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Identifying key sources of uncertainty in the modelling of greenhouse gas emissions from wastewater treatment

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ARTICLE INFO

Article history:
Received 8 February 2013
Received in revised form
13 May 2013
Accepted 16 May 2013
Available online 28 May 2013

Keywords:
Benchmark model
Greenhouse gas
Model identification
Sensitivity
Uncertainty
Wastewater treatment

ABSTRACT

This study investigates sources of uncertainty in the modelling of greenhouse gas emissions from wastewater treatment, through the use of local and global sensitivity analysis tools, and contributes to an in-depth understanding of wastewater treatment modelling by revealing critical parameters and parameter interactions. One-factor-at-a-time sensitivity analysis is used to screen model parameters and identify those with significant individual effects on three performance indicators: total greenhouse gas emissions, effluent quality and operational cost. Sobol's method enables identification of parameters with significant higher order effects and of particular parameter pairs to which model outputs are sensitive. Use of a variance-based global sensitivity analysis tool to investigate parameter interactions enables identification of important parameters not revealed in one-factor-at-a-time sensitivity analysis. These interaction effects have not been considered in previous studies and thus provide a better understanding wastewater treatment plant model characterisation. It was found that uncertainty in modelled nitrous oxide emissions is the primary contributor to uncertainty in total greenhouse gas emissions, due largely to the interaction effects of three nitrogen conversion modelling parameters. The higher order effects of these parameters are also shown to be a key source of uncertainty in effluent quality.

1. Introduction

Wastewater treatment can result in direct emissions of greenhouse gases (GHGs) such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O), as well as indirect emissions resulting from energy generation, chemical manufacture and sludge disposal, amongst other sources. Reduction of GHG emissions is a topic of global interest, and it is recognised that appropriate design and operation of wastewater treatment processes can play a significant role in mitigating the effects of global warming (Gori et al., 2011).

Models used to estimate the magnitude of GHG emissions from wastewater treatment plants (WWTPs) for inventories typically utilise empirical emission factors (e.g. IPCC, 2006b), based on the volume of wastewater treated, influent concentrations, effluent concentrations or the mass of wastewater components removed. These emission factors, however, have a high degree of variability and uncertainty (Corominas et al., 2012): for example, N_2O emissions in the range 0–90% of the nitrogen-load were reported by Kampschreur et al. (2009). As such, there has been increasing interest in the use of comprehensive process models and mechanistic models to

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http://dx.doi.org/10.1016/j.watres.2013.05.021

estimate dynamic GHG emissions. Resulting from this, it has been highlighted that significant variability can occur in GHG emissions from WWTPs with different designs (Shahabadi et al., 2009) and operating under different conditions (Flores-Alsina et al., 2011).

As wastewater utilities face the challenge of simultaneously reducing GHG emissions and improving treatment standards due to increasing regulatory pressures, the importance of including GHG emissions in addition to effluent quality and operational costs when evaluating design alternatives is clear. It has been shown that use of automatic control can reduce GHG emissions (Corominas et al., 2010), but models used are typically of hypothetical WWTPs and their results are not always validated with real data (e.g. Hiatt and Grady, 2008; Guo et al., 2012). As such, results are likely to be subject to a high degree of uncertainty; and careful calibration is therefore essential if applying the models and estimation methodologies to a real WWTP for plant design or control strategy development to reduce GHG emissions. Identification of the most significant sources of uncertainty could aid efficient calibration of models and reduce the complexity of future uncertainty analyses, yet there has been little research into the magnitude of uncertainty in GHG emission estimates resulting from uncertainty in model parameters and emission factors.

Sensitivity analysis is a useful tool for identification of the key parameters controlling model outputs (Tang et al., 2007a). However, whilst sensitivity analyses of dynamic WWTP models have previously been undertaken to investigate the effects of uncertainty in model parameters (e.g. Pons et al., 2008; Flores-Alsina et al., 2009; Ramin et al., 2012), design and operational parameters (Benedetti et al., 2008; Pons et al., 2008) and influent characteristics (Pons et al., 2008), no detailed analyses for identification of key parameters affecting GHG emissions have been carried out. Gori et al. (2011) completed a sensitivity analysis to investigate the effects of varying the pCOD/VSS ratio on the rate of GHG emissions from different sources, but no other model parameters were considered. Global sensitivity analyses (GSAs) of the Benchmark Simulation Model No. 1 (BSM1) (Sin et al., 2011) and the Benchmark Simulation Model No. 2 (BSM2) (Benedetti et al., 2008), based on Monte Carlo experiments and linear regression, enabled the identification of individual parameters with significant effects on effluent quality and operational cost, but did not consider GHG emissions. However, interactions were not investigated and output uncertainty was attributed to individual parameters only.

The aim of this research is to identify individual parameters and parameter interactions which contribute significantly to uncertainty in modelled GHG emissions from wastewater treatment, as well as the more widely used performance indicators of effluent quality and operational cost. Investigation of the relative contributions of specific parameter interactions to output uncertainty represents an advance in WWTP modelling, as previous analyses have not enabled identification of significant interactions. Sensitivity analysis of a revised BSM2, with pre-defined layout, operating conditions and influent characteristics, is carried out using the one-factor-at-a-time (OAT) method, to identify significant individual (first order) effects and inform the selection of parameters for inclusion in further analysis. GSA is then carried out using a variance-based method – Sobol's method (Saltelli, 2002) – to investigate higher order effects (interactions). This tool has not, as of yet, been extensively used in wastewater treatment, but previous applications have revealed situations and modelling scenarios in which calibration is likely to be most challenging due to the greater presence of parameter interactions (Massmann and Holzmann, 2012) and improved the efficiency of multi-objective optimisation problems by identifying important decision variable interactions (Fu et al., 2012). The results enable identification of: a) parameters that have negligible impact on uncertainty in key model outputs and can, therefore, be excluded from future uncertainty analyses; and b) parameters which contribute significantly to variance in any key model output, due to first or higher order effects, and so need to be accurately defined for model calibration and application.

2. Materials and methods

2.1. Model description

2.1.1. Model structure

The WWTP model used for parameter sensitivity analysis, which will be referred to as BSM2-e, is based on the Benchmark Simulation Model No. 2: BSM2 (Jeppsson et al., 2007), with modifications (outlined in Section 2.1.2) made to enable dynamic modelling of the emissions shown in Fig. 1. The plant layout and modelling of pre-treatment and sludge treatment processes are unaltered from those of BSM2 (as detailed by Jeppsson et al. (2007) and Nopens et al. (2010)), but adjustments have been made to the activated sludge model to enable calculation of N_2O emissions. A complete description of all equations added and modifications made to the BSM2 is provided as Supplementary information.

2.1.2. Greenhouse gas emission modelling methodologies

GHG emissions are modelled using previously published estimation methodologies, which are implemented in BSM2. Sources of GHG production and direct emissions from the modelled processing units include:

- Aerobic substrate utilisation (CO_2), biomass decay (CO_2) and denitrification (CO_2 and N_2O) in activated sludge reactors

In BSM2, the reduction of nitrate to nitrogen is modelled as a one-step process and dynamic production of N_2O (an intermediate product) cannot be determined. Modifications have therefore been made to include four-step denitrification as detailed by Samie et al. (2011). Stripping of N_2O from solution is then modelled using Henry's law. CO_2 emissions resulting from nutrient removal are calculated using emission factors derived from the stoichiometric relationships for denitrification with and without an external carbon source (Shahabadi et al., 2010).

