1 **Identifying sensitive sources and key control**
2 **handles for the reduction of greenhouse gas**
3 **emissions from wastewater treatment**

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8 **ABSTRACT**

9 This research investigates the effects of adjusting control
10 handle values on greenhouse gas emissions from wastewater
11 treatment, and reveals critical control handles and sensitive
12 emission sources for control through the combined use of local
13 and global sensitivity analysis methods. The direction of
14 change in emissions, effluent quality and operational cost
15 resulting from variation of control handles individually is
16 determined using one-factor-at-a-time sensitivity analysis, and
17 corresponding trade-offs are identified. The contribution of
18 each control handle to variance in model outputs, taking into
19 account the effects of interactions, is then explored using a
20 variance-based sensitivity analysis method, i.e., Sobol's
21 method, and significant second order interactions are
22 discovered. This knowledge will assist future control strategy

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23 development and aid an efficient design and optimisation
24 process, as it provides a better understanding of the effects of
25 control handles on key performance indicators and identifies
26 those for which dynamic control has the greatest potential
27 benefits. Sources with the greatest variance in emissions, and
28 therefore the greatest need to monitor, are also identified. It is
29 found that variance in total emissions is predominantly due to
30 changes in direct N₂O emissions and selection of suitable
31 values for wastage flow rate and aeration intensity in the final
32 activated sludge reactor is of key importance. To improve
33 effluent quality, costs and/or emissions, it is necessary to
34 consider the effects of adjusting multiple control handles
35 simultaneously and determine the optimum trade-off.

36 *Keywords:* Greenhouse gas; wastewater treatment; operation;
37 control; sensitivity

38 **1 INTRODUCTION**

39 Developing strategies for the reduction of greenhouse gas
40 (GHG) emissions is a topic of great interest and current
41 relevance, as countries have committed to emission reduction
42 targets under the Kyoto Protocol to mitigate the effects of
43 global warming. Energy use in the water industry is an
44 important source of GHG emissions; whilst in Europe it only
45 typically contributes 1% of national consumption, this is
46 predicted to increase (Olsson, 2012), and in the U.S.A. 4% of

47 electricity demand is attributable to the movement and
48 treatment of water and wastewater (Mo et al., 2010).
49 Wastewater treatment also results in the formation and direct
50 emission of the GHGs carbon dioxide (CO₂), methane (CH₄)
51 and nitrous oxide (N₂O). The wastewater sector was
52 responsible for over 5% of global non-CO₂ GHG emissions in
53 2005, and these emissions are predicted to increase by 27% by
54 2030 (U.S. Environmental Protection Agency, 2012).
55 Wastewater utilities must contribute to emission reduction
56 targets, but are faced with the challenge of simultaneously
57 improving effluent quality and managing costs.

58 Appropriate operation of wastewater treatment processes can
59 play a significant role in reducing GHG emissions (Gori et al.,
60 2011) and wastewater treatment plant (WWTP) control
61 strategies which both improve effluent quality and reduce GHG
62 emissions have been developed (Flores-Alsina et al., 2011; Guo
63 et al., 2012). However, control handles with the greatest impact
64 on GHG emissions need to be identified if significant further
65 improvements are to be made. The effects of adjusting the
66 dissolved oxygen (DO) setpoint, sludge retention time (by
67 alteration of the wastage flow rate), carbon source addition rate,
68 primary clarifier TSS removal efficiency, anaerobic digester
69 temperature and control of the digester supernatant return flow
70 on GHG emissions from different sources, as well as effluent
71 quality and operational cost, have been assessed previously

72 (Flores-Alsina et al., 2011; Flores-Alsina et al., 2014). Since
73 the effects of interactions due to simultaneous adjustments or
74 strategy implementations were not considered and variation
75 within the full range of feasible values not explored, however,
76 key findings regarding the effects of these adjustments are of
77 limited use in further control strategy development. The
78 importance of analysing a wide range of values for each control
79 handle is evidenced by the identification of non-linear
80 relationships between parameter values and effluent quality,
81 and control handle values beyond which further increase
82 produces no additional gain (Nopens et al., 2007). Previous
83 analysis (Benedetti et al., 2012) has identified control handles
84 to which effluent quality and operational cost are most sensitive
85 in the Benchmark Simulation Model No. 2 (BSM2) (Jeppsson
86 et al., 2007), taking into account simultaneous variation across
87 a range of values, but the impacts on GHG emissions have not
88 been considered. Furthermore, whilst the effects of interactions
89 are automatically considered when multiple control changes are
90 implemented, the relative significance of specific interactions
91 between control handles cannot be revealed explicitly to inform
92 control strategy development by focusing on interactions.

