

Multi-objective performance assessment of wastewater treatment plants combining plant-wide process models and life cycle assessment

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ABSTRACT

Multi-objective performance assessment of operational strategies at wastewater treatment plants (WWTPs) is a challenging task. The holistic perspective applied to evaluation of modern WWTPs, including not only effluent quality but also resource efficiency and recovery, global environmental impact and operational cost calls for assessment methods including both on- and off-site effects. In this study, a method combining dynamic process models – including greenhouse gas (GHG), detailed energy models and operational cost – and life cycle assessment (LCA) was developed. The method was applied and calibrated to a large Swedish WWTP. In a performance assessment study, changing the operational strategy to chemically enhanced primary treatment was evaluated. The results show that the primary objectives, to enhance bio-methane production and reduce GHG emissions were reached. Bio-methane production increased by 14% and the global warming potential decreased by 28%. However, due to increased consumption of chemicals, the operational cost increased by 87% and the LCA revealed that the abiotic depletion of elements and fossil resources increased by 77 and 305%, respectively. The results emphasize the importance of using plant-wide mechanistic models and life cycle analysis to capture both the dynamics of the plant and the potential environmental impacts.

Key words | LCA, mathematical modelling, performance assessment, process control, wastewater treatment

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NOMENCLATURE

AD	Anaerobic digester	CEPT	Chemically enhanced primary treatment
ADM1	Anaerobic Digestion Model No. 1	CML	Centrum voor Milieukunde, Leiden University, The Netherlands
ADP	Abiotic depletion potential	COD	Chemical oxygen demand [mg·l ⁻¹]
ANOX	Anoxic model reactors	DEOX	Non-aerated deox model reactor
AOB	Ammonia oxidizing bacteria	DO	Dissolved oxygen [mg·l ⁻¹]
ASU	Activated sludge unit	DW	Dewatering unit
ASM1	Activated Sludge Model No. 1	EQI	Effluent quality index
ASM1G	Activated Sludge Model No. 1 Greenhouse Gas	EU28	The 28 member states of the European Union
BSM	Benchmark Simulation Model	FLEX	Flexible, aerated or non-aerated model reactor
BSM2G	Benchmark Simulation Model No. 2 Greenhouse Gas	GHG	Greenhouse gas

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GU	Gas upgrade unit
GWP	Global warming potential
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
ISS	Inert suspended solids [$\text{mg}\cdot\text{l}^{-1}$]
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
OCI	Operational cost index
ODP	Ozone depletion potential
OX	Aerated model reactors
PRIM	Primary mechanical treatment
RAS	Return activated sludge
SF	Sand filter
STOR	Sludge storage
THK	Thickener unit
TN	Total nitrogen [$\text{mg N}\cdot\text{l}^{-1}$]
TSS	Total suspended solids [$\text{mg}\cdot\text{l}^{-1}$]
VS	Volatile solids [$\text{mg}\cdot\text{l}^{-1}$]
WWT	Wastewater treatment
WWTP	Wastewater treatment plant

INTRODUCTION

The holistic view applied to modern wastewater treatment plants (WWTPs) challenges the traditional methods for performance evaluation. Today, not only effluent water quality and operational cost, but also energy efficiency, resource recovery rate and global environmental impact (e.g., on climate) need to be considered when assessing plant performance (Olsson 2015). It is well known that WWTPs can emit substantial amounts of greenhouse gases (GHGs). Apart from the mostly biotic CO_2 emissions from degraded organic matter, the main direct emissions from WWTPs are typically CH_4 from the sludge treatment and N_2O from the secondary biological treatment (Foley *et al.* 2011; Mannina *et al.* 2016). Foley *et al.* (2011) also showed that the variations especially for the important N_2O emissions are large and dependent of process configuration and operational strategy. For over a decade there has been a strong focus on energy efficiency and recovery at WWTPs to reduce both costs and GHG emissions (Olsson

2015). Even if this is a commendable intention it has been shown that energy saving at the same time can lead to an overall increase in GHG emissions, for example increased N_2O emissions (Flores-Alsina *et al.* 2014). Altogether, this calls for better modelling tools to evaluate and compare treatment strategies. Mechanistic process models have been used for several decades to assess treatment efficiency, residual environmental load and operational costs for WWTPs. They have proven to be valuable tools for everything from green field design of new plants to design and evaluation of detailed control strategies (Rieger *et al.* 2012).

However, for the global environmental impact, not only the direct emissions to water, land and air of different pollutants from the plant are relevant, but also the up and downstream processes need to be taken into account. That includes external processes, such as production of input goods like power and chemicals, but also impacts of the effluent load in the recipient and utilization of bio-solids and bio-methane. These types of effects have successfully been assessed for WWTPs using life cycle assessment (LCA) (Corominas *et al.* 2013a, 2013b; Baresel *et al.* 2016; Morera *et al.* 2016; Svanström *et al.* 2016). LCA evaluates the potential environmental impact due to the product or service under study characterized in different categories (Baumann & Tillman 2004). The ISO standard for LCA (ISO 14044) gives a structured procedure for performing the LCA where, after defining the study, a life cycle inventory (LCI) is done where the environmental loads from the whole system are calculated followed by a life cycle impact assessment (LCIA) where the loads are characterized by the selected impact categories to get aggregated measures of the potential environmental impacts expressed in equivalent units (Baumann & Tillman 2004).

To capture both the dynamic performance of the WWTP and the global environmental impact, the combination of mechanistic process models and LCA has been explored on generic benchmark type WWTP layouts (Corominas *et al.* 2013b; Bisinella de Faria *et al.* 2015; Meneses *et al.* 2016). These studies use combinations of different WWTP models and LCA to assess control strategies, load variations and local recipient conditions. However, these studies are limited in their coverage of the WWTP; either only the water line is modelled mechanistically (Corominas *et al.* 2013b; Meneses *et al.* 2016) or, as in Bisinella de Faria *et al.*

(2015), the highly dynamic GHG production is modelled using static emission factors. Thereby, important aspects of the analysis may be lost. Furthermore, none of these studies apply the method to real plants. Another area of interest when analysing operational strategies is operational cost. The mechanistic models enable operational cost estimates (Gernaey et al. 2014). However, in existing studies where process models are combined with LCA, operational cost is either not included at all or not extended to include major cost items of relevance, e.g., precipitation chemicals.

