Journal of Environmental Management 128 (2013) 759-767

Contents lists available at SciVerse ScienceDirect

ELSEVIER

Environmental Management



Journal of Environmental Management

Including Life Cycle Assessment for decision-making in controlling wastewater nutrient removal systems



Lluís Corominas^{a,b,*}, Henrik F. Larsen^{c,d}, Xavier Flores-Alsina^{a,e}, Peter A. Vanrolleghem^a

^a modelEAU, Département de génie civil et de génie des Eaux, Pavillon Adrien-Pouliot, Université Laval, 1065, Avenue de la Médecine, Québec, G1V 0A6 QC, Canada

^b ICRA, Catalan Institute for Water Research, Scientific and Technological Park of the University of Girona, C/Emili Grahit 101, E-17003 Girona, Spain

^c Research and Development, Danish Road Directorate, Guldalderen 12, DK-2640 Hedehusene, Denmark

^d QSA, Department of Management Engineering, Technical University of Denmark (DTU), Building 426, DK-2800 Lyngby, Denmark

^e Div. of Industrial Electrical Engineering and Automation (IEA), Lund University, Box 118, SE-221 00 Lund, Sweden

ARTICLE INFO

Article history: Received 8 March 2013 Received in revised form 27 May 2013 Accepted 5 June 2013 Available online

Keywords: ASM3-BioP Control Decision making LCA Mathematical modelling Nutrient removal Sustainability Wastewater treatment Weighting

ABSTRACT

This paper focuses on the use of Life Cycle Assessment (LCA) to evaluate the performance of seventeen control strategies in wastewater treatment plants (WWTPs). It tackles the importance of using site-specific factors for nutrient enrichment when decision-makers have to select best operating strategies. Therefore, the LCA evaluation is repeated for three different scenarios depending on the limitation of nitrogen (N), phosphorus (P), or both, when evaluating the nutrient enrichment impact in water bodies. The LCA results indicate that for treated effluent discharged into N-deficient aquatic systems (e.g. open coastal areas) the most eco-friendly strategies differ from the ones dealing with discharging into P-deficient (e.g. lakes and rivers) and N&P-deficient systems (e.g. coastal zones). More particularly, the results suggest that strategies that promote increased nutrient removal and/or energy savings present an environmental benefit for N&P and P-deficient systems. This is not the case when addressing N-deficient systems for which the use of chemicals (even for improving N removal efficiencies) is not always beneficial for the environment. A sensitivity analysis on using weighting of the impact categories is conducted to assess how value choices (policy decisions) may affect the management of WWTPs. For the scenarios with only N-limitation, the LCA-based ranking of the control strategies is sensitive to the choice of weighting factors, whereas this is not the case for N&P or P-deficient aquatic systems.

© 2013 Elsevier Ltd. All rights reserved.

1. Introduction

Decisions about wastewater nutrient removal systems have traditionally been driven by considerations of technical aspects and cost-benefit analyses. In order to assess sustainability it is essential also to incorporate environmental and social aspects. In this line, several methods for sustainability assessment of wastewater treatment technologies have been proposed and evaluated in literature (e.g. Muga and Mihelcic, 2008; Guinée et al., 2011). Focussing on environmental performance, Life Cycle Assessment (LCA) (ISO 14040, 2006) is an accepted tool that has also been used to evaluate potential environmental impacts from environmental processes (Finnveden et al., 2009) including wastewater treatment processes throughout their whole life cycle (Guest et al., 2009; Kalbar et al., 2012). The success of this method is demonstrated by the large number of published studies which have applied LCA to the wastewater treatment field (Corominas et al., 2013). The published studies so far, have been applied to estimate the impact of different wastewater treatment plants (WWTPs) and to compare conventional and new wastewater treatment technologies.

Complex wastewater nutrient removal systems require sophisticated monitoring and control systems that optimize overall process performance and account for daily and seasonal process variability. The optimization of treatment technologies dealing with nitrogen and phosphorus removal has never been evaluated in terms of environmental performance using LCA, probably because of the technical difficulty of evaluating multiple strategies at full scale. An alternative to intensive full-scale optimization is the use of models. These, are cost-effective tools for the evaluation of control strategies as has been demonstrated by the IWA Task group of benchmarking of control strategies (Gernaey et al., 2013).

^{*} Corresponding author. ICRA, Catalan Institute for Water Research, Carrer Emili Grahit, 101, E-17003 Girona, Spain. Tel.: +34 972 183380.

E-mail addresses: lcorominas@icra.cat, llcorominas@gmail.com (L. Corominas), hfl@vd.dk (H.F. Larsen), xavier.flores@iea.lth.se (X. Flores-Alsina), peter.vanrolleghem@gci.ulaval.ca (P.A. Vanrolleghem).

^{0301-4797/\$ –} see front matter \odot 2013 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.jenvman.2013.06.002

Hence, the combination of mechanistic models for evaluating control strategies with LCA tools can bridge the gap between process control and environmental performance (Flores-Alsina et al., 2010).

Linking LCA results with decision-making is a challenging task. First, LCA has traditionally been a site and time-independent/ generic tool and for impact categories such as global warming (GW) a site-generic approach is justifiable. However, other impact categories are site-dependent (e.g. nutrient enrichment, NE) and the generic models most probably provide a far from realistic description of the impacts. Therefore, LCA research is moving towards developing methodologies that include the importance of local conditions and set country-specific or site-dependent characterization factors (e.g. Gallego et al., 2010; Basset-Mens et al., 2006; Azevedo et al., 2012). Second, LCA methodology proposes a weighting step, which allows for consideration of the relative importance of the different impact categories. Most studies present the results without weighting leaving the final interpretation to the decision-maker. We believe that incorporating an evaluation of the importance of the weighting factors in the discussion of the LCA results would facilitate the task of the decision-makers.