93 It would also be beneficial to investigate variance in GHG
94 emissions from different sources, in order that control strategy
95 development can focus on those with greatest potential for
96 improvement. For example, manufacture of material for on-site

97 usage is a key source of GHG emissions (Shahabadi et al.,
98 2010) but, given that previous studies show little variation in
99 emissions resulting from chemical consumption under different
100 control strategies (Guo et al., 2012), attempts to reduce GHG
101 emissions by reduction of carbon source addition may be
102 ineffective without introduction of alternative treatment
103 processes such as Anammox. Conversely, it has been found
104 that implementation of different control strategies can result in
105 significant variation in the magnitude of N₂O emissions from
106 activated sludge (Guo et al., 2012), suggesting that there is
107 great potential for reduction of total GHG emissions from
108 wastewater treatment by reducing N₂O emissions. It is known
109 that DO concentration and COD/N ratios, which are controlled
110 by adjustment of aeration and carbon source addition rates, play
111 a key role in controlling production of N₂O (Kampschreur et
112 al., 2009; Guo et al., 2012), yet there is a need to investigate the
113 effects on net emissions of varying these control handles
114 simultaneously, as well as the effluent quality and operational
115 cost. At present, there are conflicting observations regarding
116 the effects of WWTP control on N₂O emissions: Clippeleir et
117 al. (2014), for example, measured increased N₂O emissions
118 when operating with a high DO setpoint, whilst Guo et al.
119 (2012) found a reduction in DO setpoint to correspond with an
120 increase in N₂O emissions.

121 This research aims to detect control handles to which key
122 performance indicators (including GHG emissions, effluent
123 quality and operational cost) are sensitive and to identify the
124 most significant sources of variance in total GHG emissions,
125 taking into account interaction effects. It is important to
126 identify control handles to which GHG emissions are
127 significantly more sensitive than effluent quality or operational
128 costs, since selection of their values might be attributed little
129 importance in conventional design practices. This knowledge
130 will guide the selection of control handles for efficient and
131 effective control strategy development, based on those with
132 potential to yield the greatest improvements. Knowledge of
133 control handles to which no key model outputs are sensitive
134 will also reduce the number of decision variables required,
135 therefore reducing computational demand and improving the
136 feasibility of multi-objective optimisation for control strategy
137 development.

138 Sensitivity analysis is employed to identify important
139 parameters controlling model outputs (Tang et al., 2007a); this
140 approach can be utilised to assist system optimisation by
141 detecting the most influential control handle(s) (Naessens et al.,
142 2012), and has previously been shown to be effective (Fu et al.,
143 2012). Analysis is carried out through the combined use of a
144 local sensitivity method - one-factor-at-a-time (OAT) - and a
145 variance-based global method – Sobol’s method; this allows

trade-offs to be investigated, and reveals control handles with significant individual effects on GHG emissions, effluent quality and operational cost, as well as those with interaction effects which contribute significantly to variance in the model outputs. Model evaluations carried out with global sensitivity analysis (GSA) also reveal the most significant sources of variance in GHG emissions and, therefore, the sources from which it is most important to control and monitor GHG emissions.

2 MATERIALS AND METHODS

2.1 Wastewater treatment plant description and modelling

Wastewater treatment processes are simulated in this work using BSM2-e (Sweetapple et al., 2013), a WWTP model based on the BSM2 (Jeppsson et al., 2007) but with modifications made to enable dynamic modelling of GHG emissions (Sweetapple et al., 2013). The plant consists of a primary clarifier, an activated sludge unit containing five tanks in series (two anoxic followed by three aerobic), a secondary settler, a sludge thickener, an anaerobic digester and a dewatering unit.

Control handles included in this analysis are restricted to 14 available in BSM2 (shown in Figure S1): aeration and carbon source addition rates in each of the five reactors (KL_{a1-5} and $carb1-5$), internal recirculation flow rate (Q_{intr}), return sludge

170 flow rate (Q_r), wastage flow rate (Q_w) and reject water flow
171 rate setpoint ($Q_{storage}$). Q_r and Q_w are included despite
172 previously having been found not to have a significant effect on
173 effluent quality and operational cost (Benedetti et al., 2008),
174 since their interactions with other control handles were not
175 previously investigated, their effects on GHG emissions are
176 unknown, and the range of Q_w values considered was
177 insufficient to encompass those previously proposed for
178 operation of BSM2 (Nopens et al., 2010). It is also known that
179 wastage flow rate affects aeration requirements and sludge
180 production, both of which contribute significantly to
181 operational costs.

182 The median value for each control handles is assumed to equal
183 the BSM2 open loop default, and minimum and maximum
184 feasible values are specified in the BSM2 code (Nopens et al.,
185 2010). However, whilst a large range of values are possible, it
186 might not be realistic in practice to operate the WWTP with
187 some or all of the control handles at the extremes of their
188 allowable ranges. Therefore, for the purposes of sensitivity
189 analysis, upper and lower bounds are set to the default value
190 $\pm 10\%$ of the allowable range (with the lower limit set to zero
191 where this gives a negative number).

192 GHG emissions are modelled as detailed by Sweetapple et al.
193 (2013). Sources of direct GHG emissions include CO_2 and N_2O
194 from substrate utilisation, biomass decay and incomplete

195 denitrification in the activated sludge reactors, leakage and/or
196 combustion of CO₂ and CH₄ from the anaerobic digester, and
197 CH₄ stripped from solution in the dewatering unit. Indirect
198 emissions resulting from generation of net energy imported,
199 manufacture of chemicals used, degradation of effluent, and
200 sludge transportation and degradation are also modelled.

201 Additional CH₄ emissions, which may result from
202 unintentionally anaerobic conditions (Monteith et al., 2005), are
203 not modelled due to a lack of reliable estimation techniques.

