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1 **Potential of Interactive Multiobjective Optimization in Supporting the Design of**
2 **a Groundwater Bionitrification Process**

3
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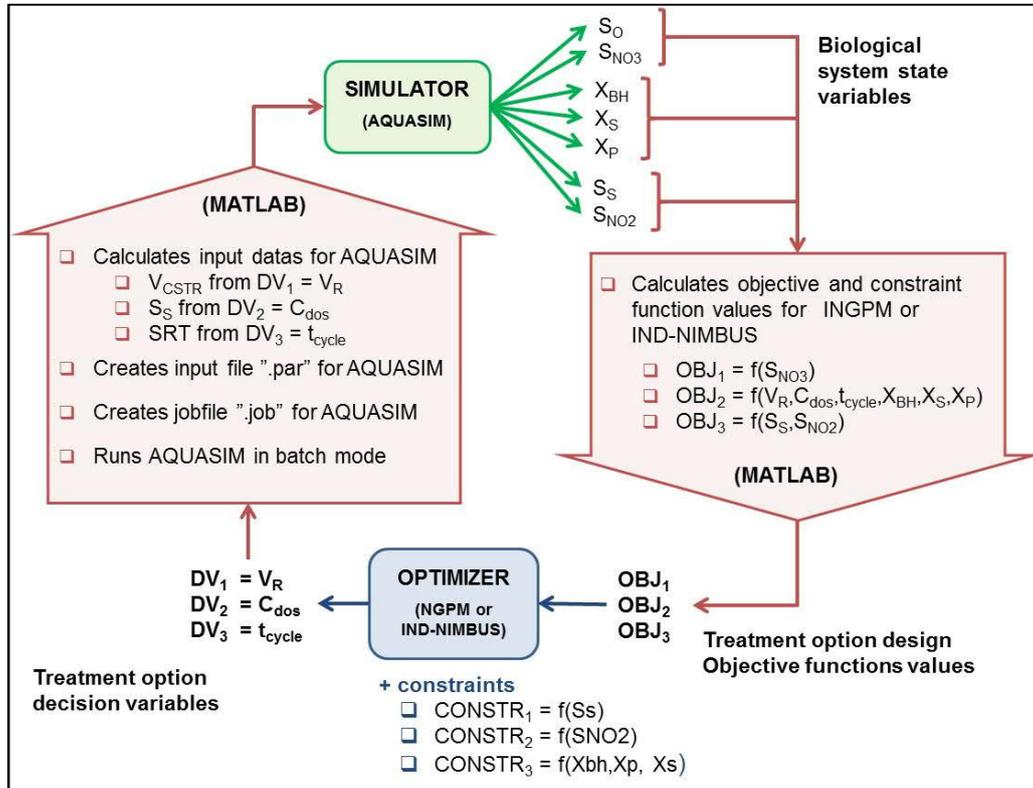
SUPPLEMENTARY MATERIAL

26

27

Text S1. Conceptual representation of the design optimization problem

29 The iterative multiobjective design optimization problem is sketched in the flowchart reported in Figure S1.



30

31

Figure S1. Flowchart representing the design optimization problem constituting units

32

Text S2. Hydraulic characterization of the biofilter

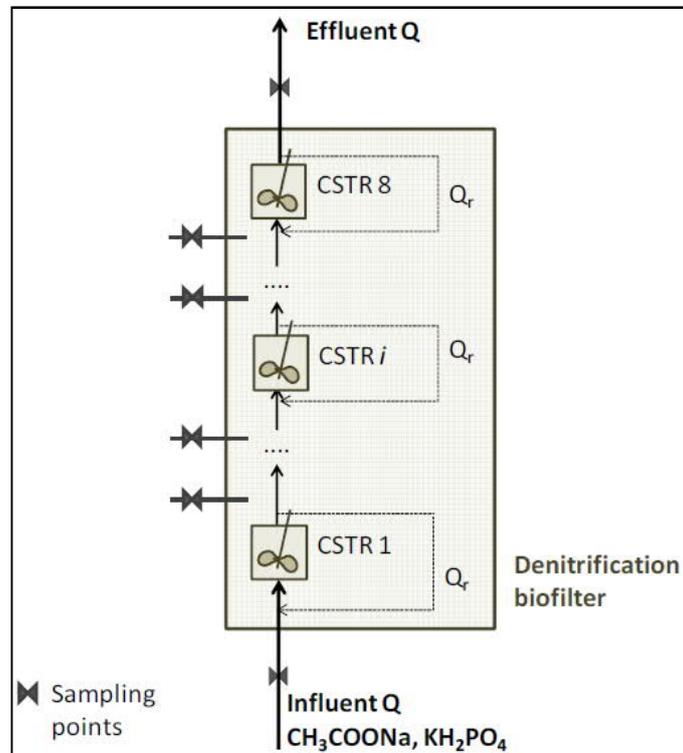
34 The hydraulic characterization has been performed by an impulsive tracer test, obtaining the Residence Time Distribution (RTD) and calculating the average hydraulic retention time (HRT). Four campaigns were performed. In the first one, before the biofilter colonization, NaCl was used (16.5 g NaCl), monitoring the conductivity of the water at the filter outlet. In the others campaigns, LiCl was used sampling effluent water at intervals of time defined by the results of the NaCl tracer tests. LiCl has been used as tracer as it doesn't present any toxicity, inhibition or accumulation effect on biomass (Séguret e Racault, 1998; Olivet et al., 2005). The first campaign with LiCl (4.2 mg LiCl) was carried out before biofilter colonization, the second and third campaigns were carried out immediately after the biofilter start-up phase (1.1 mg LiCl) and at the end of the process evaluation phase (1.0 mg LiCl). All tracer tests were performed at continuous flow rate of 0.6 m³/h.

43

Text S3. Biofilter simulation model

45 The biofilter has been hydraulically modelled as eight CSTRs in series, as in Figure S.2. Attached biomass biofilter is reported to be modelled through different kinds of biofilm models (Boltz et al., 2010; Huang et al., 1998; Morgenroth et al., 2000). However, these models have become more and more

48 complex (Morgenroth et al., 2000) and, when the main modelling purpose is the prediction of biofilter removal
 49 efficiency, simpler models can be applied (Lazarova et al., 1994, 1992; Vrtovšek and Roš, 2006) considering
 50 only the pollutant removal rate (i.e. denitrification rate) without any simulation of biomass growth and decay
 51 processes. A good trade-off among model simplicity and predictability of both dissolved and particulate matter
 52 concentrations is represented by variations of the Activated Sludge Models (ASM) that are reported to be
 53 applied for attached biomass systems as Moving Bed Bioreactors (Plattes et al., 2007, 2006) and submerged
 54 bioreactors (Huang et al., 1998; Ordaz et al., 2012). However, none of the ASM models (not even ASM2) is
 55 able to predict concentrations of nitrite produced during nitrate reduction, since it considers denitrification as
 56 a one step process (i.e., nitrate reduction to nitrogen biogas). Anyway, in drinking water treatment, nitrite
 57 accumulation cannot be neglected as it is a known carcinogen whose concentration is subjected to a stringent
 58 regulation limit. Therefore, more properly, a 2-step denitrification kinetic should be considered.
 59



60
 61 **Figure S2.** Hydraulic modelling of the pilot biofilter through 8 CSTRs in series

62
 63 **Table S1.** Biological and physical processes assumed to occur in the biofilter following ASM1 [Henze et al.,
 64 2000]

Process name	S_S	S_O	S_{NO_2}	S_{NO_3}	X_{BH}	X_S	X_P	RATE
Aerobic Growth	$-\frac{1}{Y_H}$	$-\frac{1 - Y_H}{Y_H}$		$-i_{XB}$	1			r_1
Anoxic Growth NO ₃	$-\frac{1}{Y_H}$		$+\frac{1 - Y_H}{1,14 \cdot Y_H}$	$-\frac{1 - Y_H}{1,14 \cdot Y_H}$ $-i_{XB}$	1			r_2

Anoxic Growth NO₂	$-\frac{1}{Y_H}$		$-\frac{1 - Y_H}{1,71 \cdot Y_H} - i_{XB}$		1			r ₃
Decay					-1	$1 - f_p$	f_p	r ₄
Hydrolysis	1					-1		r ₅

65

66 **Table S2.** Process rates in mg_{COD}/L·d taken from ASM1 by Henze et al. [2000]^[1], 2-step denitrification models
67 by Magrì and Flotats [2008]^[2], Kornaros and Lyberatos [1998]^[3].

