

GREENHOUSE GASES FROM WASTEWATER TREATMENT SYSTEMS

The energy versus nitrous oxide emissions nexus

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ABSTRACT

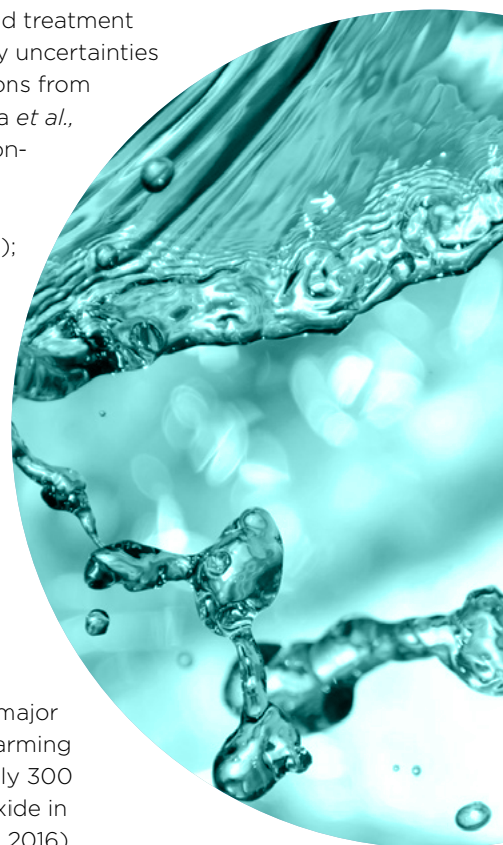
Engineering WWTPs to reduce electrical energy use and achieve nitrogen removal through 'short-cut' biological pathways, such as 'nitrification', may be misguided in terms of life cycle impacts, including greenhouse gas (GHG) emissions. Paradoxically, some of the 'novel' wastewater treatment processes receiving a lot of attention in the industry for reasons of cost, energy and/or effluent nitrogen reduction, have a significantly higher risk of nitrous oxide (N_2O) emissions. Without active mitigation measures, it is likely that increased N_2O emissions will cancel out (or exceed) the benefits of reduced Scope 2 (indirect) emissions associated with lower use of grid electricity. Furthermore, the use of a fixed emission factor for N_2O from wastewater treatment, as applied in many GHG calculation protocols (e.g. including the current NGER (2016) Scheme in Australia), is flawed. At best, the current NGER (2016) emission factor (4.9 tonnes CO_2 -e per tonne N removed) might overestimate actual emission for systems with lower actual N_2O profiles. At worst, it is likely to underestimate (potentially by a large margin) the actual N_2O for processes that typically have high N_2O emissions profiles.

INTRODUCTION

Climate change abatement and related energy efficiency drivers have seen an increased focus on greenhouse gas emissions (GHG) for more than ten years. The water industry in Australia is embracing initiatives toward improving energy efficiency and GHG reduction, both at the utility level (e.g. Donaghy, 2017; YVW, 2017; WSAA, 2016) and, in some cases, state level (e.g. DELWP, 2016). From a life cycle perspective in urban water systems,

looking at the traditional infrastructure scenario (i.e. without seawater desalination, and with limited use of rain tanks or water recycling), wastewater collection and treatment typically dominates the global warming and ozone depletion impact potentials (Lane *et al.*, 2015). However, there are a number of significant uncertainties in the estimation of greenhouse emissions from wastewater collection and treatment systems. Some of the key uncertainties include: methane emissions from sewer systems (Guisasola *et al.*, 2008; Liu *et al.*, 2015); non-biogenic carbon sources in the sewage organic material (Law *et al.*, 2013); the extent of methane direct emissions (including losses of biogas produced) in treatment processes (Foley and Lant, 2007; de Haas *et al.*, 2008); and direct nitrous oxide emissions from treatment processes (Kampschreur *et al.*, 2009; Ahn *et al.*, 2010; Foley *et al.*, 2010).

Nitrous oxide (N_2O) is a major GHG and has a global warming potential of approximately 300 times that of carbon dioxide in equivalent terms (NGER, 2016).



Citing earlier data (for 1990), Kampschreur *et al.* (2009) noted that the global emissions from human sewage treatment was estimated to be 3.2% of total anthropogenic N_2O emissions. Kampschreur *et al.* (2009) also noted that N_2O from wastewater handling was estimated (by others in 2008) to contribute 26% to the total greenhouse gas emissions (carbon dioxide, methane and N_2O) of the water supply chain (i.e. drinking water production, transport, wastewater treatment, sludge treatment and discharge). Similarly, in the Australian context, Lane *et al.* (2015) undertook a comprehensive life analysis of the urban water cycle, based on inventory data for the City of Gold Coast. Their results for the 'traditional infrastructure mix' (see above) suggested that fugitive direct emissions from wastewater treatment and discharge accounted for 15% of the global warming potential for the entire urban water system. This included fugitive methane emissions from sewers and dams and a range of indirect emissions (power and chemicals use, biosolids disposal, construction). For a water utility in Australia now, with wastewater treatment systems that are classified as 'fully aerobic', reporting obligations under the National Greenhouse and Energy Reporting Scheme (NGER, 2016) will typically be largely made up of Scope 1 (N_2O) and Scope 2 (grid electricity) components. In that case, wastewater N_2O emissions might make up a significant proportion of the reported total emissions for a water utility. For example, excluding Scope 3 emissions (for which reporting is voluntary), one major water utility in Australia (SA Water, 2014) reported Scope 1 emissions to be 23% of its total emissions for Scopes 1 and 2 in the 2013-14 period. However, the N_2O contribution to reported Scope 1 estimates will be dependent on the relevant emission factor adopted.