The goal of this paper is to present a methodology to evaluate the environmental impacts of enhanced process performance strategies applied to wastewater nutrient removal systems. The assessment of site-specific conditions for the NE impact category and the assessment of the importance of weighting factors are key issues addressed in this paper.

2. Process and nutrient removal strategies description

2.1. Process

The Neptune Simulation Benchmark (NSB) is the virtual WWTP under study (Fig. 1) which serves 80,000 population equivalents and comprises the water treatment and the sludge treatment lines. The design of the water treatment line was conducted following the Metcalf & Eddy guidelines (Tchobanoglous et al., 2003). First, there is a primary settler of 900 m³, which reaches on average 50% TSS removal. Then, biological treatment was designed for an average flow rate of 22,938 $\text{m}^3 \text{d}^{-1}$ and organic, nitrogen and phosphorous loads (just after primary settling) of 12,200 kg COD d⁻¹, 1140 kg N d⁻¹ and 215 kg P d⁻¹ respectively. The biological treatment configuration is the A_2O (Tchobanoglous et al., 2003) comprised of seven reactors in series (tank An1 & An2 are anaerobic with a volume of 1000 m³ each, tanks Ax1 and Ax2 are anoxic with a volume of 1500 m³ each and tanks Ox1, Ox2 and Ox3 are aerobic with a volume of 3000 m³ each). Ax1 and Ox3 are linked by means of an internal recycle of 108,000 $\text{m}^3 \text{d}^{-1}$. The system allows for the addition of chemicals to improve nitrogen and phosphorus removal. Sodium acetate can be added in An1 when carbon source is a limiting factor for denitrification and phosphorus removal. Ferric chloride can be added in Ox3 to promote the precipitation of phosphorus. The secondary settler has a surface area of 1500 m² and a total volume of 6000 m³. Part of the settled sludge is recycled to An1 (22,600 m³ d⁻¹) and part is wasted (400 m³ d⁻¹) for further



Fig. 1. Neptune simulation benchmark process schematic. Grey boxes correspond to a deterministic dynamic model and white boxes to simplified model using empirical factors. *Gravity thickening is assumed not to have a significant impact on the LCA results and is therefore not included in the evaluation.

sludge treatment line together with the primary settler waste. Sludge treatment starts with gravity thickening, followed by mesophilic anaerobic digestion. The biogas produced is burnt in a cogenerator to recover energy. Afterwards, the sludge goes to mechanical dewatering and the supernatant obtained is recycled just before An1. After dewatering, the sludge is burnt in an on-site incinerator that uses electricity produced in the co-generator. Finally, the ashes are transported to landfill. Several chemicals are applied during sludge treatment for promoting flocculation, precipitation and neutralization for pH adjustment. The design of the sludge treatment line has been taken from the Neptune project (EU FP6) as documented in Larsen et al. (2010).

2.2. Process models

2.2.1. Wastewater treatment line

Modelling and simulation works were performed using the WEST[®] modelling/simulation environment (mikebyDHI.com). The EAWAG Activated Sludge Model (ASM) No 3 Bio-P was chosen as bio-chemical model (Rieger et al., 2001). This model has 19 state variables and describes (bio) chemical phosphorus removal, nitrification and denitrification in activated sludge systems by means of a set of nonlinear differential equations. The model was extended to include chemical precipitation of phosphorus as in ASM2d (Henze et al., 2000). The double exponential velocity function defined in Takács et al. (1991), based on the solids flux concept was selected as a fair representation of the settling process, using a 10 layer discretization. Kinetic parameters were adjusted according to the influent temperature using the Arrhenius equation. The default parameters for the activated sludge and the settling model can be found in Rieger et al. (2001) and Gernaey et al. (2013) respectively, while the parameters for phosphorus precipitation kinetics were taken from Gernaey et al. (2002). It is important to highlight that the settling characteristics were assumed to be constant along the case study although the authors are aware that the floc characteristics may change in systems with chemical precipitation. Besides the use of the bio-chemical mechanistic model, direct greenhouse gases emissions (GHG) were estimated using simple comprehensive models (Table 1). CO₂ emissions generated during the oxidation of non-biogenic organics were modelled as described in

Table 1

Details of the NSB model. Water line.

Corominas et al. (2012). Nitrous oxide (N₂O) emissions generated during treatment were also included using the factor 0.09% of TN removed emitted as N₂O–N proposed in Ahn et al. (2010) for BNR plants. Also, energy models were considered in the biological treatment including aeration energy demand (for organic matter and nitrogen removal), mixing energy (of the activated sludge tanks) and pumping energy (for the internal and external recirculations) using the equations proposed in Gernaey et al. (2013) (Table 1). The load of sludge produced (SP) in the biological treatment that goes to the sludge line was calculated based on the amount of solids accumulated in the plant and from the solids wasted from the primary and secondary clarifiers.

2.2.2. Sludge treatment line

The sludge produced in the wastewater treatment line (in mass per day) is the input of the sludge treatment model which consists of empirical factors to estimate i) biogas production, non-methane volatile organic compounds (NMVOCs), energy consumption and energy recovery for the anaerobic digester, ii) chemicals consumption for dewatering, iii) emissions and chemicals consumption for on-site incineration and iv) emissions of organic and inorganic compounds at landfill. The sludge incineration factors were taken from the ARA Hard Sludge Incineration Plant in Winterthur, Switzerland (Larsen et al., 2010). A summary of the empirical factors is presented in Table 2.

2.3. Simulations

Plant performance evaluation was based on 609 days simulated influent data generated using the influent generator developed in Gernaey et al. (2011). All dynamic simulations (609 days) were preceded by steady state simulations (200 days). Only the data generated during the last 364 days of the dynamic simulations were used for plant performance evaluation and the output sampling was set to 15 min (Gernaey et al., 2013).