This paper explores the hypothesis that it is possible to combine a plant-wide WWTP process model – with detailed energy, cost and GHG models – and a LCA study to evaluate the overall performance of operational strategies at WWTPs, capturing both the dynamic effects at the plant, the global environmental impact due to external resource use and operational cost. A model framework is presented and tested in a case study performed at a full-scale Swedish WWTP comparing two operational strategies.

METHODS

The general method developed in the Benchmark Simulation platform (Gernaey et al. 2014) coupled to a LCA model is presented in detail. Thereafter, a case study where the model is

adjusted to the plant under study is described together with a simulation study on enhanced primary treatment.

The methodology of coupling process modelling and LCA is outlined in Figure 1. The modelling and simulation is performed in two steps. Step 1 represents detailed dynamic process modelling of on-site processes at the WWTP. The outputs of Step 1 are effluent water load, sludge load, direct GHG emissions and resource requirements. Step 2 is LCA modelling of the WWTP including off-site processes required for plant operation, the direct emissions and resource use calculated in Step 1 are used as inputs in Step 2. The output from Step 2 is environmental impact in selected impact categories.

Process model development

For the detailed process modelling, the Benchmark Simulation Model No. 2 GHG (BSM2G) presented by Flores-Alsina et al. (2014) is used as a basis. The BSM platform is a general simulation platform for benchmarking of operational and control strategies at WWTPs. It consists of: (i) a general plant layout (see Supplementary information, Figure S1, available with the online version of this paper); (ii) a set-up of sub-models for the included processes; (iii) models for sensors, controllers and actuators to allow implementation of various control strategies; (iv) a specified simulation procedure including an

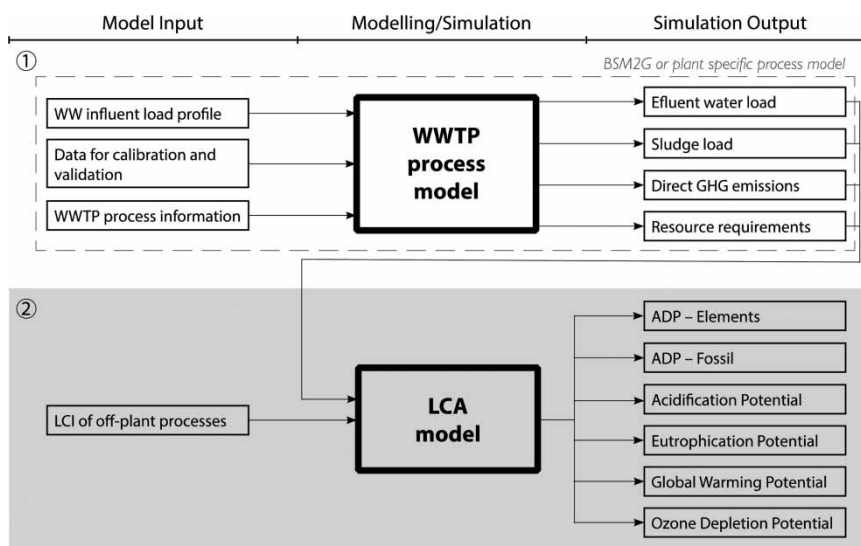


Figure 1 | Work flow diagram for modelling approach. Step 1 (white area, top): process modelling of on-site processes; Step 2 (grey area, bottom): life cycle assessment (LCA) of off-site processes using simulation outputs from Step 1.

influent profile; and (v) an evaluation procedure including two aggregated indices: Effluent Quality Index (EQI) and Operational Cost Index (OCI). The EQI measures the effluent water quality as a weighted average of effluent chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia, nitrate and total solids loads whereas the OCI provides a relative comparison for the operational costs including, power for mixing, aeration and pumping, carbon source addition, heating of the digester, utilization of biogas and disposal of sludge. More information about the BSM platform can be found in [Gernaey *et al.* \(2014\)](#).

The BSM2G plant consists of a primary clarifier (PRIM) modelled with a simplified model by [Otterpohl & Freund \(1992\)](#), followed by an activated sludge unit (ASU) in a modified Ludzack-Ettinger configuration where the initial anoxic tanks (ANOX) and last aerated tanks (OX) are connected by an internal recycle. The bioprocess model is an extended version of the Activated Sludge Model No. 1 (ASM1), described by [Guo & Vanrolleghem \(2014\)](#), including greenhouse gas emissions (ASM1G); the key modifications are the two-step nitrification – with separate nitrifier biomass states: ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria – and four-step denitrification ([Hiatt & Grady 2008](#)) along with nitrifier denitrification by AOB proposed by [Mampaey *et al.* \(2013\)](#). The secondary settler is described by a one-dimensional ten-layer settler by [Takács *et al.* \(1991\)](#). The sludge line contains a thickener (THK), an anaerobic digester (AD) and a dewatering unit (DW). The AD is modelled using the Anaerobic Digester Model No. 1 (ADM1, [Batstone *et al.* 2002](#)) and the THK and DW as ideal separation models. Dewatered sludge is stored for 12 months (STOR) before utilization.

The GHG emissions included in the BSM2G by [Flores-Alsina *et al.* \(2014\)](#) were supplemented with the following:

- direct emissions from digestion with 1% of the raw biogas ([Avfall Sverige Utveckling 2009](#));
- direct emissions from 12 months' sludge storage (open storage in piles) with 8.7 kg CH₄·ton VS⁻¹ (VS for volatile solids) and 0.36% of total nitrogen (TN) as N₂O-N ([Jönsson *et al.* 2015](#)) – corresponding amounts of carbon and nitrogen are removed from the sludge;
- direct emissions from gas utilization with 1.7% of combusted raw biogas in co-generation unit ([Liebetau *et al.* 2010](#));
- indirect emissions of N₂O from the recipient due to residual effluent nitrogen with 0.005 kg N₂O-N·kg TN_{effluent}⁻¹ ([IPCC 2013](#));
- extended GHG emissions assessed in the three different bio-solids utilization alternatives included, i.e., CH₄ and N₂O emissions (the organic matter is assumed to mineralize and carbon emitted as CO₂ ([Flores-Alsina *et al.* 2014](#)):
 - fertilization of farm land: emission factor N₂O = 0.01 kg N₂O-N·kg TN⁻¹ ([IPCC 2006](#));
 - composting; emission factor N₂O = 0.01 kg N₂O-N·kg TN⁻¹ ([IPCC 2006](#)), emission factor CH₄ = 0.0075 kg CH₄·kg TOC⁻¹ ([Kirkeby *et al.* 2005](#));
 - fertilization of forest: emission factor N₂O = 0.01 kg N₂O-N·kg TN⁻¹ (no data available, assumed equal to farm land).

