A Standardized Stoichiometric Life-cycle Inventory for Enhanced Specificity in Environmental Assessment of Sewage Treatment

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Abstract: In recent years, many life-cycle assessments (LCAs) have been applied to the field of sewage treatment (ST). However, most LCAs lack systematic data collection (DC) and processing methods for inventories of conventional ST (CST), much less for recently-developed technologies. In addition, the use of site-generic databases results in LCAs that lack the representativeness and understanding of the regional environmental impacts and trade-offs between different impact categories, especially nutrient enrichment and toxicity-related categories. These shortcomings make comparative evaluation and implementation more challenging. In order to assist in the decision-making process, a novel stoichiometric life-cycle inventory (S-LCI) for ST was developed. In the S-LCI, biochemical pathways derived from elemental analyses combined with process-engineering calculations enable steady-state comparison of the water, air, and soil emissions of any sewage and sludge sample treated through the ST configurations here analyzed. The DC required for the estimation of the foreground data for a CST is summarized in a 41-item checklist. Moreover, the S-LCI was validated for CST by comparing the S-LCI with actual ST plant operations performed in Hong Kong. A novel energy-derived ST inventory is developed and compared here with the CST. The resulting inventories are ready to be integrated into the SimaPro software for life cycle impact assessment as illustrated by the case study. Using the S-LCI not only helps to standardize the DC and processing, but it also enhances the level of specificity by using sample characterization and site-specific data. The EcoInvent database, which contains a single sample characterization per Swiss and global average ST plant class could be expanded by using the S-LCI.

Keywords: chemically enhanced primary treatment, partial nitritation/anammox fluidized-bed membrane bioreactor, stoichiometry, life-cycle assessment

1. INTRODUCTION

In 2012, sewage treatment (ST) was the eighth-largest anthropogenic source of methane (CH_4) emissions (12.8 million metric tons of $CO_{2\text{-equivalent}}$) in the United States.[1] Some countries have focused on developing more-holistic ST processes, so called emerging processes, to save energy and reduce greenhouse gas (GHG) emissions. The pursuit of sustainable ST processes requires assessments of the trade-offs between the level of ST, sludge production, and energy-related emissions.[2,3,4] However, the application of diverse assessment tools for comparison has complicated the decision-making process for their implementation.

Life cycle assessment (LCA) is the most commonly used tool to account for environmental impacts in the ST field.[2,5] As defined by the International Standards Organization 14000 series, an LCA is a methodology for evaluating or comparing the potential environmental impacts of a product, service, or activity throughout its life cycle.[2,6,7] LCA comprises of four main steps: (a) goal and scope; (b) life-cycle inventory (LCI); (c) life-cycle impact assessment, and (d) interpretation. Previous studies have identified several areas in which the LCA methodology could be improved.[2,3,7,8] In particular, challenges in LCI include data requirements, inventory coverage levels, site-specificity, regionalization, and uncertainty.

Data requirements, standardization, and quality have been identified as important factors to decrease bias and uncertainty; and, to increase representativeness[9-18] as explained in detail in Table S1. However, many authors have faced difficulties in collecting reliable data, because it was unavailable, expensive or time-consuming.[3,10,13,14,17-27] Current methodologies for data collection (DC) include measuring campaigns (i.e. GHG emissions), [9,11,15,20,25,28-32] literature, government reports, databases, modelling, and simulations. Regarding modeling and simulation, some literature can be found on plant-wide biological models applied to conventional systems: for example, the Benchmark Simulation Model series including activated sludge model No. 1 and anaerobic digestion model no. 1[33-36], BioWin,[25,37-39]

WEST®,[21,40-41] Mantis series from GPS-X,[42-45] design and simulation of activated sludge systems,[46-50] decision support system,[51-52] and dynamic supply chain system model.[53] In addition, there are biological models that have been integrated to LCA.[37] These models are excellent for research purposes, but their application in an environmental assessment (EA) is limited because their purpose differs from the LCA approach. In general, these models involve high-level data requirements; focus on conventional treatments thus neglecting emerging technologies; lack the integration of energy generation, material inputs, and/or GHG implications; and/or their design is not focused on obtaining functional units for LCA. Detailed explanations of these models are given in Table S2.

The need for site-specificity and regionalization has been identified by several authors as explained in detail in Table S1.[10,12,18,27,54-55] Site-specific and regionalized data enables understanding of the regional environmental impacts and trade-offs between different environmental indicators;[8,56] increases relevance, precision, discriminating power, and representativeness;[3,9,13,15,23,57] and helps to reduce uncertainty in the nutrient enrichment-related and ecotoxicity categories.[4,5,9,13,21,58] The available databases such as Ecoinvent, the US Life Cycle Inventory, and the Swiss Input/output contain comprehensive LCI data which represent the situation in Europe, North America or Japan. The ST inventory templates in the Ecoinvent database consists of just information on Switzerland and the rest of the world (RoW) based on a global average.[59] Thus, any other country using the databases without site-specific or regionalized data might generate results with high uncertainty.

Even though the lack of a systematic approach for DC, site-specificity, and regionalization have been recognized in several studies, only a few studies have been solely dedicated to tackling these issues. In terms of DC, some efforts have been made to provide transfer coefficients for municipal conventional sewage treatment works (STW)[59] and ST from the chemical industry.[60] However, recently developed processes were not included, and sludge management and GHG emissions (except CO_2) were excluded from the system boundaries. Regarding standardization, efforts have focused on the development of a primary data (PD) collection scheme[18] and data requirement reduction through sub-process elimination to decrease parameter uncertainty.[13] Yet no further analysis for predicting inventory was included, and a potential for increased scenario uncertainty remains. Concerning regionalization, Hernández-Padilla[9] not only identified that regionalization can be achieved through the inventory and the characterization factors (CFs) but also proposed a systematic approach for regionalization based solely on DC. Bai et al.[8] developed China-specific CFs, while Lehtoranta et al.[61] included Finland-specific CFs. Furthermore, Lorenzo-Toja et al.[11] carried out on-site measurements in two different climatic regions to provide regional emission factors as a benchmark for Spain.

This study focuses on methodological development of the LCI with standardization of DC and processing, and regionalization through site-specific data. The stoichiometric LCI (S-LCI) is a framework designed to integrate the elemental analysis from any type of sewage sample using the stoichiometric calculations for constructing the foreground LCI information. In the present study, the S-LCI is compared with data from (i) a conventional STW in Hong Kong, and (ii) an energy-derived sewage treatment (EST) system that includes recently developed processes. The main features of S-LCI are (i) its enhanced standardization of PD requirements and processing through a determined checklist and an Excel-based spreadsheet, (ii) its increased specificity through the fixed checklist, S-LCI recommendations, and a laboratory test—elemental analysis—for sample composition, and (iii) its potential for database expansion.