Name	Process	Expression
r ₁ ^[1]	Aerobic Growth	$\hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S}\right) \cdot \left(\frac{S_O}{S_O + k_{O,H}}\right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₂ ^[2]	Anoxic growth on nitrate	$\eta_{g1} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S}\right) \cdot \left(\frac{S_{NO3}}{S_{NO3} + k_{NO3}}\right) \cdot \left(\frac{k_O}{k_O + S_O}\right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₃ ^[3]	Anoxic growth on nitrite	$\eta_{g2} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S}\right) \cdot \left(\frac{S_{NO2}}{S_{NO2} + k_{NO2}}\right) \cdot \left(\frac{k_O}{k_O + S_O}\right) \cdot \left(\frac{k_{I,NO3}}{k_{I,NO3} + S_{NO3}}\right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₄ ^[1]	Biomass decay	$b_H \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₅ ^[1]	Hydrolysis of entrapped organics	$k_h \cdot \frac{X_S}{K_X \cdot X_{BH} + X_S} \cdot \left[\left(\frac{S_O}{S_O + k_{O,H}}\right) + \eta_h \cdot \left(\frac{k_{O,H}}{S_O + k_{O,H}}\right) \cdot \left(\frac{S_{NO3} + S_{NO2}}{S_{NO3} + S_{NO2} + k_{NO}}\right) \right] \cdot X_{BH}$

68

69 **Table S3.** Model parameters values taken from: ASM1 by Henze et al. (Henze et al., 2000)^[1], 2-step
70 denitrification models by Magrì and Flotats (Magrì and Flotats, 2008)^[2], Kornaros and Lyberatos (Kornaros
71 and Lyberatos, 1998)^[3] and bioenergetic evaluations by Henze et al. (Henze et al., 2008)^[4].

Parameter	Value	Unit	Meaning
X _{BH,0}	0.01	mg _{COD} /L	Initial concentration of heterotrophic active biomass
X _{P,in}	0.01	mg _{COD} /L	Influent concentration of non-biodegradable particulate matter
X _{S,in}	0.01	mg _{COD} /L	Influent concentration of slowly-biodegradable substrate
$\hat{\mu}_H$	4.16 ^[2]	1/d	Maximum specific growth rate for heterotrophic biomass
b _H	0.62 ^[1]	1/d	Decay rate for heterotrophic biomass
k _h	3.0 ^[1]	1/d	Maximum specific hydrolysis rate
θ	1.03 ^[1]	-	Correction factor for temperature effect on biomass growth and decay rates
η _{g1}	0.23 ^[2]	-	Correction factor for μ _h for anoxic growth on nitrate-nitrogen
η _{g2}	0.592 ^[3]	-	Correction factor for μ _h for anoxic growth on nitrite-nitrogen
η _h	0.4 ^[1]	-	Correction factor for hydrolysis under anoxic conditions
k _s	4 ^[2]	mg _{COD} /L	Half-saturation constant for organic substrate

$k_{O,H}$	0.2 ^[1]	mg _{COD} /L	Half-saturation constant for oxygen
k_{NO_3}	0.5 ^[2]	mg _N /L	Half-saturation constant for nitrate nitrogen
k_{NO_2}	0.28 ^[3]	mg _N /L	Half-saturation constant for nitrite nitrogen
k_x	0.03 ^[1]	-	Half-saturation constant for slowly biodegradable substrate
k_{I,NO_3}	8.75 ^[3]	mg _N /L	Nitrate inhibition constant
Y_h	0.46 ^[4]	mg _{COD} /mg _C OD	Heterotrophic biomass yield, considering NO ₃ -N uptake for protein synthesis
f_p	0.08 ^[1]	-	Fraction of biomass leading to particulate products
i_{XB}	0.086 ^[1]	mg _N /mg _{COD}	Mass of nitrogen per mass of COD in biomass

72

73 **Text S4. Optimization problem formulation**

74 The definition of design variables, objective functions and constraints for the multiobjective design
75 optimization problem has been driven by considerations based on practical experience and literature data.

76 For **design variables**, lower and upper bounds had to be indicated. Bounds for reactor volume (V_r , expressed
77 in L) have been defined by assuming acceptable values for **denitrification contact time** (Empty Bed Contact
78 Time, EBCT). Richard (Richard, 1989) reports an EBCT of 23-38 min and 22-43 min for two different full
79 scale groundwater denitrification biofilters with expanded clay as biomass carrier. These values are in
80 agreement with recommendations reported by Metcalf and Eddy (Tchobanoglous et al., 2014), who suggest
81 EBCT values in the range of 20-30 min for submerged denitrification reactors, and by Pujol et al. (Pujol et al.,
82 1994), who suggest 15-60 min as a possible EBCT range. Nurizzo and Mezzanotte (Nurizzo and Mezzanotte,
83 1992) report EBCTs in the range of 8-36 min for groundwater denitrification in a biofilter using sand as
84 biomass support media of granules size comparable to expanded clay. EBCT was then set to vary within the
85 range 5-120 min, leading to a reactor volume ranging between 50-1200 L.

86 As for the **external carbon source dosage** (C_{dos} , expressed in mg_{COD}/L), the lower bound corresponds to no
87 carbon dosage (0 mg_{COD}/L) while the upper bound ($C_{dos,ub}$) was chosen in order not to set a practical limit to
88 this variable. A value of 200 mg_{COD}/L was chosen, corresponding to a dosage four times higher than the
89 stoichiometric requirements (see equations eq. S1 - S2), calculated considering influent electron acceptor
90 concentrations reported in Table 1, stoichiometric ratios reported in Section 2.1 and 2.667 g_{COD}/g_C for sodium
91 acetate dosage. This excess value is a reasonable upper bound: it should not imply any inhibition of denitrifying
92 biomass when acetic acid is adopted as carbon source, as reported by Her and Huang (Her and Huang, 1995),
93 who reached a 1600% carbon dosage excess. Furthermore, considering that Tang et al. (Tang et al., 2011)
94 observed 19 mg_{COD}/L in the effluent just with a 25% excess of carbon dosage (as CH₃COONa), unacceptable
95 carbon concentrations in the effluent should be limiting the optimal values of this design variable before
96 reaching its upper bound, as desired.

$$C_{dos,ub} = \alpha \cdot \left[\left(\frac{C}{DO} \right) \cdot DO_{in} + \left(\frac{C}{NO_3 - N} \right) \cdot (NO_3 - N)_{in} + \left(\frac{C}{NO_2 - N} \right) \cdot (NO_2 - N)_{in} \right] \cdot 2.667 \frac{mg_{COD}}{mg_C} \quad (S1)$$

$$C_{dos,ub} = 4 \cdot \left[0.70 \frac{g_C}{g_{DO}} \cdot 6.2 \frac{mg_{DO}}{L} + 1.69 \frac{g_C}{g_{NO_3-N}} \cdot 8.9 \frac{mg_{NO_3-N}}{L} + 1.27 \frac{g_C}{g_{NO_2-N}} \cdot 0.014 \frac{mg_{NO_2-N}}{L} \right] \cdot 2.667 \frac{mg_{COD}}{mg_C} \cong 200 \frac{mg_{COD}}{L} \quad (S2)$$

97 Finally, for Sludge Retention Time (SRT, expressed in days) lower and upper bounds were set, respectively,
 98 at 1 d and 100 d considering that MetCalf and Eddy (Tchobanoglous et al., 2014) report 20-40 d as typical
 99 SRT range for denitrifying submerged biofilters.

100 As for **objective functions**, the first and the third objectives were both expressed in terms of water measurable
 101 characteristics, as expressed by eq. S3 and eq. S4.

$$OBJ_1 = NO_3 - N_{out} \left[\frac{mg_N}{L} \right] = S_{NO_3} \quad (S3)$$

$$OBJ_3 = COD_{out} \left[\frac{mg_{COD}}{L} \right] = S_S + S_{NO_2} \cdot 1.71 \frac{mg_{COD}}{mg_N} \quad (S4)$$

102 On the other hand, the second objective function, representing investment and management costs, was defined
 103 based on data from practical experience and values refer to Italian prices and conditions. Considered costs
 104 were the ones significantly varying as a function of design variable variation:

105 - Reactor volume considerably affects investments costs ($Cost_{invest}$) in terms of cost of biofilter building
 106 works (160-200 €/m³) and filling material requirements (500-630 €/m³), as expressed by equation eq. S5:

$$Cost_{invest} [\text{€}] = (0.2 + 0.6) \frac{\text{€}}{L} \cdot V_R \quad (S5)$$

107 - Carbon dosage considerably affects management costs ($Cost_{reag}$) in terms of reagent supply (850 €/t of
 108 CH₃COOH as 15% solution). The reagent supply costs have been calculated by equation eq. S6,
 109 considering 2.667 g_{COD}/g_C in case of acetic acid dosage.

$$Cost_{reag} [\text{€/yr}] = \frac{C_{dos} \cdot 14400 \frac{L}{d}}{2.667 \frac{g_{COD}}{g_C}} \cdot \frac{60 \frac{g_{AC}}{mol}}{24 \frac{g_C}{mol}} \cdot \frac{1}{0.15} \cdot \frac{365 \frac{d}{yr}}{10^6 \frac{mg}{kg}} \cdot 0.85 \frac{\text{€}}{kg} \quad (S6)$$

110 - Sludge Retention Time affects management costs ($Cost_E$ and $Cost_{sludge}$) due to energy consumption
 111 (0.10-0.20 €/kWh) and sludge disposal (150 €/t of sludge, wet weight) by defining backwashing
 112 frequency, as treatment cycle duration (t_{cycle} , expressed in hours).

