore the GHG 466 production mechanisms, and the influences of operational conditions. Thus, it is beneficial to 467 use plant-wide models to quantify GHG emissions before controlling them. However, the 468 increase or decrease in consuming resources and recovery leads to different environmental 469 outcomes that are not captured by the simulation model; it is only the LCA that can do this 470 (Arnell, Rahmberg et al., 2017). 471

472 5. Integration of LCA and plant-wide models

LCA and plant-wide models have been considered helpful tools to evaluate the environmental impacts and operational condition, respectively. However, when using LCA, the performance evaluation was limited to nitrogen and phosphorus removal. For an optimal overall assessment, models are suggested for use in the LCA research (Corominas, Larsen et al., 2013). It is essential to combine LCA with other indicators so that evaluation is more reliable and accurate; this combination can fill the gap in our knowledge concerning what happens between process control and environmental performance (Flores-Alsina et al., 2010). From Section 3, it can be 480 seen that LCA was applied to assess the impact of some control strategies demonstrated by 481 simulation models. The models were used to measure the quantity of resource inputs and 482 outputs, including GHG emissions, while LCA captured the environmental impacts caused by 483 an increase or decrease inputs and options for recovery. In these situations, the outcomes of 484 LCA could be used for deciding which strategy to use. Once again, plant-wide models are good 485 options to assess the WWTPs' performance.

The integrated LCA and plant-wide models were applied in some studies. Flores-Alsina et al. 486 (2010) first suggested adding the environmental assessment carried out by LCA with economic, 487 technical and legal criteria. The main purpose of their research was to investigate the character 488 and impact of twelve controllers. The evaluation process followed the method of LCA research. 489 Most of the inventory data were collected from the results of the dynamic simulation BSM2. 490 491 Some other data was adapted from the literature and relevant databases. GHG emissions from treatment processes were not considered in the study, and instead methane and nitrous oxide 492 emissions from sludge were tested for their application to agriculture. 493

Combination of LCA and another plant-wide model were conducted in a study of Corominas 494 et al. (2013b). They set out to compare the environmental impacts of different controllers. The 495 496 WWTP layout was simulated with Neptune Simulation Benchmark (NSB) while the direct GHG emissions were estimated using ASM3 Bio-P. The advantage of this research includes 497 the weights, which reflect the relationship of different categories and the results. The main 498 findings were (1) implementing controls for reducing energy consumption is beneficial but 499 does not lead to ideal environmental performance; and (2) nutrient enrichment is the most 500 important factor as it strongly influences selecting the best operational strategies. There are 501 some limitations in the research, such as only the water line was modelled and uncertainty in 502 the N₂O emissions factors was envident. According to the research, significant increasing in 503 the total CO₂ equivalent emissions might result in no change in the final LCA outcomes. 504

505 Multiple evaluation criteria for WWTP performance and as environmentally defined by BSM2 jointly with LCA criteria were applied in the study by Meneses et al. (2015). The main objective 506 of their analysis was to compare the environmental profile of the four control strategies 507 508 implemented by BSM2 from the environmental impact perspective. The impact categories included acidification potential, global warming, eutrophication, photochemical oxidation, 509 depletion of abiotic resources, ozone layer depletion, and terrestrial ecotoxicity. GHG 510 emissions from the biological treatment process were excluded from the study due to the 511 limitations of BSM2. The outcomes of the LCA showed that information on the environmental 512 513 impacts for each category was evaluated. Since the number of assessed parameters was large, it was difficult to determine the differences and compare them. However, by understanding the 514 relationship of each assessment to obtain the best results, it helps the decision-maker to choose 515 516 suitable strategy.