Calculation of CO_2 emissions from substrate utilisation and biomass decay is based upon the method detailed by Monteith et al. (2005), with the suspended solids mass balance equation adapted for non-steady state conditions. Required

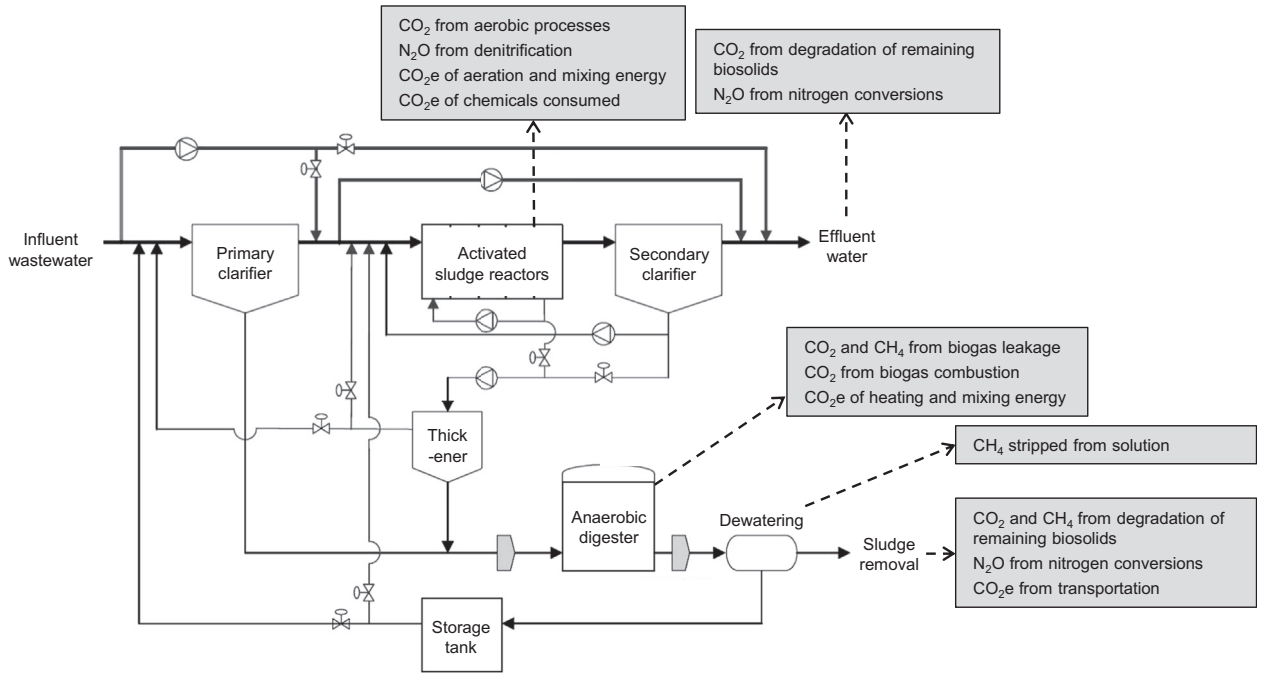

Fig. 1 – Schematic diagram of the modified BSM2 plant and sources of modelled GHG emissions (adapted from Nopens et al. (2010)).

concentrations and flow rates have been derived from the BSM2 state variables and theoretical emission factors, derived from stoichiometry, are applied.

- Biogas leakage (CO_2 and CH_4) and combustion (CO_2)

Dynamic CH_4 and CO_2 formation and stripping in the anaerobic digester and the resultant biogas composition and flow rate are modelled in BSM2. It is assumed in BSM2 that all biogas is combusted for energy recovery. However, past investigations (e.g. Shahabadi et al., 2009; Shahabadi et al., 2010), have identified biogas leakage as a potential contributor to total emissions. As it is impractical to accurately measure or model small leaks, a fixed leakage factor of 5% (Shahabadi et al., 2009) has been applied. It is assumed that the remaining biogas is fully combusted and a theoretical emission factor (Monteith et al., 2005) is used calculate CO_2 production.

- Stripping of dissolved gases (CH_4) in dewatering unit

Dissolved CH_4 concentration in the digester effluent is calculated using the BSM2 methodology. Given the negligible partial pressure of CH_4 in the atmosphere, it is assumed that all CH_4 is stripped from solution during dewatering.

Additional direct emissions may result from poorly managed treatment and unintentionally anaerobic conditions (Monteith et al., 2005); these are not modelled, however, due to a lack of reliable estimation techniques. Likewise, N_2O emissions associated with nitrifier denitrification during nitrification are omitted. There have been recent studies into the factors influencing N_2O emissions (e.g. Foley et al., 2010; Law et al., 2011; Rassamee et al., 2011), but there is little consensus on a method which can be used to estimate emissions with any degree of certainty and metabolic models of the nitrifier denitrification pathway (Mampaey et al., 2011; Ni et al., 2011) have been found unable to consistently reproduce experimental N_2O emissions data (Law et al., 2012; Ni et al., 2013). The significance of this omission is uncertain; heterotrophic denitrification is the dominant nitrogen removal process, but nitrifier denitrification yields greater N_2O emissions relative to the nitrogen converted (Kampschreur et al., 2009). Incomplete hydroxylamine oxidation can also result in N_2O emissions, but it is unclear under what conditions this process becomes dominant and current models are inadequate (Ni et al., 2013). If nitrification modelling is included in future GHG emission estimates, inclusion of the associated parameters in uncertainty analysis is recommended.

Indirect emissions result from:

- Generation of energy imported

Energy required for pumping, aeration, heating and mixing is modelled using the original BSM2 methodologies; energy recovery from biogas combustion is also calculated using the BSM2 methodology, but with allowance for biogas leakage incorporated. GHG emissions associated with net energy import are affected by the electricity generation mix, as emissions differ between energy sources. However, as electricity grid composition varies locally and nationally and the model is not linked to a specific location, a single emission factor of 0.245 kg CO_2e/kWh (Gori et al., 2011) is used but defined as uncertain.

- Manufacture of chemicals

Indirect emissions due to chemical addition have been calculated using the carbon source flow rate for each tank, as modelled in BSM2, and an emission factor of 1.54 kg CO_2e/kg MeOH (Shahabadi et al., 2010).

- Offsite degradation of effluent

Indirect CO_2 emissions are modelled based on the assumption that all BOD_5 remaining in the effluent degrades aerobically, as detailed by Shahabadi et al. (2010). Indirect N_2O emissions are calculated using an emission factor of 0.005 kg N_2O–N/kg N (IPCC, 2006b).

- Transport and offsite degradation of sludge

Emissions resulting from the transport of sludge are estimated using a fixed emission factor (Shahabadi et al., 2010). Emissions of CO_2 and CH_4 resulting from the degradation of biosolids remaining in the sludge are modelled as detailed by Shahabadi et al. (2009), based on the theoretical stoichiometric equation for biomass decay in an anaerobic environment. Dynamic N_2O emissions are calculated using the modelled sludge nitrogen content and an emission factor of 0.01 kg N_2O-/kg N (IPCC, 2006a).

All emissions are converted to CO_2 equivalent (CO_2e) units, using global warming potentials (GWPs) of 21 and 310 for CH_4 and N_2O respectively (IPCC, 1996), to enable comparison of the magnitude of emissions from each source.

2.1.3. *Simulation strategy and performance assessment*

The performance of control strategies in the BSM2 is typically assessed using a 609 day simulation, incorporating stabilisation and evaluation periods, with predefined dynamic influent data. Initial values should be determined by simulation with 200 days of constant influent data to allow the model to reach steady state (Jeppsson et al., 2007). In order to carry out a GSA of model parameters, however, it is necessary to significantly reduce the computational demand. Based on analysis of the effects of modifications in stabilisation and evaluation periods on the OAT sensitivity analysis parameter rankings, a reduced dynamic simulation period (consisting of 14 days stabilisation and 14 days evaluation, using days 322–350 of the BSM2 dynamic influent data) has been selected to follow the 200 day steady state initialisation. Whilst this shortened simulation does not reproduce the model outputs obtained with full length stabilisation and evaluation, it has been found to be suitable for assessment of the relative importance of parameters, enabling correct identification of the most sensitive model parameters in OAT sensitivity analysis and resulting in an average change in rank of just 1.1 for all 70 parameters across the three key outputs when compared with analysis using the full dynamic simulation period (609 days).