113 Thus, treatment cycle duration can be derived from the definition of SRT, which is the ratio between the
 114 total amount of particulate matter in the reactor ($M_{TOT,r}$) and the amount of sludge removed per unit time.
 115 For submerged biofilters, sludge removal is achieved through backwashing; thus, sludge removal can be
 116 expressed as sludge removal for single backwashing ($\Delta M_{TOT,bw}$) divided by treatment cycle duration
 117 (t_{cycle}). Thus, SRT definition can be expressed by eq. S7 to eq. S9.

$$SRT[d] = \frac{M_{TOT,r}}{\Delta M_{TOT,bw} / t_{cycle}} \quad (S7)$$

$$M_{TOT,r} = \sum_{i=1}^8 (X_{BH,i} + X_{P,i} + X_{S,i}) \cdot V_{cstr,i} \quad (S8)$$

$$\Delta M_{TOT,bw} = V_{bw} \cdot X_{TOT,bw} = (Q_{bw} \cdot t_{bw}) \cdot X_{TOT,bw} \quad (S9)$$

118 where $M_{TOT,r}$ is calculated in eq.S8 as the sum of particulate matter in each one of the eight reactors in
 119 series (with index i referring to the considered CSTR). At the same time, in eq. S9 $\Delta M_{TOT,bw}$ has been
 120 expressed following the backwashing procedure reported by Richard (1989) on real scale Biofor^(R) for
 121 drinking water treatment plants. The volume of backwashing water (V_{bw}) has been determined considering
 122 a backwashing cycle duration (t_{bw}) of 1 h and a backwashing filtration rate 2 times the treatment cycle
 123 filtration rate, involving a total particulate matter concentration in backwashing water ($X_{TOT,bw}$) of
 124 192 mg_{COD}/L. Thus, $\Delta M_{TOT,bw}$ can be expressed by eq. S10, where Q is the influent flow rate (in L/h),
 125 allowing the determination of t_{cycle} through eq. S11 and sludge disposal costs ($Cost_{sludge}$) through eq. S12.

$$\Delta M_{TOT,bw} = 2.1 \cdot Q \cdot 1 \text{ h} \cdot 192 \frac{\text{mg}_{COD}}{\text{L}} \quad (S10)$$

$$t_{cycle} = \frac{\Delta M_{TOT,bw}}{M_{TOT,r}} \cdot SRT \cdot 24 \text{ h/d} \quad (S11)$$

$$Cost_{sludge} [\text{€/yr}] = \Delta M_{TOT,bw} \cdot 10^{-6} \text{ kg}_{COD}/\text{mg}_{COD} \cdot \frac{24 \text{ h/d} \cdot 365 \text{ d/yr}}{t_{cycle}} \cdot 0.15 \frac{\text{€}}{\text{kg}} \quad (S12)$$

126 The energy absorbed during a backwashing cycle has been then estimated considering the backwashing
 127 procedure normally recommended by biofilter's supplier (Degrémont S. A, 2010) and installed power for
 128 a pilot plant treating 0.6 m³/h as 0.122 kWh per backwashing cycle and has been used for $Cost_E$ calculation
 129 through equation eq. S13.

$$Cost_E [\text{€/yr}] = \frac{365 \frac{\text{d}}{\text{yr}} \cdot 24 \frac{\text{h}}{\text{d}}}{t_{cycle}} \cdot 0.122 \text{ kWh} \cdot 0.15 \frac{\text{€}}{\text{kWh}} \quad (S13)$$

130 Investments and management cost contributions were grouped in a single cost function, considering an
 131 amortisation time of 10 years, through equations eq. S14.

$$OBJ_2 = Costs[\text{€}] = C_{invest} + 10\text{yr} \cdot (Cost_{reag} + Cost_E + Cost_{sludge}) \quad (S14)$$

132 As for the constraint functions of the model, the first constraint was set in order to limit biomass accumulation
 133 in the system at reasonable values considering the attachment capacity of the filter media. The maximum
 134 amount of particulate matter that can accumulate in the biofilter is equal to the volume of voids. Considering
 135 a biofilter filled with BioliteTM with average diameter 3.5 mm and density 1.6 kg/m³ (Degrémont S. A, 2010),
 136 a porosity (e) of 0.36 can be assumed [Aesoy et al., 1998]. Considering a particulate matter density (ρ_x) of
 137 25 kg_{COD}/m³ (Henze et al., 2008), the maximum particulate matter concentration has been calculated through
 138 equation eq. S15. This has led to the definition of the first constraint as expressed by eq. S17, which limits
 139 total particulate matter concentration under the value corresponding to 75% of reactor voids filled by
 140 particulate matter.

$$\frac{V_L \cdot e \cdot \rho_x}{V_L} = e \cdot \rho_x = 0.36 \cdot 25 \frac{\text{kg}_{COD}}{\text{m}^3} \cdot 10^3 = 9000 \frac{\text{mg}_{COD}}{\text{L}} \quad (S15)$$

$$CONSTR_1: (\overline{X_{TOT}}) \leq 0.75 \cdot 9000 \frac{mg_{COD}}{L} \quad (S16)$$

$$CONSTR_1: (\overline{X_{TOT}}) \leq 6750 \frac{mg_{COD}}{L} \quad (S17)$$

141 Then, the second constraint has been set to comply with the regulation limit for nitrate concentration in water
 142 treated for drinking purposes that is set at 50 mgNO₃/L, corresponding to 11.3 mg_{NO₃-N}/L. In particular, it limits
 143 nitric nitrogen concentration in the biofilter effluent under the value corresponding to 75% of the regulation
 144 limit:

$$CONSTR_2: S_{NO3} \leq 0.75 \cdot 11.3 \frac{mg_N}{L} \quad (S18)$$

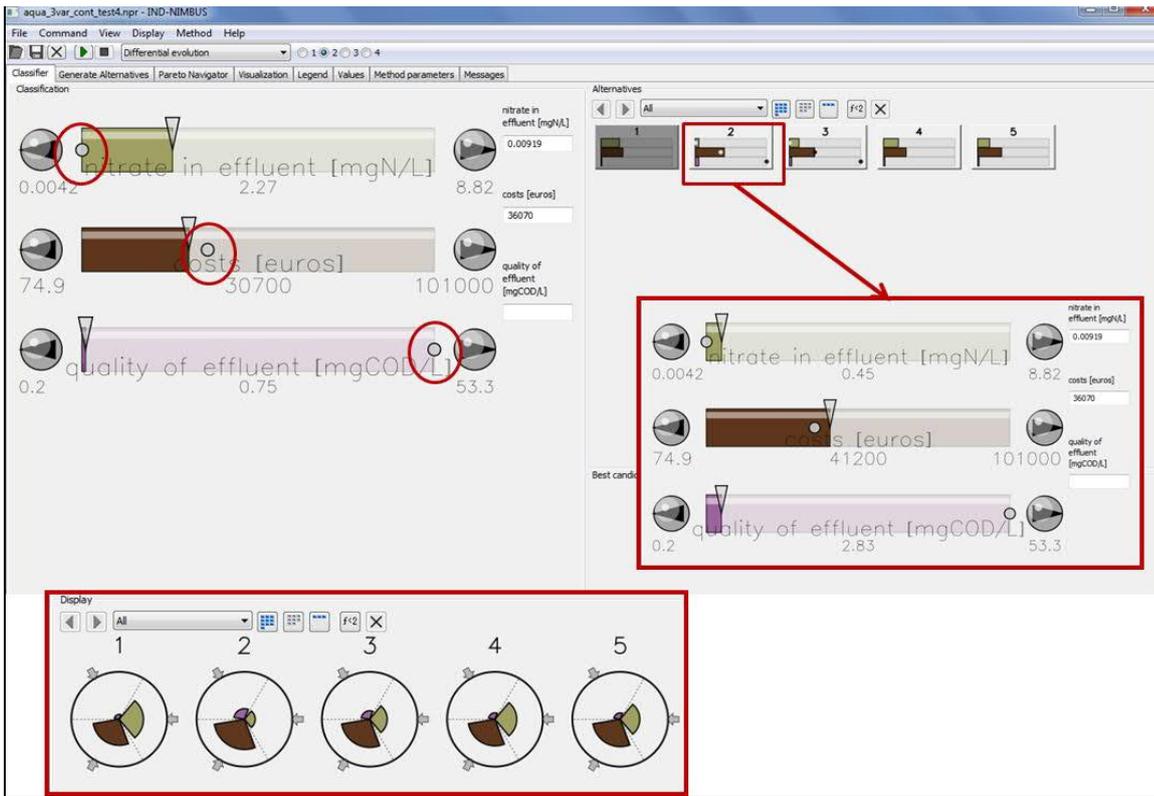
$$CONSTR_2: S_{NO3} \leq 8.5 \frac{mg_N}{L} \quad (S19)$$

145

146 Text S5. User interface of IND-NIMBUS

147 Interactive multiobjective optimization has been performed with IND-NIMBUS (Miettinen, 2006; Ojalehto et
 148 al., 2014), a software for nonlinear multiobjective optimization. An example screenshot of the graphical user-
 149 interface of IND-NIMBUS is reported in Figure S3

150



151

152 **Figure S3** Screenshot of the graphical user interface of the IND-NIMBUS software

153

154 Text S6. Simulation models considered in the sensitivity analysis

155 Biological and physical processes considered by the models proposed by Kornaros and Lyberatos (Kornaros
 156 and Lyberatos, 1998) and Magrì and Flotats (Magrì and Flotats, 2008) have been presented in Table S1, while

157 specific stoichiometric coefficients, process rates and parameters are presented in Table S4, Table S6, Table
 158 S5 and Table S7 for Kornaros and Lyberatos (Kornaros and Lyberatos, 1998) model and for the Magrì and
 159 Flotats (Magrì and Flotats, 2008) model.

