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- 1 Beyond Energy Balance: Environmental Trade-offs of Organics Capture and Low
- 2 Carbon-to-Nitrogen Ratio Sewage Treatment Systems
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- 9 Abstract: Several life-cycle assessments (LCAs) have evaluated the environmental impacts (EIs) of different
- wastewater treatment (WWT) configurations, attempting resource recovery and energy efficiency. However, a
- plant-wide LCA considering up-concentration primary treatment and low carbon-to-nitrogen (C/N) ratio sewage
- 12 at the secondary biological treatment (SBT) has not yet been conducted. This study identifies the environmental
- 13 trade-offs and hotspots for the chemically enhanced primary treatment (CEPT) and low C/N ratio SBT emerging
- 14 processes compared to conventional WWT. The life-cycle inventories were calculated using a stoichiometric
- 15 life-cycle inventory framework that couples stoichiometry and kinetics to obtain site-specific water, air, and soil
- emissions. The midpoint results of LCA show that CEPT with anaerobic digestion (AD) for sludge treatment
- 17 achieves energy self-sufficiency, but increases marine eutrophication (MEu) by one order of magnitude
- 18 compared to conventional WWT. A mainstream anaerobic fluidized-bed bioreactor and a partial nitritation-
- anammox fluidized-bed membrane bioreactor which can reduce all environmental impacts by 17%–47%,
- 20 including MEu, are proposed as the SBT of the low-carbon CEPT settled sewage. Integrating the standardized
- 21 S-LCI framework resulted in a site-specific LCA that aids decision-makers on choosing between higher
- 22 reductions in most EIs at the expense of high MEu or less but consistent reductions in all EI categories.

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- 27 **Keywords:** chemically enhanced primary treatment, partial nitritation, anammox, energy self-sufficiency, life-
- 28 cycle assessment

Abbreviatio ns/Notations	Definition	Abbreviatio ns/Notations	Definition
AD	Anaerobic digestion	LCIA	Life-cycle impact assessment
AFBR	Anaerobic fluidized bioreactors	MBRs	Membrane bioreactors
Anammox	Anaerobic ammonium oxidation	MCM	Million cubic meters
AnFMBR	Anaerobic fluidized-bed membrane bioreactors	N/D	Nitrification/denitrification
AnMBRs	Anaerobic membrane bioreactors	NEB	Net energy balance
		PN	Partial nitritation
BNR	Biological nitrogen removal	PN/A	Partial nitritation/anammox
C/N	Carbon-to-nitrogen	PMF	Particulate matter formation
СЕРТ	Chemically enhanced primary treatment	RoW	Rest of the World
СНР	Combined heat and power	SAF-MBR	Staged anaerobic fluidized-bed membrane bioreactor
COD	Chemical oxygen demand	SEM	Standard error of mean
DFE	Dual fuel engines	SM	Supporting Methods
FMBR	Fluidized-bed membrane bioreactor	STWs	Sewage treatment works
FU	Functional unit	S-LCI	Stoichiometric life-cycle inventory
GAC	Granulated activated carbon	S1	Scenario 1
GHG	Greenhouse gases	S2	Scenario 2
GWP	Global warming potential	S3	Scenario 3
HRAS	High-rate activated sludge	US	United States
IAMs	Impact assessment methodologies	WRRFs	Water resource recovery facilities
LCA	Life-cycle assessment	WWT	Wastewater treatment
LCI	Life-cycle inventory	WWTPs	Wastewater treatment plants

1. INTRODUCTION

Climate change, eutrophication, and acidification of water bodies are three of the most
pressing environmental impacts to be tackled by the United Nations' sustainable development
goals. Stricter regulations for water quality discharge from wastewater treatment plants
(WWTPs)while reducing greenhouse gases (GHG) emissions demand the implementation of
more sophisticated treatment that include energy efficiency optimization, use of reclaimed
water and nutrient recovery. ^{2–4}
Energy efficiency optimization refers to enhanced energy recovery and reduced energy
consumption, which might lead to energy self-sufficient WWTPs. ^{2,5} In theory, energy self-
sufficiency is possible given that the chemical energy potential of raw sewage 6 surpasses the
unit electricity consumption of municipal WWTPs in the United States (US) and in Europe. ²
In practice, there are several energy self-sufficient WWTPs that can be claimed as water
resource recovery facilities (WRRFs) (e.g. two WRRFs in Austria, five WRRFs in the US,
and one WRRF in Switzerland). These WRRFs achieved energy self-sufficiency through a
combination of anaerobic digestion (AD) of sludge and other process units. AD is a widely
applied biological process in which the organic components of sludge are converted into
biogas containing mainly carbon dioxide (CO ₂) and methane (CH ₄). ⁸ AD for energy
generation has been identified as one of the key processes to achieving energy self-
sufficiency. ^{3,7,9,10}
In the primary treatment of a conventional WWTP, a fraction of the organics present in
sewage is concentrated as primary sludge for AD, whereas the remaining organics in the
settled sewage assist in the biological nitrogen removal (BNR) process. In recent years, up-
concentration alternatives have emerged for primary treatment such as chemically enhanced
primary treatment (CEPT), high-rate activated sludge (HRAS), anaerobic processes, and
other advanced treatments. ^{4,5,11} Several benefits have been identified in the up-concentration

55 of organics, such as enhanced sludge, higher biogas, and energy yield, and reduction of CO₂equivalent emissions from power importation.^{3,12,13} Despite the benefits, there are still cost-56 57 benefit questions on whether the organic matter should be preserved to assist in 58 denitrification of BNR or, alternatively, should be up-concentrated in primary treatment to obtain higher sludge production for high methane yield in AD.¹⁴ 59 60 CEPT and HRAS are the most common up-concentration practices at full-scale. However, 61 there has been increasing interest in the development of mainstream anaerobic treatment processes, 15 which have been adopted at full-scale in tropical regions. 16 Submerged anaerobic 62 63 membrane bioreactors (AnMBRs) have several benefits, such as a small footprint, less sludge 64 yield, adequate control of biomass due to decoupling of solids retention time and hydraulic retention time, and production of high-quality effluents. 15,17,18 Recent studies have suggested 65 that AnMBRs are a promising technology even under low temperature, 9,17,19 but the dissolved 66 67 CH₄ capture in the effluent, electricity consumption, membrane fouling mitigation, the membrane material, its complex operation, the application of stricter pre-treatment 68 requirements, and minimum removal of soluble nutrients should be addressed before full-69 scale implementation. 4,11,15,18 70 71 Owing to low nutrient removal, up-concentration processes applied as primary treatment 72 require further BNR. Secondary treatment should consider the low carbon-to-nitrogen (C/N) ratio, which restricts the application of conventional activated sludge because of the lack of 73 organic matter as an electron donor for denitrifiers. 8 Considering that municipal sewage 74 typically has medium-to-low chemical oxygen demand (COD) content and that up-75 76 concentration further decreases the carbon content in the settled sewage, research attention is shifting to the study of emerging biological pathways for low C/N ratios, ^{4,9,19,20} such as 11, 7, 77 78 and 3 for simultaneous nitrification/denitrification (N/D), shortcut nitritation/denitritation and