Performance indicators used include an effluent quality index (EQI) and an operational cost index (OCI), calculated using the BSM2 methodology (Jeppsson et al., 2007). EQI is a weighted sum of average effluent concentrations; OCI is a measure of the average energy demand, energy recovery, carbon source dosage and sludge production for disposal. Average GHG emissions per unit of wastewater treated are also calculated, and the contribution of each gas and direct and indirect emissions to total GHG emissions are modelled to allow a more in-depth investigation into the most significant sources of uncertainty.

2.1.4. *Model validation*

The magnitude of GHG emissions per unit of treated wastewater reported in the literature differs significantly, even for WWTPs with the same or similar treatment processes and control. Total emissions in the range 19,554–22,920 kg CO_2e/d (equivalent to 0.947–1.110 kg CO_2e/m^3, based on specified flow rate) were reported by Corominas et al. (2012) in an investigation into the effects of different GHG modelling approaches for the BSM2 plant. The BSM2-e emissions model gives total GHG emissions of 1.077 kg CO_2e/m^3 when using the default BSM2 evaluation period, which is within this range.

2.2. *Sensitivity analysis methodology*

153 BSM2 parameters are used in the model (excluding those relating to the plant design and operation), and a further 64 are used for the incorporated denitrification and emissions modelling. Given the large number of evaluations required for GSA, it is not practical to include every parameter. Therefore, OAT sensitivity analysis, which requires significantly fewer model evaluations, is used to provide an indication of the importance of each parameter and identify parameters with negligible effect on uncertainty in model outputs.

OAT sensitivity analysis enables changes in model outputs to be clearly attributed to a specific parameter, with no ambiguity, but does not explore the effects of varying two or more parameters simultaneously and is unable to identify any significant interactions. As such, it is followed by GSA to obtain an understanding of second (and higher) order effects and allow exploration of the full parameter space.

2.2.1. *Parameter screening*

2.2.1.1. Parameter selection and definitions. Selection of BSM2 parameters is guided by the results of previous GSA by Benedetti et al. (2008): those identified as being not significant for EQI, OCI and effluent NH_4 violations in terms of both the standard regression coefficient and the partial correlation coefficient are excluded from this analysis. Henry's law coefficients used to model dissolution and stripping of CO_2 and CH_4 in the anaerobic digester, however, are added to the analysis, as they may affect emissions despite not having significant effects on previously considered model outputs.

All half-saturation constants added for the modelling of nitrogen conversions are included in the sensitivity analysis, because these parameters have a high degree of uncertainty (Reichert and Vanrolleghem, 2001) and affect modelled N_2O production, which has been shown to be a major contributor to GHG emissions from WWTPs (Rodriguez-Garcia et al., 2012). Also, other half-saturation constants were found to be significant by Benedetti et al. (2008).

It is assumed that median values for each parameter are equal to the BSM2 default values (where applicable). For all other parameters, median values are assumed to be those reported in the literature on which the calculations are based.

Parameters for which no feasible range is specified in the literature are classified according to the system defined by Reichert and Vanrolleghem (2001) (summarised in Table 1) and adopted in later sensitivity and uncertainty analyses (Rousseau et al., 2001; Benedetti et al., 2008).

Full details of parameters selected for screening are given in Tables 2 and 3 and Table 3. Parameters 1–26 are BSM2 parameters, 27–39 are nitrogen conversion modelling parameters and 40–70 are emissions modelling parameters.

2.2.1.2. One-factor-at-a-time sensitivity analysis. To carry out OAT sensitivity analysis, a simulation is first conducted with all parameters set at their default values; this represents the base case. Further simulations are carried out with each parameter individually set to its upper and lower bound values in turn, whilst all others are held at their default values. Percentage change in each model output with respect to the base case is calculated for each simulation, to determine which parameters cause the greatest variation in model outputs when individually varied within their feasible range.

2.2.2. Global sensitivity analysis

Sobol's method (2001) is selected for GSA despite being computationally expensive, as it enables first, second and higher order effects to be distinguished through the calculation of first, second and total order sensitivity indices for each parameter or parameter pair. It also provides more robust sensitivity rankings and a more detailed description of the impact of individual parameters and their interactions on model performance than other GSA methods such as analysis of variance (Tang et al., 2007b), and requires significantly fewer model evaluations than factorial design given the large number of parameters under investigation.

The total variance (D) of model outputs, resulting from samples of the feasible parameter space, is decomposed and attributed to specific parameters and their interactions as follows, assuming parameters are independent (Tang et al., 2007b):

$$D = \sum_{i} D_i + \sum_{i<j} D_{ij} + \sum_{i<j<k} D_{ijk} + \ldots + D_{12\ldots p} \quad (1)$$

where D_i = output variance resulting from the ith parameter; D_{ij} = output variance resulting from interaction between ith and jth parameters; p = total number of parameters.

First and second order sensitivity indices S_i and S_{ij} represent the percentage contribution of the ith parameter alone and the interaction between the ith and jth parameters to total variance, respectively; total order index S_{Ti} represents the percentage contribution related to the ith parameter, including the interactions of any order, as defined below:

$$S_i = \frac{D_i}{D} \quad (2)$$

$$S_{ij} = \frac{D_{ij}}{D} \quad (3)$$

$$S_{Ti} = 1 - \frac{D_{\sim i}}{D} \quad (4)$$

where $D_{\sim i}$ = output variance resulting from all parameters except ith parameter. A high first order sensitivity index indicates a parameter whose individual uncertainty provides a large contribution to output variance, whereas a low first order index and high total order index indicates a parameter whose interactions result in significant output variance, but individually has little effect.

Sobol's method is implemented here as follows:

1. Specify upper and lower bounds of parameters for analysis.
2. Generate $2n$ random parameter samples within the specified bounds, with quasi-Monte Carlo sampling using Sobol's sequence generator.
3. Resample parameters using Saltelli's (2002) extension to Sobol's method, holding one fixed at a time, to generate $n(2p+2)$ parameter sets.
4. Run model with each parameter set in turn, recording values of model outputs.
5. Compute first order, total order and second order sensitivity indices, and rankings for each parameter as detailed by Tang et al. (2007b).
6. Calculate 95% bootstrap confidence intervals for all sensitivity indices.

Table 1 – Parameter uncertainty classes.

Class	Description	Uncertainty (%)	Examples
1	Accurately known parameters	5	External and input parameters
2	Intermediate	20	Growth rates; temperature dependence coefficients
3	Very poorly known parameters	50	Kinetic parameters, except those listed in Class 2; half-saturation concentrations; specific death and respiration rates

3. Results and discussion

3.1. One-factor-at-a-time sensitivity analysis

OAT sensitivity analysis results are presented in Tornado diagrams, which show the percentage change in each model output with respect to the base case when each model parameter is individually set to its respective upper and lower bounds. Parameters are ranked by the greatest range of percentage change for any model output and results for the most sensitive parameters are presented in Fig. 2. For clarity, only the 28 parameters with a corresponding range of change of at least 5% in one or more model output are shown.

Variation of a single parameter within its feasible range can have particularly significant effects on modelled GHG emissions; setting the half saturation constant for readily biodegradable substrate for N_2O reduction (parameter 30) to

Table 2 – BSM2 and nitrogen modelling parameters selected for sensitivity analysis screening and global sensitivity analysis (highlighted); HSC = half saturation coefficient.