2. MATERIALS AND METHODS

The S-LCI is integrated into the second step of LCA, namely the LCI (Fig. 1). The S-LCI comprises of three main parts, namely DC, calculations, and results. The DC includes the elemental analysis of the samples, PD, and STW energy requirements. The calculations include the STW configurations and the stoichiometric calculations. The results of the elemental analysis are used to construct a specific empirical formula for microbial cells. The stoichiometric calculations are developed based on the Thermodynamic Electron Equivalents Model (TEEM).[62] The TEEM is complemented with the PD (i.e., flows, concentrations, removal efficiencies) and energy requirements to construct whole-plant process inventories including the system inputs and outputs. The results of the calculations are converted to the LCA functional unit (FU) of "1 m^3 of sewage treated".

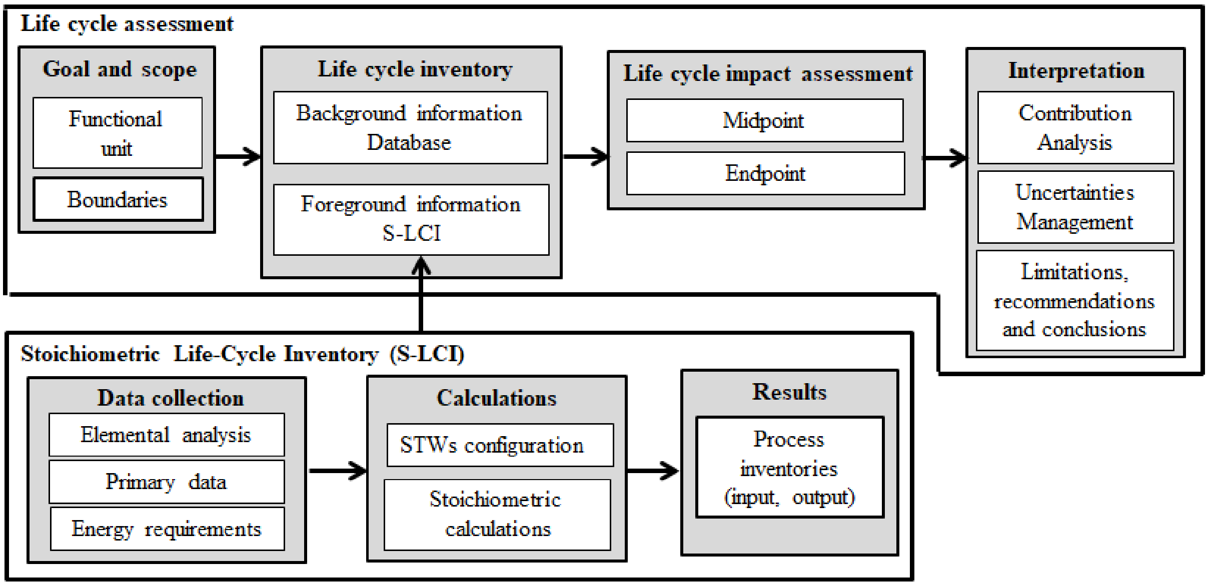

Fig. 1. The framework of stoichiometric life-cycle inventory (S-LCI).

2.1 Data Collection

2.1.1 Elemental analysis

The sewage and sludge sample collection process followed the APHA-1060B procedure for grab samples.[63] The sample preparation followed the APHA-2540G procedure for total solids (TS).[63] The samples were then pulverized as determined by ASTM-D2013[64], while the CHNS elemental analysis according to ASTM-D5373 to obtain the weight percentage of each element.[65] Oxygen was calculated by subtraction, and the ash was obtained according to ASTM-D3174.[66]

2.1.2 PD

In the S-LCI, the PD was collected from (i) the authority in charge of ST and (ii) the results of the experiments on chemically-enhanced primary treatment (CEPT) sewage and sludge. A 41-item checklist of the collected PD is provided in Table S3, which can be used as a reference for future studies. In the present study, the PD collected included the blueprints of a conventional STW and a literature review that focused on the materials used for the construction of recently-developed technologies that could serve as a template for future studies.

The experimental results on CEPT sewage and sludge can be used as default values in future work. The sample collection and TS test were performed as described for the elemental analysis. The total suspended solids (TSS) test followed the APHA-2540D procedure, while the volatile solids (VS) test was performed based on the APHA-2540G procedure.[63]

2.1.3 Energy requirements

The data collected for the energy requirements were from the authority in charge of the ST, which was complemented with a literature review as presented in Table S3.

2.2 Stoichiometric and kinetic calculations

The percentage of each organic element (C, H, O, and N) present in a sewage sample was obtained from the results of the elemental analysis. Rittman and McCarty[62] introduced the equations for the empirical microbial cells formulas $C_nH_aO_bN_c$, where n, a, b, and c are the mass distributions of the four elements in a sample. These empirical formulas were used as part of the custom organic half-reactions for TEEM. As Rittman and McCarty[62] state: "The overall biochemical equation (Eq. (1)) uses the half-reactions of the electron donor (R_d), the electron acceptor (R_a), and the carbon or nitrogen source for biomass synthesis (R_c):

$$R = f_e R_a + f_s R_c - R_d \ , \tag{1}$$

where R is the general equation for microbial synthesis and growth on an electron-equivalent basis, f_e is the electron portion for energy generation considering net yield, R_a is the electron-acceptor equation, f_s is the electron portion used for cell synthesis considering net yield, R_c is the cell synthesis equation, and R_d is the electron-donor equation. The fractions of electrons used for energy generation (f_e) and synthesis (f_s) must equal 1.0. Assuming a steady-state process, f_s can be estimated from Eq. (2):

$$f_s = f_s^0 \left[\frac{1+(1-f_d)b\theta_x}{1+b\theta_x}\right], \tag{2}$$

where f_s^0 is the portion of energy used for cell synthesis, f_d is the fraction of the active biomass, that is, biodegradable, b is the endogenous decay rate, and θ_x is the solids retention time.”[62]

The overall biochemical equations were built for conventional biological treatments, such as organic oxidation (Ox), nitrification (Nit), denitrification (Den), and methanogenesis (Meth), and for ESTs that included partial nitritation (Pn) and anammox (Anx). Pn and Anx were selected because a recent study focusing on nitrogen removal identified that mainstream anaerobic treatment followed by Anx, and anaerobic digestion (AD) for treating solids were the most environmentally friendly options compared to conventional nitrification/denitrification processes.[67] Activated sludge (AS) treatment included Ox (Eq. (3)) and proportional nitrification-denitrification (Eq. (9)). AD was based on Meth (Eq. (10)). SF-MBR included proportional partial nitritation/anammox (Eq. (16)) and Meth processes. Table 1 contains the overall biochemical equations for the conventional and EST processes.