2.4. Enhanced process performance strategies

Seventeen nutrient removal strategies were evaluated in this study. A reference strategy (A_1) was defined where the system

Process	Factor/equation/model	Source
Primary settling	Otterpohl and Freund	Otterpohl and Freund (1992)
Biological secondary treatment	ASM3 Bio-P	Rieger et al. (2001)
Sludge production (kg d^{-1})	$SP = \frac{1}{t_{ebs} \cdot 1000} \cdot (M_{TSS}(t_end) - M_{TSS}(t_ini) + \int_{t_ini}^{t_end} TSS_{prim}(t) \cdot Q_{prim}(t) \cdot dt + \int_{t_ini}^{t_end} TSS_{sec}(t) \cdot Q_{sec}(t) \cdot dt + \int_{t_ini}^{t_end} TSS_{sec}(t) \cdot dt + \int_{t_ini}^$	Gernaey et al. (2013)
Secondary settling	Takács	Takács et al. (1991)
Aeration energy for secondary treatment (kWh d ⁻¹)	$AE = \frac{So_{las}^{1.5}}{t_{obs} \cdot 1.8^{-1.000}} \int_{L_{L}ini}^{L_{L}ond} \sum_{i=1}^{i=7} (V_i \cdot K_{L} a_i^{15}(t)) \cdot dt$	Gernaey et al. (2013)
Mixing energy for secondary treatment (kWh d ⁻¹)	$ME = \frac{24}{t_{obs}} \int_{t_{inin}}^{t_{inin}} \sum_{i=1}^{t_{inin}} \left[for t \text{ when } K_L a_i(t) < 20 \text{ d}^{-1} \text{ then } ME_{unit} \cdot V_i \right] \cdot dt$ for t when $K_L a_i(t) \ge 20 \text{ d}^{-1}$ then 0	Gernaey et al. (2013)
Pumping energy for secondary treatment (kWh d ⁻¹)	$PE = \frac{1}{t_{obs}} \int_{t_{ini}}^{t_{end}} (PE_{-}Q_{int} \cdot Q_{int} + PE_{-}Q_{r} \cdot Q_{r} + PE_{-}Q_{w} \cdot Q_{w})dt$	Gernaey et al. (2013)
N ₂ O, Biological treatment	0.0009 kg N ₂ O kg TN removed ^{-1}	Ahn et al. (2010)
CO_2 , oxidation of sodium acetate (kg $CO_2 d^{-1}$)	$CO_2, BOD_{ox} = \left[BOD_{ox} - 1.42 \cdot \left(\frac{\gamma}{(1+k_d \cdot SRT)}\right) \cdot BOD_{ox}\right] \cdot f_{CO_2/O_2}$	Corominas et al. (2012)

For AE, t_{obs} is the period of simulation (d) ($t_{ini} - t_{end}$), V_i is the volume of reactor i (m³), $K_L a_1^{15}$ is the mass transfer coefficient in reactor i recalculated to 15 °C (d⁻¹), and S_{0sat}^{15} is the saturation oxygen concentration at 15 °C (8 g (-COD) m⁻³). For SP, M_{TSS} is the total suspended solids in the reactors, TSS_{prim} and TSS_{sec} are the total solids concentrations in the primary and secondary settler underflows, Q_{prim} and Q_{sec} are the wastage flow rates in the primary and secondary settlers. For ME, ME_{unit} is the mixing energy consumption for the activated sludge tanks, which is set to 0.005 kW m⁻³. PE is calculated as a weighted averaged sum of the internal recycle flow rate Q_{int} , the external recycle flow rate Q_w . PE_Q_{int}: 0.004 kWh m⁻³, PE_Q_w: 0.005 kWh m⁻³. BOD_{ox} is the BOD load (kg d⁻¹) corresponding to sodium acetate addition, Y is the yield (0.84 kg VSS kg BOD removed⁻¹), SRT is the sludge retention time (d), k_d is the endogenous decay coefficient (0.05 d⁻¹), and f_{CO_2/O_2} corresponds 1.1 kg of CO₂ produced per 1 kg O₂ consumed in the BOD oxidation.

Details of the NSB model. Sludge line.

Туре	Process	Factor/equation/model	Units	
Electricity consumption	Anaerobic digestion	70	kWh tDM ⁻¹	
	On-site incineration	330 (110 from digester + 220	kWh tDM ⁻¹	
		from external supply)		
	Dewatering	20	kWh tDM ⁻¹	
Electricity production	Anaerobic digestion	500	kWh tDM ⁻¹	
Fuel oil consumption	On-site incineration	80 ^a	kWh tDM ⁻¹	
Biogas consumption	On-site incineration	740 ^a	kWh tDM ⁻¹	
Chemicals for sludge treatment	Copolymer of acrylamide, 0.48%)	0.22	kg kg $\mathrm{D}\mathrm{M}^{-1}$	
	Quarry sand for filtration	0.028	kg kg $\mathrm{D}\mathrm{M}^{-1}$	
Chemicals for on-site incineration	Calcium chloride	0.00065	kg kg DM^{-1}	
	TMT 15/Na 3T, 15%	0.16	kg kg DM^{-1}	
	Sodium persulfate	0.00064	kg kg DM^{-1}	
	Sodium hydroxide	0.034	kg kgDM ⁻¹	
	NH ₃ , 45%	5.07	kg kg $\mathrm{D}\mathrm{M}^{-1}$	
	HCI	0.002	kg kg $\mathrm{D}\mathrm{M}^{-1}$	
	Sulphuric acid	0.00120	kg kg DM^{-1}	
	Sodium chloride	0.00059	kg kg DM^{-1}	
CO ₂	Anaerobic digestion	82.1	Nm ³ CO ₂ tDM ⁻¹	
CH4	Anaerobic digestion	115.5	Nm ³ CH ₄ tDM ⁻¹	
Organic and inorganic	Incineration	Check in Larsen et al. (2010)	—	
Residue	Slag from incineration	515	kg tDM ⁻¹	

^a Energy content.