Parameter number/name	Description	Default value		Class	Bounds		
		Value	Ref.		Lower	Upper	Ref.
1/Y_H	Heterotrophic biomass yield (g COD/g COD)	0.67	[a]	1	0.6365	0.7035	[c]
2/f_P	Fraction of biomass yielding particulate products	0.08	[b]	1	0.076	0.084	[c]
3/i_XB	Biomass nitrogen/COD mass ratio (g N/g COD)	0.08	[a]	1	0.076	0.084	[c]
4/mu_H	Heterotrophic max specific growth rate (/d)	4	[a]	2	3.2	4.8	[c]
5/K_OH	Oxygen HSC for heterotrophic biomass (g(−COD)/m^3)	0.2	[a]	3	0.1	0.3	[c]
6/ny_g	Correction factor for anoxic heterotroph growth	0.8	[a]	2	0.64	0.96	[c]
7/ny_h	Correction factor for anoxic hydrolosis	0.8	[a]	2	0.64	0.96	N/A
8/k_h	Max specific hydrolosis rate (g COD/g COD/d)	3	[a]	3	1.5	4.5	N/A
9/K_X	HSC of slowly biodegradable substrate (g COD/g COD)	0.1	[a]	3	0.05	0.15	N/A
10/mu_A	Autotrophic max specific growth rate (/d)	0.5	[a]	2	0.4	0.6	[c]
11/K_NH	Ammonia HSC for autotrophs (g NH_3–N/m^3)	1	[a]	3	0.5	1.5	[c]
12/b_A	Decay coefficient for autotrophic biomass (/d)	0.05	[a]	3	0.025	0.075	N/A
13/K_OA	Oxygen HSC for autotrophic biomass (g (−COD)/m^3)	0.4	[a]	3	0.2	0.6	[c]
14/k_a	Ammonification rate (m^3/g COD/d)	0.05	[a]	3	0.025	0.075	N/A
15/F_TSS_COD	TSS fraction of total COD (g TSS/g COD)	0.75	[a]	1	0.7125	0.7875	N/A
16/k_hyd_ch	Hydrolosis influence coefficient for carbohydrates (/d)	10	[a]	N/A	6.25	12.5	Derived from [c]
17/k_hyd_pr	Hydrolosis influence coefficient for proteins (/d)	10	[a]	N/A	6.36	13.64	Derived from [c]
18/k_hyd_li	Hydrolosis influence coefficient for lipids (/d)	10	[a]	N/A	6.36	13.64	Derived from [c]
19/K_S_ac	Monod HSC for acetate (kg COD/m^3)	0.15	[a]	3	0.075	0.225	N/A
20/K_H_co2	Henry's law coefficient for CO_2 (M_{liq}/bar)	0.035	[a]	2	0.028	0.042	N/A
21/K_H_ch4	Henry's law coefficient for CH_4 (M_{liq}/bar)	0.0014	[a]	2	0.00112	0.00168	N/A
22/frxs_adm	Anaerobically degradable fraction biomass	0.68	[a]	1	0.646	0.714	N/A
23/v0	Maximum Vesilind settling velocity (m/d)	474	[a]	2	379.2	568.8	[c]
24/r_h	Hindered zone settling parameter (m^3/g SS)	5.76E-04	[a]	2	0.00046	0.00069	[c]
25/r_p	Flocculent zone settling parameter (m^3/g SS)	0.00286	[a]	2	0.00229	0.00343	[c]
26/f_ns	Non-settleable fraction	0.00228	[a]	2	0.00182	0.00274	[c]
27/K_S2	HSC for S_S for NO_3- reduction (g COD/m^3)	20	[d]	3	10	30	N/A
28/K_S3	HSC for S_S for NO_2- reduction (g COD/m^3)	20	[d]	3	10	30	N/A
29/K_S4	HSC for S_S for NO reduction (g COD/m^3)	20	[d]	3	10	30	N/A
30/K_S5	HSC for S_S for N_2O- reduction (g COD/m^3)	40	[d]	3	20	60	N/A
31/K_NO3	HSC for SNO_3 for heterotrophs (g N/m^3)	0.2	[d]	3	0.1	0.3	N/A
32/K_NO2	HSC for SNO_2 for heterotrophs (g N/m^3)	0.2	[d]	3	0.1	0.3	N/A
33/K_NO	HSC for SNO for heterotrophs (g N/m^3)	0.05	[d]	3	0.025	0.075	N/A
34/K_N2O	HSC for SN_2O for heterotrophs (g N/m^3)	0.05	[d]	3	0.025	0.075	N/A
35/ny_g2	Anoxic growth factor for NO_3^- reduction	0.28	[d]	2	0.224	0.336	N/A
36/ny_g3	Anoxic growth factor for NO_2^- reduction	0.16	[d]	2	0.128	0.192	N/A
37/ny_g4	Anoxic growth factor for NO reduction	0.35	[d]	2	0.28	0.42	N/A
38/ny_g5	Anoxic growth factor for N_2O reduction	0.35	[d]	2	0.28	0.42	N/A
39/ny_Y	Anoxic yield factor for heterotrophs	0.9	[d]	1	0.855	0.945	N/A

a Alex et al. (2008).
b Henze et al. (1987).
c Benedetti et al. (2008).
d Hiatt and Grady (2008).

its upper bound, for example, results in a 244% increase in reported GHG emissions. Individual variation of a further eight parameters is shown to result in a range of at least 25% change in GHG emissions.

A maximum range of variation in total GHG emissions of 260%, resulting from uncertainty in just one parameter (No. 30), is observed, whereas maximum changes in EQI and OCI are significantly lower at 22.0% (No. 12) and 17.9% (No. 64) respectively. This confirms that accurate calibration of the model with regards to GHG emissions modelling is extremely important. The nine parameters shown to have greatest individual effects on GHG emissions are all used in the modelling of nitrogen conversions, suggesting that uncertainty in GHG emissions corresponds primarily to uncertainty in the rate of N_2O production. The three parameters to which GHG emissions are shown to be most sensitive result in negligible change in EQI and OCI and ought, therefore, to be relatively simple to calibrate if significant higher order effects are not identified in GSA.

The greatest changes in EQI arise due to uncertainty in the original BSM2 parameters, and nitrogen modelling parameters have comparatively little impact. Uncertainty in

Table 3 – Emissions modelling parameters selected for sensitivity analysis screening and global sensitivity analysis (highlighted); EF = emission factor.