204 N₂O emissions from nitrifier denitrification during nitrification
205 are also omitted due to a lack of suitable modelling techniques
206 – metabolic models exist (Ni et al., 2011; Mampaey et al.,
207 2013) but have been found unable to accurately and
208 consistently reproduce experimental data (Law et al., 2012; Ni
209 et al., 2013; Sperandio et al., 2014). The significance of these
210 omissions is uncertain, as previous field studies have identified
211 CH₄ emissions from every processing unit (Wang et al., 2011)
212 and nitrifier denitrification is known to yield high N₂O
213 emissions relative to the mass of nitrogen emissions converted,
214 although the proportion of nitrogen removal attributed to this
215 pathway is hard to determine (Kampschreur et al., 2009). If
216 these sources are included in future GHG emission estimates
217 for control strategy development, further work to investigate
218 their variance resulting from the choice of control handle
219 values is recommended.

220 Further details on the control handles included in this analysis
221 are provided in the supplementary information.

222 **2.2 Preliminary investigation using OAT**

223 Preliminary investigation is carried out using OAT sensitivity
224 analysis, which allows changes in model outputs to be
225 attributed to a specific control handle, with no ambiguity: two
226 WWTP performance evaluations are carried out for each
227 control handle (one with the value at its lower bound and
228 another with the value at its upper bound, whilst all other
229 control handles are held at their default value) and the
230 percentage change in each model output with respect to the
231 base case is calculated. The results are then used to identify
232 control handles with the highest control authority, and to
233 determine the direction of change in each model output
234 resulting from an increase or decrease in control handle value.

235 **2.3 Global sensitivity analysis of control handles**

236 Sobol's method (2001) is selected for GSA, as it enables the
237 impacts of interactions between specific control handles pairs,
238 as well as those of individual control handles and higher order
239 interactions, on key model outputs to be distinguished. It is
240 more effective at identifying interactions between variables in
241 highly non-linear models than alternatives such as analysis of
242 variance, gives a more detailed description of the effects of

243 individual control handles and their interactions, and provides
244 more robust sensitivity rankings (Tang et al., 2007b).

245 Sobol's method is variance-based and centres upon the
246 decomposition of total variance in a model output into
247 components resulting from specific control handles and control
248 handle interactions; Sobol's sensitivity indices of different
249 orders are then a measure of the output's sensitivity to each
250 individual control handle or control handle interaction. In this
251 study, first and total order indices are calculated for each
252 individual control handle and second order indices for each
253 control handle pair. Total order indices (S_{Ti}) represent the
254 percentage contribution of control handle i to output variance,
255 taking into account the effects of interactions of all orders.

256 Second order indices (S_{ij}) represent the contribution of
257 interaction between control handles i and j only, and first order
258 indices (S_i) the effects of control handles i alone. A high total
259 order sensitivity index, therefore, indicates a control handle
260 whose adjustment can affect model outputs significantly, and if
261 the corresponding first order index is low, the contribution to
262 output variance is predominantly due to interaction effects.

263 To implement Sobol's method, random control handle samples
264 are generated and WWTP performance evaluated using each set
265 of values in turn. The total variance of each model output is
266 calculated, and the first, second and total order sensitivity
267 indices for each control handle or control handle pair and their

268 corresponding 95% bootstrap confidence intervals are
269 computed as detailed by Tang *et al.* (2007b). Further details on
270 control handle sampling are provided in the supplementary
271 information Section 1.3.

272 **2.4 Simulation strategy and performance assessment**

273 The importance of developing GHG emission mitigation
274 strategies based on dynamic simulations has been highlighted
275 previously (Corominas *et al.*, 2012; Guo *et al.*, 2012), and
276 significant differences in N₂O emissions modelled under
277 steady-state and dynamic conditions have been identified (Guo
278 *et al.*, 2012). Sensitivity analysis, therefore, uses dynamic
279 simulations to calculate key performance indicators.
280 Performance assessment for OAT sensitivity analysis is based
281 on a one-year evaluation period, using the BSM2 simulation
282 strategy and influent data. However, given the high
283 computational demand of extended simulations and the number
284 of evaluations required, a reduced simulation period is used for
285 GSA. This consists of 200 days of constant influent to allow
286 the system to reach steady state, followed by 56 days of
287 dynamic influent, of which the final 14 are used for
288 performance evaluation. Although not fully replicating model
289 outputs from the full length simulation (since the model may
290 not reach quasi steady state with the reduced period of dynamic
291 influent preceding the evaluation, and performance will differ
292 throughout the year), this was found to be sufficient for

293 assessing the relative importance of each control handle in
294 terms of their effects on each output. Further details on the
295 choice of simulation strategy are available in the supplementary
296 information.

297 Use of a shortened evaluation period provides additional
298 benefits: if change in a specific control handle can have
299 opposite effects depending on the state of the system (e.g. due
300 to interaction with temperature), the resultant variance in mean
301 performance over an extended period may be small, despite the
302 control handle potentially being of importance. Such control
303 handles are less likely to be overlooked with a short evaluation
304 period and are of great interest since their dynamic control
305 could be particularly advantageous. For sensitive control
306 handles it is still important that potentially differing effects
307 throughout the year are considered in control strategy
308 development, however, since assumption that their behaviour
309 remains as reported in this study could lead to process control
310 related problems.