79 combined partial nitritation (PN) and anaerobic ammonium oxidation (anammox) (PN/A), 80 respectively.5,21 81 PN/A works under the lowest C/N ratio, consumes the least oxygen (thus electricity consumption) and carbon, and reduces sludge production and CO₂ emissions.^{8,22,23} In PN, the 82 83 two-step nitrification process is interrupted by inhibiting denitrifiers after the first step, where ammonium (NH₄⁺) is only oxidized to nitrite (NO₂⁻).²⁴ Anammox is the process where NH₄⁺ 84 and NO₂⁻ are simultaneously converted into dinitrogen gas. Combined PN/A can be achieved 85 through suspended and attached growth systems, and single-stage or two-stage reactors. ^{21,23,25} 86 87 In practice, the majority of full-scale PN/A systems operating worldwide work as single-stage suspended growth systems for sidestream treatment.²⁵ Furthermore, Strass WWTP in Austria 88 and Changi water reclamation plant in Singapore carry out mainstream PN/A. 5,23,26 However, 89 90 given that out-selection of nitrite-oxidizing bacteria and heterotrophic denitrifiers is more challenging in the mainstream, 11 the slow growth rate of anammox bacteria, and the stringent 91 92 operational conditions, ²⁷ system configurations that minimize anammox-biomass washout, such as membrane bioreactors (MBRs), have gained attention. ^{28–30} 93 94 Since energy self-sufficiency is unlikely achieved through a single technology, combined 95 configurations for primary and secondary treatment, including these emerging technologies, 96 have been evaluated in terms of net energy balance (NEB). Net energy production was 97 predicted for a staged anaerobic fluidized-bed membrane bioreactor (SAF-MBR) with granulated activated carbon (GAC) as the fouling mitigation strategy. 31-33 In addition, the use 98 99 of PN/A in anaerobic fluidized-bed membrane bioreactors (AnFMBR) has been proposed, 100 which would lead to lower nitrous oxide (N₂O) formation and increased polishing of residual carbon and nitrogen.^{28,34} Mainstream PN/A combined with AD,⁹ a combined submerged 101 AnMBR with a completely autotrophic nitrogen removal over nitrite MBR, ¹¹ mainstream 102 anaerobic fluidized-bed bioreactor (AFBR) followed by PN/A,⁴ and up-concentration 103

followed by mainstream anaerobic and PN/A treatment²⁰ showed higher energy recovery than 104 105 other emerging technologies. 106 Even though the results of NEB greatly contribute to the evaluation of the total environmental 107 impacts of WWTPs and WRRFs, additional information on air and soil emissions, materials 108 and fuel inputs, and waste, should be included to assess the environmental performance of a process.^{2,9} Life-cycle assessment (LCA) is a management technique that enables the 109 evaluation and comparison of the environmental impact of different systems by considering a 110 cradle-to-grave approach.³⁵ It consists of four steps: i) goal and scope, (ii) life-cycle inventory 111 112 (LCI), (iii) life-cycle impact assessment (LCIA), and (iv) interpretation. LCA has been 113 widely used for environmental performance comparison in wastewater treatment (WWT), 114 including plant-wide LCA analyses of emerging technologies. ^{36,37} LCAs for mainstream AnMBR using gas sparging, ¹⁹ up-concentration processes combined with sidestream PN/A 115 for urine source-separation, ³⁸ mainstream PN/A, ³⁹ and sidestream two-step anammox-single 116 117 reactor for high-activity ammonia removal over nitrite system⁴⁰ have already been conducted. However, a plant-wide LCA analysis for low-strength wastewater in a subtropical climate 118 119 considering different primary treatments and a mainstream AFBR followed by a PN/A 120 fluidized-bed membrane bioreactor (FMBR) with GAC as the membrane fouling mitigation 121 technique, has not yet been investigated. Existing LCAs are an excellent step toward identifying the WWT configurations with the 122 123 best environmental performance. However, data collection and processing for LCI differ in 124 each LCA, making comparison and decision-making more challenging. A standardized 125 stoichiometric life-cycle inventory (S-LCI) framework combines biochemistry and process engineering to obtain steady-state and site-specific air, water, and soil emissions for a 126 WWT. 41 Studies integrating the S-LCI framework are expected to enhance standardization 127 128 and specificity and improve decision-making.

The goal of this paper is to identify the environmental trade-offs of CEPT and emerging BNR processes for low C/N ratio sewage in comparison with conventional WWT systems. Three scenarios are analyzed through LCA. Scenario 1 (S1) represents a conventional system with activated sludge. Scenario 2 (S2) consists of CEPT is the only process to obtain settled sewage. Scenario 3 (S3) proposes further BNR after CEPT using a SAF-MBR. The LCIs of S1 and S3 have been presented elsewhere using the S-LCI framework.⁴¹ However, the S-LCI results for S1 and S3 need further analysis through impact assessment methodologies (IAMs) and interpretation methods to obtain a clearer trend of their environmental impacts. This paper has a three-fold objective: (i) to compare the environmental trade-offs of CEPT to those of conventional primary treatment, (ii) to determine environmental hotspots in different plant-wide configurations for low-strength wastewater in a subtropical city, and (iii) to demonstrate the integration of the S-LCI framework into LCA. This study adds to our understanding of the environmental impacts of low-carbon WWT, whose characteristics are likely to become more frequent because of diluted sewage from high-intensity precipitation as a consequence of climate change.^{42–44}

2. MATERIALS AND METHODS

LCA consists of four main steps that are integrated into the software SimaPro v.8.2.3. The first step is setting the goal and scope of the LCA, which includes the definition of a functional unit (FU) and the different boundaries (Figure 1; data flow 1). Although researchers agree that the selected FU should cover the different influent qualities and different removal rates, most still use the "treatment of 1 m³ of sewage to be treated". ^{36,37,45,46} The S-LCI adopts this unit to facilitate further comparison between studies. The Ecoinvent database, the most widely used database for WWT, also uses this FU. ⁴⁷