160

161 **Table S1** Process rates in $\text{mg}_{\text{COD}} \cdot \text{L}^{-1} \cdot \text{d}^{-1}$ as described in the model by Kornaros and Lyberatos (Kornaros and
 162 Lyberatos, 1998).

Name	Process	Expression
r ₁	Aerobic Growth	$\hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_O}{S_O + k_{O,H}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₂	Anoxic growth on nitrate	$\eta_{g1} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO3}}{S_{NO3} + k_{NO3}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₃	Anoxic growth on nitrite	$\eta_{g2} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO2}}{S_{NO2} + k_{NO2}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot \left(\frac{k_{I,NO3}}{k_{I,NO3} + S_{NO3}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₄	Biomass decay	$b_H \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₅	Hydrolysis of entrapped organics	$k_h \cdot \frac{X_S}{K_X \cdot X_{BH} + X_S} \cdot \left[\left(\frac{S_O}{S_O + k_{O,H}} \right) + \eta_h \cdot \left(\frac{k_{O,H}}{S_O + k_{O,H}} \right) \cdot \left(\frac{S_{NO3} + S_{NO2}}{S_{NO3} + S_{NO2} + k_{NO}} \right) \right] \cdot X_{BH}$

163

164 **Table S2** Process rates in $\text{mg}_{\text{COD}} \cdot \text{L}^{-1} \cdot \text{d}^{-1}$ as described I the model by Magrì and Flotats (Magrì and Flotats,
 165 2008).

Name	Process	Expression
r ₁	Aerobic Growth	$\hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_O}{S_O + k_{O,H}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₂	Anoxic growth on nitrate	$\eta_{g1} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO3}}{S_{NO3} + k_{NO3}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₃	Anoxic growth on nitrite	$\eta_{g2} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO2}}{S_{NO2} + k_{NO2}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₄	Biomass decay	$b_H \cdot X_{BH} \cdot \vartheta^{T-20}$
r ₅	Hydrolysis of entrapped organics	$k_h \cdot \frac{X_S}{K_X \cdot X_{BH} + X_S} \cdot \left[\left(\frac{S_O}{S_O + k_{O,H}} \right) + \eta_h \cdot \left(\frac{k_{O,H}}{S_O + k_{O,H}} \right) \cdot \left(\frac{S_{NO3} + S_{NO2}}{S_{NO3} + S_{NO2} + k_{NO}} \right) \right] \cdot X_{BH}$

166

167 **Table S3** Model parameters and assumed values for the model by Kornaros and Lyberatos (Kornaros and
 168 Lyberatos, 1998).

Parameter	Value	Unit	Meaning
$\hat{\mu}_H$	6	1/d	Maximum specific growth rate for heterotrophic biomass

b_H	0.62	1/d	Decay rate for heterotrophic biomass
k_h	3.0	1/d	Maximum specific hydrolysis rate
θ	1.03	-	Correction factor for temperature effect on biomass growth and decay rates
η_{g1}	0.345	-	Correction factor for μ _h for anoxic growth on nitrate-nitrogen
η_{g2}	0.411	-	Correction factor for μ _h for anoxic growth on nitrite-nitrogen
η_h	0.4	-	Correction factor for hydrolysis under anoxic conditions
k_s	20	mg _{COD} /L	Half-saturation constant for organic substrate
k_{O,H}	0.2	mg _{COD} /L	Half-saturation constant for oxygen
k_{NO3}	0.77	mg _N /L	Half-saturation constant for nitrate nitrogen
k_{NO2}	0.28	mg _N /L	Half-saturation constant for nitrite nitrogen
k_X	0.03	-	Half-saturation constant for slowly biodegradable substrate
k_{I,NO3}	8.75	mg _N /L	Nitrate inhibition constant
Y_h	0.46	mg _{COD} /mg _{COD}	Heterotrophic biomass yield, considering NO ₃ -N uptake for protein synthesis
f_p	0.08	-	Fraction of biomass leading to particulate products
i_{XB}	0.086	mg _N /mg _{COD}	Mass of nitrogen per mass of COD in biomass

169

170 **Table S4** Model parameters values assumed following the model presented by Magrì and Flotats (Magrì and
171 Flotats, 2008).

Parameter	Value	Unit	Meaning
$\hat{\mu}_H$	4.16	1/d	Maximum specific growth rate for heterotrophic biomass
b_H	0.17	1/d	Decay rate for heterotrophic biomass
k_h	3.0	1/d	Maximum specific hydrolysis rate
θ	1.03	-	Correction factor for temperature effect on biomass growth and decay rates
η_{g1}	0.23	-	Correction factor for μ _h for anoxic growth on nitrate-nitrogen
η_{g2}	0.62	-	Correction factor for μ _h for anoxic growth on nitrite-nitrogen
η_h	0.4	-	Correction factor for hydrolysis under anoxic conditions
k_s	4	mg _{COD} /L	Half-saturation constant for organic substrate
k_{O,H}	0.1	mg _{COD} /L	Half-saturation constant for oxygen
k_{NO3}	0.5	mg _N /L	Half-saturation constant for nitrate nitrogen
k_{NO2}	0.12	mg _N /L	Half-saturation constant for nitrite nitrogen
k_X	0.03	-	Half-saturation constant for slowly biodegradable substrate

Y_h	0.65	$\text{mg}_{\text{COD}}/\text{mg}_C$ OD	Heterotrophic biomass yield, considering $\text{NO}_3\text{-N}$ uptake for protein synthesis
f_p	0.08	-	Fraction of biomass leading to particulate products
i_{XB}	0.086	$\text{mg}_N/\text{mg}_{\text{COD}}$	Mass of nitrogen per mass of COD in biomass

172

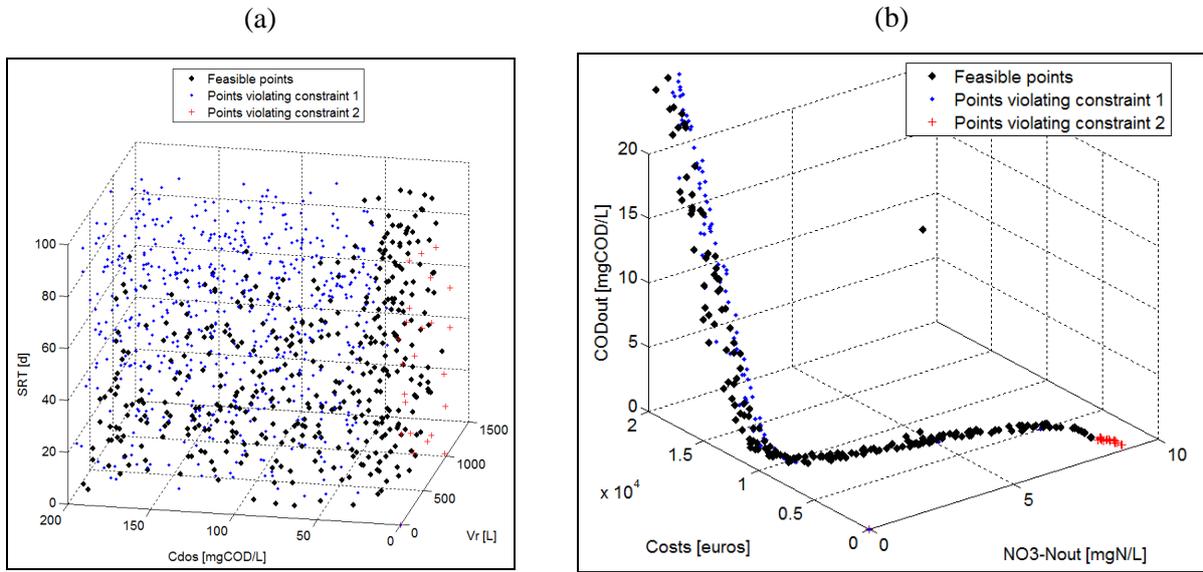
173 **Text S7. Investigation of the feasible region**

174 For the analysis of the feasible region (Section 2.4), 3000 random points were generated and corresponding
 175 constraint and objective function values were evaluated. The performed simulations showed that 41% of the
 176 points generated were feasible with respect to the constraints defined in Section 2.3. Constraint 1, on the total
 177 particulate matter concentration, turned out to be the main reason of infeasibility, as it was violated by 96% of
 178 the infeasible points. By plotting the generated random points in the design space (Figure S4a), it is possible
 179 to notice that points violating the first constraint are spread in the whole design space, and:

- 180 - in 61% of cases they are related to reactor volumes up to 600 L (the lower half of DV_1 range), while only
- 181 39% of points belong to the upper half of the DV_1 range (600 - 1200 L);
- 182 - in 58% of cases they correspond to a carbon dosage between 2 and 4 times the stoichiometric dosage (with
- 183 only 42% occurring in case of C_{dos} lower than 2 times the stoichiometric dosage);
- 184 - in 69% of cases they are related to SRT higher than 50 d (upper half of DV_3 range) with only 31% occurring
- 185 in case of SRT lower than 50 d.