311 Average total GHG emissions per unit of wastewater treated
312 are calculated to enable identification of control handles with
313 the greatest overall effects on GHG emissions. Emissions of
314 each individual gas from each individual source are also
315 calculated, to allow more in-depth investigation into the
316 greatest sources of variability and identification of critical
317 sources. Emissions are expressed in units of CO₂ equivalent

318 (CO₂e) to take into account the differing effects of each GHG
319 on global warming and enable the relative significance of
320 emissions from different sources to be assessed. Global
321 warming potentials of 21 g CO₂e/g CH₄ and 310 g CO₂e/g N₂O
322 (IPCC, 1996) are used for CH₄ and N₂O respectively.

323 Given that design of a WWTP control strategy must also ensure
324 that an acceptable effluent quality is achieved at a reasonable
325 cost, performance is assessed using an effluent quality index
326 (EQI) and an operational cost index, as defined by Jeppsson *et*
327 *al.* (2007). The EQI is a weighted measure of the effluent loads
328 of compounds with major effects on receiving water quality;
329 the OCI is a measure of average energy use, energy recovery
330 from biogas combustion, chemical usage and production of
331 sludge for disposal.

332 **3 RESULTS AND DISCUSSION**

333 **3.1 Impacts of adjusting control handles individually**

334 The results of OAT sensitivity analysis of the control handles
335 with respect to EQI, OCI and total GHG emissions are
336 presented in Tornado diagrams (Figure 1). The percentage
337 changes in each model output with respect to the base case,
338 resulting from adjustment of each control handle to its upper
339 and lower bounds individually, are shown and effects of
340 increasing and decreasing control handle values are
341 distinguished.

342

Figure 1

343 It is shown that considering the effects on GHG emissions
344 when developing control strategies to improve effluent quality
345 and/or reduce cost is vital, since trade-offs are identifiable and,
346 in some instances (such as *KLa1* and *KLa2*), small changes in
347 EQI and/or OCI resulting from the first order effects of
348 adjusting a control handle correspond with a significant change
349 in GHG emissions.

350 OAT sensitivity analysis suggests that GHG emissions are
351 affected predominantly by aeration intensities and that
352 increasing aeration in any of the reactors would result in an
353 increase in emissions with respect to the base case. On average,
354 101% of this observed increase in net total GHG emissions is
355 attributed to increases in direct N₂O emissions: this is as
356 expected since high DO concentrations due to over aeration
357 contribute to high N₂O emissions during denitrification
358 (Kampschreur et al., 2009) and N₂O has a high GWP. Reducing
359 aeration intensities *KLa3*, *KLa4*, and *KLa5* significantly
360 reduces GHG emissions; however, there is a trade-off between
361 performance indicators, and EQI is increased by over 35%.

362 The greatest change in total GHG emissions (32%) is achieved
363 when *KLa1* is set to its upper bound. This knowledge may not
364 enable development of improved control strategies, since
365 adjustment of *KLa1* is shown only to worsen all three key

366 performance indicators, but the fact that adjustment of KL_{a1}
367 has such a significant impact on GHG emissions compared
368 with that on EQI and OCI highlights the importance of
369 selecting suitable aeration intensities when developing control
370 strategies. It may not be reasonable to actually operate the
371 WWTP with control handles at the values tested, as satisfactory
372 effluent quality would not be achieved – for example, KL_{a1} is
373 typically set to zero since the first reactor is anoxic, but an
374 increase would introduce aerobic conditions and severely
375 reduce the denitrification capacity of the plant. Decreasing
376 aeration rates in the aerobic reactors to reduce emissions could
377 also substantially increase the EQI. The relative significance of
378 each control handle in terms of each model output may differ
379 when varied only within a range that provides an acceptable
380 level of treatment. However, trade-offs must be considered and
381 in some cases, although undesirable, it may be that a deliberate
382 reduction in nitrogen removal is a possible means of reducing
383 emissions in an affordable manner.

384 EQI and OCI are affected most significantly by Q_w : reducing
385 Q_w to its lower bound (giving a SRT of 46 days, within the
386 range of an extended aeration system) results in an 85%
387 increase in EQI and an 18% reduction in OCI. It is only ranked
388 6th based on its impact on GHG emissions, but a decrease in
389 emissions corresponds with a decrease in OCI, suggesting that
390 the most cost effective choice of flow rate to achieve the

391 required effluent quality will also perform favourably in terms
392 of GHG emissions. Change in energy consumption associated
393 with pumping provides negligible (<0.2%) contribution to the
394 observed net change in emissions resulting from decreased Q_w ,
395 whilst direct emissions from activated sludge and the digester
396 contribute 58% and 32% respectively. It is not, however,
397 proposed that Q_w be decreased to the extent modelled here, due
398 to the significant adverse effects on effluent quality.