The second step is constructing the LCI based on the background and foreground
information. The background information can be obtained from databases. In this study, the
foreground information follows the S-LCI methodology for enhanced standardization and
specificity. ⁴¹ The S-LCI consists of three main steps: data collection, calculations, and results.
In the S-LCI, the results of the elemental analysis of the samples (Figure 1; data flow 2) help
in constructing the specific empirical formulas for microbial cells. Then, the empirical
formulas are used for the stoichiometric and kinetic calculations following the
Thermodynamic Electron Equivalents Model (TEEM). ⁸ The TEEM is complemented by
primary data (i.e., flows, concentrations, and removal efficiencies) and energy requirements
of the municipal WWTP or WRRF of interest to construct the process inventories with inputs
and outputs of the system (Figure 1; data flow 3).
The third step of the LCA is evaluating the environmental impacts of the resulting LCI
through midpoint or endpoint methodologies (Figure 1; data flow 4). It has been found that
the use of more than one characterization model validates the robustness of the results and
provides concise information for decision-making. 46,48 In this study, Recipe Midpoint is the
methodology used for midpoint analysis, while Recipe Endpoint and Ecological Scarcity
2013 are the methodologies used for endpoint analyses.
Recipe is the successor of the methods Eco-Indicator 99and CML-IA. ⁴⁹ Recipe has been the
preferred method for its midpoint and endpoint results in recent years. 50-55 In addition, Recipe
is a methodology that includes conceptual sub-compartments to allocate the receiving body
for emissions into air, water, and soil. ⁴⁷ Even though the midpoint results have lower
uncertainty, endpoint indicators are perceived as more relevant by decision-makers. ^{37,47} The
normalization factors in Recipe are based on updated emission and extraction data acquired
from 28 European countries. ⁵⁶ The population of those 28 countries are used to recalculate
the normalization figures per citizen in SimaPro. ⁴⁷

Given that the principal function of WWTPs and WRRFs is related to water emissions,
Ecological Scarcity 2013 is chosen because its integration of water pollution is a key factor
for its single score. Ecological Scarcity 2013 uses "eco-factors" whose normalization
involves the quantification of the current pressure/load from pollutant emissions or resource
consumption in Switzerland per year, and weighting consists of the current substance
emissions in the reference area per year divided by the environmental law limits or political
targets. 47,57
The assumptions for the value choices include the selection of the attributional model, the
allocation approach for multifunctional processes, and the hierarchist perspective. ⁵⁸ The
allocation procedure is by default the "Allocation at the point of substitution" integrated into
Ecoinvent v.3.2. ⁵⁹ The hierarchist perspective is selected because it was built in accordance
with policy principles to model environmental mechanisms through time. ⁴⁹
The results of the LCIA (Figure 1; data flow 5) are used for interpretation, which is the last
step of the LCA. Interpretation of the results includes contribution analysis, uncertainties
management, limitations, recommendations, and conclusions. Contribution analysis indicates
the percentage to which main processes contribute to the total environmental impact of each
category. Contribution analysis can be calculated from SimaPro8 to identify the main
contributors and hotspots of each scenario. Uncertainties management focuses on conducting
sensitivity analysis and Monte Carlo simulations

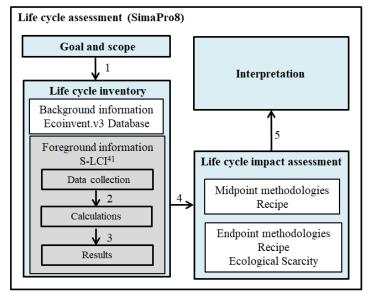


Figure 1. Stoichiometric life-cycle inventory (S-LCI) integration to life-cycle assessment (LCA). The blue boxes represent the steps of the LCA. The white boxes show further descriptions for the life-cycle inventory and life-cycle impact assessment. The gray boxes refer to the S-LCI framework.

2.1 Treatment of low C/N ratio sewage in subtropical climate: Hong Kong

Hong Kong is a subtropical city that treats ~2.8 million cubic meters (MCM) of municipal sewage per day. 60 Treated seawater for toilet flushing is the common practice in Hong Kong, thus 80% of municipal sewage contains high salinity. The majority of the saline sewage is treated at Stonecutters Island sewage treatment works (STWs) which is the largest STWs treating ~1.65 MCM of municipal sewage per day. Shatin STWs is the second major STWs in Hong Kong treating ~0.24 MCM of municipal sewage per day.

2.2 Goal and scope

The LCA goal was to compare the plant-wide environmental trade-offs of treatments for low-strength wastewater in a subtropical-climate area considering different primary treatments and mainstream BNR emerging processes and to identify the hotspots associated with each treatment. The processes under study were conventional primary sedimentation, CEPT, conventional activated sludge, SAF-MBR (includes AFBR followed by PN/AFMBR), which were divided into three scenarios (Figure 2).

214	S1 consisted of a conventional system based on the current operation and influent
215	characterization at Shatin STWs, which benefited from the organic presence for the N/D
216	process in activated sludge. Shatin STWs consists of conventional primary sedimentation
217	followed by activated sludge, and thickening followed by AD, dewatering, incineration and
218	landfill for sludge management. 61-63 Shatin STWs produces energy from AD through
219	combined heat and power (CHP) and dual fuel engines (DFE). The energy and chemical
220	consumption and transportation distance are based on Shatin STWs primary data.
221	S2 harnessed the organics in sewage for AD purposes by using CEPT without further BNR,
222	which illustrated the advantages of up-concentration over conventional primary treatment.
223	The primary treatment and raw CEPT sludge characterization in S2 are based on the current
224	operation at Stonecutters Island STWs. The influent characterization is based on Shatin
225	STWs. CEPT is the only treatment to obtain settled sewage in Stonecutters Island STWs,
226	while dewatering, incineration, and landfill are the processes for sludge management. 61,64
227	Thickening, AD with CHP and DFE for energy generation were added to S2 as an alternative
228	short-term strategy for sludge management. The AD process assumed in S2 is based on the
229	laboratory-scale anaerobic digester fed with raw CEPT sludge obtained from Stonecutters
230	Island STWs by Ju et al. ⁶⁵ The CHP and DFE performances are based on the operation at
231	Shatin STWs. The electricity consumption for sludge handling and dewatering, and
232	chemicals consumption is based on Stonecutters Island STWs. The consumption of electricity
233	for thickening and thermal energy for AD are based on Shatin STWs. The transportation
234	distance is based on Shatin STWs location. Given that the current water effluent from the
235	Stonecutters Island STWs has high COD and nitrogen concentrations, the Government has
236	encouraged further studies on different BNR processes.
237	S3 considered the need for BNR but discarded conventional activated sludge because of the
238	low C/N ratio after CEPT. 41 Instead, S3 explored emerging technologies by using a SAF-

MBR consisting of an AFBR and a PN/AFMBR to enhance water effluent quality. The
sludge management practices included thickening, AD with CHP and DFE for energy
generation, dewatering, incineration, and landfill. The primary treatment, raw CEPT sludge
characterization, AD process, and chemical consumption are based on the Stonecutters Island
STWs. The electricity consumption for sludge dewatering was updated with new data from
Stonecutters Island STWs. The influent characterization, energy production and consumption,
and transportation distance are based on Shatin STWs.
The geographical boundaries were taken from Europe (including Switzerland) and Rest of the
World (RoW) average values from the Ecoinvent database. However, the concepts were
regionalized as much as possible to China. The temporal boundary representing the STWs
lifetime was 30 years. The system boundaries included the following: construction, operation
and maintenance, and disposal. The system boundaries in the S-LCI framework included
GHG emissions from biological treatment, sludge management, treatment and disposal, and
energy recovery.