Table 1. Overall reactions to conventional and energy-derived sewage treatments

Process	Overall reaction
Organic oxidation	$R_{Ox}: C_nH_aO_bN_c + \frac{df_{e_{Ox}}}{4}O_2 \rightarrow \left(n - c - \frac{df_{s_{Ox}}}{5}\right)CO_2 + \frac{f_{s_{Ox}}}{20}C_5H_7O_2N + \left(c - \frac{df_{s_{Ox}}}{20}\right)NH_4^+ + \left(c - \frac{df_{s_{Ox}}}{20}\right)HCO_3^- + \left(-2n + b - c + \frac{df_{e_{Ox}}}{2} + \frac{9df_{s_{Ox}}}{20}\right)H_2O$ (3)
Nitrification	$R_{Nit}: \left(\frac{f_{s_{Nit}}}{20} + \frac{1}{8}\right)NH_4^+ + \frac{f_{e_{Nit}}}{4}O_2 + \frac{f_{s_{Nit}}}{5}CO_2 + \frac{f_{s_{Nit}}}{20}HCO_3^- \rightarrow \frac{f_{s_{Nit}}}{20}C_5H_7O_2N + \frac{1}{8}NO_3^- + \left(\frac{5}{4} - f_{e_{Nit}} - f_{s_{Nit}}\right)H^+ + \left(\frac{f_{e_{Nit}}}{2} + \frac{9f_{s_{Nit}}}{20} - \frac{3}{8}\right)H_2O$ (4)
Denitrification	$R_{Den}: \frac{1}{d}C_nH_aO_bN_c + \left(\frac{f_{e_{Den}}}{5} + \frac{f_{s_{Den}}}{28}\right)NO_3^- + \left(\frac{6f_{e_{Den}}}{5} + \frac{29f_{s_{Den}}}{28} - 1\right)H^+ \rightarrow \frac{f_{s_{Den}}}{28}C_5H_7O_2N + \frac{f_{e_{Den}}}{10}N_2 + \frac{c}{d}NH_4^+ + \left(\frac{n}{d} - \frac{c}{d} - \frac{5f_{s_{Den}}}{28}\right)CO_2 + \frac{c}{d}HCO_3^- + \left(\frac{-2n}{d} + \frac{b}{d} - \frac{c}{d} + \frac{3f_{e_{Den}}}{5} + \frac{11f_{s_{Den}}}{28}\right)H_2O$ (5)
Proportional Nitrification-Denitrification	Step 1: Normalization to one mole of ammonium $R_{Nit_{norm}}: R_{Nit} * \frac{1}{\left(\frac{f_{s_{Nit}}}{20} + \frac{1}{8}\right)}$; $R_{Den_{norm}}: R_{Den} * \frac{d}{c}$ (6) Step 2: Determine the proportional factor for NO_3^- $f_{prop_{D-N}}: \frac{\frac{\frac{1}{8}}{\left(\frac{f_{s_{Nit}}}{20} + \frac{1}{8}\right)}}{\left(\frac{f_{e_{Den}}}{5} + \frac{f_{s_{Den}}}{28}\right) * \frac{d}{c}}$ (7) Step 3: Include the proportional factor in the denitrification process $R_{Den_{prop}}: R_{Den_{norm}} * f_{prop_{D-N}}$ (8) Step 4: Summation of nitrification and denitrification, and normalize to one mole of ammonium $R_{D-N}: \left[R_{Den_{prop}} + R_{Nit_{norm}}\right] * \frac{1}{\left(1 - f_{prop_{D-N}}\right)}$ (9)
Anaerobic digestion (as developed by Rittman and McCarty, 2001)	$R_{Meth}: C_nH_aO_bN_c + \left(2n + c - b - \frac{9df_{s_{Meth}}}{20} - \frac{df_{e_{Meth}}}{4}\right)H_2O \rightarrow \frac{df_{e_{Meth}}}{8}CH_4 + \left(n - c - \frac{df_{s_{Meth}}}{5} - \frac{df_{e_{Meth}}}{8}\right)CO_2 + \frac{df_{s_{Meth}}}{20}C_5H_7O_2N + \left(c - \frac{df_{s_{Meth}}}{20}\right)NH_4^+ + \left(c - \frac{df_{s_{Meth}}}{20}\right)HCO_3^-$ (10)
Partial nitritation	$R_{Pn}: \left(\frac{f_{s_{Pn}}}{20} + \frac{1}{6}\right)NH_4^+ + \frac{f_{e_{Pn}}}{4}O_2 + \frac{f_{s_{Pn}}}{5}CO_2 + \frac{f_{s_{Pn}}}{20}HCO_3^- \rightarrow \frac{f_{s_{Pn}}}{20}C_5H_7O_2N + \frac{1}{6}NO_2^- + \left(\frac{4}{3} - f_{e_{Pn}} - f_{s_{Pn}}\right)H^+ + \left(\frac{f_{e_{Pn}}}{2} + \frac{9f_{s_{Pn}}}{20} - \frac{1}{3}\right)H_2O$ (11)
Anammox	$R_{Anx}: \left(\frac{f_{e_{Anx}}}{3} + \frac{f_{s_{Anx}}}{20}\right)NH_4^+ + \left(\frac{f_{e_{Anx}}}{3} + \frac{f_{s_{Anx}}}{2}\right)NO_2^- + \frac{f_{s_{Anx}}}{5}CO_2 + \frac{f_{s_{Anx}}}{20}HCO_3^- \rightarrow \frac{f_{s_{Anx}}}{20}C_5H_7O_2N + \frac{f_{s_{Anx}}}{2}NO_3^- + \frac{f_{e_{Anx}}}{3}N_2 + \left(\frac{2f_{e_{Anx}}}{3} - \frac{f_{s_{Anx}}}{20}\right)H_2O$ (12)

Proportional partial nitritation/anammox	Step 1: Normalization to one mole of ammonium $R_{Pn_{norm}}: R_{Pn} * \frac{1}{\frac{f_{s_{Pn}}}{20}+\frac{1}{6}} \quad ; \quad R_{Anx_{norm}}: R_{Anx} * \frac{1}{\frac{f_{e_{Anx}}}{3}+\frac{f_{s_{Anx}}}{20}}$ (13) Step 2: Determine the proportional factor for NO_2^- $f_{prop_{F-MBR}}: \frac{\left(\frac{\frac{f_{e_{Anx}}}{3}+\frac{f_{s_{Anx}}}{2}}{\frac{f_{e_{Anx}}}{3}+\frac{f_{s_{Anx}}}{20}}\right)}{\left(\frac{\frac{1}{6}}{\frac{f_{s_{Pn}}}{20}+\frac{1}{6}}\right)}$ (14) Step 3: Include the proportional factor in the partial nitritation process $R_{Pn_{prop}}: R_{Pn_{norm}} * f_{prop_{F-MBR}}$ (15) Step 4: Summation of partial nitritation and Anammox, and normalize to one mole of ammonium $R_{F-MBR}: \left[R_{Pn_{prop}} + R_{Anx_{norm}}\right] * \frac{1}{\left(1+f_{prop_{F-MBR}}\right)}$ 16)

The approach to calculating the overall biochemical formulas is explained in detail in the Supporting Method (SM) S1. The overall biochemical reactions include the kinetic parameters that can be substituted with the typical values as discussed in the literature (Table 2).