(Source: Larsen et al., 2010).

operates at a constant aeration flow rate, constant wastage flow rate and no chemicals are added in the biological treatment. Although the performance of this system is not optimal, it would accomplish with current legislation in terms of effluent nitrogen. However, phosphorous removal is minimal because of the limited available COD at the influent of the biological reactor, which results in competition between denitrification and phosphorous removal. Then, sixteen combinations of seven control algorithms $[A = (A_2, ..., A_{17})]$ were implemented (See details in the Supporting Information and find comprehensive information about control of WWTPs in Olsson et al., 2005). The dissolved oxygen (DO) control at 2 gO₂ m⁻³ aims at optimizing aeration energy consumption by maintaining the DO concentration in Ox1, Ox2 & Ox3 constant. The ammonium (NH_4^+) cascade control sets NH_4^+ concentration in the last aerobic reactor (Ox3) to 1 gN m^{-3} by manipulating the DO control set-point. Thus, during high influent NH_4^+ loads the DO setpoint established by the controller is higher and during low influent loads it is lower. The third controller is based on oxygen uptake rate (OUR) measures. The controller switches the aeration in Ox1, 2 & 3 Off when the system reaches endogenous conditions (an indication that there is no nitrogen or organic matter to oxidize), which are assumed achieved at a measured OUR of $650~\text{gO}_2~\text{m}^{-3}~\text{d}^{-1}$ in Ox1. When the aeration is On, the DO control at $2 \text{ gO}_2 \text{ m}^{-3}$ is activated. The mixed liquor suspended solids (MLSS) controller maintains the desired biomass concentration in the reactors (during winter 3500 gTSS m^{-3} and during summer actors (during winter 5500 grss in and during summer 4000 grSs m⁻³) by adjusting the wastage flow rate (Qw). The phosphate (PO₄⁻⁻) control aims at maintaining the PO₄³⁻ concentration in Ox3 at 1 gPO₄-P m⁻³ by adding ferric chloride. Finally, the last set of controllers aims at maintaining the nitrate (NO₃⁻) in Ax2 at 1 gN m⁻³ by manipulating either the internal recirculation (Q_{int}) or the flow of sodium acetate addition (Q_{carb}).

Fig. 2 illustrates the behaviour of some of the proposed controllers. Fig. 2a demonstrates that enhanced P precipitation with chemicals allows significantly reducing the PO_4^{3-} concentration down to 1 g P m⁻³ Fig. 2b shows that NO_3^- in Ax2 can be maintained at around 1 gN m⁻³ with proper manipulation of the internal recycle flow. Fig. 2c illustrates the benefits of including DO control by optimizing the use of aeration (which in the default strategy is poor at daytime and excessive at night time with values up to 5 gO₂ m⁻³). Finally, Fig. 2d shows that implementing NH_4^+ control has a smoothing effect on the peaks as higher DO concentrations in the reactor are imposed during NH_4^+ peaks.

3. LCA goal and scope

The goal of the LCA part is to compare the environmental impacts of different control options modelled with the Neptune Simulation Benchmark (NSB). The scope of the LCA involves the construction and operation of the WWTP presented in Fig. 1 including the disposal of the residues. The infrastructure (material stage, construction etc.) is considered in the analysis although we assumed that no extra equipment was needed to implement the different controllers. The functional unit is the treatment of one cubic metre wastewater. The transport of the final residue to landfill was considered as well as the transport from the production site of the materials/chemicals to the WWTP. The variables for the LCA evaluation include direct gaseous (e.g. CO2, CH4, N2O), and aqueous (e.g. TN, TP) and solid (slag) emissions from the biological treatment and the sludge handling. The emissions and resources required for e.g. generating electricity to run the entire WWTP, for producing the chemicals applied to the biological treatment (sodium acetate and ferric chloride) and to sludge treatment (see Table 2) are also incorporated into the evaluation. The CO₂ emissions from the biological treatment step include only the nonbiogenic carbon dioxide emissions from sodium acetate dosing, and not the biogenic carbon dioxide emissions (e.g. the CO₂ emissions generated from the oxidation of influent wastewater organics) as recommended by IPCC (2006).

4. Life cycle inventory

The inventory data is shown in Table 3 and was obtained after running simulations with the benchmark model NSB and applying the controllers described above. Emissions and consumptions related to the sludge handling can be estimated by using the sludge production- and emission factors listed in Table 2. It was assumed that sodium acetate was produced by mixing acetic acid (based on butane) with sodium hydroxide. The production of ferric chloride was based on the reaction of iron and hydrochloric acid. Data



Fig. 2. Behaviour of the different controllers studied: Phosphate controller with chemical addition (a), Nitrate controller by means of internal recycle (b), DO controller (c), Ammonium controller (d).

regarding the production of the other chemicals were taken from the Ecoinvent database (Ecoinvent, 2007).

The inventory shows that by implementing a DO control, electricity consumption in the water line is reduced slightly (5%) from 0.22 to ~0.21 kWh m⁻³ (values expressed per 1 m³ of treated wastewater), while maintaining similar nutrient loads in the effluent (TN_e and TP_e). With the NH₄⁺ controller, both electricity

consumption and TN_e are decreased compared to the reference situation (by 18% and 13% respectively). The OUR controller allows reducing energy consumption by 9%. The MLSS controller does not always show a clear benefit since the sludge retention time is not a limiting factor for nutrient removal in the studied system. The PO_4^{3-} control achieves a large reduction in TP_e from 9.47 to 1.20 gP m⁻³. These controllers not only increase denitrification rates but also

Table 3Results of some of the evaluation criteria for the 17 evaluated control strategies.