LITERATURE REVIEW

Accurately defining the extent to which N_2O emissions from wastewater treatment are significant on a global scale is difficult. There are uncertainties around the N_2O emission factors and historical ambiguities over how these factors were derived or have been applied in calculation protocols (Kampschreur *et al.*, 2009). Moreover, research has shown that measured N_2O emission rates vary widely according to type of treatment systems or unit processes, and can also vary widely both spatially and temporally within a given process (Kampschreur *et al.*, 2008; Foley *et al.*, 2009; Law *et al.*, 2012; Ye *et al.*, 2014; Ni *et al.*, 2015; Pan *et al.*, 2016).

N_2O emission factors for wastewater treatment have been variously reported in the literature as: percent of influent nitrogen load (Total Kjeldahl N or Total N); percent of Total N (TN) removed; or normalised to unit population equivalent on flow rate ($g N_2O/PE/year$). Expressing the emission factor as % influent TN is probably the simplest and most commonly reported approach. The IPCC (2006) method applied a value of 0.5% of nitrogen load, regardless of whether the nitrogen is converted in the treatment plant or in effluent-receiving rivers and estuaries (Kampschreur *et al.*, 2008). In Australia, the NGER (2016) determination, like its predecessors, uses an emission factor for wastewater treatment (4.9 tonnes CO_2 -e per tonne N) that is based on 1% of TN removed across a treatment plant. A separate (lower) emission factor is applied for the nitrogen discharged in the form of treated effluent, depending on the type of receiving waters. If the extent of TN removal for a given plant is known, then it is a simple matter to convert emission factors from % TN removed to % influent TN. However, the extent of N removal across different treatment processes is not always reported. When interpreting wastewater treatment data, care needs to be taken to ensure that mass balances are appropriately considered.

Kampschreur *et al.* (2009) reviewed N_2O emission factors from full-scale and lab-scale systems. Some lab-scale systems had extremely high emission factors (e.g. 16% to 90% of influent TN). The range for full-scale systems was 0.001% to 15% of influent TN, with typical averages being approximately 0.01-0.6% (Kampschreur *et al.*, 2009). Kampschreur *et al.* (2008) undertook a detailed analysis of a large (620,000 EP) plant in Rotterdam (Netherlands) with nitrification-anammox sidestream sludge liquor treatment and mainstream separate nitrification activated sludge systems.

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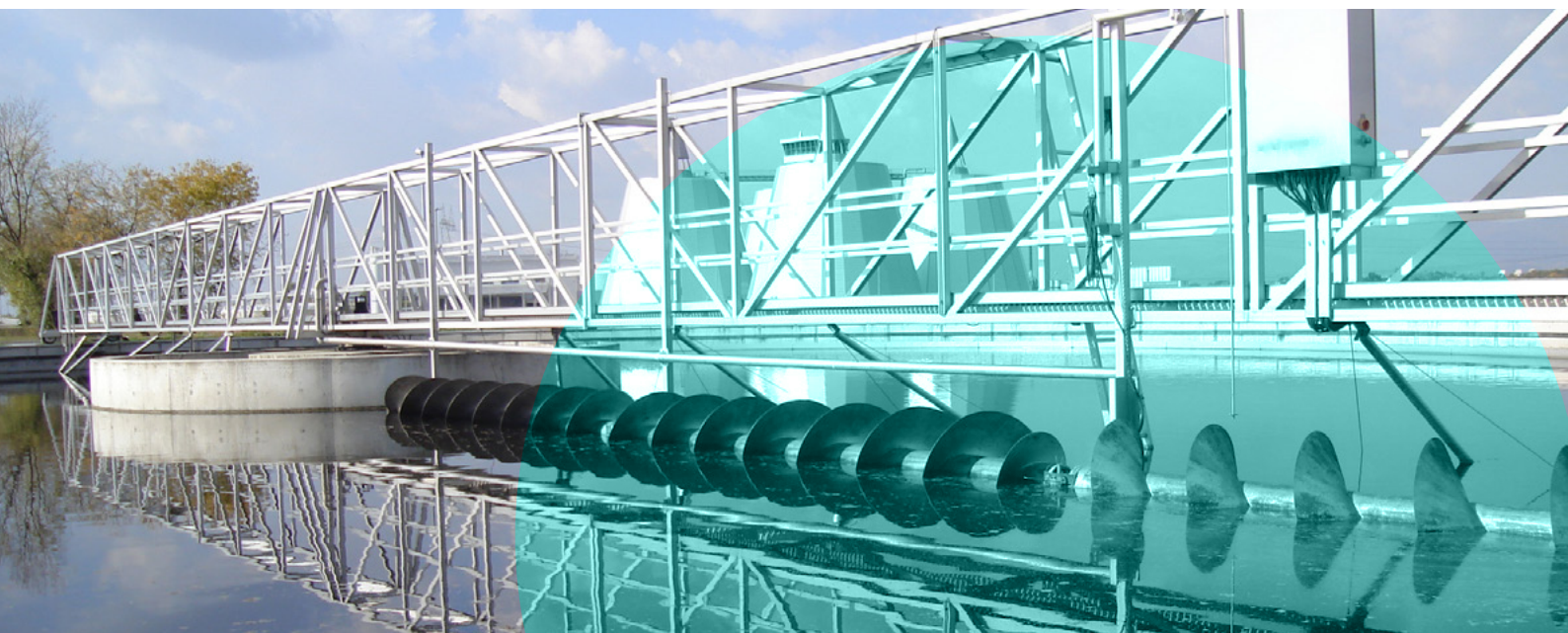
They found that these systems had significantly higher total emission rates, in the order of 5% of influent TN, with the majority (83% of average total N_2O produced or 4.2% of influent TN) coming from the 'B-stage' of the so-called 'A-B' activated sludge process. Other process units made smaller but nevertheless significant contributions (7-8% of average total N_2O produced; and similarly, 0.3-0.4% each from the A-stage and sidestream nitrification reactors).