Table 2. Typical kinetic values for different biological processes

Parameter	**Organic oxidation (Ox)**	**Nitrification (Nit)**	**Denitrification (Den)**	**Partial nitritation (Pn)**	**Anammox (Anx)**	**Methanogenesis (Meth)**
f_s^0	0.6[62]	0.127[62]	0.52[62]	0.065[69]	0.080[67]	0.11[62]
f_d[62]	0.8	0.8	0.8	0.8	0.8	0.8
b	0.15[62]	0.11[62]	0.04[68]	0.15[62]	0.05[67]	0.05[62]

Notes: f_s^0 is the portion of energy used for cell synthesis; f_d is the fraction of the active biodegradable biomass; b is the endogenous decay rate.

The combustion of biogas for energy production by combined heat and power (CHP), and dual fuel engines (DFE) was calculated using the overall biogas combustion equation:[70]

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \qquad (17)$$

Analysis of the link between the microscopic and macroscopic levels for engineering applications comprised of using the calculated moles (n_{RX}) of the reactants and products to obtain the daily concentrations as inputs and outputs of the system. In general, the flows of reactants, organics, or ammonium treated were determined by their concentrations at the influent point and the removal efficiency of the process (as stated by the current STW operation or legislation). The process products, namely the outputs (required as the foreground information in the LCI), were calculated as shown in Eq. (18) based on Rittmann and McCarty[62] and Bisinella de Faria et al.[37]:

$$M_i = M_{RX} * (MW_i * n_i)/(MW_{RX} * n_{RX}), \qquad (18)$$

where

M_i is the mass of product i (kg/d),

M_{RX} is the mass flow of the organics or inorganics (reactants) to be treated, which includes their concentrations (mg/L) times and flow (L/d),

MW_i is the molecular weight of product i (g/mol),

n_i is the number of moles of product i obtained from the overall biochemical equation (mol),

MW_{RX} is the molecular weight of the reactants treated (g/mol), and

n_{RX} is the number of moles of the reactants obtained from the overall biochemical equation (mol).

In the case of gas production, the volume was calculated based on the ideal gas equation. The S-LCI process inventories contain the inputs and outputs of the water effluent quality, solids generation, gas production, energy consumption, and production. The detailed assumptions and justifications of the S-LCI are explained in SM S2.

2.3 Results

To increase the S-LCI standardization, the S-LCI uses the existing “Wastewater, average {RoW}| treatment of, capacity #L/year| Alloc Def, U” inventory from Ecoinvent v.3.2 as a template, where the #L/year (number of liters of sewage treated per year) vary for different classes of STW. The Ecoinvent database has five classes of STW that represent five different annual flows. Apart from Switzerland, the rest of the countries may only collect the missing data from the RoW template in the Ecoinvent database. This database was chosen over other options because of its comprehensive coverage.[71] To select the STW class, the flow per year calculations were made based on the Ecoinvent database.

The Ecoinvent inventory comprises of seven categories that include the concept, amount, unit, distribution, and standard deviation. First, the concepts are taken directly from the RoW template. The Ecoinvent template requires the highest level of inventory data according to the collection scheme by Yoshida et al.[18] However, some new concepts were added into the S-LCI from biogas combustion and incineration, transport of sludge and incineration products, the specific infrastructure, and the dissolved CH_4 in the water effluent. Second, the amounts correspond to the stoichiometric results which are in kg/d, m^3/d or kWh/d. Therefore, conversion factors and consideration of the influent flow are included to fulfill the FU. Third, the units represent the FU for each concept. Fourth, the distribution is assumed to be a log-normal based on the current Ecoinvent database. Fifth, the standard deviation is calculated by following the Pedigree Matrix, given that the uncertainty estimations are unknown as explained in detail in SM S3.[18,72]

The “S-LCI” file in “Supplementary information” already has a built-in Pedigree Matrix. The values 1–5 must be updated for each case study. The standard deviations for other concepts in the template are taken from the Ecoinvent v.3.2 values.[73] To construct the LCI, the results of the process inventories are closely related to the LCA assumptions as explained in detail in SM S4.

2.4 Performance measurement analysis

The performance of the S-LCI is evaluated by comparing the estimated data with the originally collected data using the mean percentage error (MPE) as performed by Hou et al.[71]. In addition, the S-LCI framework is compared to an inventory calculated with current DC practices. LCIA and interpretation are the third and fourth steps of the LCA, thus out of the scope of this study. Nevertheless, the inventories are compared by using impact assessment methodologies (IAM) to illustrate the methodology and the possible results that can be achieved. ReCiPe Endpoint and CML 2 baseline 2000 were the IAM evaluated in SimaPro 8.

3. RESULTS AND DISCUSSION

The S-LCI was tested with two different STW configurations: System 1 represents a conventional treatment and system 2 represents an EST system.

3.1 System 1: conventional treatment

System 1 is a conventional treatment that includes the AS, AD, CHP, and DFE for energy production. This configuration is the same as the Shatin STW, which is the second-biggest secondary treatment STW in Hong Kong. The effluent from Shatin STW is transported to Victoria Harbor. The sludge generated is sent to the T-Park Sludge Treatment Facility.[74] The sludge management method that includes thickening, AD, dewatering, and incineration has been found to have the best environmental and economic performance in China.[75]

3.1.1 DC

For the elemental analysis, the sewage and sludge sample collection technique involved using plastic containers for manual single-grab sampling from different points of the Tai Po STW that contained saline sewage treated by the same processes as at Shatin STW. Within 2 h of collection, approximately 2 L of samples were stored in a refrigerator at 4°C. Evaporating crucibles were prepared and weighed. The samples were mixed and added to the crucibles to dry at 105°C until 5 mg of dried sample were yielded. The dry samples were pulverized with a mortar and pestle. The samples were prepared to pass a 250-µm (no. 60) sieve size and were taken out of the oven right before the elemental analysis, which was performed with vario MICRO cube (Elementar).[66] Listed in Table 3, the results are within the ranges of CHNS of other STW sludge.[76] In another crucible, 1 g of sample was added and placed in a cold furnace. The temperature was increased gradually to 500°C in 1 h and then increased to 750°C for another 1 h, which was maintained for 2 h. The crucibles were then cooled in a 105°C oven. Lastly, the crucibles were placed in a desiccator until room temperature was reached, whereupon they were weighed.