Stra	t	TN _e	TPe	Sodium acetate	Ferric chloride	Sludge production	Electricity water line	Electricity water line	Total CO ₂ emissions
Unit		${ m gN}~{ m m}^{-3}$	$\rm gP \ m^{-3}$	kg m^{-3}	$kg m^{-3}$	kgDM m ⁻³	kWh m ⁻³	kgCO ₂ e m ⁻³	$kgCO_2 e m^{-3}$
A ₁	Reference	13.19	9.47	0.00	0.00	0.39	0.22	0.14	0.15
A_2	DO	12.99	9.48	0.00	0.00	0.39	0.21	0.13	0.15
A_3	$DO + Q_{intr}$	12.70	9.09	0.00	0.00	0.39	0.20	0.12	0.14
A_4	$Q_{intr} + NH_4$	11.48	8.15	0.00	0.00	0.40	0.18	0.11	0.14
A_5	$Q_{intr} + NH_4 + M_{LSS}$	11.26	8.02	0.00	0.00	0.40	0.18	0.11	0.14
A_6	$DO + Q_{carb}$	9.25	5.73	0.08	0.00	0.42	0.24	0.15	0.32
A_7	$Q_{carb} + NH_4$	8.66	5.31	0.07	0.00	0.43	0.23	0.14	0.29
A ₈	$Q_{carb} + NH_4 + M_{LSS}$	9.44	5.84	0.05	0.00	0.43	0.22	0.14	0.26
A ₉	$DO + PO_4$	12.88	1.20	0.00	0.049	0.40	0.21	0.13	0.30
A ₁₀	PO ₄ +NH ₄	11.94	1.19	0.00	0.045	0.41	0.20	0.12	0.28
A ₁₁	$PO_4 + NH_4 + M_{LSS}$	11.87	1.19	0.00	0.044	0.41	0.20	0.12	0.28
A_{12}	$Q_{intr} + OUR$	12.42	8.88	0.00	0.00	0.39	0.20	0.12	0.14
A ₁₃	$Q_{intr} + OUR + M_{LSS}$	12.61	8.86	0.00	0.00	0.39	0.20	0.12	0.15
A ₁₄	$Q_{carb} + OUR$	9.27	5.77	0.10	0.00	0.42	0.24	0.15	0.37
A ₁₅	$Q_{carb} + OUR + M_{LSS}$	10.15	5.58	0.09	0.00	0.43	0.24	0.15	0.34
A ₁₆	PO ₄ +OUR	12.37	1.19	0.00	0.047	0.40	0.21	0.13	0.29
A ₁₇	$PO_4 + OUR + M_{LSS}$	12.74	1.18	0.00	0.046	0.40	0.21	0.13	0.29

Strat: strategy; TN_e : effluent total nitrogen; TP_e : effluent total phosphorus; DO: dissolved oxygen control; Q_{intr} : internal recycle control, NH_4 : cascade ammonium control; M_{LSS} : mixed liquor suspended solids control; Q_{carb} : carbon addition control; PO₄: phosphate addition control; OUR: oxygen uptake rate control; DM: Dried Matter. Values are expressed per m³ of treated wastewater.

improve phosphorus removal. While Q_{int} coincides with reductions of TN_e and TP_e of ~ 15%, Q_{carb} achieves reductions down to ~ 50% for TN_e and down to 40% for TP_e. The addition of sodium acetate (Q_{carb}) increases sludge production up to 0.43 kgDM m⁻³, increases electricity consumption in the water line and provokes the largest CO₂ emissions (0.37 kgCO₂e m⁻³ in strategy A₁₄). From the inventory data we can observe that high nutrient removal efficiencies increase the overall CO₂ emissions as a trade-off.

5. Life cycle impact calculation

The impacts were calculated by using a version of the LCA modelling tool GaBi 4 (PE, 2008) with the integrated LCA database EcoInvent 2.0. The EDIP 97 methodology (Hauschild and Wenzel, 1998) was used considering the following impact categories: acidification (AC), soil ecotoxicity (SE), water ecotoxicity (WE), global warming (GW), human toxicity via air (HTA), human toxicity via soil (HTS) human toxicity via water (HTW), nutrient enrichment (NE), stratospheric ozone depletion (OD) and photochemical ozone formation (PO).

The impact potentials for each variable were calculated based on the principles of life cycle impact assessment (LCIA), i.e. classification, characterisation, normalisation and weighting, as described in the LCA standard (ISO 14040, 2006). In order to evaluate the impacts for the site-dependent conditions for the NE category (further information in the following section), the characterization, normalization and weighting steps were calculated for three scenarios: N&P-deficient. P-deficient and N-deficient ecosystems. Nutrients that were not limiting for each scenario were not taken into account in the characterization of NE. For instance, in the case of freshwaters, characterization factors for ammonium and nitrates were 0 as they are P-limited ecosystems. For global warming calculations, the marginal technology for energy production selected was based on natural gas. All normalization references used in this study are based on data for Europe (EU-15) except for global warning and ozone depletion which are based on global data (Stranddorf et al., 2005) (see Supp Information). For the first part of this study it has been chosen to give the same weight to all impact potentials, i.e. 1. Since the EDIP 97 methodology is used, the weighted person-equivalents are called targeted, i.e. personequivalents-targeted (PET) and the weighted impact potentials are therefore expressed in the unit milli-person-equivalentstargeted regarding one year (mPET year). Further details on the quantifications can be found in the Supplementary Material. Inventory data (Table 3) combined with the impact potentials (Table 3 in Supplementary Material) were then used to calculate the induced and avoided impacts for the evaluated control strategies compared to the reference situation A_1 (see Fig. 3).