Parameter number/name	Description	Default value		Class	Bounds		
		Value	Ref.		Lower	Upper	Ref.
40/f	Ratio of BOD_5 to BODu (g BOD_5/g BODu)	0.68	[b]	1	0.646	0.714	N/A
41/EF_AerOxi	EF for aerobic oxidation of BOD (kg CO_2/kg O_2)	1.1	[b]	1	1.045	1.155	N/A
42/EF_AerAutoOxi	EF for endogenous respiration of VSS (kg CO_2/kg VSS)	1.947	[b]	1	1.850	2.044	N/A
43/EF_CO2denitWCarb	EF for CO_2 emissions from denitrification with external carbon source (g CO_2/g N_2_N)	2.62	Derived from [c]	1	2.489	2.751	N/A
44/EF_CO2denitWOCarb	EF for CO_2 emissions from denitrification without external carbon source (g CO_2/g N_2_N)	2.83	Derived from [c]	1	2.689	2.972	N/A
45/K_H_n2o_base	Henry's law constant for N_2O (mol/l/bar)	0.025	[d]	2	0.02	0.03	N/A
46/kLa_n2o	Gas transfer coefficient for N_2O (/d)	2	[e]	3	1	3	N/A
47/pgas_n2o	Partial pressure of N_2O in atmosphere (bar)	3.20E-07	[f]	2	2.56E-07	3.84E-07	N/A
48/EF_AnaerBODremCH4	CH_4 emissions from anaerobic carbonaceous substrate utilisation (g CH_4/g BOD)	0.25	[c]	1	0.238	0.263	N/A
49/EF_AnaerBODremCO2	CO_2 emissions from anaerobic carbonaceous substrate utilisation (g CO_2/g BOD)	0.27	[c]	1	0.257	0.284	N/A
50/EF_AnaerVSSdecCH4	CH_4 emissions from anaerobic biomass decay (g CH_4/g VSS)	0.35	[c]	1	0.333	0.368	N/A
51/EF_AnaerVSSdecCO2	CO_2 emissions from anaerobic biomass decay (g CO_2/g VSS)	0.58	[c]	1	0.551	0.609	N/A
52/leak_frac	Fraction of biogas leaked	0.05	[g]	3	0.025	0.075	N/A
53/CH4toCO2_combust	Combustion emission factor (g CO_2/g CH_4)	2.75	[b]	1	2.613	2.888	N/A
54/CH4_conversioneff	Energy conversion efficiency for heating	0.5	[h]	2	0.4	0.6	N/A
55/PF_Qintr	Pumping energy factor, internal AS recirculation (kWh/m^3)	0.004	[a]	2	0.0032	0.0048	N/A
56/PF_Qr	Pumping energy factor, AS sludge recycle (kWh/m^3)	0.008	[a]	2	0.0064	0.0096	N/A
57/PF_Qw	Pumping energy factor, AS wastage flow (kWh/m^3)	0.05	[a]	2	0.04	0.06	N/A
58/PF_Qpu	Pumping energy factor, pumped underflow from primary clarifier (kWh/m^3)	0.075	[a]	2	0.06	0.09	N/A
59/PF_Qtu	Pumping energy factor, pumped underflow from thickener (kWh/m^3)	0.06	[a]	2	0.048	0.072	N/A
60/PF_Qdo	Pumping energy factor, pumped underflow from dewatering unit (kWh/m^3)	0.004	[a]	2	0.0032	0.0048	N/A
61/mixenergyunitreac	Energy for activated sludge mixing (kW/m^3)	0.005	[a]	2	0.004	0.006	N/A
62/mixenergyunitAD	Energy for anaerobic digester mixing (kW/m^3)	0.005	[a]	2	0.004	0.006	N/A
63/cp	Specific heat capacity for water (Wd/gC)	4.84E-05	[a]	1	4.60E-05	5.09E-05	N/A
64/O2TransferEff	Aeration oxygen transfer efficiency (kg O_2/kWh)	1.80	[i]	2	1.44	2.16	N/A
65/EF_Elec	EF for electricity generation (kg CO_2e/kWh)	0.245	[h]	2	0.196	0.294	N/A
66/EF_EmbodiedCarb	EF for methanol usage (kg CO_2e/kg)	1.54	[c]	2	1.232	1.848	N/A
67/EF_SludgeTransport	EF for transport of sludge (kg CO_2e/tonne)	24	[c]	2	19.2	28.8	N/A
68/EF_SludgeN2O	EF for sludge applied to managed soils (kg N_2O/kg N)	0.016	[j]	2	0.013	0.019	N/A
69/EF_AerBODreml	EF for carbonaceous BOD removal (kg CO_2/kg COD)	0.33	[c]	1	0.314	0.347	N/A
70/EF_EffN2O	EF for N_2O emissions from effluent (kg N_2O/kg N)	0.008	[k]	2	0.006	0.009	N/A

a Alex et al. (2008).
b Monteith et al. (2005).
c Shahabadi et al. (2010).
d Lide and Frederiske (1995).
e Samie et al. (2011).
f European Environment Agency (2011).
g Shahabadi et al. (2009).
h Gori et al. (2011).
i Nopens et al. (2010).
j IPCC (2006a).
k IPCC (2006b).

emissions modelling parameters has no effect on EQI. Uncertainty in BSM2 parameters contributes to uncertainty in all three of the key model outputs, although OCI is affected to a lesser degree (maximum 3.2% change, compared with 22.0% and 19.0% for EQI and GHG emissions respectively). It is, therefore, important to take into account the effects of BSM2 parameter values on GHG emissions as well as on conventional performance assessment measures when calibrating the model.

The OCI is affected predominantly by uncertainty in the oxygen transfer efficiency (parameter 64) during OAT sensitivity analysis, suggesting that this is particularly important to consider when carrying out uncertainty analyses with regard to operational costs.

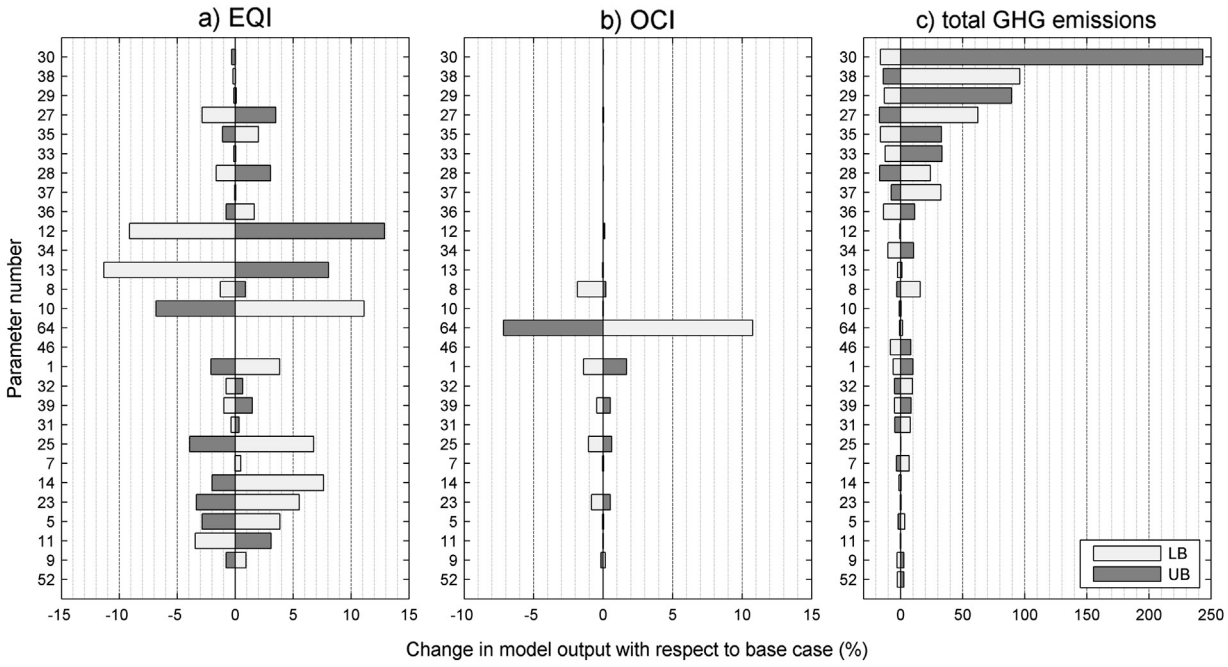

Fig. 2 – Percentage change in model output resulting from variation of individual parameter values.