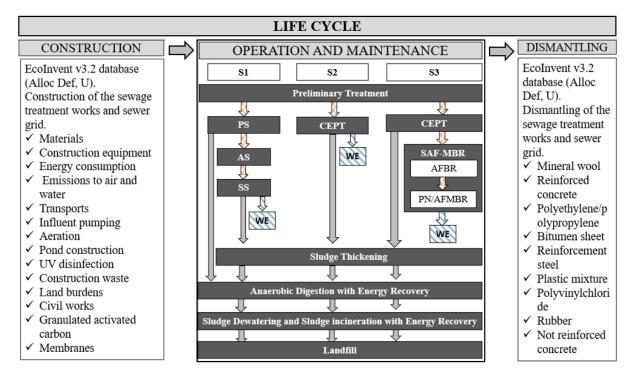


Figure 2. Sewage treatment works scenarios and system boundaries for scenario 1 (S1), scenario 2 (S2), and scenario 3 (S3). All the scenarios include energy and greenhouse gas implications. The squared orange arrows represent the mixed sewage streams, the gray arrows represent the sludge streams, and the blue diagonal arrows show the water streams. PS: Primary sedimentation; AS: Activated sludge; SS: Secondary sedimentation; WE: Water effluent; CEPT: Chemically enhanced primary treatment; SAF-MBR: Staged anaerobic fluidized-bed membrane bioreactor consisting of an anaerobic fluidized-bed bioreactor (AFBR) followed by a partial nitritation anammox fluidized-bed membrane bioreactor (PN/AFMBR).

2.3 LCI

The calculation of the LCI for S1 and S3 is explained in detail elsewhere. The S-LCI for S1 was based on the operation of Shatin STWs, whereas that for S3 was based on data from the Shatin and Stonecutters Island STWs. The differences between the previous and updated LCI for S1 and S3 are that COD and NH₄⁺ removal efficiencies from the final sedimentator were modified to 0% for S1; and, S3 was updated with newly collected data for electricity consumption for sludge dewatering.

Following the S-LCI framework, the LCI for S2 was calculated in this study based on the Shatin and Stonecutters Island STWs. The degritted sewage characterization for S2 is based on the previously collected data for S3 from Shatin STWs (Table S5-S6). The degritted sewage flow is 230,150 m³/d. The degritted sewage COD, total suspended solids (TSS) and

265	volatile suspended solids (VSS) concentrations are 310 mg/L, 270 mg/L, and 50 mg/L,
266	respectively. The NH ₄ ⁺ and total kjeldahl nitrogen (TKN) concentrations were assumed from
267	the effluent of the primary clarifier in Shatin STWs as 31 mg/L and 42 mg/L, respectively.
268	The nitrate concentration (11 mg/L) was assumed as the difference between TKN and $\mathrm{NH_4}^+$
269	concentration. Empirical data for S2 was collected from the previous experimental sludge
270	characterization in S3 which is based on Stonecutters Island STWs. ⁴¹ The empirical total
271	solids (TS), TSS, volatile solids (VS), VS/TS and TSS/TS for raw CEPT sludge is 38.65 g/L,
272	26.08 g/L, 17.09 g/L 0.44 and 0.67, respectively (Table S2). Further details are explained in
273	the supporting method (SM) S1.
274	The "Wastewater treatment facility, capacity #l/year {GLO} market for Alloc Def, U"
275	inventory from the Ecoinvent database was modified to include the considerations of S2. The
276	infrastructure considerations were based on the infrastructure inventories of S1 and S3,41 as
277	detailed in SM S1. The full inventories for the materials and the WWT facility for S2 are
278	shown in Tables S3-S4.
279	The water emissions were calculated from the data provided by the government authority in
280	charge of the STWs. The C/N ratio after primary sedimentation was 5.47, and the estimated
281	C/N ratio after CEPT is 2.76, as presented in Table S7. Other water emissions are explained
282	in detail in SM S1.
283	The NEB included electricity and heat consumption and production. The electricity and heat
284	production at the STWs and incineration facility were considered avoided products. S1 and
285	S3 showed net electricity consumption of 0.1604 and 0.0478kWh/m³, respectively, whereas
286	S2 indicated net electricity production of 0.1180 kWh/m³, as explained in SM S1 and Table
287	S12. The electricity consumption and production were accounted separately to enable clear
288	visualization of the saved impacts (electricity production) and the harmful impacts (electricity

289 consumption). The heat balance at the STWs reflected higher production than consumption, 290 as detailed in Table S12. 291 The thickened CEPT sludge production was previously calculated in S3 as 904 m³/d. The thickened sludge is anaerobically digested during 10 d with a VS destruction of 61%;65 thus, 292 the digested flow in S2 is 782.44 m³/d. The percentage of TS and VS in CEPT digested 293 294 sludge is obtained from a laboratory scale anaerobic digester as 2.58% of dry solids, and 75.32% of TS, respectively. 41 The dry solids content after dewatering was assumed as 31%, 295 296 hence the dewatered sludge production for incineration in S2 is 65.12 m³/d as shown in 297 Figure S1. Similar to S3, the biogas production in S2 was based on the elemental analysis of Shao et al., ⁶⁶ which measured the carbon, hydrogen, nitrogen, sulfur, oxygen and ash content 298 299 in raw CEPT sludge as 33.00%, 4.35%, 2.02%, 0.50%, 36.72%, and 23.41%, respectively. 300 The biogas production in S2 was 17,550 m³/d from which 8,531 m³/d was CH₄ gas (including 301 dissolved CH₄). S-LCI enables the quantification of biogenic emissions to replace the 302 existing default values in the Ecoinvent database for WWT. Otherwise, the calculations 303 would be based on generic data rather than site-specific or regionalized data. Other non-GHG 304 emissions were obtained from the Ecoinvent database which involves biogas combustion and 305 incineration as shown in Table S13 (e.g. carbon monoxide, sulfur dioxide, nitrogen oxides, 306 NMVOC, etc.). Material and fuel consumption for S2 are explained in SM S1. In S2, the emissions to the soil 307 308 were deleted because the sludge was not used for agricultural spreading. 309 The typical kinetic values and design criteria for anaerobic digestion by methanogenesis in 310 scenario 2 are detailed in Table S11. A summary of the operation, kinetic and design 311 parameters for the three scenarios are presented in Table S14.