399 Adjustment of carbon source addition rates may offer potential
400 for reducing GHG emissions, based only on their individual
401 effects – it is known that a low COD/N ratio can increase N_2O
402 emissions from denitrification (Shahabadi et al., 2009), and it is
403 found that increasing *carb1* or *carb2* to their upper bound value
404 results in a 4.9% reduction in GHG emissions with negligible
405 (up to 0.8%) trade-off in EQI. This is, however, at the expense
406 of OCI, which increases by 7.0% (predominantly due to costs
407 of providing the additional carbon). No single control handle
408 can be adjusted to improve all three performance indicators
409 simultaneously, reinforcing the importance of considering
410 interaction effects in control strategy development and
411 suggesting that trade-offs may be necessary.

412 **3.2 Relative significance of first, second and total order** 413 **effects of control handles**

414 Control handles are classified as ‘highly sensitive’, ‘sensitive’
415 or ‘not sensitive’ based on their first, second and total order
416 contributions to output variance: a sensitivity index greater than
417 0.1 (i.e. a contribution of at least 10%) denotes a highly
418 sensitive control handle and a sensitivity index greater than
419 0.05 (i.e. a contribution of at least 5%) a sensitive control
420 handle. Any small discrepancies observed between
421 first/second/total order indices are fully resolved if confidence
422 intervals are considered.

423 For clarity, confidence intervals are only presented for first and
424 total order indices greater than 0.05. It is noted that some
425 confidence intervals are large, however, the impact on control
426 handle classification is small: all control handles classed as
427 highly sensitive based on any of the key model outputs retain at
428 least a sensitive classification if lower confidence bounds are
429 used. No key control handles could have been overlooked due
430 to uncertainty in the sensitivity indices, since no control
431 handles currently classed as not sensitive have an upper
432 confidence bound above the highly sensitive limit.

433 Total order sensitivity indices calculated based on EQI, OCI
434 and total GHG emissions are presented in Figure 2, with the
435 contribution of first and higher order effects shown.

436 Figure 2

437 In terms of their total order effects on GHG emissions, three
438 control handles are classified as highly sensitive: Q_w , $KLa1$ and
439 $KLa5$. Q_w is also the greatest contributor to output variance in
440 EQI and OCI and appropriate control of this control handle is,
441 therefore, vital. The importance of wastage flow rate in terms
442 of its effects on effluent quality and operational costs is already
443 recognised, but by showing the sensitivity of GHG emissions to
444 this control handle, this study highlights the necessity to
445 consider all three performance indicators when selecting an
446 appropriate value. EQI and OCI are also both either sensitive or
447 highly sensitive to variation in $KLa5$, suggesting that selection
448 of an appropriate aeration intensity is key to the reduction of
449 GHG emissions whilst maintaining an acceptable effluent
450 quality and cost. This appears intuitive, since energy
451 requirements for pumping and aeration contribute to both costs
452 and emissions, yet it has been established in OAT sensitivity
453 analysis that these control handles have a much greater effect
454 on direct emissions than on those associated with energy
455 consumption.

456 The aeration intensities $KLa1-4$ all have a significant impact on
457 GHG emissions but provide a greater contribution to output
458 variance in emissions than in EQI, suggesting that a reduction
459 in emissions with comparatively little impact on effluent
460 quality should be achievable. Furthermore, reducing emissions
461 without incurring additional costs may be possible since all

462 control handles to which GHG emissions are sensitive, except
 463 Q_w , have a higher total order sensitivity index based on GHG
 464 emissions than on OCI.

465 It is also found that interactions between control handles have a
 466 significant impact on both GHG emissions and EQI, accounting
 467 for 15% of variance in each output. As such, effective design of
 468 control strategies to reduce GHG emissions will need to
 469 consider the effects of using multiple control handles
 470 simultaneously and may require complex control algorithms.

471 Model predictive control of the DO setpoint and external
 472 carbon flow rate, for example, has been shown to enable
 473 reduced operating costs and improved effluent quality (Stare et
 474 al., 2007), although GHG emissions have not been considered.

475 GSA results show that neither EQI, OCI nor GHG emissions
 476 are sensitive to adjustment of Q_{intr} , Q_r , $Q_{storage}$, $carb2$ or
 477 $carb3$ values, so optimisation of their values is of low priority
 478 and can be omitted to simplify the design problem.

479 Reduction of OCI – or correlation of OCI with chosen control
 480 handles values – ought to be straightforward since GSA reveals
 481 no significant interaction effects and shows variance to be
 482 predominantly (62%) attributable to variation in Q_w .

483 Second order indices are presented in Figure 3, in which the
 484 darkest colours denote control handle pairs to which the
 485 corresponding output is most sensitive. Control handle pairs

486 individually accounting for more than 5% are identified and,
487 whilst no specific pairs are classified as sensitive based on
488 more than one model output, all sensitive pairs (for any model
489 output) are found to include *KLa5*. This reinforces the
490 importance of controlling *KLa5* if GHG emissions are to be
491 reduced and an acceptable effluent quality maintained, and
492 shows that interactions of *KLa5* with *Q_w*, *KLa3*, *KLa4*, *carb1*,
493 *carb2* and *carb3* must be taken into account. This appears
494 reasonable since it is known, for example, that a low SRT,
495 insufficient COD availability and low DO concentrations can
496 lead to nitrite accumulation, which in turn can contribute to
497 high N₂O emissions (Kampschreur et al., 2009). It must be
498 noted, however, that the impacts of *KLa5* adjustments and
499 interactions may differ in practice due to model limitations; in
500 this study, changes in *KLa5* have a large impact on conditions
501 in the first reactor due to the use of a standard non-reactive
502 clarifier model, but creation of anoxic conditions due to oxygen
503 consumption can occur in a reactive clarifier (Guerrero et al.,
504 2013), thereby preventing or reducing recirculation of oxygen.