Foley *et al.* (2009) reported N_2O emissions measured in seven wastewater treatment plants (WWTPs) in Australia. The main process systems included an oxidation ditch, a sequencing batch reactor (SBR) and several compartmentalised continuous-flow configurations ("Johannesburg", "A2/O" or 3-Stage Phoredox", and three different "MLE" types). Their results were expressed as $\text{kgN}_2\text{O-N/kgN}$ denitrified (i.e. percent TN removed) but average TN removal performance was also reported. Converting the Foley *et al.* (2009) results to % influent TN, average N_2O emissions factors were in the range 0.6-1.3% for the oxidation ditch; 0.7- 5.0% for the SBR; 1.0- 2.0% for the "Johannesburg" process; 1.0- 1.7% for the "A2/O" process; and 0.5- 8.5% for the MLE types. However, the MLE-types were more widely represented in the survey and in two measurement rounds showed extremely high emissions. One MLE process gave an average emission factor of 20% of influent TN (or 25% of TN removed) in one round, but 0.5- 0.8% of influent TN in the other two rounds. Similarly, another MLE process gave an average emission factor of 7.0- 8.5% of influent TN in two rounds but 0.6-2.0% in a further two rounds. Foley *et al.* (2009) did not specifically investigate the underlying causes of the variation in emission factors. However, process loading (particularly specific ammonia oxidation rate and influent COD:N ratio) as well as operation (including aeration rate,

dissolved oxygen control, and recycles) are known to contribute to instantaneous N_2O emission rates (Law *et al.*, 2012; Ni *et al.*, 2015; Andalib *et al.*, 2017).

Ahn *et al.* (2010) quantified N_2O emissions from twelve WWTPs in the USA and reported average emission factors in the range 0.01- 1.8% of influent TKN (or 0.01- 3.3% of TN removed). Plant configurations with step-feed activated sludge processes were reported by Ahn *et al.* (2010) to be amongst the highest emitters (up to 2.6% of influent TKN or 4.8% of TN removed). Pan *et al.* (2015) measured N_2O emissions from a step-feed 'plug-flow' activated sludge plant in Australia that achieves, on average, 75% TN removal. The emission factor for the plant overall was 1.9% of influent TN, with the majority (80% of this) emitted from the second 'feed step' in the process. Ni *et al.* (2015) applied a mathematical model to explain the mechanism underlying the large variations (spatial and temporal) in emissions from the same plant. Depending on the sample location, Ni *et al.* (2015) attributed most of the N_2O emissions to the ammonia oxidising bacteria (using nitrifier denitrification and/or hydroxylamine pathways) in the aerated zones.

Schaubroeck *et al.* (2015) applied life cycle assessment (LCA) to data from the Strass WWTP in Austria. This plant is well known as an example of energy-efficient wastewater treatment with nutrient removal. It achieves close to 'net energy neutral' operation by maximising the recovery of heat and electricity from biogas via on-site cogeneration (Wett, 2007). The plant incorporates a two-stage (A-B) activated sludge process. According to Schaubroeck *et al.* (2015), the mainstream B-Stage process at the plant was reported to emit very little N_2O (0.0073% of influent TN) when operating in nitrification-denitrification mode.



The N₂O emission factor of the overall plant for this baseline configuration was approximately 0.3% of influent TN (inventory input data from Schaubroeck *et al.*, 2015). When sidestream treatment for digestate (sludge treatment return liquors) using a nitrification-denitrification system was implemented, overall plant N₂O emissions increased to approximately 1.4% of influent TN (data from Schaubroeck *et al.*, 2015). The sidestream treatment process was subsequently changed to a nitrification-anammox (DEMON™) configuration and Schaubroeck *et al.* (2015) reported somewhat lower overall plant N₂O emissions for this scenario (0.42% of influent TN, i.e. closer to the baseline scenario). When mainstream deammonification (anammox) was implemented, the inventory data showed an increased level of N₂O emissions (2.2% of TN load for the mainstream or 2.5% of influent TN load for the plant overall, including sidestream nitrification-anammox/DEMON™). The increase in N₂O emissions was linked with possible nitrite accumulation, as a result of nitrification (Schaubroeck *et al.*, 2015). Citing Ahn *et al.* (2010), Schaubroeck *et al.* (2015) noted that lower emissions might be possible by biological adaptation and improved process control.

From their LCA study, for a notionally 'best practice' energy-self-sufficient plant in Austria that recycles nutrients to agriculture through stabilised biosolids (digestate), Schaubroeck *et al.* (2015) found that wastewater treatment processes might have a number of environmental benefits (e.g. preventing resource extraction) but also lead to damaging effects via climate change and heavy metal toxicity. Schaubroeck *et al.* (2015) concluded that it was not yet possible to consider the studied system as 'environmentally friendly'. They specifically identified N₂O emissions as a major contributor of impact potential for climate change that would need to be restrained through process optimisation.