186 These observations highlight the fact that, in a biodenitrification reactor, high values of total particulate matter
 187 concentration can be reached through different operating conditions, mainly related to low values of reactor
 188 volume, or high values of dosed carbon (non-limiting carbon substrate conditions) and high values of SRT
 189 (good biomass retention in the system). Considering the second constraint (on nitric nitrogen concentration),
 190 it was violated on the average by 2.5% of points. When looking at Figure S4a, one can notice that these points
 191 correspond to the lowest carbon dosages (C_{dos} lower than 5 $\text{mg}_{\text{COD}}/\text{L}$), meaning a carbon supply under 9.6% of
 192 the stoichiometric needs (calculated to be 52 $\text{mg}_{\text{COD}}/\text{L}$ considering data reported in Section 2.1). These results
 193 highlight that carbon dosages under the stoichiometric requirements can be acceptable as far as the partial
 194 removal efficiency assures an acceptable output concentration and as far as endogenous carbon can sustain
 195 metabolic reactions and thus biological degradation. Below a certain level, represented by the 9.6% of
 196 stoichiometric dosage, the endogenous carbon is not enough and $\text{NO}_{3,\text{out}}$ concentration is no more acceptable.
 197 Figure S4b reports the generated random points in the objective space, for values of COD in the effluent lower
 198 than 20 $\text{mg}_{\text{COD}}/\text{L}$, thus, zoomed on the more informative portion of the space. However, points related to values
 199 of COD_{out} higher than 20 $\text{mg}_{\text{COD}}/\text{L}$ have the same trend. The lowest nitrate removal efficiencies correspond to
 200 infeasible points because of the violation of constraint 2, indicating a carbon dosage not sufficient for the
 201 respect of 75% of the regulation limit on nitrate concentration. Increasing nitrate removal efficiencies
 202 corresponds to increasing costs and COD concentrations in the treated water, both following an exponential
 203 increase as the nitrate removal efficiency exceeds 90%. In this region, points violating constraint 1 correspond
 204 to lower costs. In case of partial removal of nitrate, both costs and COD concentrations in the effluent present

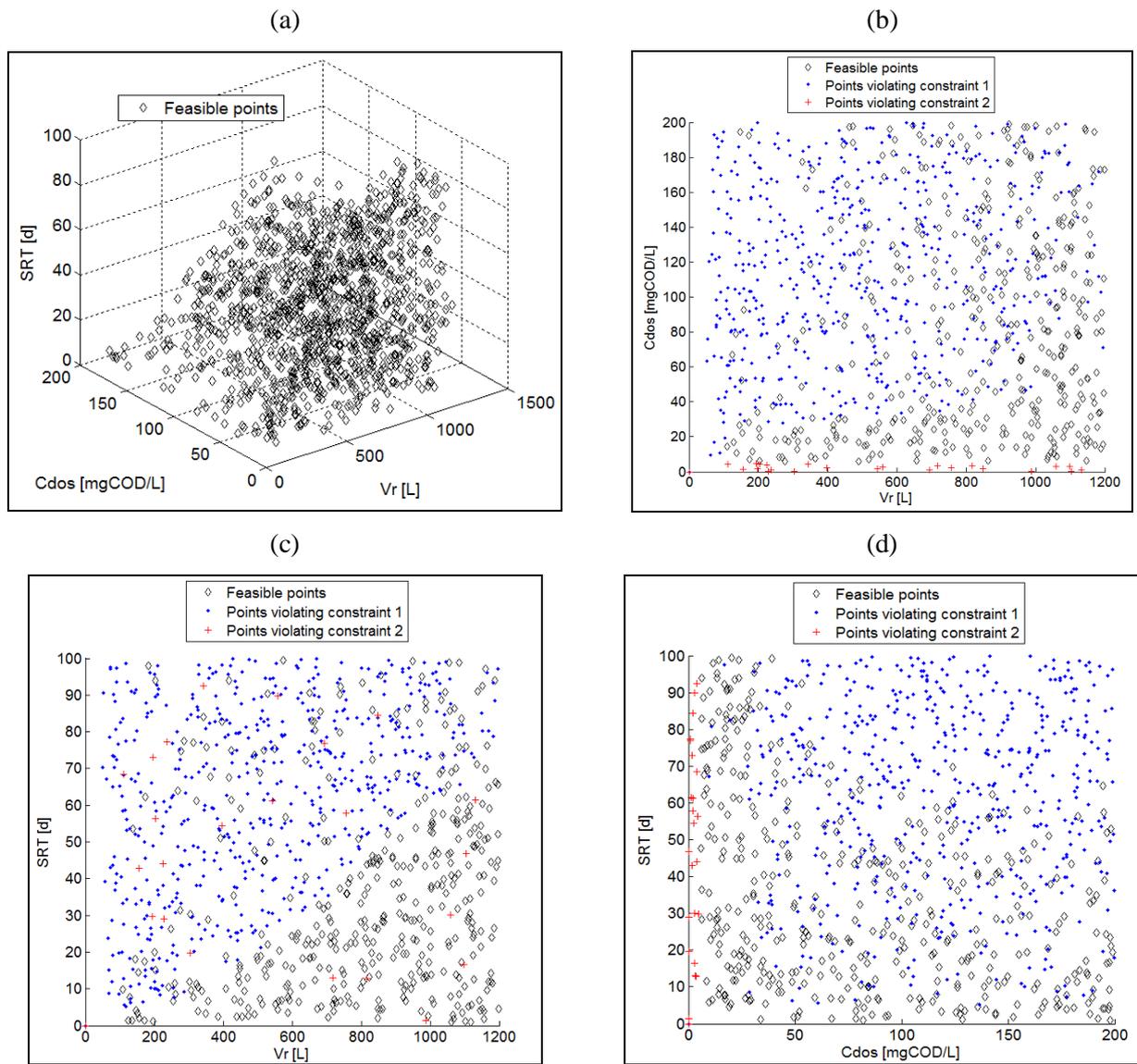
205 low values and could represent acceptable trade-offs, even if COD concentrations in the effluent should be
 206 mainly due to nitrite leaks, involving the systematic application of specific oxidation post-treatments. In order
 207 to limit the post-treatments to be applied, a Pareto optimal design should be found in the portion of feasible
 208 region characterized by an almost complete nitrate removal but further analysis needs to be performed and
 209 optimization is needed for it.
 210



211 **Figure S4.** Feasible points (a) in the design space, and (b) their counterparts in the objective space (portion
 212 $COD_{out} \leq 20 \text{ mgCOD/L}$).

213
 214 More in details, Figure S5a represents generated feasible points in the design space with Figure S5b-d
 215 projecting them in 2D design space graphs. Figure S6a reports their representation in the objective space.
 216 Conflicts among objectives can be seen more clearly by plotting projections in 2D objective space graphs, as
 217 presented in Figure S6b-d. In particular, graphs in Figure S6b and Figure S6c show that points with lower
 218 values of objective 2 (Costs) and objective 3 (COD_{out}) involve the highest values of objective 1 (NO_3-N_{out}).
 219 On the contrary, Figure S6d shows that points with lower values of objective 2 (Costs) are also characterized
 220 by lower values of objective 3 (COD_{out}). From these observations, it can be concluded that the second and the
 221 third objective are not in conflict within each other, being on the other hand, both in conflict with the first
 222 objective.