The numbers suggested are to be used together with the generic BSM2G. For specific case studies these should be updated (transport distances, % distribution) or if a coupled LCA model is used then off-plant processes should be replaced by LCI data.

The model was implemented in Matlab/Simulink (MATLAB 8.4, The MathWorks Inc., Natick, MA, USA, 2014) and dynamic simulations for a full year evaluation period were run according to the procedures in [Gernaey *et al.* \(2014\)](#). The simulation results were used as input for the static LCA modelling of global environmental impact potential.

LCA model description

The LCA was performed according to the standard [ISO 14044](#). The goal and scope of the LCA was to perform a comparative assay of the operational strategies simulated using the process model. The system boundaries for the LCA were therefore chosen to be the WWTP itself with direct emissions to water, soil (micropollutants are not considered in this study) and air and the production and transport of power and chemicals from resource extraction to the plant. The benefit of utilizing the produced bio-methane was accounted for by expanding the system to include the benefitting process, i.e., co-generation of heat and power (in BSM2G) or replacing diesel as vehicle fuel. For the purpose of comparing operational strategies,

one year of operation was considered and construction and demolition phases were excluded as they have been shown to have a limited impact for most traditional advanced WWTPs (Corominas et al. 2013a). To manage the LCI and LCIA, the Gabi software tool was used (Gabi software 6.3, Thinkstep, Leinfelden-Echterdingen, Germany, 2013). The process model simulation outputs were used together with generic data from the Ecoinvent database (Weidema et al. 2013). Six impact categories were selected based on similar previous studies (Corominas et al. 2013a) and expected impacts calculated following the procedures developed by Centrum voor Milieukunde at Leiden University, The Netherlands (CML) (Guinée et al. 2002): abiotic depletion potential of elements (ADP elements) and fossil (ADP fossil) resources, eutrophication potential, acidification potential, global warming potential (GWP) and ozone depletion potential (ODP). For GWP emissions, CH₄, N₂O and abiotic CO₂ were considered with GWP factors according to IPCC (2013). The procedures of CML were used also for the characterization in the LCIA.

Modelling the Käppala WWTP

The Käppala WWTP in Lidingö outside Stockholm, Sweden receives wastewater from 11 municipalities in the northern part of the greater Stockholm area. The plant is mainly an underground facility built into the mountain on the island

Lidingö in the archipelago. The effluent requirements are 10 mg N·l⁻¹ TN (annual average) and 0.3 mg P·l⁻¹ total phosphorus (quarterly average). This is achieved with a treatment process based on primary mechanical treatment, secondary biological treatment and tertiary filtration (Figure 2). The plant is built in two parallel parts, where the original part has biological phosphorus removal in an A₂O process while the newer part is a pre-denitrification configuration with simultaneous precipitation using ferrous sulphate. For the purpose of the modelling study only, the new part of the plant is modelled. Where applicable, the volumes and flows are reduced to correspond to the part of influent wastewater that is treated in the new part of the plant. The description below refers only to the new part of the plant, if not otherwise stated. The BSM2G is used as the model base and modifications are highlighted outlining the plant model below.

The treatment process in detail consists of the following.

Primary treatment

3 mm bar screens followed by pre-aeration and an aerated grit chamber. Primary sedimentation in five parallel lines with a removal efficiency of 51% COD and a primary sludge total suspended solids (TSS) concentration of about 6%. The primary sludge is pumped to the sludge treatment. Only the primary settlers were modelled.

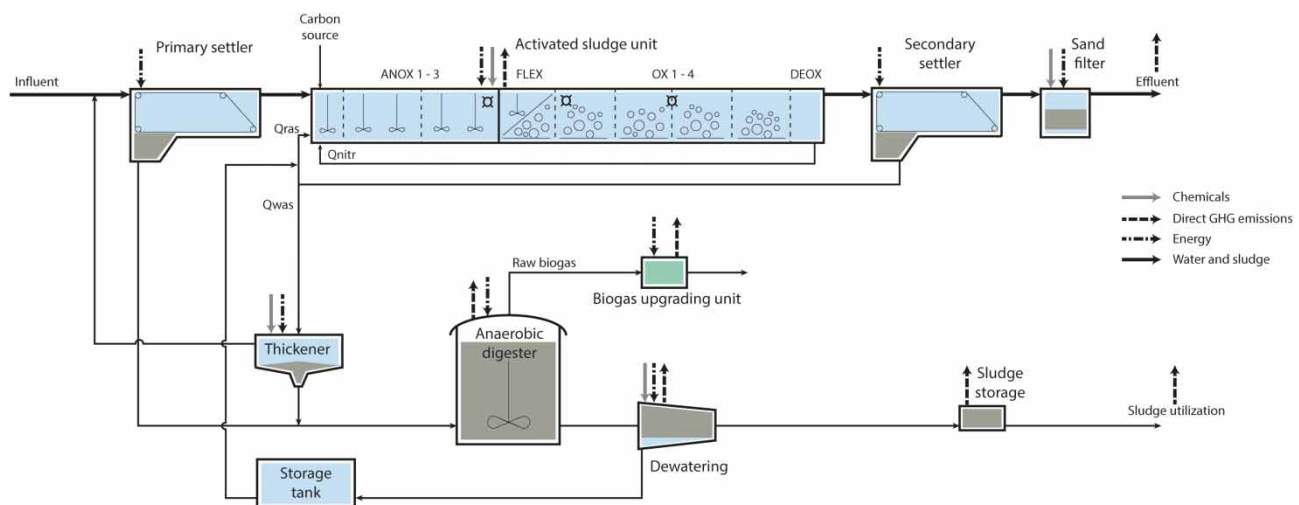


Figure 2 | Process flow diagram of the modelled parts of the plant. Arrows indicate flows of water and sludge (solid black), chemical input (solid grey), direct GHG emissions (dashed black) and energy input (dot-dashed black). The measurement points for aqueous N₂O are indicated in the ASU (α).