OBJECTIVE

A review of the literature therefore raises an intriguing question. For a given level of nitrogen removal, what are the trade-offs, in terms of the configuration of wastewater treatment, between conserving and recovering energy versus the risk of N₂O emissions? This desktop study aimed to use a life cycle inventory approach to address this question by attempting to quantify likely direct (Scope 1) N₂O emissions and those related to imported (grid) electricity use (Scope 2). Uncertainties in key assumptions and the method of accounting for N₂O emissions (e.g. NGER, 2016) were also examined.

METHODOLOGY

Six different wastewater treatment process configurations were simulated using the BioWin™ commercial software (Envirosim, 2017). All configurations had a common set of variable raw influent parameters (flow and concentration) over a 7-day period, based on actual historical data for a full-scale plant in eastern Australia. The average flow rate was 50 ML/d and average (steady-state) COD and TKN concentrations were 539 mg/L and 48 mgN/L respectively. All configurations incorporated primary treatment (sedimentation), followed by one of three possible activated sludge process configurations, with co-digestion of thickened primary sludge and waste activated sludge via anaerobic digestion. Apart from the activated sludge processes, the plant configurations differed in terms of sidestream/ mainstream processes applied (refer to Table 1). All mainstream activated sludge processes were simulated with a nominal sludge age in the range 10 to 12 days. Ideal secondary sedimentation (99.8% solids capture) was applied with RAS rate paced at 70% of influent flow. Anaerobic digestion solids retention time was deliberately conservative (40 ± 2 days) so as to optimise biogas production, and hence on-site energy production, for each scenario. All simulations were carried out at the following temperatures: mainstream reactors at 20°C; mesophilic anaerobic digestion at 35°C; sidestream nitrification reactors at 32°C and sidestream denitrification or anammox reactors at 31°C. Effluent (TN) predictions were modelled and typically averaged 8 to 16 mgN/L. Aeration (including dissolved oxygen) and solids capture model parameters were adjusted to reflect likely operational values for a given process configuration, based on experience. All other model parameters were set at default values.

The six overall plant configurations simulated are summarised in Table 1. Configurations 1 to 3 were set up to be similar to the scenarios for the Strass WWTP modelled by Schaubroeck *et al.* (2015).

BioWin™ simulation outputs were used as the inventory data for calculation of nitrogen and solids mass balances, including biogas production. Electrical energy use, energy production from biogas, net electricity import from the grid and GHG emissions (N₂O Scope 1; and grid electricity use, Scope 2) were all calculated outside BioWin™ using a dedicated spreadsheet tool developed for this study. The spreadsheet tool used @RISK™ add-on software for combined uncertainty calculations. The key model input uncertainties are given in Table 2 (see

Supplementary Information). Specific electrical energy use for Configuration Nos. 1 to 3 was selected to closely match the breakdown (into 'mainstream', 'sidestream' and 'other' process units) of flow-specific average values (in kWh/ML) stated by Schaubroeck *et al.* (2015) for comparable configurations. On the same basis, in the breakdown energy use for Configuration Nos. 3 and 4, commonality of flow-specific average values for sidestream process units was assumed. Specific electrical energy use for the mainstream in Configuration Nos. 4 to 6 was based on experience and informed by WWTP energy benchmarking in Australia (de Haas *et al.*, 2015). Electrical energy production from biogas was calculated, assuming reciprocating engines with overall electrical efficiency as a model uncertainty (refer to Table 2 in **Supplementary Information**). Waste heat recovery from co-generation was assumed to be partially used

for digester heating, but no further heat recovery was included in the model.

Uncertainty was modelled using the @RISK™ software add-on in MS-Excel™. The variable model parameters are listed in Table 2 (**Supplementary Information**). Distributions for the variable parameters were generated in the model using the PERT function in @RISK™. Somewhat similar to a triangular distribution, the PERT distribution emphasises the "most likely" value over the minimum and maximum estimates, but is designed to more closely resemble a realistic probability distribution. Depending on the values provided, the PERT distribution can provide a close approximation of normal or lognormal distributions. For comparative purposes, GHG emissions were also calculated by following the NGER (2016) Method 2 determination, using average values from the inventory data.

Table 1. Summary of WWTP configurations simulated using BioWin™ in this study

Configuration No.	Short Description	Key process units in treatment train	
		Mainstream	Sidestream
1	A-B, Mainstream & Sidestream DEMON	Two-stage (A-B) activated sludge. B-Stage MLE-like process with 35% anoxic volume fraction, internal mixed liquor recycle (5:1). Hydro-cyclone on sludge wasting line to enrich Anammox bacteria in Mainstream reactors.	Nitritation-Anammox (DEMON™ or similar concept) with alkalinity supplement to Nitritation reactor. Seeding of Anammox bacteria from Sidestream to Mainstream reactors. Similar to Schaubroeck <i>et al.</i> (2015) 'Scenario 3'.
2	A-B, Sidestream DEMON	Two-stage (A-B) activated sludge. B-Stage MLE-like process with 35% anoxic volume fraction, internal mixed liquor recycle (5:1).	Nitritation-Anammox (DEMON™ or similar concept) with alkalinity supplement to Nitritation reactor. Similar to Schaubroeck <i>et al.</i> (2015) 'Scenario 2'.
3	A-B, Sidestream N/DN		Nitritation-Denitritation with alkalinity supplement to Nitritation reactor and methanol supplement to Denitritation reactor. Similar to Schaubroeck <i>et al.</i> (2015) 'Scenario 1'.
4	Step Feed, Side-stream N/DN	Step Feed (3 pass) activated sludge. 1 st and 2 nd pass with 25% and 50% anoxic volume fraction respectively; overall 25% anoxic volume fraction. Flow split 50% each to 1 st and 2 nd pass.	As for Configuration No. 3 (above).
5	Step Feed, no side-stream	As for Configuration No. 4 (above).	None
6 (Base Case)	MLE, no sidestream	MLE-activated sludge with 38% anoxic volume fraction, internal mixed liquor recycle (5:1).	None