3.2. *Sobol's method global sensitivity analysis*

GSA was carried out using the highlighted parameters in Tables 1 and 2, selected based on OAT sensitivity analysis screening results. In addition to the 28 parameters shown in Fig. 2, these include a further 11 of the highest ranked parameters. First order, second order and total order sensitivity indices computed using a sample size of 4000 are presented,

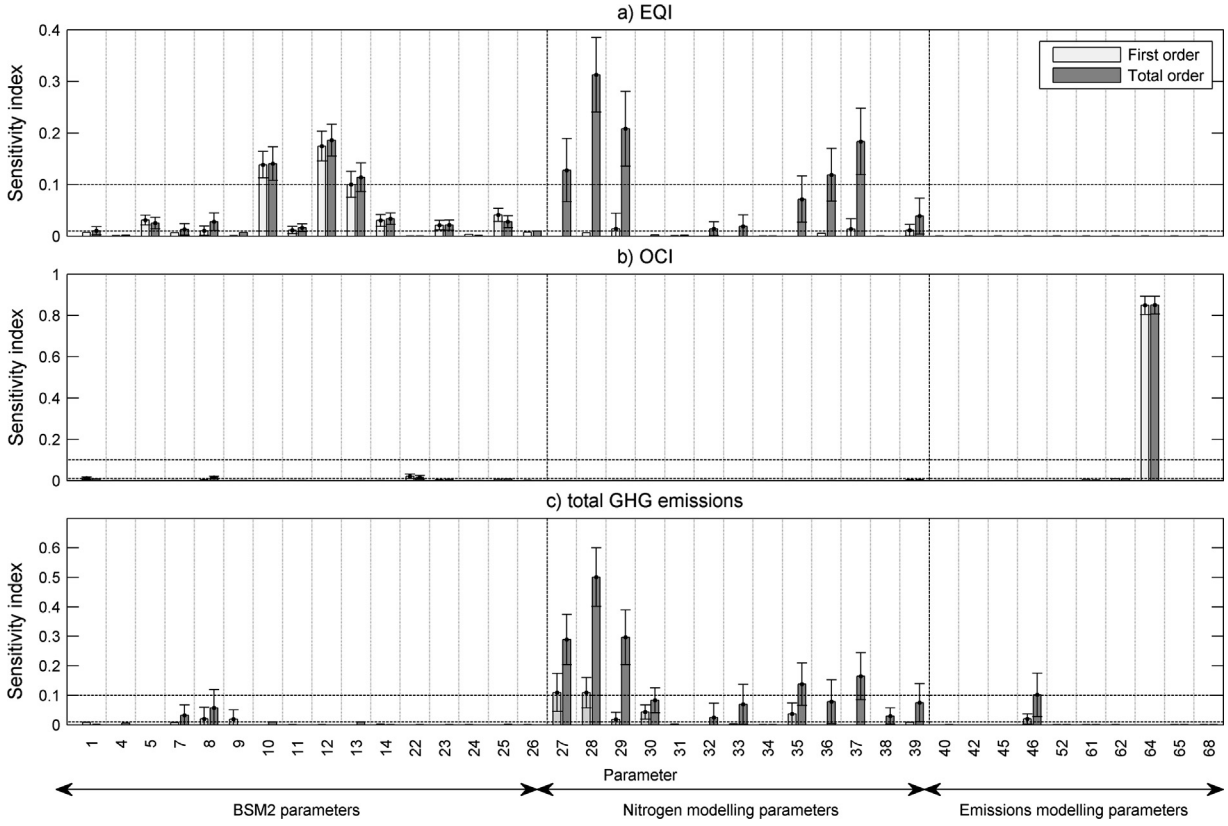

Fig. 3 – First and total order sensitivity indices calculated using Sobol's method.

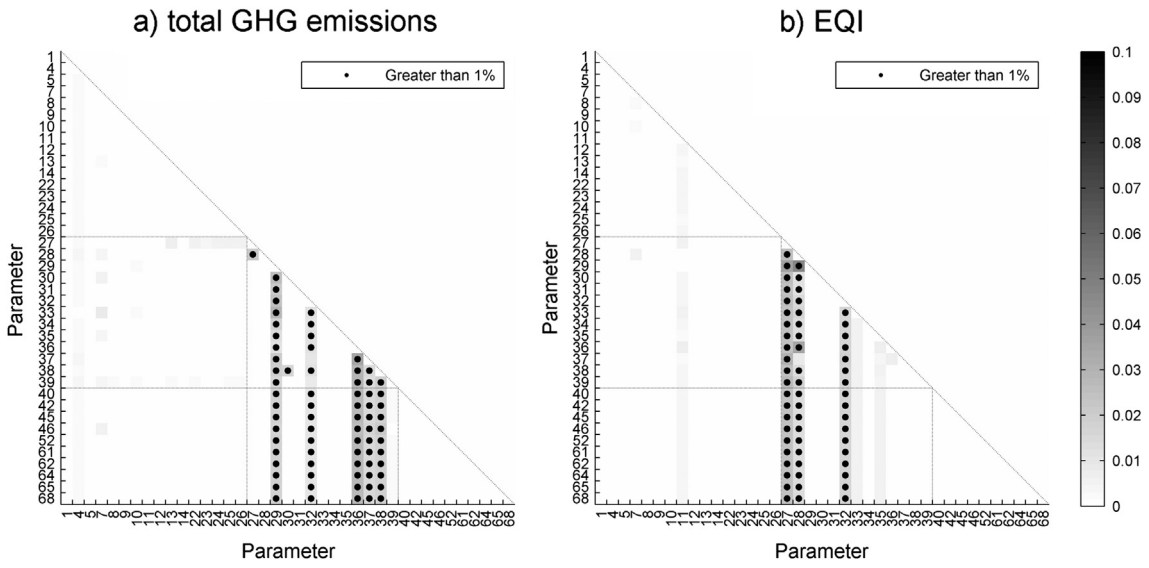

Fig. 4 – Second order sensitivity indices calculated using Sobol's method.

and parameters are classified as either 'not sensitive', 'sensitive' or 'highly sensitive' based on their contribution to output variance. A threshold of 1% contribution to output variance (i.e. a sensitivity index of at least 0.01) is used to define sensitive parameters, and a 10% contribution (i.e. a sensitivity index of at least 0.1) for highly sensitive parameters.

It is known that small numerical errors can result from the truncation of Monte Carlo approximations used in Sobol's method for calculation of integrals (Tang et al., 2007b), so slightly negative indices are assumed to equal zero. Instances in which the total order index is slightly greater than one or the total order index is less than the sum of the first and second order indices are also attributed to such errors. For the OCI, total order indices sum to less than one; this apparent error, however, is fully accounted for by the 95% confidence intervals.

Bootstrapped confidence intervals, calculated using 1000 resamples, are presented for all first and total order indices greater than 0.01. It is noted that some sensitivity indices have a high degree of uncertainty, with the greatest confidence interval being 0.501 ± 0.099. The number of samples generated for analysis was quadrupled from preliminary analyses in an attempt to reduce confidence intervals, but further increase in the number of samples is impractical due to the high computational demand. Large uncertainties are not unexpected for Sobol's method, however, due to random number generation effects (Tang et al., 2007b), and confidence intervals in excess of 20% of the corresponding sensitivity indices have been reported for previous analyses (Tang et al., 2007a,b). Despite large confidence intervals, the sensitivity indices can still be used to provide an indication of the relative significance of uncertainty in each modelling parameter in terms of its effects on model output uncertainties.

3.2.1. *Sensitivity indices based on EQI, OCI and total GHG emissions*

3.2.1.1. First and total order indices. First and total order sensitivities calculated based on EQI, OCI and total GHG emissions are presented in Fig. 3.

The EQI is shown to be sensitive or highly sensitive to twenty BSM2 and nitrogen modelling parameters, with emissions modelling parameters (predictably) having no effect. Uncertainty in the BSM2 parameters results primarily in first order effects, but it is shown that higher order effects are dominant for nitrogen modelling parameters, and that some important parameters cannot be identified based on their individual effects alone. For example, OAT sensitivity analysis suggests that EQI is not sensitive to parameters 28 and 29 (ranked 11th and 25th), but investigation into their interactions using Sobol's method shows that they are the greatest contributors to output variance.