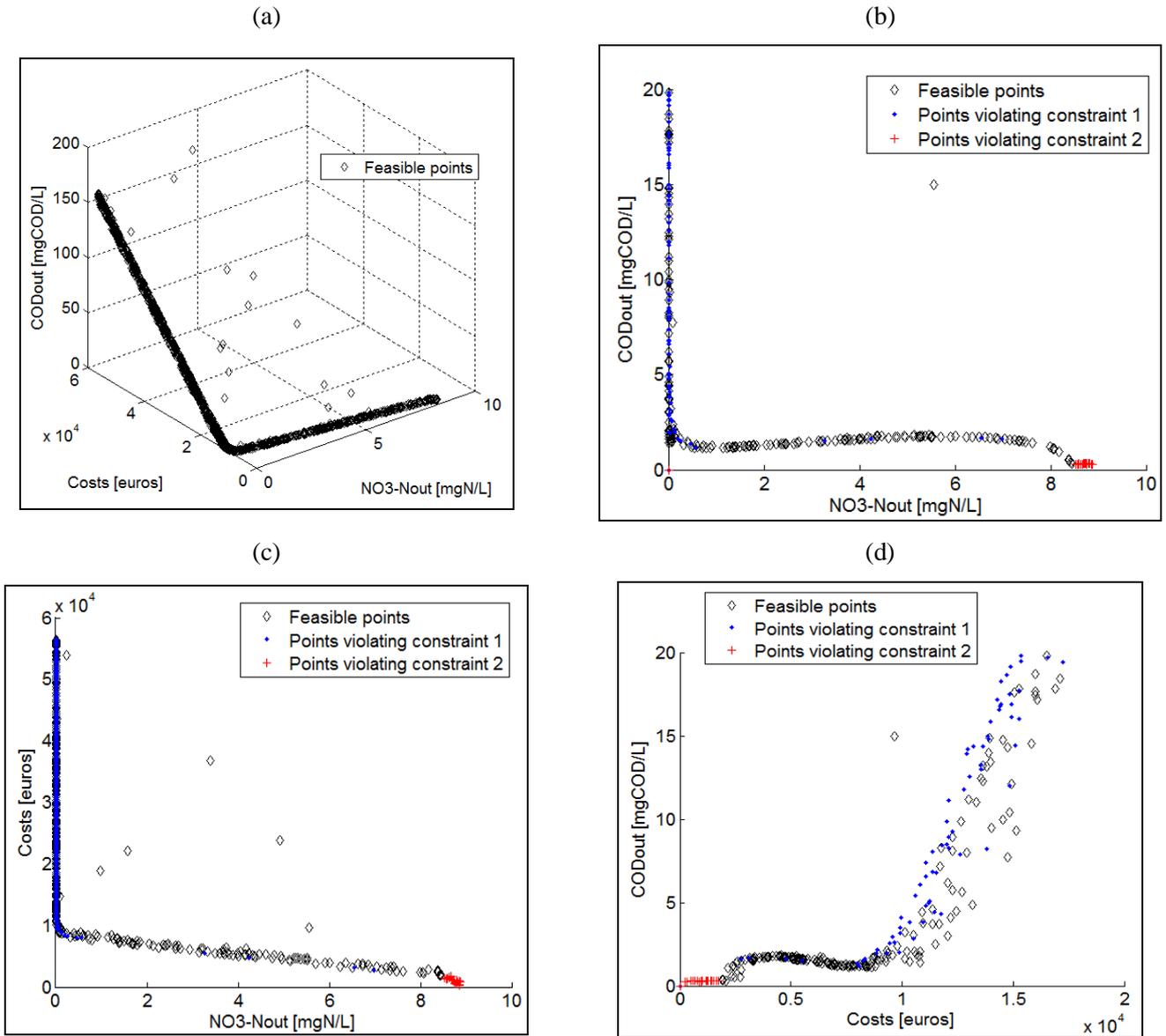
223
 224



225 **Figure S5.** Representation of the results of the analysis of the feasible region in the design space: (a) Feasible
 226 points in the 3D plot; (b) Projection of feasible points in the 2D space (b) DV₂ as ordinate and DV₁ as abscissa;
 227 (c) DV₃ as ordinate and DV₁ as abscissa; (d) DV₃ as ordinate and DV₂ as abscissa.

228

229



230 **Figure S6.** Projection of feasible points in the objectives space: (a) Feasible points in the 3D plot. Projection of
 231 feasible points in the 2D space (b) OBJ_2 as ordinate and OBJ_1 as abscissa; (c) OBJ_3 as ordinate and OBJ_1 as
 232 abscissa; (d) OBJ_3 as ordinate and OBJ_2 as abscissa.

233

234 **Text S8. Sensitivity analysis**

235 A sensitivity analysis has been performed varying few case-study specific parameters and the simulation model
 236 as presented in Section 2.6. The variation of feasible points and constraint violations, with respect to the case
 237 study results, is presented as bar charts in Figure S7.

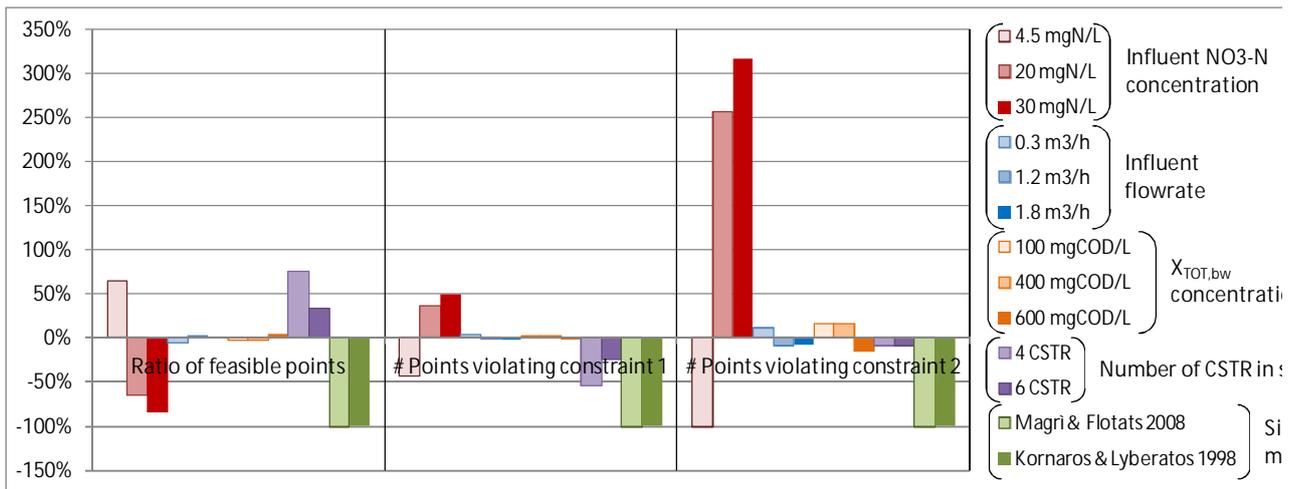
238 As far as NGPM results are concerned, non-dominated solutions are represented in the design and objectives
 239 spaces with case study results: Figure S2 reports results obtained when varying NO_3-N_{in} concentration and
 240 influent flow rate, while Figure S3 reports results obtained when varying the concentration of biomass in
 241 backwashing water, the number of CSTRs in series considered in the biofilter hydraulic model and the
 242 simulation model. The distribution of obtained non-dominated solutions are compared in the box plot reported

243 in Figure S4, while the variation of mean and standard deviation with respect to the case study results presented
 244 in the manuscript, is presented in Figure S5.

245 As far as IND-NIMBUS results are concerned, the sensitivity analysis has been conducted performing five
 246 optimization steps that replicates considerations that guided the DM's choices in the case study:

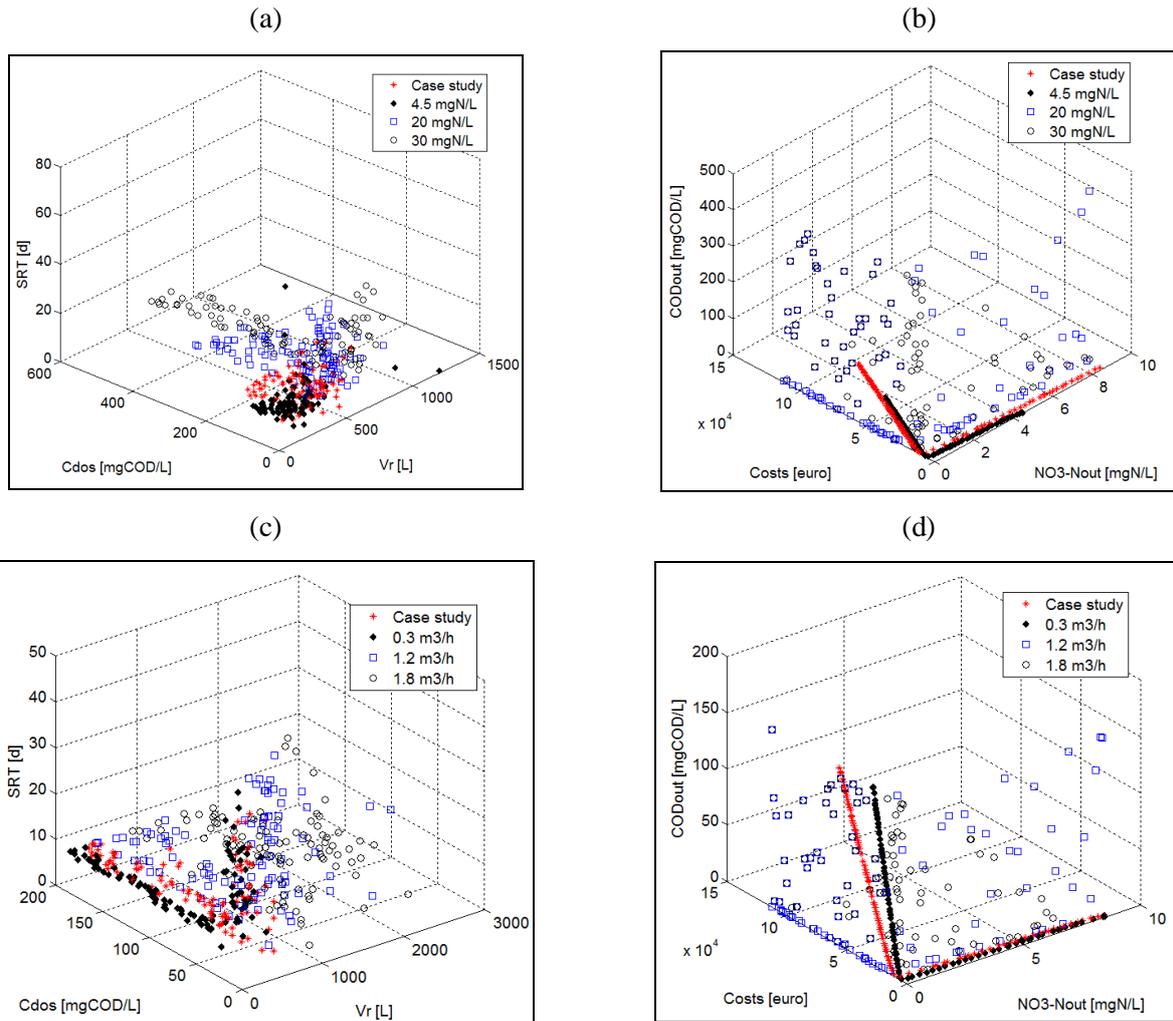
- 247 1. problem initialization;
- 248 2. optimization looking for a similar value of OBJ₁, the best value for OBJ₃ and letting OBJ₂ change
 249 freely;
- 250 3. from the solution with lower OBJ₁ value, optimization looking for the best OBJ₁ value, letting both
 251 OBJ₂ and OBJ₃ change freely;
- 252 4. optimization to an acceptable level of OBJ₂ and OBJ₃ letting OBJ₁ change freely;
- 253 5. optimization to an acceptable level of OBJ₁, OBJ₂ and OBJ₃;
- 254 6. identification of the most preferred design among proposed Pareto optimal solutions (i.e. designs)