6. Life Cycle Assessment interpretation

The selection of the more appropriate nutrient control strategy highly depends on the site-specific conditions. Looking at the NE impact category it is important to discriminate the exposed ecosystems (where WWTPs discharge) according to the limiting nutrient (Hauschild et al., 2008), i.e. the one which limits the growth of primary producers. Phosphorus is the limiting factor for plant growth in many freshwater ecosystems, whereas nitrogen is more commonly the key limiting nutrient of marine waters. Other ecosystems such as coastal zones can be limited by either nitrogen or phosphorous or both (Gallego et al., 2010). Therefore, decisions about wastewater nutrient removal might change depending on the type of receiving water body. Therefore, the LCI analysis is repeated for three different scenarios: i) N&P-deficient, ii) P-deficient and iii) N-deficient systems.

6.1. N&P-deficient scenario (Fig. 3a)

In N&P-deficient systems any control action leads to reduced environmental impact since the avoided impacts of all strategies are larger than the induced impacts. The strategies including chemical addition (from A_6 to A_{11} and from A_{14} to A_{17}) present better results compared to the ones without chemical addition. This is mainly due to the significant reductions in the effluent N and P loads (see Table 3). The strategies showing best environmental performance comprise the addition of ferric chloride (A₉, A₁₀, A₁₁, A₁₆, A₁₇), reaching the highest difference between avoided and induced impacts (around 2 mPET year m⁻³). Implementing control for reducing energy consumption (strategies A₂, A₃, A₄, A₅, A₁₃ and A₁₄) is beneficial but does not lead to optimal environmental performance. The highest avoided impact is NE and the highest induced impact is on HT (on soil and water) categories. HT increase is caused by i) the increased sludge production which increments the sludge treatment impacts and ii) the production of sodium acetate and ferric chloride.

6.2. P-deficient scenario (Fig. 3b)

Avoided impacts are an order of magnitude larger compared to the N&P-deficient scenario as a result of the normalization step. For N&P-deficient systems nIP_{ne,1} = 269 and nIP_{ne,2} = 37.2, whereas for P-deficient systems nIP_{ne,1} = 2464 and nIP_{ne,2} = 0 (see Supplemental Material for the specific calculations), i.e. nIP_{ne,1} for Pdeficient systems is eight times larger. Again, any control actions promoting both energy optimization and nutrient removal result in better environmental performance. The best performance is achieved by the strategies adding ferric chloride to increase phosphorus removal at the WWTP with absolute differences between induced and avoided impacts of ~20 mPET year m⁻³.

6.3. N-deficient scenario (Fig. 3c)

This last scenario does not show large environmental benefits from the implementation of the proposed controllers. The best strategy is A₅, with an absolute difference of 0.08 mPET year m⁻³. This strategy includes DO, internal recycle, NH₄⁺ and MLSS controllers, which reduce energy consumption and total nitrogen discharges (compared to A₁). Implementing an OUR controller (A₁₂ and A₁₃) is not as effective as the NH₄⁺ controller for this case. The strategies with sodium acetate addition do not always result in larger avoided impacts compared to the induced ones. Therefore, it is shown that there is not always environmental benefit in adding sodium acetate to decrease total nitrogen loads in the effluent. In the same way, the addition of ferric chloride for P removal has a negative impact on the environment in N-deficient scenarios.

Overall, NE is a dominating factor which has a strong influence on the selection of best operating strategies. This can be explained by the relatively high share of the NE normalization reference, which is related to WWTP emissions, whereas other normalization references (e.g. GW) are dominated by other societal activities like transport. As there is high uncertainty in the N₂O emission factors LCIA calculations were also conducted using the much higher factor 0.03 kg N₂O kgTN removed⁻¹ (Foley et al., 2008). The obtained results (see supporting Information) showed that although the total CO₂ equivalent emissions were almost doubled, no change in the final LCA outcomes was observed.

7. The importance of LCA weighting in decision-making

In order to reflect the relative importance of different impact categories on the results, weights can be assigned after the



Fig. 3. Impact for each of the categories considered for the strategies A1 to A17. a) N&P-deficient scenario, b) P-deficient scenario, c) N-deficient scenario.

normalization step. In general terms the criterion for weighting addresses the assumption that potential impacts in one impact category may be considered as more severe than potential impacts in other impact categories. Using a general Weighting Factor (WF) of 1 for all impact categories means that the severity of all categories is considered equal. However, weighting factors may also be determined by a method called 'distance to political reduction targets', i.e. a ratio between the potential impact in the reference year (i.e. the year of normalisation) and the potential impact in the target year (i.e. the year at which the reduced impact should be achieved) as described for the EDIP method (Hauschild and Wenzel, 1998).

In this study we test whether it is important or not to care about the weighting by performing a global sensitivity analysis (GSA, Neumann, 2012) on the weights of the impact categories. For the GSA, Monte Carlo calculations (200 iterations) were conducted by specifying ranges for the weighting factors, sampling from these ranges and propagating the sampled values through the LCIA model. The weighting factors were characterized using uniform probability density functions, with the minimum values set at 1 and the maximum values set at the values provided by Stranddorf et al. (2005) using the 'distance to political reduction targets' approach. For global impacts (GW and OD) the worldwide weighting factors were used, whereas for regional and local impacts the EU-15 weighting factors were applied. The specific GSA method used is based on calculating standard regression coefficients (SRCs) between the GSA inputs (the weighting factors) and the outputs, i.e. the Net Environmental Impact (NEI). The NEI is the difference between the induced and the avoided impacts. Negative values indicate that the impact is reduced compared to A₁ while positive values correspond to increased impacts. The higher the absolute values of the SRC, the stronger the influence of the corresponding weight in determining the NEI.