Biological treatment

Secondary treatment in activated sludge. Five parallel 10 m deep reactors with a modified Ludzak-Ettinger configuration with a total anoxic volume of 34,130 m³ utilizing influent COD as carbon source followed by aerated zones of in total 55,000 m³ and a final non-aerated zone of 3,300 m³ for lowering the effluent dissolved oxygen (DO). The final non-aerated zone and the first denitrification zone are connected by an internal recycle. Five secondary settlers with a depth of 6.1 m separate the sludge and the return activated sludge (RAS) is pumped back in a joint RAS channel. The waste activated sludge is pumped to centrifugal thickeners. The ASU was modelled as one line – with five times the volume – with nine zones in series (Fujie *et al.* 1983): three anoxic zones (ANOX 1 to 3), one swing zone (FLEX), four aerated zones (OX 1 to 4) and one final non-aerated zone (DEOX). The ASM1G model was used. To capture the effluent TSS at high flow rates the secondary clarifiers were described using the Bürger-Diehl model (Bürger *et al.* 2013) implemented in Matlab by Arnell (2015). The simultaneous precipitation was not modelled as such, but an addition of inorganic suspended solids (ISS) was introduced. For DO control the zones OX 1 to 4 have individual DO measurements and a PI regulator controls the airflow towards a DO set-point. The DO set-point, in turn, is controlled through ammonia feedback, i.e., a PI regulator controlling the DO set-point against a set-point of NH₄-N in the effluent of DEOX. The aeration model was extended according to the principles described in Arnell & Jeppsson (2015) for converting K_L α to airflow.

Filtration

A tertiary filtration step with chemical precipitation (using ferric chloride) on two media downstream filters (SF). The filters are intermittently backwashed and the sludge is recycled to the plant influent. A simple cut-off model was constructed taking away all particulates above 5 mg TSS·l⁻¹ in the filter influent to the sludge phase, which was recycled to the plant influent.

Sludge treatment

The sludge from both the old and the new part of the plant is treated in two ADs in series, each 9,000 m³. The primary

sludge is fed to the first digester and before entering the second digester mixed with the thickened secondary sludge. The digested sludge is dewatered in centrifuges to a dry solids content of 27%, which is stored for 12 months before application. The produced biogas is up-graded to vehicle fuel quality in a gas-upgrading unit (GU) and sold to the local public transport company to be used in city buses. The bio-solids are transported and utilized for fertilization of farmland or for landscaping. The dewatering supernatant is pumped back to the joint RAS channel before entering the anoxic zones. The sludge line was modelled with BSM2G default models but the reactor configuration was adjusted to the Käppala plant layout and volumes adjusted according to the modelled portion of the flow.

Consumption of resources, such as energy and chemicals, were either modelled dynamically (i.e., carbon source addition) or calculated based on specific consumption numbers. Most of the specific data were retrieved from the utility but some generic BSM2G data were used when specific data were missing. Values are tabulated in Supplementary information, Table S1 (available with the online version of this paper). The specific consumption factors have, when possible, been coupled to dynamically modelled flows and volumes but for a few energy-related processes static consumptions per day represented the most reliable information.

A dynamic influent profile was created from plant data for the years 2012 and 2014 using a five-step influent generation methodology (Arnell 2016). The actual flow measurements were used along with a synthetic diurnal load pattern created from the averaged and normed diurnal flow rate during dry weather. For influent fractionation and calibration, the procedures in Rieger *et al.* (2012) were followed. The year 2012 was selected for calibration of the model for all but N₂O production and emission, for which specific measurement data from 2014 were used. The simulation results from 2012 were chosen for results presentation as this was a year with several severe rain events challenging the treatment process. For calibration of the N₂O production in the bio-process model additional measurements were made in the ASU. N₂O gas concentration was measured in the ventilation channel from one of the five parallel ASU lines and, at the same time, the aqueous N₂O concentration was measured at three different positions

(one at a time) – beginning of OX, end of ANOX and mid OX. The N_2O measurements continued for more than 100 days. During this period, aqueous grab samples were also analysed for dissolved NO_2-N 11 times.

For the LCA model, the WWTP was split up into the main treatment steps supplemented with extraction of resources for and production and transport of power and chemicals needed for the process. The LCA model was also extended with the use of produced bio-methane as vehicle fuel. System boundaries are illustrated in Supplementary information, Figure S2 (available online).

LCI data with references are given in Supplementary information, Table S2 (available online), but some important assumptions were:

- for power production, the average Swedish electricity grid mix was used (mainly hydro and nuclear power, together about 90% of total);
- the methanol used in the scenario was assumed to have fossil origin;
- when used as vehicle fuel the bio-methane was assumed to replace 0.765 MJ of diesel for each MJ of bio-methane;
- for transport, the actual transport distances were used (see Supplementary information, Table S2) with Euro 4 class trucks running on diesel containing 10 ppm of sulphur. Unloaded return trips were assumed.

For the interpretation of the LCA results, the impact equivalence numbers – in functional unit, per m^3 – were normed to the base case for internal comparison and to the total EU28 emissions (Guinée et al. 2002) (values given in Supplementary information, Table S3, available online) in each respective impact category for evaluation of the relative importance of the different categories.

Global performance assessment of CEPT

The interest regarding enhanced mechanical treatment is growing as focus is shifted towards multiple objectives for WWTPs (Bachis et al. 2015). If the reduction over the different steps of the mechanical treatment can be increased several objectives can be improved: (i) the load on the ASU is decreased leading to lower aeration requirements and increased treatment capacity in the ASU; and (ii) the biogas production is increased leading to less GHG

emissions if the gas is utilized to replace fossil resources. A common way to achieve this is to implement chemically enhanced primary treatment (CEPT), where pre-precipitation with metal salts are used to improve the removal efficiency over the primary clarifiers. However, there are risks with this practice; extensive pre-precipitation can easily lead to deficiency of available organic matter for denitrification and even phosphorus in the ASU. Moreover, recovery of P becomes more difficult. Thus, there is conflicting competition for the influent organic matter, whether it is best used for denitrification in the ASU or for biogas production in the AD. For denitrification, external carbon sources, such as methanol or ethanol, can be used but that comes with a cost and environmental impact from production and transport of the chemicals.