2.4 LCIA

Eutrophication, acidification, freshwater ecotoxicity, marine ecotoxicity, human toxicity, global warming potential (GWP), and climate change are the impact categories that have received much attention in WWT. 13,15,17,19,37,40,46,48,67-72 Thus, particulate matter formation (PMF), metal depletion, fossil depletion, terrestrial ecotoxicity, abiotic depletion, and photochemical oxidation are quantified but not discussed further in this study.

2.5 Interpretation

In this study, the total environmental impacts include positive and negative values. Positive values represent harmful impacts on the environment, whereas negative ones correspond to those impacts that have been avoided because of energy production (i.e., electricity and heat). The avoided impacts are included to reflect the benefits of energy self-sufficiency for the environment.

The sensitivity analysis evaluates the sensitivity of the emissions in the impact categories to sensitive parameters. Table S16 lists the 9 sensitive parameters, which represent variations in membrane performance, energy efficiency, and GHG fugitive emissions. SAF-MBR is an emerging process for which less data is available relative to activated sludge and CEPT, thus almost half of the sensitive parameters were associated with S3. In particular, a sensitivity analysis was done assuming the membrane lifetime in S3 as 5, 10 and 15 years. ¹⁹ Similarly, the sensitivity to the membrane cleaning frequency uncertainty parameter was evaluated by assuming 4.33 times/month instead of 0.027 times/month. ¹⁹ S3 assumes 0.227 kWh/m³ of electricity consumption for the SAF-MBR, thus the sensitivity analysis assumed future reductions to the electricity consumption as 0.140 kWh/m³ and 0.053 kWh/m³. ^{33,73} The sensitivity to dissolved CH₄ recovery was performed for 0%, 50% and 90%. ¹⁹ Vacuum extraction with degassing membranes is the process assumed for dissolved CH₄ recovery, and the design parameters are explained in SM S3. ^{19,74-76} Spearman's rank correlation coefficient

was determined between the sensitive parameters and the impact categories results, and an arbitrary level of a 5% change in the impact values were chosen as the sensitivity threshold to determine if the influence of the sensitive parameter is significant.⁷⁷

The Monte Carlo analysis evaluated the aggregate impacts of data uncertainty with a 95% confidence interval and 1000 iterations for reliability test. The probability distributions were obtained through the Pedigree Matrix in the "S-LCI" file.⁴⁴

The results of the S-LCI for S2 were compared to S1 and S3, as summarized in Table S15.

3. RESULTS AND DISCUSSION

3.1 LCI

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346 The complete LCI for S2 is shown in Table S13. The highest COD, NH₄⁺, and NO₃⁻ 347 concentrations in the effluent resulted from using CEPT as the only treatment to obtain 348 settled sewage in S2. CEPT followed by SAF-MBR in S3 resulted in reductions of ~94%, 349 94% and 75% in COD, NH₄⁺, and NO₃⁻ compared to S2, respectively. However, the low-350 voltage electricity consumption within the WRRF in S3 was ~2.66 times higher than S2. 351 Despite the use of membranes, the conventional system in S1 consumed ~26% more low-352 voltage electricity than S3. In addition, S3 reduced COD and NH₄⁺ by ~89% and 44% 353 compared to S1, respectively. Yet, NO₃⁻ emissions increased about one order of magnitude 354 for S3 compared to S1 due to the PN/A process consideration. 355 The highest sludge production resulted from activated sludge in S1. CEPT followed by SAF-356 MBR in S3 and sole CEPT in S2 produced ~31% and 35% less sludge compared to S1, respectively. Even though S3 produced only ~4% more sludge than S2 compared to S1, the 357 358 generation of low-voltage electricity through AD and high-voltage electricity through 359 incineration was ~17% and 5% higher than S2, respectively. However, the NEB showed that 360 S3 had net electricity consumption, whereas S2 had net electricity production. In terms of

GHG emissions, S1 produced the highest amount of biogenic CO₂, whereas S2 and S3 produced more biogenic CH₄ than S1. S3 had the lowest direct CH₄ emissions because of the low COD concentration in the effluent. S2 produced the highest N₂O emissions because of the untreated NH₄⁺ in the effluent.

The main trade-off that could be identified among the scenarios was related to electricity consumption and water effluent quality. Even though S2 reached energy self-sufficiency, the water effluent quality was the worst among the three scenarios. In order to obtain a clearer idea of each system's environmental performance, an LCIA was conducted.

3.2 LCIA

The results of Recipe Midpoint (Figure 3) identified marine ecotoxicity, marine eutrophication, and freshwater ecotoxicity as the environmental impact categories with the highest values for the three scenarios. S1 has the highest impacts on marine and freshwater

marine eutrophication. S3 has the lowest impact on marine eutrophication. S1 exhibits the highest impact in all other impact categories, and S2 exhibits the lowest, except for metal depletion.

S-LCI enables site-specific inventories particularly for nitrogen emissions in the water effluent, which is an important factor for marine eutrophication and ecotoxicity. In addition, S-LCI encourages the collection of site-specific data regarding electricity consumption in WWTPs and WRRFs, which is key in freshwater and marine ecotoxicity, and freshwater eutrophication. Thus, the implementation of S-LCI helps in obtaining and processing site-

specific data that are related to the categories with the highest impacts in the LCIA.

ecotoxicity. S2 has the lowest impacts in marine and freshwater ecotoxicity, but the highest in

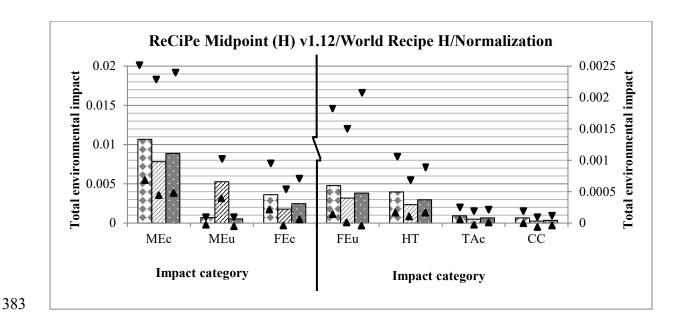
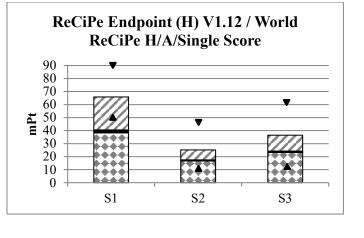