Fig. 4 shows the absolute SRC values (from 0 to 1) for the three scenarios and for each strategy (from A_2 to A_{17}). It can be seen clearly that NE is the impact category that is most sensitive to the weighting used (SRC values close to 1, in the outer circle), especially for N&P and P-deficient systems. However, for N-deficient systems and in



Fig. 4. Standard Regression Coefficients from the GSA on the weights of the impact categories. Ranges for the weights are: AC = [1-1.27], SE = [1-1], WE = [1-1.18], GW = [1-1.05], HTA = [1-1.4], HTS = [1-1.23], HTW = [1-1.3], NE = [1-1.22], OD = [1-63], PO = [1-1.33]. Lines which are not visible obtained SRCs close to 0.



Fig. 5. Sensitivity of Net Environmental Impact to weighting factors for the strategies A1 to A17. a) N&P-deficient scenario, b) P- deficient scenario, c) N-deficient scenario.

particular the strategies with ferric chloride addition (A₉, A₁₀, A₁₁, A₁₆ and A₁₇) changes in the NE weighting do not imply changes in the NEI. For these strategies, HTW and HTS impacts become the most sensitive to the weighting factors. As the evaluated LCIA model is linear all r^2 values obtained from the regression were 1.

Fig. 5 shows the NEI for the three different scenarios, after propagating variability in the weighting factors through the LCIA model. It can be seen that the choice of weighting does not influence the selection of the best strategy for N&P and P-deficient scenarios since the interquartile ranges of the box plots do not overlap with competing groups of strategies. It is confirmed that best strategies involve ferric chloride addition. These interquartile ranges are larger for the strategies leading to largest reductions in NE, which is the most sensitive impact category. For N-deficient scenarios the selection of the weights has an effect on the ranking of some strategies (i.e. A_6 , A_7 , A_8). In these cases, some combinations of weights lead to negative NEI values (being better strategies compared to A_1), but for other combinations positive NEI values are obtained. It is clear though, that A_4 and A_5 are the best strategies for N-deficient scenarios.

8. Conclusions

Decision-making in controlling wastewater nutrient removal systems can be assessed using a combination of mechanistic process models together with LCIA models. The use of site-specific conditions for the NE impact category is essential to define best environmental performance strategies. In the evaluated case-study, strategies that promote increased nutrient removal and/or energy savings in WWTPs present an environmental benefit for N&P and Pdeficient systems. This is not the case in N-deficient systems where the use of chemicals (even for improving N removal efficiencies) would not always be beneficial for the environment. A sensitivity analysis applied to the weights of the impact categories showed that the choice of weighting factors would only influence decision making for N-deficient systems.

Acknowledgements

This research is supported by the European Union 6th framework project NEPTUNE and the NSERC Special Research Opportunities grant as part of the Canadian contribution to NEPTUNE (Contract-No. 036845). Lluís Corominas received funding from the Government of Spain (jci-2009-05604; CTM-2009-13018) and the Career Integration grant (PCIG9-GA-2011-293535) from EU. Peter Vanrolleghem holds the Canada Research Chair on Water Quality Modelling. We acknowledge Leiv Riger for the helpful discussions in the initial stages of this research.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2013.06.002.

References

- Ahn, J.H., Kim, S., Park, H., Rahm, B., Pagilla, K., Chandran, K., 2010. N2O emissions from activated sludge processes, 2008-2009: results of a national monitoring survey in the United States. Environ. Sci. Technol. 44, 4505–4511.
- Azevedo, L.B., Cosme, N., Elshout, P.M.F., Larsen, H.F., Leuven, R.S.E.W., Hauschild, M.Z., Hendriks, A.J., Huijbregts, M.A.J., van Zelm, R., 2012. Recommended Assessment Framework, Method and Characterisation and Normalisation Factors for Non-toxic Pollutant Impacts: Phase 2 (Report, Model and Factors): Ecosystem Impacts of Eutrophying Emissions. DRAFT. Deliverable 3.2 (T3.1: Aquatic Eutrophication). EC Project "LC-IMPACT", Project No.: 243827 FP7-ENV-2009-1. www.lc-impact.eu.