505 Figure 3

506 For the EQI, no significant second order effects involving *Q_w*
507 are identified, showing that interaction effects visible in Figure
508 2 must be due to higher order effects. Selection of appropriate
509 control handle values to improve effluent quality will be

510 challenging, therefore, since Q_w is the greatest source of output
511 variance and must interact with multiple control handles.

512 Analysis of the first and total order indices shows interaction
513 effects to have negligible impact on the OCI, with only Q_w
514 involved in any identifiable interactions. This corresponds with
515 the second order indices, in which no sensitive control handle
516 pairs are found and the only interactions of note involve Q_w .

517 **3.3 Key control handles for control strategy design**

518 The results of OAT sensitivity analysis are used in conjunction
519 with those of GSA to identify key control handles for the
520 design of control strategies to reduce GHG emissions, since
521 they give an indication of the likely direction of change whilst
522 GSA explores the whole control handle space. To enable
523 comparison, control handle rankings derived from the two
524 analyses are summarized in Table 1. Results are also compared
525 to identify important control handles which may be overlooked
526 based on OAT sensitivity analysis alone. Control handles found
527 to be most important in OAT sensitivity analysis are found to
528 have significant effects in GSA, confirming that sensitive
529 control handles have not been overlooked due to the reduced
530 model stabilization and evaluation periods.

531 Table 1

532 OAT sensitivity analysis correctly identifies control handles
533 classified as highly sensitive based on EQI and OCI in GSA as
534 having the most significant effects. However, it does not enable
535 identification of all control handles to which GHG emissions
536 are highly sensitive due to the greater significance of
537 interaction effects: Q_w is ranked only 6th in OAT sensitivity
538 analysis, but GSA shows it to be the second most important
539 control handles, with its interactions contributing 7.7% of
540 output variance. Simultaneous manipulation of Q_w (to adjust
541 SRT) and other control handles (such as aeration intensities) is
542 an established approach to WWTP control, and the potential for
543 improvements in effluent quality and operational costs has been
544 demonstrated (e.g. Guerrero et al., 2012), but these results
545 highlight the importance of considering interaction effects on
546 GHG emissions also. No control handles which enable
547 simultaneous improvement in EQI, OCI and GHG emissions
548 through their first order effects alone were found, but trade-offs
549 may be lessened or avoided when interactions are considered.

550 In this study, the impact of Q_w on EQI is shown to be
551 predominantly due to first order effects and OAT sensitivity
552 analysis results suggest that adjustment is only likely to worsen
553 effluent quality. It is also shown, however, that GHG emissions
554 and OCI can both be reduced through the first order effects of
555 Q_w . Given that interaction effects with Q_w do contribute to
556 variance in EQI, and significantly to variance in GHG

557 emissions, simultaneous improvements which are not revealed
 558 through OAT sensitivity analysis alone might be possible
 559 through appropriate control of Q_w and its interacting control
 560 handles.

561 All three outputs are sensitive or highly sensitive to adjustment
 562 of $KLa5$. However, OAT sensitivity analysis shows that a
 563 decrease in $KLa5$ corresponds with a significant reduction in
 564 GHG emissions and OCI but an increase in EQI, so adjustment
 565 to reduce emissions whilst maintaining acceptable effluent may
 566 not be straightforward. An increase in $KLa5$ results in a small
 567 improvement in EQI but significantly worsens GHG emissions;
 568 this reinforces the necessity to consider the effects on GHG
 569 emissions when control is modified to improve effluent quality
 570 and supports previous recommendation that GHG emissions
 571 should be included as an evaluation criterion to provide a
 572 clearer picture of the overall suitability of WWTP control
 573 strategies (e.g. Flores-Alsina et al., 2014). GSA also shows
 574 $KLa5$ to be involved in significant interaction effects, further
 575 complicating the design problem. In particular, the effects of
 576 interaction with Q_w on GHG emissions and interaction with
 577 $KLa3$ on EQI should be considered.

578 GHG emissions are found to be highly sensitive to $KLa1$ and
 579 sensitive to $KLa2$, whilst effects of these control handles on
 580 EQI and OCI are insignificant. This might imply that
 581 adjustment of $KLa1$ and $KLa2$ could be used to reduce

582 emissions without incurring trade-offs; however, the base case
583 value for both is zero and OAT sensitivity analysis shows only
584 a significant increase in emissions resulting from change in
585 *KLa1* and *KLa2*. Therefore, although they have a significant
586 impact on GHG emissions, there may be no benefits from
587 altering the base case values as performance would only be
588 worsened. Given the high sensitivity of *KLa1*, however, it is
589 recommended that the effects of small alterations are
590 investigated since these would be missed in OAT sensitivity
591 analysis and may be beneficial.