Table 3. Results of elemental analysis for grab samples from Tai Po sewage treatment works

Sample	C (%)	H (%)	N (%)	S (%)	O (%)*	Ash (%)
Effluent primary clarifier	1.2	2.4	0.1	1.8	18.5	76
Primary sludge	29.8	5.4	2.2	0.9	20.9	40.8
Thickened activated sludge	40.1	6.6	8.3	1.2	26.4	17.4

Note**:** Average for each sample from duplicates on a dry basis. * Calculated by the difference.

In the present study, the PD was collected from Shatin STW. Figure 2 shows the specific data collected from the Drainage Services Department (DSD).[77] The 41-item checklist is given in Table S3. Additional data collected to validate the S-LCI are shown in Table S4.

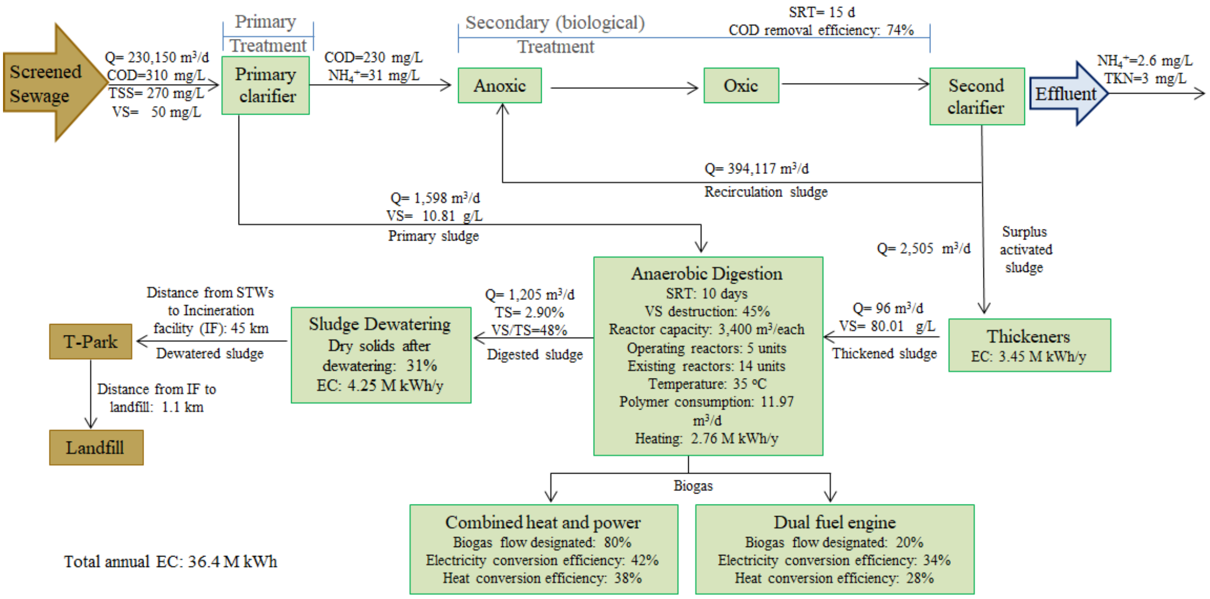

Fig. 2. Primary data (PD) from Shatin STW provided by the Drainage Services Department.[77] Notes: Q = flow rate; COD = chemical oxygen demand; TS = total solids; VS = volatile solids; NH_4^+ = ammonium; SRT = solids retention time; TKN = total Kjeldahl nitrogen; EC = electricity consumption.

Moreover, the infrastructure inventory for major materials was counted manually in 560 blueprints from a conventional STW in Hong Kong. The current land-use concept in the Ecoinvent database template was obtained by quantifying the areas of three plants in Switzerland.[78] Thus, three plants, namely the Shatin STW, the Shek Wu Hui STW, and the Stonecutters Island (SCI) STW were quantified to regionalize the inventory to Hong Kong as given in Tables S5–S8.

In Hong Kong, 7,347,900 inhabitants generated 1,048 million cubic meters of wastewater in 2016,[79] revealing that they generated 143 cubic meters of wastewater per capita equivalent (PCE) annually. The capacity classification chosen was similar to the Swiss wastewater-treatment-plant classification to make an effective comparison. The biochemical oxygen demand (BOD_5) concentrations were obtained from the chemical oxygen demand (COD) concentrations of the three main STWs in Hong Kong that represent 69% of the total annual flow of sewage treatment. The BOD_5 PCE for Hong Kong was 0.04 kg/inhab/day as given in Tables S9 and S10. In Hong Kong, many of the plants are either larger plants (class 1) or small-scale plants (class 5). The average capacity per plant for each class was calculated for Hong

Kong, with the results shown in Table 4. The infrastructure units per sewage cubic meter were calculated based on Doka.[59] These data can be used in other Asian cities.

Table 4. Classification, average capacity, and plant infrastructure for life-cycle inventory in Hong Kong

Capacity class	**Unit**	**1**	**2**	**3**	**4**	**5**
Capacity range	PCE/a	over 100,000	40,000 to 100,000*	10,000 to 40,000*	2,000 to 10,000	10 to 2,000
Number of plants	unit	10	1	4	6	28
Total treatment capacity	PCE/a	12,414,722	43,278	100,561	17,912	18,648
Average capacity per plant in class	PCE/a*unit	1,241,472	43,278	25,140	2,985	666
Annual sewage volume	m^3/a	177,066,997	6,172,569	3,585,662	425,793	94,989
Lifetime plant	A	30	30	30	30	30
Lifetime sewage volume	m^3	5,312,009,922	185,177,067	107,569,873	12,773,788	2,849,669
Plant infrastructure	$unit/m^3$	$1.882E^{-10}$	$5.400E^{-09}$	$9.296E^{-09}$	$7.828E^{-08}$	$3.509E^{-07}$
Comparison to Ecoinvent values[59]	$unit/m^3$	$7.075E^{-10}$	$2.320E^{-09}$	$6.637E^{-09}$	$3.101\ E^{-08}$	$2.047E^{-07}$

Notes: * In the Ecoinvent database for Switzerland, STW in capacity class 2 has a capacity range of 50,000 to 100,000 PCE/a, and those in capacity class 3 have a range of 10,000 to 50,000 PCE/a.