Figure 3. Recipe Midpoint results for S1: PS + AS (scenario 1, including primary sedimentation and activated sludge), S2: CEPT (scenario 2, including chemically enhanced primary treatment), and S3: CEPT + SAF–MBR (scenario 3, including CEPT and staged anaerobic fluidized membrane bioreactor). MEc: Marine ecotoxicity (kg 1,4-DB eq); MEu: Marine eutrophication (kg N eq); FEc: Freshwater ecotoxicity (kg 1,4-DB eq); FEu; Freshwater eutrophication (kg P eq); HT: Human toxicity (kg 1,4-DB eq); TAc: Terrestrial acidification (kg SO2 eq); CC: Climate change (kg CO2 eq). Triangles (▼,▲) represent 95% confidence interval of the total environmental impact per impact category from the Monte Carlo simulations.

The results of Recipe Endpoint (Figure 4) identify S1 with the highest environmental impact, followed by S3 and S2. Among the three impact categories of Recipe Endpoint, human health has the highest contribution to the single score results. The human health category is linked to human toxicity and air pollution categories such as PMF, climate change, and photochemical oxidation.⁴⁷ In Recipe Midpoint, S1 has the highest impacts on human toxicity, climate change, and PMF, whereas S2 has the lowest impacts. The resources category focuses on fossil fuel and mineral depletion. In Recipe Midpoint, S1 has the highest impacts for fossil and metal depletion, whereas S2 has the lowest impacts for fossil depletion and S3 for metal depletion. The ecosystems category includes terrestrial, marine water, and freshwater damage. Marine and freshwater ecotoxicity, were identified as the impact categories with the highest damage in midpoint methodologies. However, the contribution of these water damage categories identified in the midpoint results is barely reflected in the endpoint results.



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Human health Resources Ecosystems \$\times 2.5\% \neq 97.5\%

Figure 4. Recipe endpoint single score results for S1 (scenario 1, including primary sedimentation and activated sludge), S2 (scenario 2, including chemically enhanced primary treatment (CEPT)), and S3 (scenario 3, including CEPT and staged anaerobic fluidized membrane bioreactor). Triangles (▼, ▲) represent 95% confidence interval of the total environmental impact per impact category from the Monte Carlo simulations.

In contrast to Recipe Endpoint, the results of Ecological Scarcity 2013 identify S2 with the highest environmental impact, followed by S1 and S3 (Figure 5). In Ecological Scarcity 2013, the categories with the highest values were water pollutants and heavy metals into the water. For the water pollutants category, the total nitrogen and COD concentrations in the effluent of S2 do not comply with the standards for discharged effluents established by the Hong Kong government. 78 Even though the Ecological Scarcity 2013 methodology links substance emissions to European instead of Hong Kongnese environmental policy, it is reasonable that water pollutants have the highest contribution to the single score and that S2 has the worst environmental performance. The heavy metals (e.g. zinc, copper, cadmium) into the water category represent the background emissions obtained from the Ecoinvent database for sewer overload discharge, direct WWTP emissions, and sludge incineration. The discrepancy among different methodologies lies in their different priorities. Specifically, Recipe Endpoint assigns higher weights to human toxicity and air pollution, whereas Ecological Scarcity 2013 focuses on water impacts. A detailed comparison of the contribution analysis for endpoint methodologies clearly shows that the main contributor of Ecological Scarcity 2013 is direct emissions (e.g. COD, NH₄⁺ and dissolved CH₄ into the

water and GHG emissions into air), whereas the main contributors for Recipe Endpoint are the electricity consumption and sewer construction emissions (Figure S3).

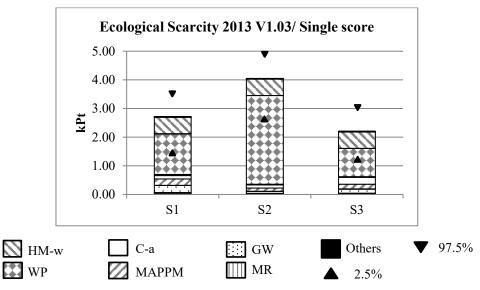


Figure 5. Ecological Scarcity 2013 single score results for S1 (scenario 1, including primary sedimentation and activated sludge), S2 (scenario 2, including chemically enhanced primary treatment (CEPT)), and S3 (scenario 3, including CEPT and staged anaerobic fluidized membrane bioreactor). HM-w: Heavy metals into the water; WP: Water pollutants; C-a: Carcinogenic substances into the air; MAPPM: Main air pollutants and particulate matter; GW: Global warming; MR: Mineral resources. Triangles (▼, ▲) represent a 95% confidence interval of the total environmental impact per impact category from the Monte Carlo simulations.

3.3 Interpretation

3.3.1 Contribution analysis

Ten processes contribute to over 92% of the impacts in midpoint methodologies for each scenario. The contribution analysis results identify that the main environmental hotspots are direct emissions and electricity consumption at the WWTPs or WRRF. Electricity use has been previously identified as one of the main contributors to MBRs processes. ¹⁸ Other less dominant hotspots recognized are the use of chemicals, sewer grid, CHP, and infrastructure. In addition, the impacts avoided from electricity recovery in S2 are higher than the harmful impacts from electricity consumption. Figure 6 shows the contribution analyses of Recipe Midpoint for freshwater and marine eutrophication and ecotoxicity. Direct emissions are the highest contributor for freshwater and marine eutrophication in the three scenarios. For freshwater eutrophication in S1 and S3, direct emissions are closely followed by low-voltage