255 Pareto optimal solutions obtained through the sensitivity analysis are reported in Figure S6 and Figure S7.
 256 Then, Figure S1.8 reports the variation of most preferred designs compared to the case study one, detailing the
 257 evaluation for each design variable and objective function.

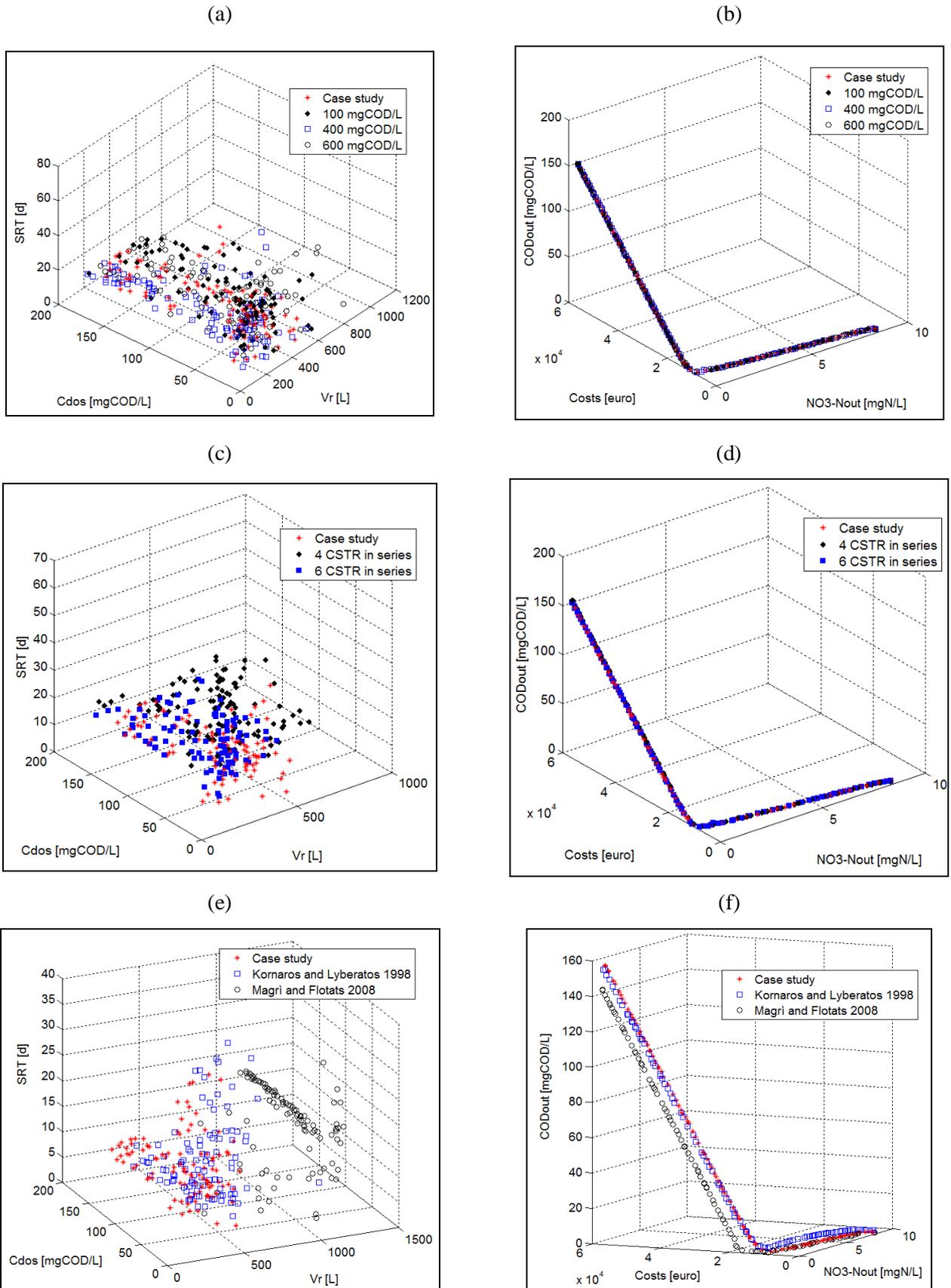


258 **Figure S1.** Sensitivity analysis on feasible region results: variations observed on feasible point ratio and on
 259 the number of points violating each constraint.

260
 261



262 **Figure S2.** NGPM results obtained varying nitric nitrogen concentration in raw water and influent flowrate
 263 represented with case study results respectively in the design (a, c) and the objectives spaces (b, d).
 264
 265



266 **Figure S3.** NGPM results obtained varying the concentration of biomass in backwashing water, the number
 267 of CSTRs in series considered in the biofilter hydraulic model and the simulation model represented with case
 268 study results respectively in the design (a, c, e) and the objectives spaces (b, d, f).