592 Interaction effects involving *KLa3* are shown to be particularly
593 important, as GHG emissions would not be classified as
594 sensitive to this control handle based on its first order effects
595 alone. Given that neither EQI nor OCI are sensitive to *KLa3*
596 and OAT sensitivity analysis shows that adjustment to reduce
597 emissions is possible, suitable control of aeration in the first
598 aerobic reactor is likely to be key to the development of control
599 strategies to reduce GHG emissions – although complex, given
600 interactions mostly involve at least three control handles.

601 Appropriate control of *KLa4* is also important, since it is
602 classified as sensitive based on both EQI and GHG emissions.
603 OAT sensitivity analysis reveals a trade-off: a reduction in
604 GHG emissions due to individual adjustment of *KLa4*
605 corresponds to an increase in EQI, but because GSA shows the
606 effects of interactions to involving *KLa4* to be significant, it is

607 likely that the comparative magnitude of effects on each output
608 differs across the range of feasible values and an optimum can
609 be identified.

610 In GSA, *carb1* is classified as sensitive based on OCI only and,
611 as such, might be adjusted in an attempt to reduce cost with
612 little impact on effluent quality or emissions. However, OAT
613 sensitivity analysis shows that a decrease in OCI due to
614 reduction of *carb1* corresponds with an increase in GHG
615 emissions. Therefore, if *carb1* is lowered to reduce operational
616 cost, it is vital that the impact on GHG emissions is considered
617 and, if necessary, countered with other measures.

618 EQI, OCI and GHG emissions are not sensitive to Q_{intr} , Q_r ,
619 $Q_{storage}$, *carb2* and *carb3*, suggesting that dynamic control of
620 these control handles would be of little benefit. It is, therefore,
621 recommended that optimisation of internal recirculation flow
622 rate, return sludge flow rate, anoxic reactor carbon source
623 addition rates (except in first reactor) and storage tank control
624 is of low priority when developing new WWTP control
625 strategies. It has been demonstrated that control strategy
626 optimisation using this knowledge can enable substantial
627 emission reductions whilst maintain an acceptable effluent
628 quality and without increasing operational costs (Sweetapple et
629 al., 2014).

630 **3.4 Key emission sources for reduction of greenhouse gas**
631 **emissions**

632 Based on simulations undertaken for GSA, the base case value,
633 mean and variance of emissions from different sources are
634 detailed in Table 2. Total GHG emissions are decomposed into
635 direct emissions of each gas and indirect emissions from each
636 source, as well as those resulting from the wastewater line and
637 sludge line. Wastewater line emissions include all direct
638 emissions associated with the activated sludge reactors and
639 indirect emissions resulting from effluent degradation and
640 energy demand for reactor aeration and mixing, chemical
641 consumption; sludge line emissions include those from biogas
642 leakage, combustion and energy recovery, dewatering, energy
643 for digester heating and mixing, and transport and offsite
644 degradation of sludge. It is noted that variances reported are
645 small in comparison with those resulting from model parameter
646 uncertainties (Sweetapple et al., 2013), and future work should
647 investigate the impact of modelling uncertainties on control
648 strategy design.

649 Table 2

650 It is notable that, whilst direct CO₂ emissions are the greatest
651 contributor to total GHG emissions (at 48%), their output
652 variance is just 1.7% of that of direct N₂O emissions, which
653 contribute only a comparatively small 24% of mean total GHG

emissions. Indirect emissions and direct CH₄ emissions contribute 28%, yet are found to have negligible variance. This shows that the source of emissions with the greatest scope for improvement does not necessarily correspond with the overall greatest source of emissions, and suggests that any reduction in GHG emissions resulting from modified control will be primarily due to a reduction in N₂O emissions. Control strategy development and optimisation should, therefore, focus on reduction of direct N₂O emissions, all of which result from wastewater processes (specifically, activated sludge), and it is important that N₂O emissions are carefully monitored to ensure that they are not unintentionally increased as a result of actions to improve effluent quality and/or reduce operational costs. Existing knowledge that a reduction in DO setpoint to reduce costs can result in an increased risk of N₂O production (Porro et al., 2014) supports this recommendation. A potential strategy for mitigating the risk whilst maintaining cost savings may include better control and distribution of the aeration (Porro et al., 2014).

Further sensitivity analysis is used to investigate key control handles affecting wastewater line and sludge line GHG emissions, and OAT sensitivity analysis results are presented in Figure 4.

Figure 4

678 In OAT sensitivity analysis it is shown that changes in total
679 GHG emissions are predominantly due to variation in
680 wastewater line emissions, with only Q_w resulting in a change
681 of emissions of more than 0.7% in the sludge line. In GSA also,
682 variance in sludge line emissions is negligible in comparison
683 with that of wastewater line emissions and is found to be
684 primarily due to the first order effects of Q_w . The ranking of
685 each control handle based on total order effects on wastewater
686 line emissions is identical to that for total GHG emissions, but
687 an additional sensitive control handle, *carb1*, is identified. The
688 significance of first order effects of variation in *KLa3* is also
689 greater on wastewater line emissions than on total emissions,
690 with the control handle classified now classified as sensitive
691 based on its first order index.