RESULTS

Energy use and 'energy neutrality'

Following a benchmarking approach for WWTPs, it is important to distinguish energy use from energy production (de Haas *et al.*, 2015; DWA, 2015). Figure 1 summarises the modelled WWTP total electrical energy use and production on a flow-specific basis (kWh/ML). This is a valid metric given that all the model configurations shared the same set of influent characteristics (flow and load). Refer to Figure 6 (**Supplementary Information**) for a breakdown of electricity use (main vs. sidestream).

Figure 1 also shows the extent of electrical energy self-supply (by on-site cogeneration) from the predicted biogas production, for each of the configurations considered. Benchmarking results for typical WWTPs are in the range approximately 30-60% electrical energy self-supply (ESS) without optimisation, and approaching 100% with optimisation (DWA, 2015). The difference between total electrical energy use and electrical energy production reflects the extent to which electricity will need to be imported from the grid. Where ESS exceeds 100%, the potential exists for electricity export ('energy positive'), but no resultant GHG credits were assumed here.

The approach taken in this study was to assume that the WWTPs modelled were reasonably well optimised, including energy use, for each of the configurations. That is, the adopted average energy use reflected the reference data (Schaubroeck *et al.*, 2015) and was close to benchmark guide values, or better, for the configurations considered (Table 2). Similarly, assuming reasonably optimised and well-operated systems, the model input uncertainty range for variable energy use

was relatively small. On this basis, in terms of energy use, the most significant differences between the configurations considered was the extent of energy self-supply. The systems with the most energy-efficient sidestream processes (i.e. Configuration Nos. 1 and 2, being those incorporating nitrification-anammox processes - refer to Table 1), along with optimal capture of organics for biogas production, had energy self-supply approaching or exceeding 100% (Figure 1). This reflects actual performance of the reference full-scale plant (Schaubroeck *et al.*, 2015). Configurations that used energy less efficiently, or did not capture organics as well and produced less biogas, performed less well against the energy self-supply metric (Configurations 3 to 6). However, the model still predicted around 60- 80% self-supply for these configurations, which compares well with actual WWTP data in Australia (de Haas *et al.*, 2015), where most plants lack advanced sidestream treatment and may not be fully optimised.

Nitrogen removal

Alongside energy efficiency, nitrogen (N) removal is another common requirement for WWTPs, usually driven by environmental protection. Given that anaerobic sludge digestion is one way of recovering energy from wastewater (as biogas), WWTPs configured with sidestream processes for removal of ammonia from nutrient-rich digester liquors can be optimised for both energy and N removal. The extent of N removal predicted for configurations modelled in this study is shown in Figure 2. Based on full-scale data from their reference plant, Schaubroeck *et al.* (2015) reported similar or slightly better N removal (averages ranging 83-91%) for their model configurations, which were comparable to Configuration 1 to 3 in this study.

Taken broadly, the results in Figure 2 can be compared for configurations defined within the framework and assumptions of this study. The results suggest that, with side-stream processes in place (Configuration Nos. 1 to 3), N removal performance comparable to that of the base case (Configuration No. 6, without sidestream treatment) can be achieved. This reflects the trade-off between maximising carbon (organics) capture for biogas and energy production, versus retaining sufficient carbon in the mainstream for N removal without sidestream treatment.

Comparing Step-Feed configurations (Configuration Nos. 4 and 5) in Figure 2, sidestream treatment again helps to bolster N removal, but performance was somewhat lower in both of these configurations compared with the MLE base case (No. 6). The main reason is that the Step-Feed configuration lacks internal (mixed liquor) recycles to promote N removal processes. Optimisation of flow splitting (feed ratios), anoxic fractions and internal recycles (if used) will largely determine the differences in N removal between alternatives when selecting activated sludge process configurations.

Greenhouse gas emissions

Refer to Figure 3 for the GHG emissions predictions modelled in this study. This figure includes Scope 1 N₂O emissions, calculated with variable emission factors, which differed between the respective configurations modelled. It also includes on-site cogeneration of electrical energy that provides significant (to near complete) energy self-supply (Figure 1). Figure 7 (see **Supplementary Information**) gives the comparable GHG emissions predictions without cogeneration (i.e. higher Scope 2 emissions for all electricity imported from the grid).

Figure 4 compares the total GHG emissions profile with electrical energy self-supply for the six configurations modelled. Configurations using side-stream treatment (such as Nos.

1 to 3) that attempt to optimise energy self-supply (and minimise energy use without sacrificing N removal) might be at most risk of higher *total* GHG emissions, mainly due to N₂O. Other configurations that aim to minimise energy use, with optional sidestream treatment to minimise the sacrifice in N removal (such as Nos. 4 and 5), might also be at risk of higher *total* GHG emissions due to N₂O.

Similarly, Figure 9 (see **Supplementary Information**) shows the total greenhouse gas emissions profiles of Configuration Nos. 1 to 5 on a relative basis with combined uncertainties, compared with the base case (Configuration No. 6).