Shatin STW belongs to class 1 with a capacity of 1,045,880 PCE/a, an annual sewage volume of 149,170,415 m^3/a, and a plant infrastructure value of $2.234E^{-10}$ (Table S11). The materials inventory for the construction concept was based on the Shek Wu Hui STW (Tables S12–S14) with some modifications (Table S15). For energy requirements, an extract of the energy-related concepts from the 41-item checklist was complemented with the literature as listed in Table 5.

Table 5. Energy requirements for system 1 (conventional system) based on Shatin STW

Concept	Value	Unit
Total annual electricity consumption (EC)[80]	36.4	MkWh/y
Sludge treatment EC[80]	10.5	MkWh/y
Waste activated sludge thickening EC [80]	3.45	MkWh/y
Digester heating[80]	2.76	MkWh/y
Sludge dewatering[80]	4.25	MkWh/y
Heat and power units (including CHP and DFE)[77]	2	unit

Aeration electricity consumption from the total*	60	%
Incineration electricity efficiency[81]	20	%

Notes: *Assumed from the difference between the annual electricity consumption and sludge-management electricity consumption.

3.1.2 Calculations

The design criteria for the stoichiometric calculations were obtained from the literature and the real operation of the Shatin STW as given in Table 6. The key parameters based on the S-LCI assumptions are summarized in Table S16. The detailed calculations are given in the "S-LCI" Excel file as supporting information.

Table 6. Design criteria for different biological processes

Parameter	Organic oxidation (Ox)	Nitrification (Nit)	Denitrification (Den)	Partial nitritation (Pn)	Anammox (Anx)	Methanogenesis (Meth)
θ_x	15	15	15	20[69]	20[67,69]	10[82*]
f_s	0.268	0.064	0.364	0.026	0.048	0.081
f_e	0.732	0.936	0.636	0.974	0.952	0.919

Notes: θ_x is the solids retention time; f_s is the electron portion used for cell synthesis considering net yield; f_e is the electron portion for energy generation considering net yield. * 20 d considered for NO_3^- formation around 4% in AFBR[69], and 7 d is the hydraulic retention time for CEPT sludge.[83]

3.1.3 Results

The complete S-LCI inventory for the Shatin STW is given in Table S17.

3.2 System 2: EST system

System 2 represents an EST process that includes CEPT followed by a staged fluidized membrane bioreactor (SF-MBR) for the water stream. The solid stream includes the same sludge treatment and energy production technologies as in the conventional system. The SF-MBR involves two reactors.[84] The first reactor is an anaerobic fluidized-bed bioreactor (AFBR), followed by a partial nitritation/anammox fluidized-bed membrane bioreactor (PN/A-FMBR).

3.2.1 DC

The results of the elemental analysis from the primary clarifier effluent were assumed to be the same as the influent for the SF-MBR. However, for the AD process, the elemental analysis values were taken from Shao et al.[85]

The PD was collected from the SCI STW, which is a CEPT plant that treats ~ 1.7 million cubic meters of municipal sewage per day in Hong Kong and complemented with the values from the literature as shown in Table 7.

Table 7. PD collected for system 2 (energy-derived sewage treatment system)

Concept	Value
Staged fluidized membrane bioreactor (SF-MBR) considerations	
- Bulk wasting ratio (%) [86]	1
- Biosolids production (g VSS/ g COD removed) [86]	0.026
- Biofilm in granulated activated carbon (GAC) (mg VSS/L) [86]	704
- Chemical oxygen demand (COD) converted to dissolved CH_4 (%) [86]	15
Chemically enhanced primary treatment (CEPT) considerations	
- Iron chloride consumption (m^3/d) [74]	29.4
Removal efficiencies (%)	
Water stream:	
- CEPT COD removal [87]	62.55
- CEPT total suspended solids (TSS) removal [87]	87.18
- CEPT volatile suspended solids (VSS) removal [87]	87.50
- Anaerobic fluidized-bed bioreactor (AFBR) COD removal [86, 88, 89]	46
- Anaerobic fluidized-bed membrane bioreactor (AFMBR) COD removal [86, 88, 89]	48
- AFMBR total nitrogen removal [69]	94.4
Solids retention times (d)	
- SF-MBR [69]	20
Anaerobic digestion	
- Total solids in raw saline CEPT sludge (%) [*]	3.87
- TSS for raw saline CEPT sludge (g/L) [*]	26.08
- Volatile Solids (VS) for raw saline CEPT sludge (%) [*]	17.09
- CEPT saline VS destruction (%) [83]	61
- Digested CEPT sludge VS (%) [*]	75.32
- Digested CEPT sludge TS (%) [*]	2.58
- Polymer consumption (kg/d) [74]	899

Notes [*] Based on a laboratory-scale anaerobic digester as explained in SM S5.

The average capacity for an SF-MBR was obtained from the literature as given in Table S18. The bioreactors tested by Shin et al.[86,90] were assumed to be in the higher class 1 capacity, whereas the bioreactors by Wu et al.[88] and Kim et al.[91] were assumed to be in the smallest capacity class 5.

Other materials were included from the literature to account for the SF-MBR construction.[86,88,90,91] The membrane quantification from the literature is given in Table S19. The granulated activated carbon (GAC) is given in Table S20. The estimated chemical requirement for cleaning the membranes is given in Table S21. The total weight per system considered for the LCI was an average between the capacity classes 1 and 5. The materials inventory for constructing the EST is given in Table S22.

Three values of raw CEPT sludge samples from SCI STW were obtained by experiments to determine the TS, TSS, and VS. The sludge samples are the manual sampling of the single-grab samples in plastic containers at the SCI STW. The samples were collected biweekly from June 2016 to March 2017. Evaporating crucibles were prepared and weighed. These tests are explained in detail in SM S6. The VS of digested CEPT sludge was obtained from an anaerobic digester fed with the CEPT sludge from the SCI STW and seeded with anaerobic sludge from the Tai Po STW.[92] The anaerobic digester is described briefly in SM S5.

The energy requirements were postulated according to several assumptions. It was assumed that the value of AFBR electricity consumption for GAC fluidization and recirculation was 0.016 kWh/m^3.[86] The AFMBR electricity consumption for GAC fluidization, recirculation, and permeate was 0.211 kWh/m^3.[86] The microaeration energy consumption was omitted.

3.2.2 Calculations

The key parameters based on the S-LCI assumptions are summarized in SM S2 and S4. The detailed calculations are given in the “S-LCI” Excel file with the supporting information. The EST STW configuration, main design criteria, and removal efficiencies are shown in Fig. 3.