A simulation study was designed to assess the total environmental impact of the above strategy using the proposed methodology. For realism and validation, the calibrated model of the Käppala WWTP was used. The base case represents the current operation of the treatment plant and the simulation results from 2012 were used as the base case. The alternative strategy with CEPT was modelled with the following model-wise simple modifications: The reduction over the PRIM was increased to 60% COD by increasing the parameter f_{corr} to 0.8 and moving the modelling of precipitation (ISS increase) to before the PRIM. A reduction over the PRIM of that magnitude is possible if the plant has effective primary settling, which is the case at Käppala. The goal with the simulation study was to show the change in environmental impact from the strategy assuming the same effluent quality. Therefore, the operation was tuned to match the same effluent standards as for the base case. To achieve this, it was necessary to add methanol to the anox-zones to match the denitrification in the base case. This was done by a 50/50 addition to ANOX 1 and 2 with a PI regulator controlling the methanol flow towards a concentration set-point of $6.7 \text{ mg N-l}^{-1} NO_3-N$ in the effluent from the DEOX. For the evaluation, the OCI was updated to cover the major impacts of the simulated case. The power for heating was updated to electrical power supply for heat pumps (default OCI factor for electrical power), the gas utilization was exchanged to vehicle fuel with an assumed OCI factor of 7.5 per m^3 of fuel and the addition of precipitation chemicals were included with an

assumed OCI factor of 13 per kg of Fe. The price of vehicle fuel at Käppala WWTP is not publically available but considering retail price and reasonable distribution costs and margins, 7.5 OCI is reasonable. Likewise, the actual contracted price for ferric chloride was not available but an OCI of about 13 is similar to the published price by several manufacturers taking the actual transport distance into account.

RESULTS AND DISCUSSION

Model calibration

The model calibration shows good fit for the standard water quality variables. Priority during the calibration was to calibrate the sludge age in the ASU to match the nitrification. This was achieved as the sludge age in the model is 13.0 compared to data 13.4 d. The resulting effluent TN and $\text{NH}_4\text{-N}$ differ by only $0.1 \text{ mg N}\cdot\text{l}^{-1}$ between model outputs and data. To achieve this, the TSS in the ASU had to be set slightly lower than actual values, 1,700 compared to the measured value of $2,100 \text{ mg}\cdot\text{l}^{-1}$, yielding a slightly lower overall bio-solids' production. Comparisons between model calibration results and plant data are tabled in Supplementary information, Table S4 (available online). Default parameter values were used in the process model.

From the measured N_2O concentration in the reactor, it was evident that no N_2O production occurred in the anoxic

zones (see Figure 3). The in-tank N_2O concentration was very low at the end of the last anoxic zone even if no forced stripping had occurred. However, in the aerated zones the N_2O concentrations were higher and increasing along the reactor. Taking the stripping, due to aeration into account, it could be concluded that the major part of the production and emission of N_2O occurred in the aerated zones. The reason for this is assumed to be the relatively high $\text{NO}_2\text{-N}$ concentrations measured, around $0.3 \text{ mg N}\cdot\text{l}^{-1}$. This relationship has been reported previously in the literature (Foley *et al.* 2011). To calibrate the $\text{NO}_2\text{-N}$ concentrations and succeeding N_2O emissions to air, two half-saturation parameters were adjusted: $K_{\text{FA}} = 0.002 \text{ g N}\cdot\text{m}^{-3}$ and $K_{\text{FNA}} = 0.00035 \text{ g N}\cdot\text{m}^{-3}$. As seen in Figure 4, the average level of the modelled emissions is in line with measured values. However, the full dynamics of the measured emissions were not well predicted by the model. The model predicts a constant base line for the emissions of about $30 \text{ kg N}_2\text{O}\cdot\text{N}\cdot\text{d}^{-1}$ even when the measured emissions decrease around day 425 (Figure 4). The model behaviour follows from the model equations for the AOB denitrification. However, recent publications emphasize the importance of using a two-pathway model – including both hydroxylamine oxidation and AOB denitrification – under dynamic conditions (Ni & Yuan 2015; Mannina *et al.* 2016). These models predict that the N_2O production relates to the rate of nitritation rather than DO. These new two-pathway models were not available at the time of conducting this study but the present results support that additional

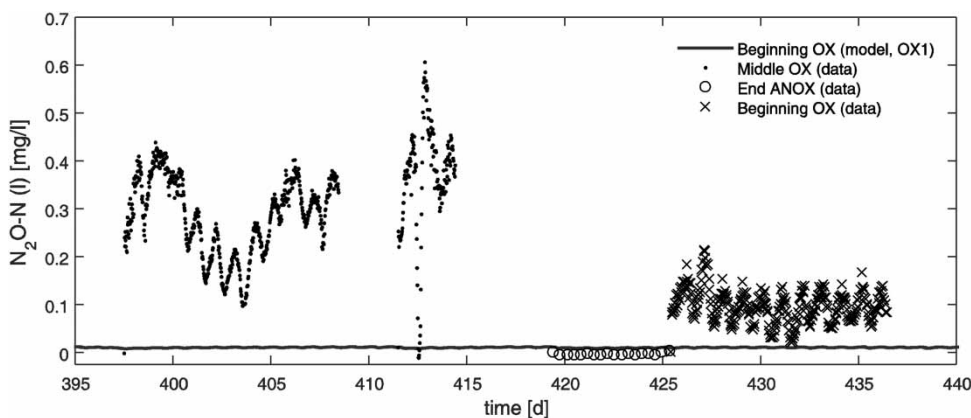


Figure 3 | $\text{N}_2\text{O}\text{-N}$ in aqueous phase at Käppala WWTP. Measurements in the three different locations indicated in Figure 2 (black markers) during three sequential time periods and modelled $S_{\text{N}_2\text{O}}$ in the corresponding zones (grey line, same time period as data). The time scale represents simulation days, where 395 corresponds to 29th May.