Figure 5 compares the GHG emissions predicted using variable vs. fixed (NGER, 2016) emission factors for Scope 1 N₂O. Given the marginal differences in predicted N removal (Figure 2), there is little difference between the configurations in Scope 1 N₂O emissions calculated using the NGER method. The differences in total GHG emissions calculated using the NGER method arise mainly from Scope 2 (imported grid electricity). Refer to Figure 8 (see **Supplementary Information**) for a comparable chart of GHG emissions excluding on-site cogeneration.

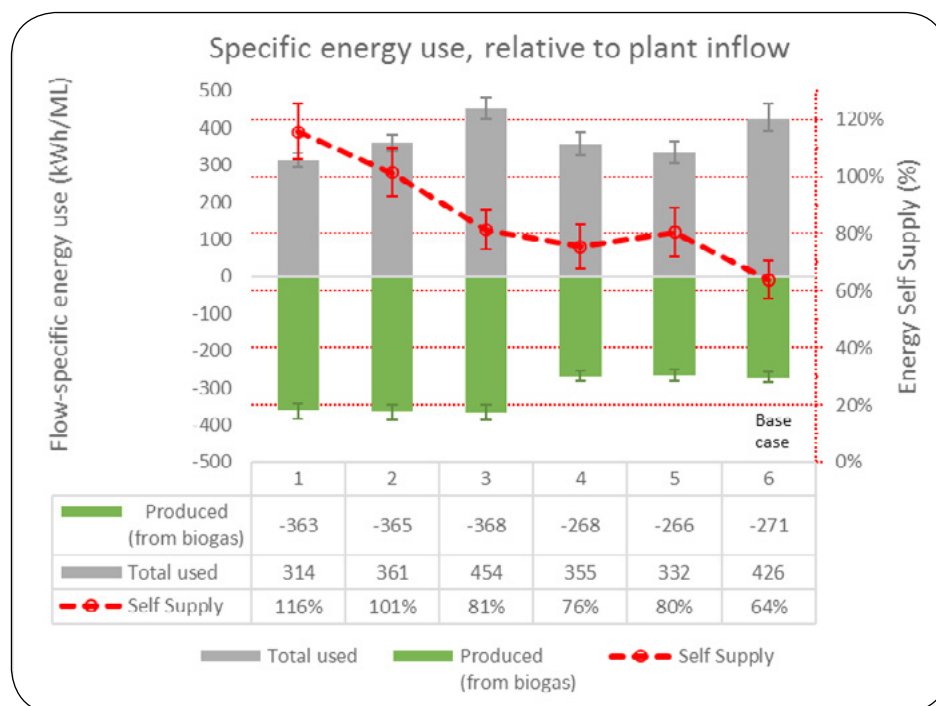


Figure 1. Model results of total electrical flow-specific energy use and production, showing extent of energy self-supply (right axis) for the six WWTP configurations studied. Values plotted are predicted means. Error bars indicate the predicted range from 5th to 95th percentile.

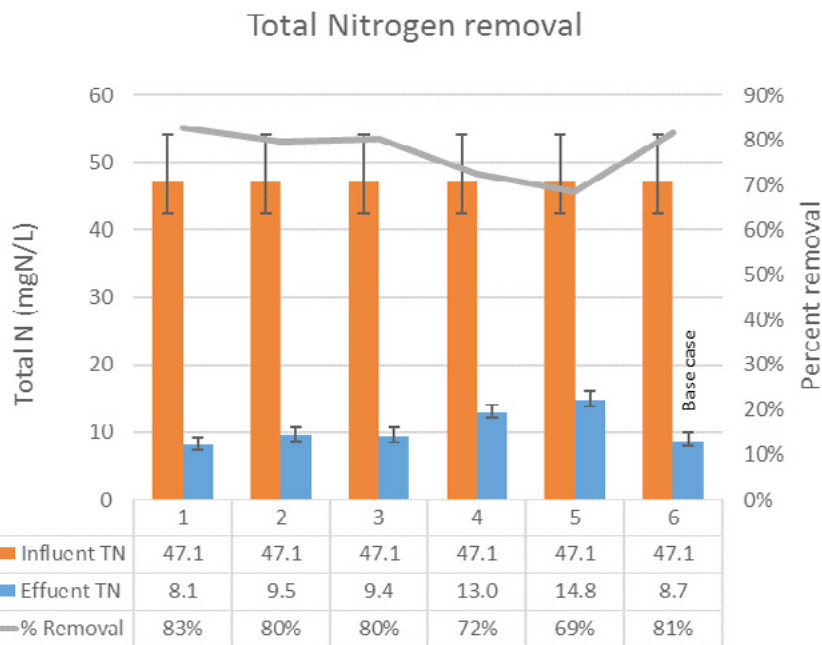


Figure 2. Model results of total nitrogen removal for the six WWTP configurations studied. Values plotted are means. Error bars indicate the range from 5th to 95th percentile.