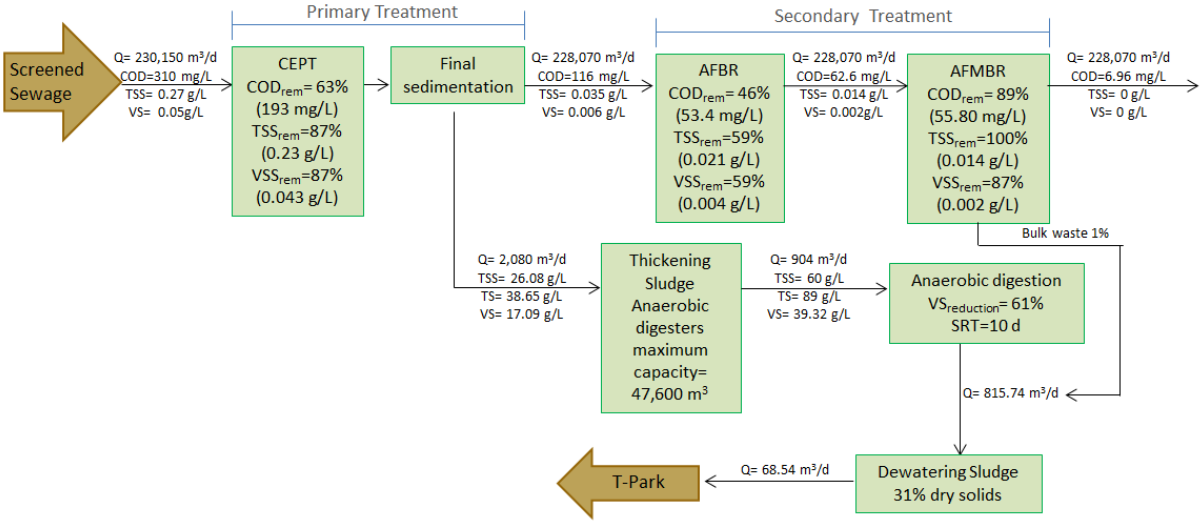

Fig. 3. Energy-derived from sewage treatment configuration, main design criteria, and removal efficiencies. Notes: Q = flow rate; COD = chemical oxygen demand; TSS = total suspended solids; TS = total solids; VS = volatile solids; CODrem = COD removal efficiency; TSS_{rem} = TSS removal efficiency; VS_{rem} = VS removal efficiency; OLR = organic loading rate.

3.2.3 Results

The complete S-LCI inventory in the EST system is given in Table S23.

3.3 Performance measurement analysis

The results for the conventional systems were compared between the actual operation of the Shatin STW and system 1. In addition, systems 1 and 2 were compared to evaluate the performance of the EST system. The comparisons were based on the water effluent quality, solids generation, energy production, and GHG emissions as given in Table 8. The MPE was used to evaluate the accuracy of the results from the S-LCI and the actual data from the Shatin STW. The step-by-step calculation of the MPE is presented in Table S24.

Table 8. Comparison by mean percentage error (MPE) of the S-LCI of system 1 (conventional treatment) to the actual performance of the Shatin STW, and the S-LCI results of system 2 (energy-derived sewage treatment system).

Water stream Concept	Shatin STW data	System 1	MPE (%)	System 2
Oxygen consumption (ton/d)	-	76.81	NA	4.46
COD concentration in the effluent (mg/L)	<59.00[77]	45.90	0	6.97
COD in the effluent (ton/d)	<13.58	10.56	0	0.79
NH_4^+ concentration in the effluent (mg/L)	3.10	2.59	17	1.75
NH_4^+ in the effluent (ton/d)	0.71	0.60	16	0.20
NO_3^- concentration in the effluent (mg/L)	0.40	0.40	0	2.72
NO_3^- in the effluent (ton/d)	0.092	0.09	0	0.31
Solids stream with energy generation	Shatin STW	System 1	MPE (%)	System 2

Concept	data			
Sludge production (m^3/d)	117.8	99.46	16	68.54
Biogas production (m^3/d)	10,410.96	9,810.32	6	17,838.50
Methane production (m^3/d)	6,246.58	6,526.06	4	10,148.57
CHP:				
Electricity recovery with an efficiency of 42% (kWh/d)	26,381.43	23,396.69	11	35,881.51
Heat recovery with an efficiency of 38% (kWh/d)	-	21,168.44	NA	32,464.22
DFE:				
Electricity recovery with an efficiency of 34% (kWh/d)	5,856.22	4,735.05	19	7,261.73
Heat recovery with an efficiency of 28% (kWh/d)	-	3,899.45	NA	5,980.25
GHG emissions (ton/d) Concept	Shatin STW data	System 1	MPE (%)	System 2
Biogenic CO_2 emissions	-	79.72	NA	17.27
Biogenic CH_4 emissions	-	0.041	NA	0.062
Direct CH_4 emissions	-	0.26	NA	0.04
Direct N_2O		0.02	NA	0.01

Notes: - No information was provided. NA: not applicable.

Overall the results obtained for the S-LCI for system 1 are in agreement with the reported data of the actual performance of the Shatin STWs, supporting the specificity of the S-LCI approach developed in this work. In terms of water effluent quality, the similarity of S-LCI to actual performance was 84% (MPE=16%) for NH_4^+ concentration and 100% (MPE=0%) for COD. Systems 1 and 2 had close results for NH_4^+ removal. System 1 considered total Kjeldahl nitrogen (TKN) removal efficiency of 93%, whereas the total NH_4^+ removal efficiency of the SF-MBR from system 2 was assumed as 94.4%.

Solids generation is closely related to biogas and energy production. The S-LCI results from system 1 were similar to the current operation of the Shatin STW of 84% (MPE= 16%) for sludge generation, 94% (MPE=6%) for biogas production, 96% (MPE=4%) for CH_4 production, 89% (MPE= 11%) for electricity generation from CHP, and 81% (MPE=19%) for electricity generation from one DFE. The differences in electricity generation may be due to the dynamic operational conditions for the ratio of biogas between CHP and DFE. In the actual operation of the Shatin STW, the biogas flow ratio of CHP and DFE changes constantly, whereas S-LCI assumes a constant ratio (80:20). System 2 had lower sludge production than the conventional system, with more biogas generated and thus more energy recovered.

The benefits of calculating the inventory with the S-LCI framework are demonstrated through its comparison with the conventional approach, which is the use of the database with available disclosed data. The data available from the DSD for Shatin STW only included the electricity production on-site; iron consumption; sludge production and distance from the Shatin STW to the incineration plant; polyacrylamide consumption; number of co-generation units, and COD and NH_4^+ in the water effluent. No data could be gathered for GHG emissions. Details of the inventories are shown in Table S25. The results of the IAM show that the conventional DC methods translated into an overestimation of the environmental impacts compared to the site-specific S-LCI as shown in detail in Figure S1.