The effects of parameter interactions on OCI uncertainty are negligible, and there is only one highly sensitive parameter: the oxygen transfer efficiency (parameter 64). OCI is also sensitive to three BSM2 parameters, although their contribution to output variance is insignificant in comparison.

Table 4 – Characteristics of total and component GHG emission results used for Sobol's method sensitivity analysis.

	Direct CO_2	Direct CH_4	Direct N_2O	Total indirect	Total GHGs
Base case (kg CO_2e/m^3)	0.4795	0.0595	0.1426	0.1872	0.8688
Mean (kg CO_2e/m^3)	0.4736	0.0596	1.1725	0.1913	1.8970
Variance ((kg $CO_2e/m^3)^2$)	0.0006	0.0003	9.6585	0.2047	9.7978

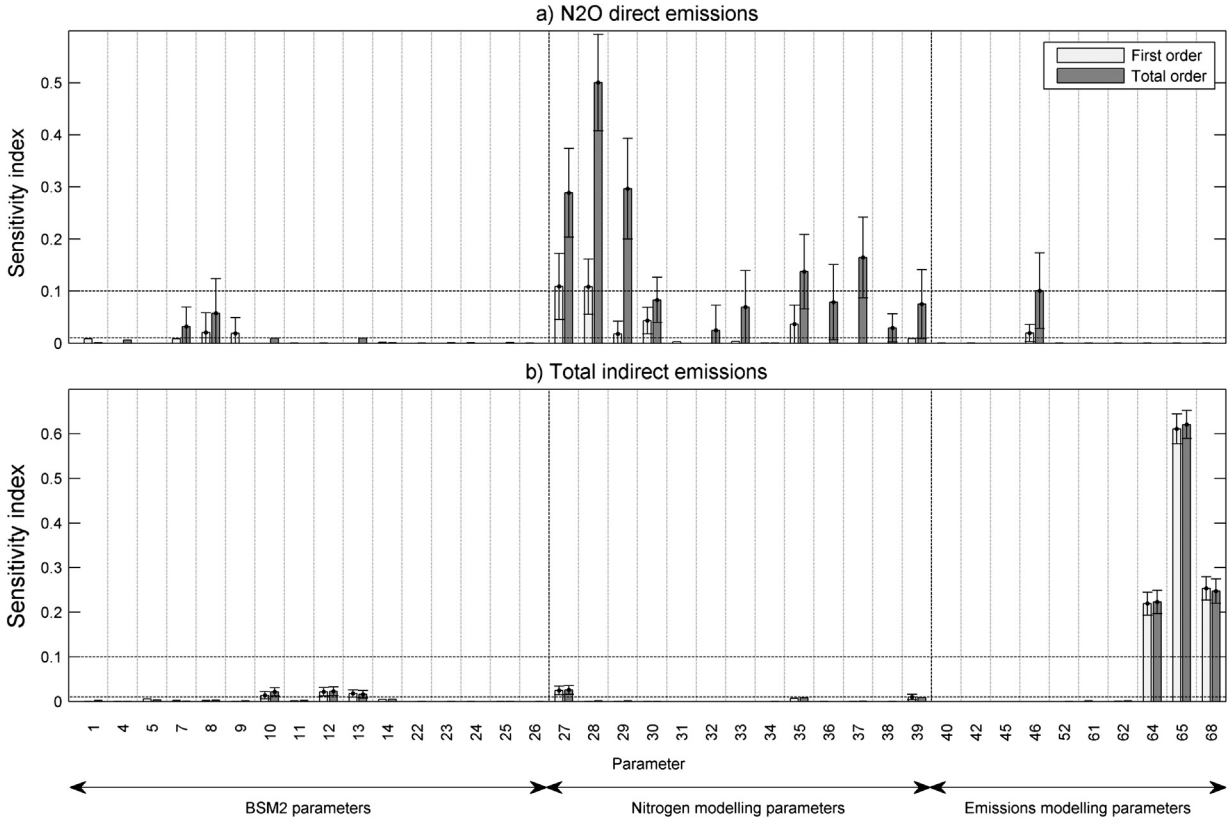

Fig. 5 – First and total order sensitivity indices based on direct N_2O emissions and total indirect GHG emissions.

All parameters classed as highly sensitive based on GHG emissions are used in the modelling of N_2O production and emission, supporting the earlier suggestion that, due to their high GWP, uncertainty in the rate of N_2O emissions is a significant contributor to uncertainty in total GHG emissions. Variance in modelled GHG emissions is predominantly due to interactions, although first order effects are still significant for some nitrogen modelling parameters: parameter 28, for example, contributes 50.1% of output variance to total output variance, with 10.9% from the parameter itself and 39.2% from its interactions with other parameters. It would, therefore, be beneficial to investigate the effects of specific interactions, to ensure that suitable allowance is made in future analyses and model calibration.

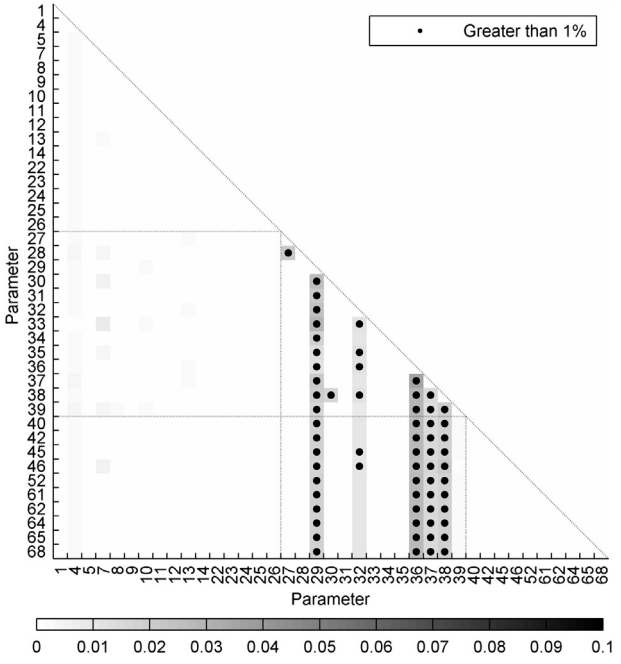

Fig. 6 – Second order sensitivity indices calculated using Sobol's method, based on direct N_2O emissions.

It can be seen that there is only one parameter to which all three key model outputs are sensitive (parameter 8), although both EQI and GHG emissions are highly sensitive to the half saturation coefficients for readily biodegradable substrate for NO_3, NO_2 and NO reduction. Fourteen parameters are not classed as sensitive based on any of the three key outputs; it is suggested that these need not be included in future uncertainty analyses.

3.2.1.2. Second order indices. Second order sensitivity indices calculated based on output GHG emissions and EQI are presented in Fig. 4 (second order indices based on OCI are not calculated since it has been shown that the effect of interactions is negligible): the shade of grey represents the sensitivity index magnitude for the corresponding parameter pair. Whilst no interactions due to individual parameter pairs can be classed as highly sensitive, there are numerous parameter pairs which have a significant impact on output variance in GHG emissions and EQI (index $\geq$ 0.01, shown with a circle).

Table 5 – Ranking of model parameters to which at least one key model output is sensitive.