electricity consumption. Even if S2 has the lowest effluent water quality, its electricity balance translates into lower freshwater eutrophication impacts than S1 and S3. In marine eutrophication, direct emissions represent more than 87% of the impacts. Thus, the high content of macro-nutrients in the water effluent of S2 makes it the indisputable worst scenario in this category. In addition to direct emissions, the use of polyacrylamide in S1 contributes to marine eutrophication. The main contributor of freshwater ecotoxicity is low-voltage electricity consumption, followed by direct emissions for S2 and S3, and infrastructure for S1. S2 has the best environmental performance in freshwater ecotoxicity because of the fewer background emissions from low-voltage electricity consumption compared to S1 and S3. Even if marine ecotoxicity is mainly dominated by direct emissions and the highest contribution of direct emissions is for S2, the electricity balance of S2 translates into the best environmental performance also in this category. The contribution analyses of terrestrial acidification and ecotoxicity and human toxicity for Recipe Midpoint are presented in Figure S2a. For terrestrial acidification, direct emissions and low-voltage electricity consumption are the two key contributors. The main contributors to terrestrial ecotoxicity are chemical consumption, sewer grid construction and WRRF infrastructure for S1, S2, and S3, respectively. For human toxicity, the key contributor for S1 and S3 is low-voltage electricity consumption, whereas it is the sewer grid for S2. The contribution analysis of air pollution and material depletion categories is presented in Figure S2b. The main contributors to climate change are the indirect emissions from lowvoltage electricity consumption, and direct GHG emissions intrinsic to the treatment, dissolved CH₄, and untreated COD and NH₄⁺ in the water effluent. S2 has the highest direct emissions because of the concentrations of COD and NH₄⁺ in the effluent that are transformed into CH₄ and N₂O by natural processes. ⁷⁹ Nevertheless, the electricity implications give S2 the best environmental performance in this category. N₂O emissions

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have been a concern in two-stage PN/A systems,²⁰ yet S3 considers a single-stage PN/A system in an enclosed system, which is expected to control N₂O emissions.²³ Electricity consumption followed by direct emissions is the main contributor to climate change in S3. Similarly, previous studies identified dissolved CH₄ in the effluent and electricity use of AnMBrs as the key contributors in GWP.^{15,19}

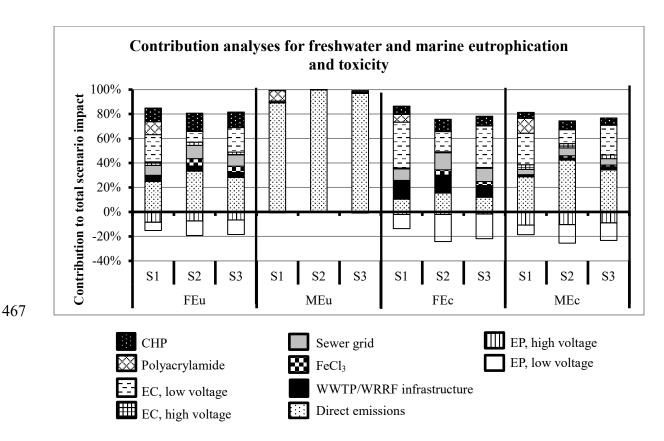


Figure 6. Contribution analyses for freshwater and marine eutrophication and ecotoxicity categories of Recipe Midpoint. S1 (scenario 1, including primary sedimentation and activated sludge), S2 (scenario 2, including chemically enhanced primary treatment), and S3 (scenario 3, including CEPT and staged anaerobic fluidized membrane bioreactor). FEu: Freshwater eutrophication; MEu: Marine eutrophication; FEc: Freshwater ecotoxicity; MEc: Marine ecotoxicity; CHP: Combined heat and power; EC: Electricity consumption; WWTP: Wastewater treatment plant; WRRF: Water resource recovery facility; EP: Electricity production.

3.3.2 Uncertainties management

The results of the sensitivity analysis with the SAF-MBR sensitive parameters show a negative correlation for membrane lifetime and impact emissions, thus increasing the membrane lifetime translates in less emissions. Yet, the increase of membrane lifetime from 5 to 10 and 15 years only decreases emissions by maximum 3% to 4% (Figure S4). In contrast,

membrane cleaning frequency, SAF-MBR electricity consumption, and dissolved methane recovery show a positive correlation to impact emissions (Table S17), thus increasing these parameters result in increased emissions. Increasing the frequency of membrane cleaning represented an increase in up to one order of magnitude for marine ecotoxicity, freshwater eutrophication, human toxicity, terrestrial acidification and climate change (Figure S4). Decreasing the SAF-MBR electricity consumption from 0.227 kWh/m³ to 0.14 kWh/m³ decreases emissions in the range of ~6% (marine ecotoxicity) to 25% (climate change) while decreasing to 0.053 kWh/m³ decreases emissions in the range of ~13% (marine ecotoxicity) to 50% (climate change) (Figure S4). Marine eutrophication was negligibly influenced by SAF-MBR electricity consumption. The recovery of 50% and 90% of dissolved CH₄ with a degassing membrane resulted in increased emissions (Figure S4). Yet, when the recovery is from 0% to 50% the emissions increase in a range of ~<1% (marine ecotoxicity and marine eutrophication) to ~5% (human toxicity). Furthermore, increasing the recovery from 50% to 90% has neglecting increased emissions (<0.02%). On the contrary, GWP and marine eutrophication decreased when CH₄ and nutrient were recovered for a single-stage AnMBR without previous primary treatment.¹⁵ The results of the sensitivity analysis with the sludge and energy production sensitive parameters (Table S18) show that increasing the electrical efficiency for CHP results in fewer emissions (freshwater ecotoxicity, terrestrial acidification, and climate change) for S2 and S3. Increasing the thermal efficiency for CHP in S3 shows a positive correlation to emissions in climate change. The electrical and thermal efficiency in DFE has no significant impact on emissions. Lastly, more fugitive emissions represent more climate change emissions for all scenarios. The Monte Carlo results show that the standard error of the mean (SEM) were $<0.01, \le 0.33$, and ≤0.02 for Recipe Midpoint, Recipe Endpoint and Ecological Scarcity 2013, respectively,

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as detailed in Table S19-S21. The highest SEM in Recipe Midpoint, Recipe Endpoint, and Ecological Scarcity are for marine ecotoxicity, human health, and heavy metals into water, respectively.

3.3.3 Policy recommendations

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The analysis of the case study for Shatin and Stonecutters Island STWs in Hong Kong and the general methodological contributions of the site-specific S-LCI integration to the LCA enable the recommendation of two tailor-made policies and three general policies for decision-makers. Current policy related to WWTPs and WRRFs focuses on water discharge emissions, but more holistic considerations should be included in the legislation to ensure sustainable processes. First, the addition of AD for sludge management to the current CEPT process in Stonecutters Island STWs as presented in S2 is recommended as a policy in the short-term which translates in energy self-sufficiency and overall environmental impacts reduction (range of 27%–61 %), except for the marine eutrophication impact, compared to conventional treatment, as supported by Recipe Midpoint. The low-voltage electricity consumption shows a positive linear relationship to the freshwater eutrophication and ecotoxicity, marine ecotoxicity and climate change categories, where higher electricity consumption translates into higher impacts. Similarly, Smith et al.¹⁹ found a linear relationship where higher energy consumption translates into higher GWP for systems including HRAS, AD, activated sludge and aerobic MBRs. In contrast, low-voltage electricity consumption had a negative correlation with COD, NH₄⁺ and NO₃⁻ emissions to water (thus marine eutrophication), where higher electricity consumption resulted in fewer water pollutants emissions. These results agree with the environmental trade-offs previously identified between eutrophication, toxicity, and global warming categories.³⁷