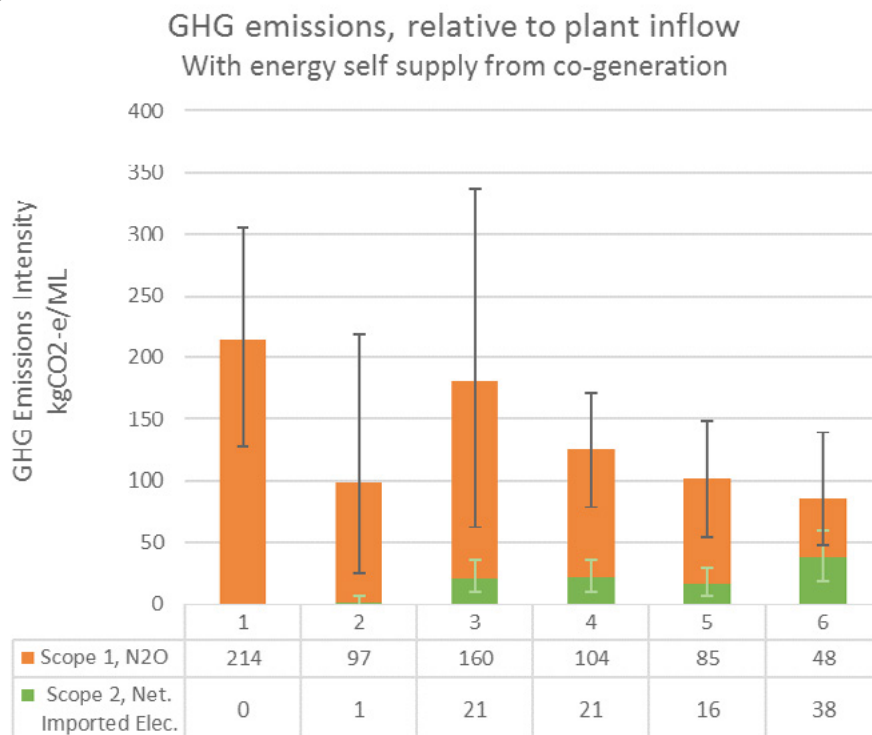


Figure 3. Model results of flow-specific greenhouse gas emissions for the six WWTP configurations studied. Values plotted are predicted means. Error bars indicate the predicted range from 5th to 95th percentile.

GHG vs. Electrical Energy Self-supply

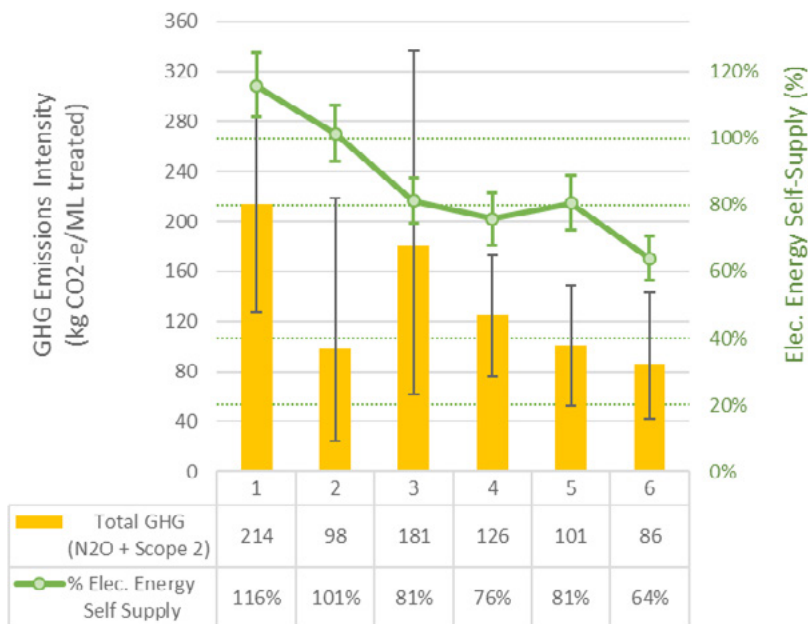


Figure 4. Model results of total flow-specific greenhouse gas emissions, compared with extent of electrical energy self-supply (right axis) for the six WWTP configurations studied. Values plotted are predicted means. Error bars indicate the predicted range from 5th to 95th percentile.

GHG emissions, relative to plant inflow With energy self supply from co-generation