According to Arnell et al. (2017), older studies are limited in choosing their boundaries and 517 analysing the operational costs. The dynamic character of GHG production is based on static 518 emission factors that could underestimate the outcomes. The study indicated that it is possible 519 to combine a simulation model and LCA to explore the dynamic effects, operational cost and 520 521 global environmental impact, including GHG emissions (Arnell, Rahmberg et al., 2017). The goal of their study was to evaluate the change in environmental impacts from the different 522 523 strategies where the same effluent quality was assumed. Findings showed that adding 524 chemically enhanced primary treatment reduced the volume of total direct GHG emissions due to the significant reduction of N₂O emitted from the activated sludge unit. The limitation of 525 this research is that the weighting step was not undertaken, but nonetheless it pointed out that 526 527 combining the simulation model and LCA can both describe the processes, and evaluate the environmental impact (Arnell, Rahmberg et al., 2017). 528

529

6. Discussion and future research

530 6.1. Discussion on combined LCA and plant-wide models

LCA and plant-wide models are widely used in assessing different WWTPs' treatment 531 strategies. However, when considering GHG emissions from WWTPs, each type still has some 532 disadvantages due to the complex treatment processes and environmental conditions. For 533 studies conducted with LCA only, the overall environmental impacts were analysed but 534 evaluation was lacking for operation cost, technical issues and effluent standards. The 535 optimisation of treatment technologies dealing with nitrogen and phosphorus removal has 536 never been evaluated through LCA due to technical difficulty of assessing multiple strategies 537 at full-scale WWTPs. It can be seen from Section 3 that there are few in number of research 538 considers GHG as the main indicator for assessing the WWTPs' performance. 539

Furthermore, CO₂ from the biological process is usually neglected while N₂O, CH₄ emissions 540 541 are normally collected from the sludge treatment process only. Lack of this kind of information leads to insufficient data for building the control framework, and consequently affects the 542 performance of the WWTPs. The quantification of GHG emissions from BNR systems is 543 uncertain. The usual way to estimate GHG emissions is based on IPCC guidelines. Proposed 544 strategies by LCA give the options to reduce the quantity of GHG emissions, but the input data 545 is mostly based on the literature while the uncertainty range correlates with the number of 546 input data. Hence, it is vital to create a complete and reliable database to improve the overall 547 quality of LCA. 548

549 The following research questions were developed by reviewing these LCA studies:

- 550 (1) Which processes of wastewater treatment (inside/ outside the WWTPs) are assessed?
- 551 (2) Which type of effects are considered?
- 552 (3) Which specific LCA methods are applied?
- 553 (4) How are GHG emissions quantified?

554 Exiting studies with plant-wide models have their limitations due to the outcomes of the control strategies selected to give the best effluent quality which may increase GHG emissions. 555 Benchmark simulations are used for effluent quality indicator and operational cost indicators 556 while a potential environmental assessment is not considered. According to the study by Barbu 557 et al. (2017b), when evaluating multiple criteria, each controller achieves goods results in one 558 of the indicators, but it has a different impact on the others. Moreover, sometimes increasing 559 or reducing the input/output may not lead to any change in total environmental impact, which 560 only is captured by LCA (Arnell, Rahmberg et al., 2017). 561

The combination of plant-wide models and LCA results represents an innovation in the environmental evaluation of WWTPs' operations (Meneses, Concepción et al., 2015). Simulating the performance criteria on plant-wide models complements the environmental evaluation based on LCA and offers a better overall assessment of WWTPs as can be seen in Table 2. Barbu et al. (2017b) emphasised the importance of analysing the LCA contribution in order to get a broader view of the environmental consequences. With LCA, it is possible to add extra criteria that complement the evaluation criteria provided by plant-wide models.

569 Table 2. Variables in each analysis methods

Method(s)	Input	Output	Local/	Boundary
			regional/ global	
			impacts	
LCA	Electrical	Abiotic depletion	Include	Construction
	energy	Eutrophication		Operation
	Resource	Global warming		Maintenance
	consumption for	Acidification		Demolition
	water treatment	Human toxicity		

	Resource			
	consumption for			
	transport			
Plant-wide	Electricity	GHG	Exclude	Operation
models	Chemical	Effluent		
	Fuel oil	Operating cost		
	Flow rate			
	Influent			
	characteristics			
LCA + plant-	Electrical	GHG	Include	Construction
wide models	energy	Effluent		Operation
	Resource	Operating cost		Maintenance
	consumption for	Environmental		Demolition
	water treatment	impacts		
	Resource			
	consumption for			
	transport			
	Flow rate			
	Influent			
	characteristics			