Second, the integration of the SAF-MBR for BNR and AD for sludge management to the current CEPT process in Stonecutters Island STWs as presented in S3 is recommended as a policy in the long-term to comply with discharge standards and reduce all impact categories (range of 17%–47 %) compared to S1 as supported by Recipe Midpoint, even if energy selfsufficiency is not achieved. The implementation of S3 for compliance with discharge levels would significantly (~90%) reduce the impact to marine eutrophication, but would increase (range of 12%–28%) freshwater eutrophication, freshwater ecotoxicity, marine ecotoxicity, terrestrial acidification, climate change, and human toxicity compared to S2 due to the addition of the SAF-MBR. These results agree with previous conclusions stating that stricter discharge levels reduce local eutrophication, but hinder overall environmental performance.⁷⁰ The reduction of marine eutrophication in mainstream one-step anammox of the SAF-MBR is ~7% higher than the reduction previously identified (16%) in sidestream two-step anammox. 40 Climate change has been found to increase when marine eutrophication is reduced, 40 whereas the configuration of S3 decreases climate change impacts because of the enhanced electricity recovery and the reduced polyacrylamide consumption. The endpoint methodologies show opposing results for S2, whereas consistent reductions of 45% and 21% are identified for S3 compared to S1 by Recipe Endpoint and Ecological Scarcity 2013, respectively. Thus, the selection of S3 represents reduced impacts for the two endpoint methodologies, without compromising marine eutrophication impacts. Third, when planning a new WRRF or upgrading a WWTP with CEPT process, the integration of AD for sludge treatment and BNR should be mandatory, so that the benefits of organic capture through a low energy process can be translated into high energy recovery without compromising water effluent quality. CEPT process has been considered as an interesting option for megacities⁵ and energy recovery due to COD capture,⁴ low footprint,

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547 low energy requirement, and high removal efficiencies. In recent decades, China and the US have adopted the CEPT process in full-scale plants. 80-82 548 549 Fourth, when the technology transfer is ready for adoption of mainstream PN/AFMBR for 550 low C/N sewage, more stringent effluent discharge limits could be set through legislation. By 2014, 100 full-scale PN/A installations were predicted to be operating worldwide. 25 Even 551 552 though more than 50% of all PN/A systems are sequencing batch reactors instead of MBRs, 553 88% of all plants are operated as single-stage systems as the PN/AFMBR presented in S3. 554 The data collected and the S-LCI framework in this study complemented with site-specific 555 data of these existing PN/A systems could be applied to obtain the holistic environmental 556 impacts of these installations. In addition, upgrading of these installations by implementing 557 MBR technology for PN/AFMBR operation might shift their application from sidestream to 558 mainstream WWT. The resulted water effluent would then have reduced carbon, ^{73,83} nitrogen,³⁴ pharmaceuticals,⁸⁴ and heavy metals.⁸⁵ 559 560 Lastly, if Governments encourage the pursuit of energy self-sufficient WRRFs, then a site-561 specific LCA analysis should be performed to ensure that the environmental impacts are not 562 shifted from one category to another. The overall environmental impacts should be studied 563 and reported, even for WRRFs that are currently energy self-sufficient (e.g. Austrian 564 WWTPs). The S-LCI is a tool that enables enhanced specificity in the quantification of 565 environmental impacts of WWTPs and WRRFs by standardizing the data collection and 566 processing of life-cycle inventories. For example, generic data from databases are substituted 567 by the site-specific air emissions calculated by S-LCI because data on GHG emissions from 568 WWTPs are not available in Hong Kong. In addition, the collection of site-specific data 569 regarding low-voltage electricity consumption translates into a more accurate quantification 570 of indirect GHG emissions. Site-specific data collection for electricity consumption and 571 sample characterization integrated into the stoichiometric calculations of S-LCI to obtain the

water, air, and soil emissions result in site-specific inventories that reduce uncertainty. Similarly, material quantification provided in the S-LCI for the WWTPs infrastructure enabled the estimation for infrastructure in S2, which would otherwise have a generic inventory.

3.3.4 Limitations and future work

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There are inherent limitations to LCA, S-LCI, and emerging technologies. Regarding LCA, several areas of improvement have been identified: data requirements, lack of specificity and regionalization, difficulties in performing comparative analyses across different studies, and IAM inconsistencies and simplified models to describe complex environmental impacts. 46,86 Data requirements, site specificity, regionalization, and standardization can be tackled using S-LCI for the calculation of the inventories. However, the data for SAF-MBR is based mainly on a steady-state pilot- and laboratory-scale studies. In addition, emerging pollutants and heavy metals are not included in the foreground emissions calculation of S-LCI. Despite these limitations, the plant-wide analysis of different primary treatments and low C/N ratio processes helps in identifying their environmental trade-offs and hotspots. This study confirms the viable integration of S-LCI into LCA as a tool for enhanced standardization and specificity, which decreases uncertainty. Further scenario analysis could be performed on S3. SAF-MBR could be integrated into a single-stage MBR to reduce materials, footprint, GHG emissions, and electricity consumption. 83 The benefits of high effluent quality might be enhanced with the integration of S3 into the urban water management system for water reuse. 87 Further development for mainstream full-scale implementation of SAF-MBR can be incorporated as data become available. Advances in the removal of emerging pollutants and heavy metals in MBR, AD, and incineration could be integrated. Sewer grid quantification can be studied in further detail.88

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6. SUPPORTING INFORMATION

- The "Supporting Information" file contains the stoichiometric life-cycle inventory for
- scenario 2, contribution analyses and uncertainty and sensitivity analysis details.

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871 HIGHLIGHTS

- CEPT must be complemented with nutrient removal processes and AD for sludge to leverage up-concentration with high energy recovery without compromising water effluent quality.
- When pursuing energy self-sufficient WRRFs, a site-specific LCA should evaluate potential environmental impacts shifts.
- CEPT followed by AFBR and PN/AFMBR with anaerobic digestion for sludge management consumes 0.0478 kWh/m³ and reduces all environmental impacts (range 17-47 %).
- The main environmental trade-offs between different primary treatments and low carbon systems were triggered by electricity consumption, water effluent quality and air emissions.
- Integrating the S-LCI framework enhanced specificity, regionalization and standardization of the air, water and soil emissions of conventional and emerging technologies.

GRAPHICAL ABSTRACT