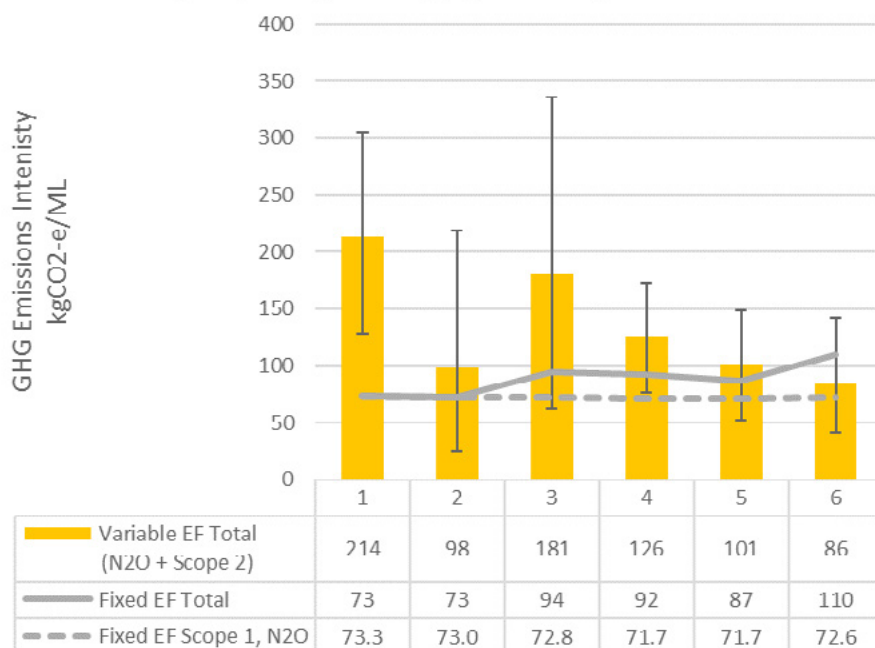


Figure 5. Model results of flow-specific greenhouse gas emissions for the six WWTP configurations studied. Values plotted are predicted means (or calculated fixed values). Where applicable, error bars indicate the predicted range from 5th to 95th percentile. The values calculated using fixed emission factors as per NGER (2016) are also shown (grey lines). Fixed EF Total (NGER, 2016) plotted as solid line (Scope 1 N2O plus Scope 2 for net electricity imported from grid, after self-supply, without credits for any surplus electrical energy potentially exported). Fixed EF Scope 1 (NGER, 2016) plotted as dashed line (N2O only).

DISCUSSION

Minimising energy use, to save on the cost of electricity imported from the grid, is a common efficiency driver in many industries, including wastewater treatment. Aiming further for 'energy neutral', 'low emissions' or 'carbon neutral' WWTPs has become a notional target for many water utilities both in Australia and worldwide, particularly within the broader environmental/social paradigms of resource recovery and sustainability (Schaubroeck *et al.*, 2015; DELWP, 2016; Donaghy, 2017). At the same time, achieving treated effluent quality targets (including nutrient removal in many cases) is co-requisite.

Taken as a whole, the modelling results from this study (refer to Figure 3, Figure 4 and also **Supplementary Information**) highlight the relativities in Scope 1 N₂O and Scope 2 electricity GHG emissions and illustrate the risks associated with uncertainty in actual N₂O emissions for WWTPs. This study examined WWTP configurations designed to recover energy from biogas whilst achieving significant N removal, in the range approximately 70-83%. The energy recovery vs. N removal trade-offs become more acute for WWTPs required to achieve more advanced N removal (>85%) and the greenhouse gas emissions profile may be different for such plants. However, a similar nexus around choice of plant configuration, including N removal performance, and the risk of increased N₂O emissions can be expected.

Based on literature data, vastly different Scope 1 N₂O emissions might occur between WWTPs that have different process configurations. Indicatively, the results of this study suggest that processes more at risk of emitting N₂O (e.g. those incorporating deliberate formation of nitrite via 'nitrification', or step feed systems) could result in total GHG emissions that are on average approximately 30-190% higher than processes that inherently have lower N₂O emissions risks. The uncertainty in N₂O emission factors is large, and greater (or lesser) differences might occur when comparing processes at across the range of predicted emissions distributions. However, reduced Scope 2 (electrical energy-related) emissions, resulting from promoting so-called 'short-cut' N removal pathways via nitrite, are unlikely to cancel out the risk of higher N₂O emissions profiles associated with such treatment processes.

Furthermore, applying a constant N₂O emission factor (e.g. in the existing NGER (2016) or similar calculation protocols) does not reflect the reality of differences

in actual N₂O emissions between various WWTP processes. The existing NGER (2016) N₂O emission factor for wastewater treatment is likely to result in underestimation of Scope 1 N₂O emissions (potentially by a large margin) for processes that tend to be at risk of emitting most N₂O. In other cases, where the processes may be designed to have lower N₂O emissions profiles, the existing NGER (2016) method is likely to overestimate the actual emissions.

CONCLUSIONS

The following conclusions can be drawn from this study:

1. There are large uncertainties associated with Scope 1 N₂O emission factors for wastewater treatment plants. The literature suggests that higher N₂O emission profiles are associated with treatment processes that are reliant on: (1) 'nitrification' (or so-called 'shortcut' or 'novel' pathways for nitrogen removal), which involves deliberate formation of nitrite; or (2) have large temporary or spatial variation in ammonium oxidation rate. The former is often applied in sidestream systems for processing ammonia-rich dewatering liquors from anaerobic digestion, including deammonification (or anammox) processes, but is also an emerging technology for incorporation into mainstream systems. The latter can occur in a range of commonly applied activated sludge configurations, including compartmentalised (particularly step-feed) and intermittent (batch) processes, especially at higher instantaneous reactor loading rates.
2. Engineering WWTPs with processes that have high N₂O emission risks in an effort to promote nitrogen removal and/or reduce electrical energy use (either total or net imported electricity, such as in a drive to notional 'energy neutrality') might be misguided in terms of life cycle GHG emissions. Without active mitigation measures, it is likely for such systems that increased Scope 1 (direct) N₂O emissions will occur and cancel out (or exceed) the benefits of reduced Scope 2 (indirect) emissions associated with lower use of grid electricity.
3. The use of a fixed emission factor for N₂O from wastewater treatment, as applied in many GHG calculation protocols (e.g. including the NGER (2016) Determination in Australia), is flawed. At best, the current NGER (2016) emission factor (4.9 tonnes CO₂-e per tonne N removed) might overestimate actual emission for systems with lower actual N₂O profiles. At worst, it is likely to underestimate (potentially by a large margin) the actual N₂O for processes that typically have high N₂O emissions profiles.

It provides no disincentive against the design and implementation of such systems, some of which are currently promoted as 'novel' or 'emerging'. Ironically, such systems may be less environmentally sustainable, particularly in situations (such as in Australia) where electricity generation from renewable energy sources is an emerging trend.

4. Improved practical methods for measurement and accounting for N₂O emissions from WWTPs are required to support better decision making within water utilities around greenhouse gas reduction.

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