570

571 Judging by the results of existing studies, it is evident that multi-criteria evaluation is possible 572 but complex. The simulation models make it possible to describe the process in detail while 573 LCA helps to evaluate the overall outcomes, including the side effects and reflexion effects. 574 Plant-wide models capture both direct and indirect GHG emissions from electricity and chemical consumption. When comparing the strategies, numerous indicators were involved, and their influences with other assessments at different levels were explored by conducting sensitivity analysis in LCAI. The combination of LCA and plant-wide models provide information on the environmental impacts for each impact category (Meneses, Concepción et al., 2015). It helps the policy or operations decision-makers choose the best strategy. LCA when combined with plant-wide models gives more realistic information than steady state assessments (Bisinella de Faria et al., 2015).

582 When evaluating the control strategies, plant-wide models look at the working scenarios while

583 LCA focus on the total environmental impacts. The simulation platform consists of a plant

584 layout, sub-models for water line and sludge line, a model for controllers, and influent profile

585 (Arnell, Rahmberg et al., 2017). The simulation model is conducted for the on-site processes

586 by providing a list of material, energy and other resouces; then the semi-outputs include solid

587 discharge, GHG emissions and effluent are measured. That information will be used as input

588 to calculate the potential effects on human health, the availability of resouces, and the natural

589 environment in LCAI as presented in Figure 4. Base on these steps, the influence of different

590 input elements will present variety in environmental impacts.

591



Figure 4. Frame work of the combination between LCA and plant-wide models.



Existing research has encountered some uncertainties as follows. The simulation models could not assess the full dynamics of N₂O production and emissions based on real data (Mannina, Rebouças et al., 2019). The trajectory and relationships between impact categories were studied in reviewed articles. However, it is difficult to compare values between studies due to differences in choosing boundaries, functional units, impact categories, and specific scopes. The best control strategy is the one that can balance EQI, OCI, GHG emissions, and results in environmental impacts on all categories.

612 6.2.Future research

Due to the limitations of this combination, there are two main things that need to be considered. 613 (1) Weighting step helps to evaluate different impact categories, which is vital for decision-614 making. The role of weighting is to explore the level of potential impacts in different impact 615 616 categories. Existing articles studied the relationship between EQI, OCI, GHG emissions and total environmental impacts. However, using a general weighting factor equal for all impact 617 categories makes the weighting process more complex and difficult to balance. There is a need 618 to determine the suitable weighting factor for sensitivity analysis when combining LCA and 619 plant-wide models. (2) The current challenge in mitigating GHG is the lack of information 620 about N₂O generation and emissions from treatment processes. Although the volume of N₂O 621 produced in WWTP is smaller than CO₂ and CH₄, its GWP is 9.5 times higher than methane 622 and 265 times higher than carbon dioxide (IPCC, 2014). A comprehensive model that is able 623 to include all the mechanism related to N₂O formation is critical for understanding the nature 624 of GHG emissions and how to manage them better. 625

626 7. Conclusion

627 WWTPs are complex operational processes that consist of transportation, treatment, and628 resources consumption. Regarding the overall performance of a WWTP, four aims need to be

629 considered and balanced when proposing the control strategy: (1) mitigate the pollutant 630 discharge including GHG emissions; (2) ensure the quality of effluent ; (3) maintain costs for 631 the lifetime of a WWTP; and (4) minimise the global environmental impact. Evaluating 632 strategies is a difficult task regarding the number of parameters and variables that can affect 633 the results at different levels. This paper has presented a comprehensive overview of the 634 integrated plant-wide models and LCA. The use of models coupled with LCA to develop the

- analytical framework provides a more complete evaluation and more detailed description than
- a single method could. When these two methods are combined, WWTPs can achieve very
- 637 efficient performance with minimal environmental impacts.
- 638

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