692 OAT sensitivity analysis shows a decrease in Q_w , the only
693 control handle to which sludge line emissions are sensitive, to
694 correspond with a decrease in both sludge line and wastewater
695 line emissions (and vice versa). WWTP modelling used during
696 control strategy development for the reduction of GHG
697 emissions could, therefore, justifiably omit sludge line
698 emissions in order to reduce computational demand, since there
699 is little potential for their reduction from improved control
700 alone and any small change observed is likely to be a decrease
701 if Q_w is manipulated to aid reduction of wastewater line
702 emissions.

703 **4 CONCLUSIONS**

704 This research has investigated the impact of adjusting 14
705 WWTP control handles, including flow rates, aeration rates and
706 carbon source addition rates, to enable identification of key
707 control handles and sensitive sources for the reduction of GHG
708 emissions. Based on the results of OAT sensitivity analysis and
709 Sobol's method GSA, the following conclusions are drawn:

- 710 • It is vital to consider the effect on GHG emissions when
711 developing control strategies to improve effluent quality
712 and/or reduce cost as, in some instances, a small change
713 in EQI and/or OCI resulting from the individual effects
714 of adjusting a control handle corresponds with a
715 significant change in GHG emissions, and trade-offs
716 between objectives have been identified.
- 717 • Selection of suitable values for aeration intensity in the
718 final tank and wastage flow rate in the activated sludge
719 process is of key importance, and active control of these
720 control handles may be beneficial, but it is essential that
721 their impacts on GHG emissions are considered. Both
722 have a significant individual impact on variance in all
723 three model outputs, and EQI and GHG emissions are
724 also sensitive to interaction effects involving the
725 aeration intensity.

- 726 • Unless effluent quality and/or operational cost are to be
727 sacrificed, it is necessary to consider the effects of
728 adjusting two or more control handles together when
729 developing control strategies to reduce GHG emissions,
730 since no control handles enabling simultaneous
731 improvement in EQI, OCI and GHG emissions through
732 their individual effects alone were identified.
- 733 • Formation of N₂O in the activated sludge process is the
734 source of GHG emissions with the greatest scope for
735 improvement, and from which it is important that
736 emissions are carefully monitored to ensure that they
737 are not unintentionally increased as a result of actions to
738 improve effluent quality and/or reduce operational
739 costs.
- 740 • Dynamic control of internal recirculation and return
741 sludge flow rates, reject water flow rate set point and
742 carbon source addition in second and subsequent anoxic
743 reactors would be of little benefit and it is
744 recommended that optimisation of these control handles
745 is of low priority since they were not classified as
746 sensitive based on EQI, OCI or GHG emissions.

747 It is hoped that this knowledge will assist future development
748 of WWTP control strategies to reduce GHG emissions whilst
749 maintaining acceptable effluent quality and operating costs, and
750 aid an efficient design and optimisation process.

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933

934

935 **FIGURE CAPTIONS**

936 *Fig. 1 – Percentage change in model outputs resulting from*
937 *individual variation of control handles*

938 *Fig. 2 – First and total order indices calculated using Sobol’s*
939 *method, based on EQI, OCI and total GHG emissions*

940 *Fig. 3 – Second order sensitivity indices calculated using*
941 *Sobol’s method based on EQI and total GHG emissions*

942 *Fig. 4 – Change in wastewater line and sludge line GHG*
943 *emissions resulting from variation of individual control*
944 *handles, as a percentage of base case total GHG emissions*

945 **TABLE CAPTIONS**

946 *Table 1 – Ranking of control handles based on OAT sensitivity*

947 *analysis and GSA*

948 *Table 2 – Characteristics of GHG emissions from key sources*

949

951 *Table 1 – Ranking of control handles based on OAT sensitivity analysis and GSA*

952

Control handle	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	
Qintr	10	13	6	13	13	13	8	9	8
Qr	8	11	8	12	10	10	12	13	12
Qw	1	1	1	1	1	1	2	2	6
Qstorage	6	14	7	14	14	14	14	14	14
KLa1	14	5	5	10	12	11	3	3	1
KLa2	4	6	11	11	11	13	7	6	4
KLa3	12	4	3	9	9	4	5	4	3
KLa4	3	3	4	6	8	5	4	5	5
KLa5	2	2	2	3	3	3	1	1	2
carb1	13	8	14	2	2	2	6	7	7
carb2	7	9	13	8	7	7	9	8	9
carb3	9	12	10	7	6	6	13	12	13
carb4	5	10	9	5	4	6	10	11	13
carb5	11	7	12	4	5	8	11	10	10
Light grey shading denotes sensitive control handles, based on corresponding index									
Dark grey shading denotes highly sensitive control handles, based on corresponding index									

953 *Table 2 – Characteristics of GHG emissions from key sources*

954

	Direct emissions				Indirect emissions			Total wastewater line	Total sludge line	Total GHGs
	Direct CO ₂	Direct CH ₄	Direct N ₂ O	Net energy	Carbon source	Reactor effluent offsite GHGs	Sludge offsite GHGs			
Base case (kg CO ₂ e/m ³)	0.47	0.06	0.15	0.06	0.04	0.03	0.06	0.55	0.32	0.87
Mean (kg CO ₂ e/m ³)	0.47	0.06	0.24	0.06	0.06	0.03	0.05	0.66	0.32	0.98
Variance ((kg CO ₂ e/m ³) ²)	0.00	0.00	0.03	0.00	0.00	0.00	0.00	0.02	0.00	0.03

955