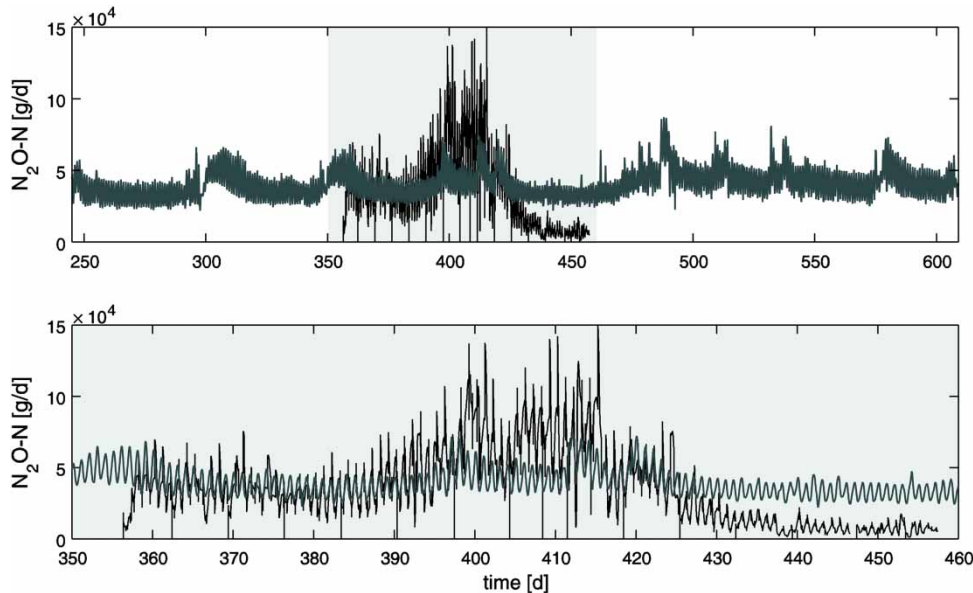


Figure 4 | N_2O measurements at Käppala WWTP and model calibration result. Data – black, model – grey. Full year simulation (top) and day 350 to 460 covering the N_2O measurement campaign (bottom).

reaction pathways need to be considered in future work. One trade-off made in favour of getting proper emissions to air was that the N_2O concentration in the aqueous phase had to be calibrated to lower than measured values in the aerated zones. The reason for this is that the modelled stripping of N_2O – as previously reported by Lindblom *et al.* (2015) – is too effective. A higher liquid N_2O concentration would lead to too high emissions. The N_2O measurements in both off-gas and aqueous phase show a high variability, approximately four to ten times from low to high values (Figures 3 and 4). This pattern is repeated in the model for the off-gas. The highly dynamic pattern motivates the use of dynamic models over averages or static emission factors.

Performance assessment of CEPT

The most relevant simulation results from the process model are presented in Table 1. The ambition that the effluent quality should be the same for the two cases was achieved and both the general EQI and the specific nitrogen parameters were approximately similar for the two simulations.

The primary objective, to increase the production of bio-methane with CEPT, was reached since the mixed sludge for digestion has a higher proportion of primary sludge, which has a higher biogas potential (Arnell *et al.* 2016). The

bio-methane production increased by almost 14%. Along with that the energy for aeration decreased as expected. However, the change was small, due to the addition of methanol to the ANOX that kept up the total COD load on the ASU. The use of chemicals for precipitation increased substantially, which was expected, just as the additional methanol. Although, the use of precipitation chemicals increased the sludge production does not differ significantly for the two scenarios. This is due to a very small difference in ISS addition when switching from simultaneous precipitation in the base case to pre-precipitation in the CEPT case. In the simulation study this was based on lab-scale jar tests and might show to be somewhat different in the full-scale application. However, Ødegaard (1998) showed that CEPT does not necessarily have to lead to excessive sludge production. The OCI is tabulated in Table 1. The total cost for operation increases by 87%. The increased revenue for selling vehicle fuel is almost fully compensated by the additional carbon source dosing. However, the major increase in OCI is due to the increase in precipitation chemicals (700% higher OCI). This cost increase would be less pronounced if a cheaper metal salt could be used, for example, residual ferrous sulphate from other industrial processes. The performance of such an alternative was not known in this case and could not be evaluated.

Table 1 | Simulation results from the process model for the base case and CEPT

	Unit	Base case	CEPT
Effluent quality			
EQI	–	17,470	17,040
NH ₄ -N	mg N·l ⁻¹	1.34	1.13
NO _x -N	mg N·l ⁻¹	6.22	6.59
TN	mg N·l ⁻¹	8.50	8.61
Sludge production	kg TS·d ⁻¹	14,694	14,530
Resource performance			
Bio-methane production	kg CH ₄ ·d ⁻¹ /kWh·d ⁻¹	4,930/68,400	5,610/77,900
Power for aeration	kWh·d ⁻¹	6,590	6,480
Consumption precipitation chem.	kg Fe·d ⁻¹	178	1,260
Consumption methanol	kg COD·d ⁻¹	0	2,060
Direct GHG emissions			
N ₂ O ASU	kg CO ₂ e·d ⁻¹	19,040	14,400
CH ₄ and N ₂ O sludge treatment	kg CO ₂ e·d ⁻¹	6,520	6,700
Operational cost			
OCI	–	13,600	25,400
Precipitation chem. cost index	–	2,320	16,400
Carbon source cost index	–	0	6,170
Vehicle fuel revenue index	–	53,600	61,100

The direct emissions of GHGs from the plant were affected by the operational strategy. The methane slip from the AD increased with CEPT due to increased gas production since it was calculated proportionally. On the contrary, a much larger decrease in GHG emissions was achieved as the N₂O emissions from the ASU decreased. It is hypothesized that this is due to the reduced load on the ASU with CEPT; the TN in the primary effluent was reduced and methanol was dosed, which increases the C to N ratio in the ASU.

The on-site effects of CEPT as described would have been more pronounced if the differences in primary removal efficiencies were greater. However, the current primary clarifiers at Käppala WWTP are well functioning already without pre-precipitation. Furthermore, it could be argued that an even higher removal can be achieved with CEPT than tested here (Ødegaard 1998) but it was found appropriate to use the actual removal efficiency established in jar tests at the plant.

The LCA results based on the two scenarios are presented in Table 2 and Figures 5 and 6. From Table 2 it can

be concluded that all except ODP is within two orders of magnitude related to the total emissions for 28 European countries for the year 2000; ODP is significantly smaller. Also, in the LCA results, striving to keep effluent quality unchanged is evident as the difference in the impact category Eutrophication is small (Figure 5). The sought reduction in Climate impact of the operations was achieved and did decrease by 28% (Table 2). This is the result of several combined effects: a decrease of the direct plant GHG emissions, decreased power consumption as well as increased bio-methane production. The latter leads to decreased GWP as a credit is made when it substitutes fossil vehicle fuels (see Figure 6). Altogether, the Climate impact is reduced by almost a third despite the increased emissions from production and transport of methanol and precipitation chemicals.