3.4 Water–energy–gas implications

The present study included the CO_2 and CH_4 biogenic and direct emissions, and N_2O emissions. According to the IPCC, the CO_2 biogenic gas emissions are recommended as GHG-neutral[93] and have been omitted in other studies.[19,32,37,94,95] The S-LCI results show that system 2 had the lowest performance solely in CH_4 biogenic emissions. The main CH_4 biogenic emissions were due to the assumed leakage from the anaerobic digesters. These emissions are highly related to the higher sludge production of system 2 from CEPT. The S-LCI also considered the direct CH_4 produced from the untreated COD released into the environment.[97] System 1 had a higher COD concentration in the water effluent, meaning that there were more direct CH_4 emissions from system 1 than from system 2. Unlike the biogenic emissions, the direct CH_4 emissions were closely related to the water effluent quality. The N_2O emissions were estimated from the literature.[78] These emissions have been recognized as essential to the calculation of GHG emissions because of the high global-warming potential of N_2O.[11,29] Moreover, other studies have noted N_2O emissions' importance when introducing anammox to the AS treatment.[32,97] In this study, denitrification was not considered to be taking place in the SF-MBR as shown by the proportional partial nitritation/anammox biochemical reaction (Eq. (16)). The SF-MBR is an enclosed system, therefore the operational conditions could be optimized through the control strategies. In the case of N_2O production, only 1% of the total production would be assumed to be in a leakage form, which would be significantly lower than the current N_2O emissions from the AS.[93,98]

It has been recognized that advanced treatments for better effluent quality lead to higher energy consumption.[2,3,7,99,100] Therefore, the trade-offs between energy consumption and contaminant removal must be analyzed carefully. Contrary to the results of other studies on membrane bioreactors where higher water quality and energy recovery imply higher energy consumption,[7,99] the results from the S-LCI for system 2 show that the higher energy production from AD, optimized AD operation, and similar energy production from incineration in systems and 1 and 2 compensated for the high energy consumption for membrane fouling control. This finding is similar to the results for anaerobic fluidized-bed membrane bioreactor (AFMBR) systems that had better energy performance than the activated sludge process,[100] and AFMBR systems that recovered more energy than high-rate activated sludge coupled with AD.[44] In this study, the plant-wide low-voltage electricity consumption decreased from 0.4004 kWh/m^3 (system 1) to 0.3298 kWh/m^3 (system 2).

3.5 Implications for LCA

S-LCI is a tool that links biochemistry and management in a user-friendly way that can help the decision-making process. Four main implications were identified for LCA research and application. First, existing LCA approaches involve high DC variability. There is no standard checklist for DC for foreground information of the existing processes. S-LCI includes a 41-item checklist of data requirements. The legislative/water authority could include parameters from the checklist into the monitoring programs to be disclosed by law. Furthermore, S-LCI provides different ranges for the variable parameters, which could facilitate sensitivity analysis.

Second, there is no standard process for estimating or predicting foreground information with higher specificity. A major original aspect of the S-LCI is its use of stoichiometry coupled with kinetics to obtain the water, air, and soil emissions, which increases the specificity of the LCI and enables its replication for similar STW configurations for any other sewage sample. S-LCI encourages using elemental analysis of the sample of interest to construct the specific empirical microbial formula for increasing the representativeness and specificity for the local conditions. Despite increased uncertainty from the use of grab samples, this approach is more specific than using current databases that involve global averages.

Third, a full set of infrastructure inventories, including EST systems, was calculated for Hong Kong and which contribute to the "wastewater treatment facility" concept in the Ecoinvent database for Asian cities. The listed inputs/outputs in the EST inventory in the present study could be used as default values for other studies that focus on inputs/outputs in which sample composition has no influence. The wastewater facility construction concept for Ecoinvent class 1 and systems 1 and 2 are compared in Table S26.

Lastly, foreground information for the ST LCI can be extended from the S-LCI beyond RoW and Switzerland to other countries. Adding data analysis of the recently developed processes contributes to new unit processes, such as CEPT, AFBR, and PN/AFMBR. A considerable amount of biological data has been collected for the recently developed processes. Thus, fewer parameters need to be calculated or tested to reduce the costs and time for DC. The S-LCI can be applied by pioneers of the emerging-technologies who might not yet understand all the kinetic breakdowns of the processes as required in state-of-the-art biochemical models (hydrolysis rates and inhibition effects) but whose technologies must be compared with the conventional systems to determine whether the trade-offs in the systems have good environmental performance.[101]

3.6 Limitations

Although adopting the S-LCI could extend the databases' inventories for sewage treatment with enhanced specificity and standardization, several limitations should be addressed before it is applied in practice. First, there are two main sources of uncertainty, namely (i) data collected from the laboratory and pilot-scale studies for the EST system, and (ii) the steady-state mass balance compared to a dynamic approach.

Second, the S-LCI could be considered to be over-simplified compared to state-of-the-art models, such as BSM2. Yet S-LCI requires DC for 41 items. In addition, between the conventional and the EST systems combined, the S-LCI encompasses 69 variable parameters for water, solids, energy generation, and gas emissions considerations. Even though the DC proposed is extensive, the acceptable ranges and experimental results are already provided in a clear way.

Third, the trade-offs involved in removing emergent pollutants and heavy metals from water to sewage sludge must be studied in further detail.[7,11,18] CEPT tends to have higher heavy-metal removal than conventional clarifiers. In particular, scenarios that involve spreading sludge for land application or agriculture must include the fate of heavy metals into the soil.[2] The optimization of the membrane reactors for energy consumption,[27,100] and strategies for dissolved methane recovery[44,50,100] should also be included when data becomes available.

Despite these limitations, S-LCI provides a clear step toward more specific and regionalized LCA. In addition, the data of recently-developed systems in EST are not from full-scale systems, but their inclusion in a whole-plant analysis helps to identify the water-energy-gas trade-offs to tackle before scaling up.

3.7 Future work

Further studies based on S-LCI are expected to reduce uncertainty, increase robustness, and improve decision-making. To reduce uncertainty, new developments regarding the mainstream full-scale application of SF-MBR and the integration of emerging pollutants and heavy metals should be included. S-LCI represents a relatively generic methodology yet other system configurations should be conducted case by case. The S-LCI robustness could be enhanced by adding the biochemical equations of other emerging sewage treatment processes. In addition, the results of the S-LCI should be evaluated by IAM and interpretation methods to provide a clearer picture of the environmental impacts of the different systems. Instead of initially spending time and money on laboratory and pilot-scale studies for each of the processes for each specific sample, further studies and more advanced models could be assessed after the environmental impacts are identified.

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5. SUPPORTING INFORMATION

The “Supporting information” file contains detailed information on the stoichiometric calculations, the assumptions for the stoichiometric life-cycle inventory (S-LCI), the information relevant to the systems analyzed, and the mean percentage error calculations.

The “S-LCI” file is an Excel tool developed to generate life-cycle inventories from stoichiometric calculations (XLSX). The tool is ready to be used with different samples.

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