- Basset-Mens, C., Anibar, L., Durand, P., van der Werf, H.M.G., 2006. Spatialised fate factors for nitrate in catchments: modelling approach and implication for LCA results. Sci. Total Environ. 367 (1), 367–382.
- Corominas, L.L., Foley, J., Guest, J.S., Hospid, A., Larsen, H.F., Morera, S., Shaw, A., 2013. Life cycle assessment applied to wastewater treatment: state of the art. Water Res. http://dx.doi.org/10.1016/j.watres.2013.06.049.
- Corominas, L., Flores-Alsina, X., Snip, L., Vanrolleghem, P.A., 2012. Comparison of different modeling approaches to better evaluate greenhouse gas emissions from whole wastewater treatment plants. Biotechnol. Bioeng. 109 (11), 2854–2863.
- Ecolnvent, Ecolnvent v. 2.0 (2.01), 2007. Database on Life Cycle Inventory Data. Swiss Centre for Life Cycle Inventories. The Ecoinvent Centre – a Competence Centre of ETH, PSI, Empa and ART. http://www.ecoinvent.org/home.
- Finnveden, G., Hauschild, M.Z., Ekvall, T., Guinée, J., Heijungs, R., Hellweg, S., Koehler, A., Pennington, D., Suh, S., 2009. Recent developments in life cycle assessment. J. Environ. Manage. 91 (1), 1–21.
- Flores-Alsina, X., Gallego, A., Feijoo, G., Rodriguez-Roda, I., 2010. Multiple-objective evaluation of wastewater treatment plant control alternatives. J. Environ. Manage. 91 (5), 1193–1201.
- Foley, J., Lant, P., Donlon, P., 2008. Fugitive greenhouse gasemissions from wastewater systems. Water J. Aust. Water Assoc. 38 (2), 18–23.
- Gallego, A., Rodríguez, L., Hospido, A., Moreira, M.T., Feijoo, G., 2010. Development of regional characterization factors for aquatic eutrophication. Int. J. Life Cycle Assess. 15, 32–43.
- Gernaey, K.V., Jeppsson, U., Vanrolleghem, P.A., Copp, J.B., Steyer, J.P., 2013. Benchmarking of Control Strategies for Wastewater Treatment Plants. IWA Scientific and Technical Report. IWA Publishing, London, ISBN 9781843391463.
- Gernaey, K., Mussati, M., Yuan, Z., Nielsen, M.K., Jørgensen, S.B., 2002. Control strategy evaluation for combined N and P removal using a benchmark wastewater treatment plant. In: Proceedings of 15th IFAC World Congress for Automatic Control. Barcelona, Spain.
- Gernaey, K.V., Flores-Alsina, X., Rosen, C., Benededtti, L., Jeppsson, U., 2011. Phenomenological modelling approach to generate WWTP disturbance scenarios. Environ. Modell. Softw. 26 (11), 1255–1267.
- Guest, J.S., Skerlos, S.J., Barnard, J.L., Beck, M.B., Daigger, G.T., Hilger, H., Jackson, S.J., Karvazy, K., Kelly, L., Macpherson, L., Mihelcic, J.R., Pramanik, A., Raskin, L., van Loosdrecht, M.C.M., Yeh, D., Love, N.G., 2009. A new planning and design paradigm to achieve sustainable resource recovery from wastewater. Environ. Sci. Technol. 43, 6126–6130.
- Guinée, J.B., Heijungs, R., Huppes, G., Zamagni, A., Masoni, P., Buonamici, R., Ekvall, T., Rydberg, T., 2011. Life cycle assessment: past, present, and future. Environ. Sci. Technol. 45 (1), 90–96.
- Hauschild, M.Z., Goedkoop, M., Guinée, J., Heijungs, R., Huijbregts, M., Jolliet, O., Margni, M., De Schryver, A., Bersani, R., 2008. Identification of best practice. Development of basis for a recommended LCIA methodology for the European commission. In: 18th SETAC Europe Annual Meeting, Warsaw, 26–29 May.
- Hauschild, M.Z., Wenzel, H., 1998. Environmental Assessment of Products, Scientific Background, vol. 2. Chapman & Hall, UK.
- Henze, M., Gujer, W., Mino, T., van Loosdrecht, M.C.M., 2000. Activated Sludge Models: ASM1, ASM2, ASM2d and ASM3. Scientific and Technical Report No. 9. IWA Publishing, London, UK.
- IPCC, 2006. Wastewater treatment and discharge. In: Eggleston, H.S., Buendia, L., Miwa, K., Ngara, T., Tanabe, K. (Eds.), 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Prepared by the National Greenhouse Gas Inventories Programme, Waste, vol. 5. IGES, Japan. (Chapter 6).
- ISO 14040, 2006. Environmental Management Life Cycle Assessment Principles and Framework: International Standard 14040. International Standards Organisation, Geneva.
- Kalbar, P.P., Karmakar, S., Asolekar, S.R., 2012. Selection of an appropriate wastewater treatment technology: a scenario-based multiple-attribute decisionmaking approach. J. Environ. Manage. 113, 158–169.
- Larsen, H.F., Hansen, P.A., Boyer-Souchet, F., 2010. Decision Support Guideline Based on LCA and Cost/Efficiency Assessment. Deliverable 4.3. EC Project "NEPTUNE", contract No.: 036845. www.eu-neptune.org.
- Muga, H.E., Mihelcic, J.R., 2008. Sustainability of wastewater treatment technologies. J. Environ. Manage. 88 (3), 437–447.
- Neumann, M.B., 2012. Comparison of sensitivity analysis methods for pollutant degradation modelling: a case study from drinking water treatment. Sci. Total Environ. 433, 530–537.
- Otterpohl, R., Freund, M., 1992. Dynamic Models for clarifiers of activated sludge plants with dry and wet weather flows. Water Sci. Technol. 26 (5–6), 1391–1400.
- Olsson, G., Nielsen, M.K., Yuan, Z., Lynggaard-Jensen, A., Steyer, J.P., 2005. Instrumentation, Control and Automation in Wastewater Systems. Scientific and Technical Report No. 15. IWA Publishing, London, UK.
- PE, 2008. GaBi 4. Software-system and Databases for Life Cycle Engineering. Echterdingen, Copyright, TM. Stuttgart. www.gabi-software.com.
- Rieger, L., Koch, G., Kühni, M., Gujer, W., Siegrist, H., 2001. The EAWAG bio-P module for activated sludge model No. 3. Water Res. 35 (16), 3887.
- Stranddorf, H., Hoffmann, L., Schmidt, A., 2005. Impact Categories, Normalisation and Weighting in LCA. Environmental News no. 78 from the Danish Environmental Protection Agency, Copenhagen.
- Takács, I., Patry, G.G., Nolasco, D., 1991. A dynamic model of the clarificationthickening process. Water Res. 25 (10), 1263.
- Tchobanoglous, G., Burton, F.L., Stensel, H.D., 2003. Wastewater Engineering: Treatment, Disposal and Reuse. McGraw-Hill Higher Education, New York.