The improvement in Climate impact comes with a cost in terms of resource use. The elemental and fossil ADP are increased by 77 and 305%, respectively (see Table 2 and Figure 5). This increase is completely explained by the increased use of chemicals for precipitation and

Table 2 | LCA results for the base case and modified control strategy (CEPT) for the six selected impact categories

Impact category	Unit	Absolute values		Normalized values EU28 [-]	
		Base case	CEPT	Base case	CEPT
ADP elements	[kg Sbe·m ⁻³]	3.7×10^{-8}	6.5×10^{-8}	2.3×10^{-7}	4.1×10^{-7}
ADP fossil	[MJ·m ⁻³]	0.29	0.90	3.2×10^{-7}	9.8×10^{-7}
Acidification	[kg SO ₂ e·m ⁻³]	4.4×10^{-4}	4.9×10^{-4}	1.0×10^{-6}	1.1×10^{-6}
Eutrophication	[kg PO ₄ e·m ⁻³]	2.4×10^{-3}	2.2×10^{-3}	4.9×10^{-6}	4.5×10^{-6}
GWP	[kg CO ₂ e·m ⁻³]	0.15	0.11	1.1×10^{-6}	8.1×10^{-7}
ODP	[kg R11e·m ⁻³]	2.0×10^{-10}	9.2×10^{-10}	7.6×10^{-10}	3.5×10^{-9}

The units only apply to absolute values, normalized values are dimensionless. The unit for ADM elements refer to antimony (Sb) equivalents and the ODP unit to equivalents of the trichlorofluoromethane R11.

denitrification (Figure 6). The origin of the methanol was here assumed to be fossil based, which has a major impact on ADP fossil. In a parallel project using the same base model a sensitivity analysis on the origin of the carbon source was done and it showed a significant impact. The increase in fossil depletion was reduced significantly if recycled glycerol was used instead (Åmand et al. 2016).

The impact category with the largest relative change was ODP; with CEPT it increased by 450% (Table 2). From the table, it is clear that the absolute values are very small and even normalized to the EU28 emissions the ODP was two to four orders of magnitude lower than that of the other categories. Thus, even if the increase was relatively large, it was from a low initial level.

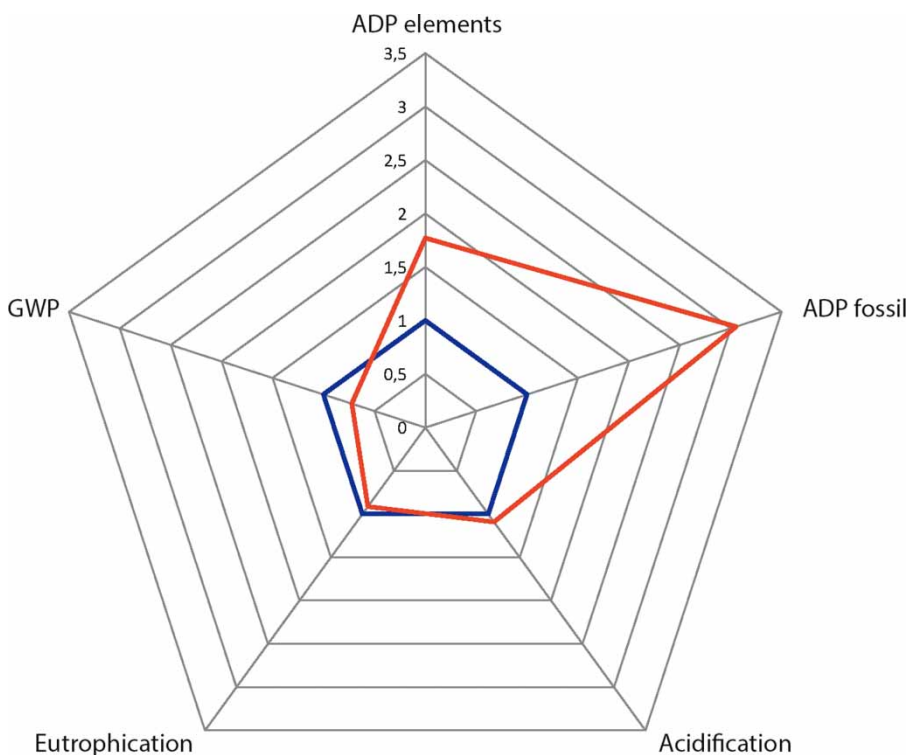


Figure 5 | Comparison between the LCA results for the base case (inner) and CEPT (outer). The results are normalized to the values of the base case. For clarity the ODP is omitted due to very low absolute levels.

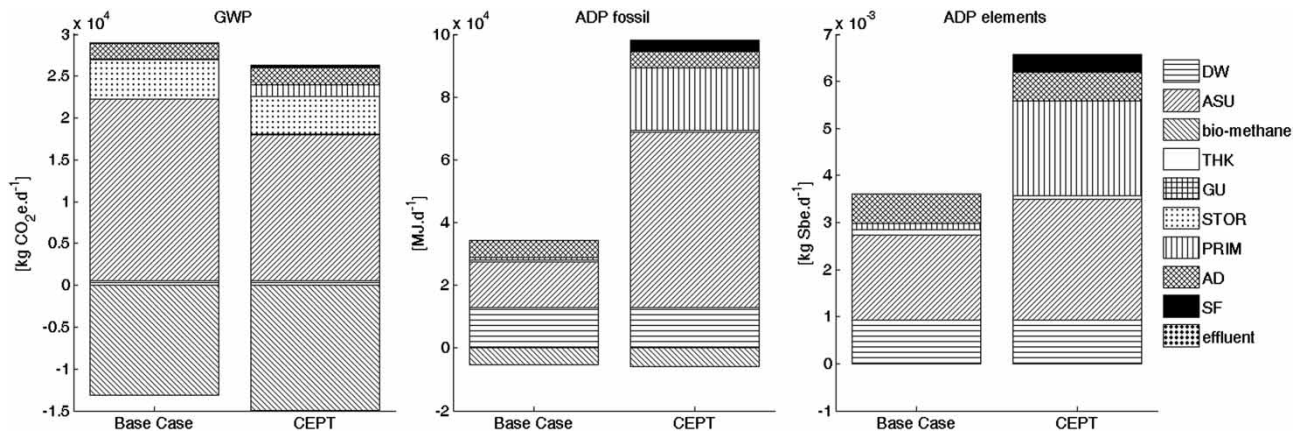


Figure 6 | Bar chart of the contribution from the different plant sub-processes to the impact categories: GWP (left), ADP fossil (middle) and ADP elements (right).