Parameter number	Sensitivities based on EQI			Sensitivities based on OCI			Sensitivities based on total GHG emissions		
	GSA sensitivity rank		OAT rank	GSA sensitivity rank		OAT rank	GSA sensitivity rank		OAT rank
	First order	Total order		First order	Total order		First order	Total order	
1		20	10						
5	5	14	7						
7		19	25					12	17
8	12	12	17		3	4	6	11	11
10	2	5	3						
11	10	17	8						
12	1	3	1						
13	3	8	2						
14	6	11	5						
22				2	2	2			
23	7	15	6						
25	4	13	4						
27		6	9				1	3	4
28		1	11				2	1	7
29	8	2	25				7	2	3
30							3	7	1
32		18	19					14	14
33		16	30					10	6
35		9	13				4	5	5
36		7	15					8	9
37	9	4	28					4	8
38								13	2
39	11	10	14					9	15
46							5	6	12
64				1	1	1			

Light grey shading denotes sensitive parameters, based on corresponding index.
Dark grey shading denotes highly sensitive parameters, based on corresponding index.

Not all parameters identifiable as having significant interactions, based on the difference between their total and first order sensitivity indices, are found to have sensitive parameter pairs, and the second order effects of some parameters account for only a small proportion of total output variance resulting from their interactions. Second order effects involving parameter 28, for example, contribute to 3.1% of variance in total GHG emissions, but all interactions with this parameter contribute 39.2% of output variance, showing that higher order interactions are significant; calibration of such parameters is, therefore, likely to be challenging.

In terms of both GHG emissions and EQI, all sensitive parameter pairings include at least one nitrogen modelling parameter and the most significant second order interactions are between two nitrogen modelling parameters. This provides further support to the earlier suggestion that careful calibration of nitrogen modelling parameters is vital if model output uncertainty is to be reduced.

3.2.2. *Sensitivity indices based on component GHG emissions*

Having identified parameters to which total GHG emissions are sensitive, the effects of uncertainty in these parameters on emissions of different gases and from different sources are explored, and the contribution of uncertainty in different emission components to uncertainty in total GHG emissions is investigated.

The characteristics of GHG emissions resulting from the 160,000 parameter sets modelled for GSA are summarised in Table 4, from which it can be seen that variance in direct N_2O emissions contributes greatly to variance in total GHG emissions. Indirect emissions provide a comparatively small (12%) contribution to mean total GHG emissions, but are the second greatest contributor to total variance. Variance in direct CO_2 and CH_4 emissions provides negligible contribution to total variance, despite contributing 33% of mean total GHG emissions. This suggests that, unless uncertainty in direct N_2O emissions is significantly reduced by reduction of relevant parameter uncertainties, inclusion of parameters to which only direct CO_2 and CH_4 emissions are sensitive is unnecessary when calculating uncertainty in total GHG emissions. Further GSA therefore focuses on sources of uncertainty in direct N_2O and total indirect emissions.

First and total order sensitivity indices based on emission components are presented in Fig. 5. There is negligible difference between those based on total GHG emissions and those based on direct N_2O emissions only, confirming that reducing uncertainty in N_2O emissions is key to reducing uncertainty in total GHG emissions.

Uncertainty in indirect GHG emissions is primarily attributed to first order effects of the oxygen transfer efficiency and emission factors for carbonaceous BOD removal and N_2O from the WWTP effluent and sludge (parameters 64, 65 and 68). A further five sensitive parameters are also identifiable. Given

that the effects of interactions are negligible and the highly sensitive parameters are not classed as sensitive based on any other model output, calibration with regards to indirect emissions ought to be straightforward.

As parameter interactions are shown to contribute significantly to variance in direct N_2O emissions, second order sensitivity indices are calculated and are shown in Fig. 6. Again, the indices based on direct N_2O emissions are very similar to those based on total GHG emissions, although there are differences: whilst all sensitive parameter pairs still include at least one nitrogen modelling parameter, nine pairs involving the half saturation coefficient for NO_2 for heterotrophs (parameter 32) are no longer classified as sensitive. This suggests that their second order interactions impact primarily on other GHG emissions. All emissions modelling parameters are involved in significant second order interactions with parameters 29, 36, 37 and 38 and are, therefore, particularly important to reduce uncertainty in and consider simultaneously during calibration. Also important is the interaction between parameters 28 and 27, which alone contributes 2% of variance in direct N_2O emissions.

3.3. *Key sources of uncertainty and comparison of results*

Model parameters to which at least one of the key model outputs (EQI, OCI and total GHG emissions) is sensitive, based on the corresponding sensitivity indices, are detailed in Table 5. Shading is used to distinguish sensitive and highly sensitive parameters for each output, and rankings based on OAT sensitivity analysis results as well as first and total order indices are provided. The maximum specific hydrolysis rate (parameter 8) is classified as sensitive based on all three key model outputs, showing that it is necessary to simultaneously consider its impacts on each output during calibration. A further ten parameters are classified as sensitive based on both EQI and OCI; their effects on both effluent concentrations and GHG emissions must be taken into account during calibration. The remaining fourteen parameters are classified as sensitive based on just one model output.

OAT sensitivity analysis results provide a good indication of the most significant individual sources of uncertainty in output EQI and OCI: parameters classified as highly sensitive based on their first order indices are also the highest ranked in OAT sensitivity analysis. For GHG emissions, however, OAT sensitivity analysis did not enable correct identification of any parameters classified as highly sensitive in GSA and there are significant discrepancies between the first order index rankings and OAT sensitivity analysis rankings for all parameters. This shows that a full GSA is an important tool even when identification of only significant first order effects is required.

GSA using Sobol's method also enables identification of parameters involved in interactions with significant effects on uncertainty in the model output. As such, highly sensitive parameters have been identified which have comparatively low first order sensitivity indices and contribute to output uncertainty primarily through higher order effects. These are not all identifiable by OAT sensitivity analysis – uncertainty in parameter 28, for example, provides the greatest contribution to uncertainty in output EQI, but is ranked only 11th based on the results of OAT sensitivity analysis. This highlights the importance of including the effects of interactions when identifying and prioritising sources of uncertainty.

4. Conclusions

This research uses sensitivity analysis tools to assess the contribution of uncertain parameters in the modelling of GHG emissions from wastewater treatment to uncertainty in model outputs, and to identify parameters to which the outputs are most sensitive. Sensitivity analyses are carried out using both the OAT method (also used for screening) and Sobol's method (to enable identification of significant interactions), from which the following conclusions can be drawn:

- Parameters used in the modelling of nitrogen conversions have negligible first order (individual) effects on the EQI and, based on OAT sensitivity analysis, have a low significance rank. Use of Sobol's method, however, enables identification of parameters involved in interactions that contribute greatly to uncertainty in EQI. This highlights the importance of considering parameter interactions using a variance-based global sensitivity analysis method such as Sobol's method.
- Uncertainty in total GHG emissions from the modelled WWTP result primarily from uncertainty in direct N_2O emissions, due to their high GWP. Key sources of uncertainty in direct N_2O emissions include the half saturation coefficients for readily biodegradable substrate for NO_3, NO_2 and NO reduction. As such, further work to reduce uncertainty in these parameter values would be beneficial in order to reduce uncertainty in total GHG emissions.
- GSA reveals that parameters used in the modelling of nitrogen conversions are key sources of uncertainty in both EQI and total GHG emissions – therefore, when calibrating the model, it is important to consider the effects on both of these outputs.
- Uncertainty in the OCI is shown to be predominantly due to first order effects resulting from uncertainty in the oxygen transfer efficiency. Neither EQI or GHG emissions are sensitive to this parameter, thus calibration of model outputs used in calculation of the OCI is expected to be relatively straightforward if this knowledge is taken into account.

In summary, this study has enabled the identification of parameters that contribute significantly to uncertainty in one or more model outputs and require careful calibration, as well as those that provide negligible contribution and can be omitted from future uncertainty analyses.

Acknowledgements

The authors would like to thank Dr Ulf Jeppsson for providing the MATLAB code of BSM2 and Dr Patrick Reed for providing the C++ code of Sobol's method. Christine Sweetapple gratefully acknowledges financial support provided by the University of Exeter in the form of a studentship.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2013.05.021.

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