The impact of CEPT on Acidification potential is negative but very small. The change is solely caused by production and transport of chemicals, especially methanol, which penalizes CEPT in this case.

Overall, the LCA results in this case study are in line with other studies using LCA to assess the environmental impact from altering operational strategies at WWTPs. It is difficult to compare absolute values between studies due to varying scope, functional units and other choices as these have been shown to have a significant impact on results (Corominas *et al.* 2013a; Åmand *et al.* 2016). However, the relative change when comparing operational strategies and specifically with and without CEPT are in the same range as in other studies (Corominas *et al.* 2013b; Bisinella de Faria *et al.* 2015; Hadjimichael *et al.* 2016). Bisinella de Faria *et al.* (2015) have also evaluated CEPT as one alternative and found similar benefits regarding energy and total climate impact. Due to differences in system boundaries and scope of the studies they show a reduction in eutrophication (not fixed effluent standard as in this study) and resources (higher dose of Fe and methanol in base case).

The results from this case study show the great variability that arises in many respects when changing operations at a WWTP. Large variations are also seen between different plants in, for example, GHG emissions (Foley *et al.* 2011). This motivates the novel model additions to the method framework presented in this study – capturing both dynamic N_2O production and other, fugitive, GHG emissions. The models and method framework have here been proven to be applicable to a full-scale plant. This has previously only

been presented by Hadjimichael *et al.* (2016), however, for a different purpose. While the presented results are specific for this plant they are in some parts also general when compared to the literature. The conclusions for impact of increased resource use when implementing CEPT were shown to be similar for three plants analysed by Åmand *et al.* (2016) and the general conclusions on CEPT (e.g., increased gas production) have been reported previously (Ødegaard 1998; Olsson 2015).

General impact of results and limitations of method

The results above show the multidimensional, and often conflicting, impacts that arise from different operational strategies at WWTPs. When the operations are evaluated, not only based on effluent quality and cost but also on resource efficiency and global environmental impact, the evaluation grows complex. From the results, it is evident that detailed process models are necessary to assess, for example, N_2O production in the ASU or overall effects of return streams. This confirms findings in previous studies, i.e., Flores-Alsina *et al.* (2014), Guo & Vanrolleghem (2014) and Corominas *et al.* (2013b); these effects can rarely be calculated with traditional spreadsheet methods (Rieger *et al.* 2012). At the same time, the increase or decrease in resource use and recovery leads to a different environmental impact off-plant that is not captured by the process model but only by LCA. To combine process modelling and LCA, as in this study, to describe the included processes and calculate the global environmental

impact is useful for comparing alternative operational strategies at WWTPs and provides a thorough base for decision-making.

The general method developed suggests also including utilization of bio-solids and expanding the system to include replacement of commercial fertilizer. This is done for completeness and consistency since effluent load is included as well as bio-methane utilization with crediting when used as vehicle fuel. In the case study, there was unfortunately limited information available on the sludge utilization. Moreover, the data quality was poor for the expanded system of fertilizer replacement. Therefore, the sludge utilization was excluded. However, since the evaluated operational strategies produce very similar amounts of sludge the exclusion in this study will have basically no impact on the conclusions. Considering replacement of commercial fertilizer will be included in future studies.

The different impact categories in LCA measure different things expressed in different units and are not comparable per se. The standard for LCA studies, [ISO 14044](#), states that weighted comparison of the impact categories is an optional part of the LCA conducted separately after the LCIA. This is good praxis since the subjective process of grading and weighting the results should not be mixed with the objective impact assessment. The weighting step was not performed in this study since it focuses on developing the method for impact assessment rather than decision-making. However, if the results are to be used for decision-making such weighting and comparison of the different impact categories' mutual importance should be done at a later stage. This exercise can be performed by skilled decision-makers involved in conducting the LCA. Principles and guidance on weighting and multi-criteria analysis can be found in the literature ([ISO14044](#); [Baumann & Tillman 2004](#); [Malmqvist *et al.* 2006](#)).

CONCLUSIONS

The model platform Benchmark Simulation Model No. 2 GHG was extended to – in addition to effluent quality and operational cost – also evaluate fugitive GHG emissions, more detailed energy efficiency models, extended OCI and global environmental impact to a level of detail not

presented before. To assess the global environmental impact, the dynamic process models have been coupled with a life cycle assessment (LCA) model. The approach to couple a detailed plant-wide model including the novelty of using dynamic energy and GHG calculations captures both direct emissions and off-site impacts, for example, from production of electricity and chemicals. This model concept was calibrated to a real plant, the Käppala WWTP in Lidingö, Sweden. The model calibration under current operational conditions shows that the model is suitable for representing this type of municipal treatment process. However, it is not possible to capture the full dynamics in N₂O production and emission based on real data. From this fact, it is concluded that a model containing N₂O production only from autotrophic and heterotrophic denitrification is not sufficient. Additional production pathways need to be considered.

A simulation study on altering the operational strategy at the studied treatment plant was performed. The current strategy with simultaneous precipitation and pre-denitrification utilizing internal COD was compared to a strategy with extensive pre-precipitation with ferric chloride and adding methanol in the anoxic zones. The comparative simulations show that several of the initial goals with the strategy are fulfilled, i.e., higher gas production, lower aeration energy requirement and an overall decrease in GWP (28%). However, at the same time, the LCA reveals that the abiotic depletion of both elemental and fossil resources increases by 77% and 305%, respectively. Also, the ozone layer depletion increases many fold but from a very low initial level. By expanding the OCI with precipitation chemicals and alternative bio-methane utilization the results show that the cost of operation increases by 87%, although the energy for aeration decreases and the increased gas production means higher revenue for bio-methane utilization.

The study shows that the method is applicable for use at WWTPs. The coupled process and LCA modelling methodology captures plant-wide and global effects that would not only be hard to show with standard spreadsheet calculations but would also remain hidden using any of the models separately. This novel method for multi-criteria evaluation of operational strategies including on-site process changes, global environmental impact and operational cost

provides a good basis for choosing operational strategies at WWTPs.

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