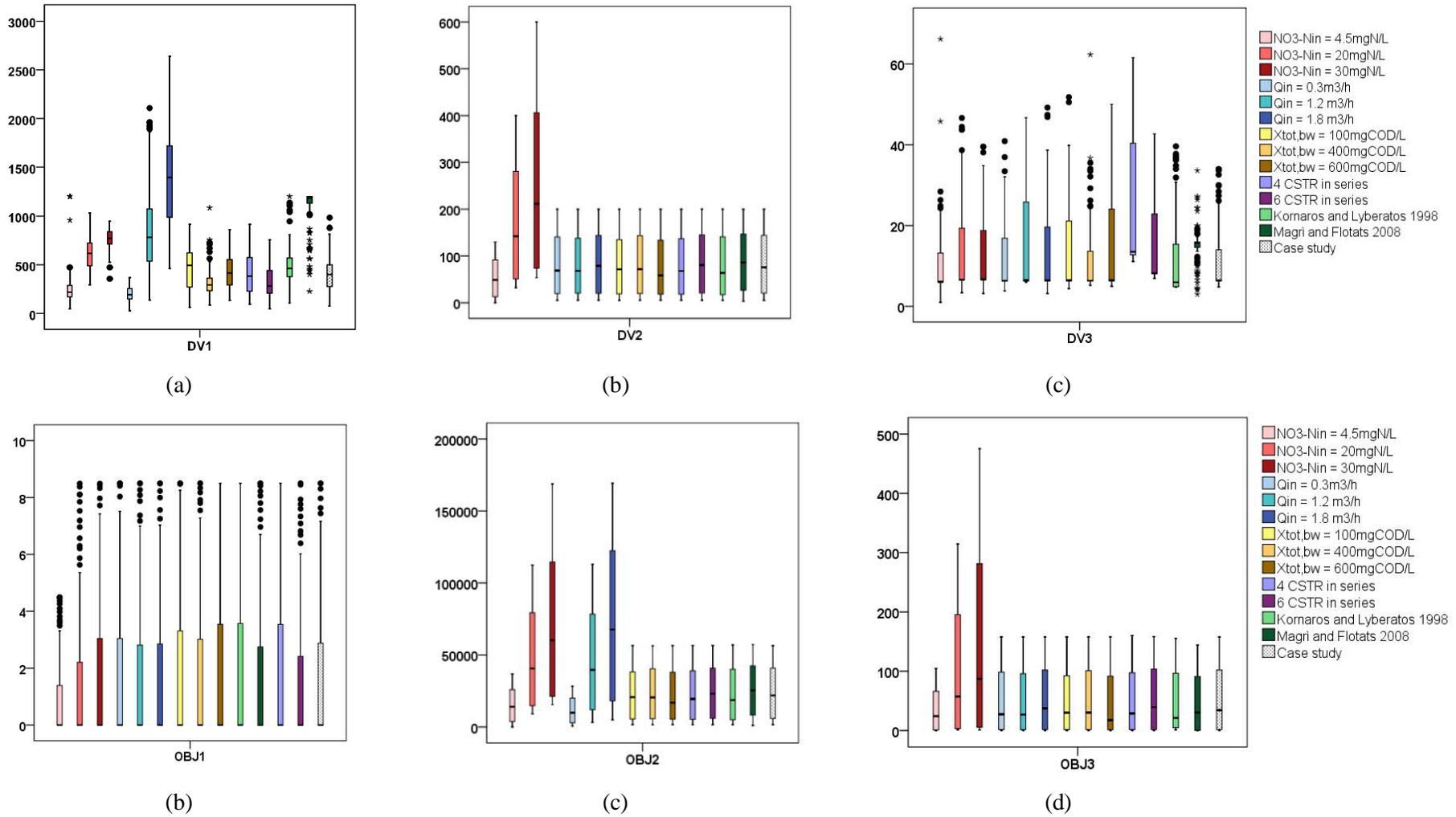
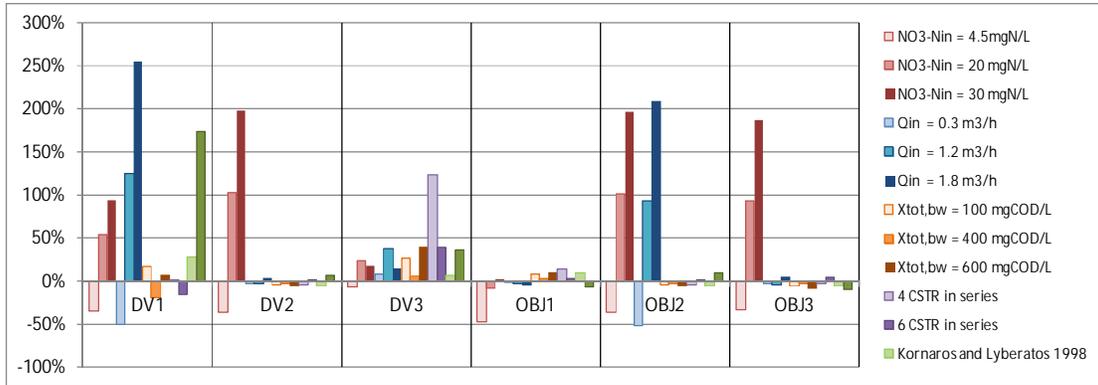
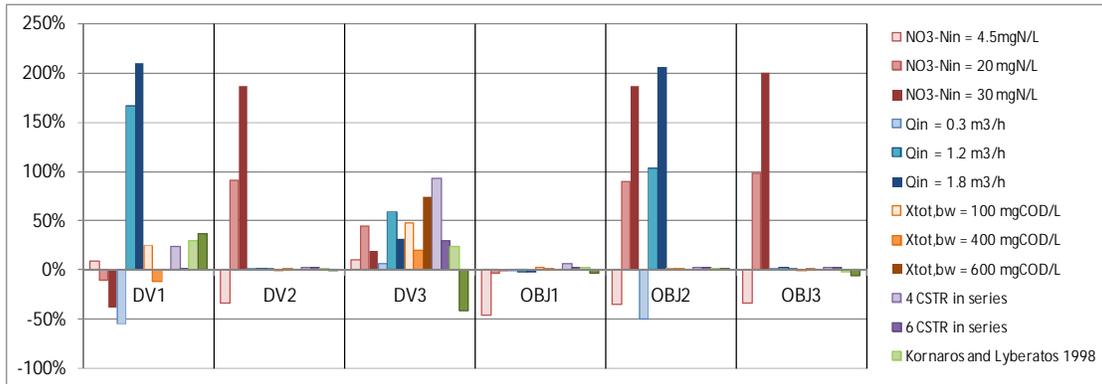


Figure S4. Distributions of NGPM results for different values of selected case specific parameters and for different simulation models, compared to case study results. Values for (a) design variable 1, (b) design variable 2, (c) design variable 3, (d) objective function 1, (e) objective function 2, (f) objective function 3.



(a)



(b)

Figure S5. Variation of mean (a) and standard deviation (b) values for design variables and objective functions of NGPM results obtained through sensitivity analysis.

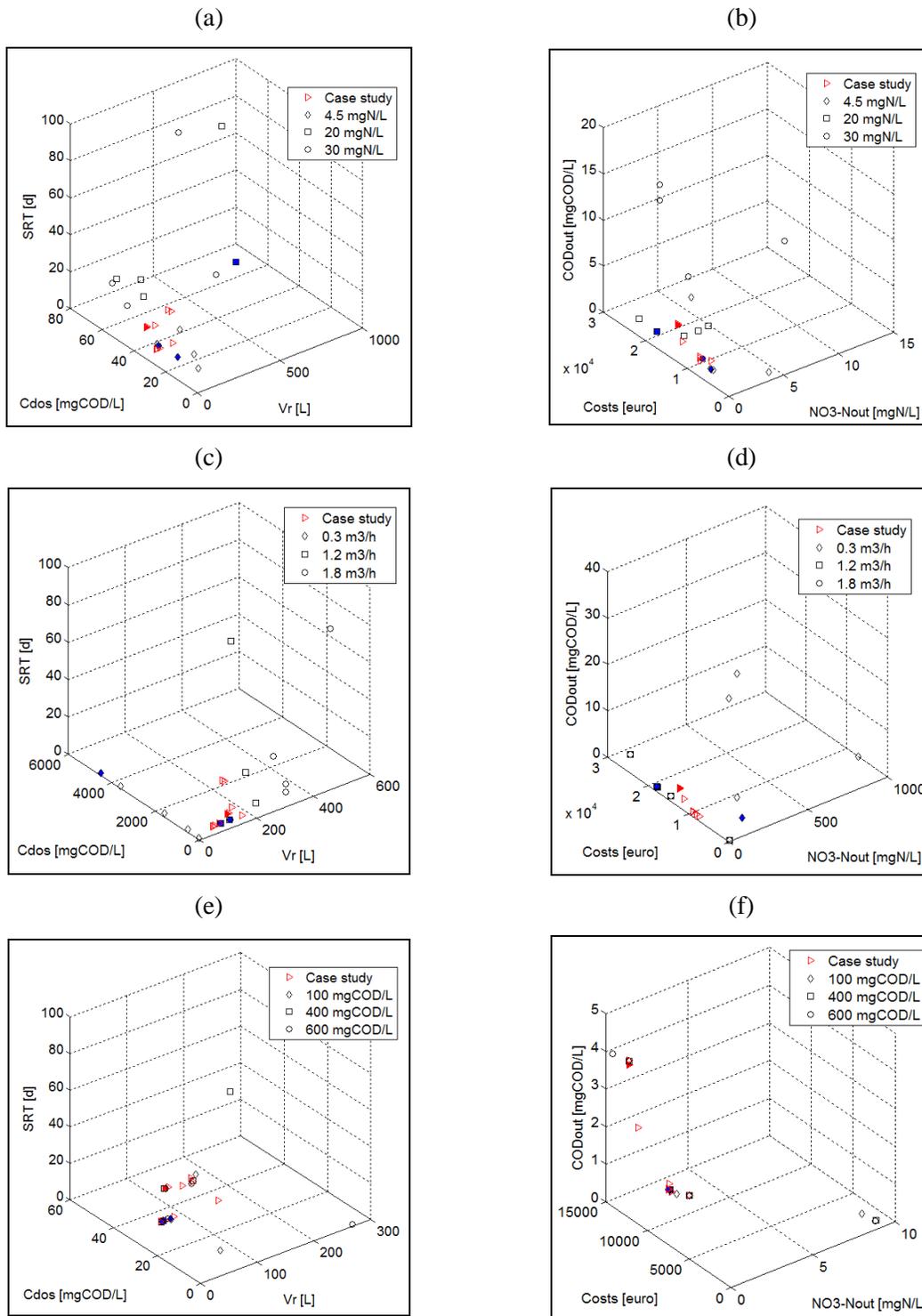


Figure S6. Pareto optimal solutions generated by IND-NIMBUS by varying nitric nitrogen concentration in raw water (a,b), influent flow rate (c,d) and concentration of biomass in backwashing water (e,f). Solutions represented with case study results in the design (a, c, e) and the objectives spaces (b, d, f). Most preferred solutions have been represented as filled points.

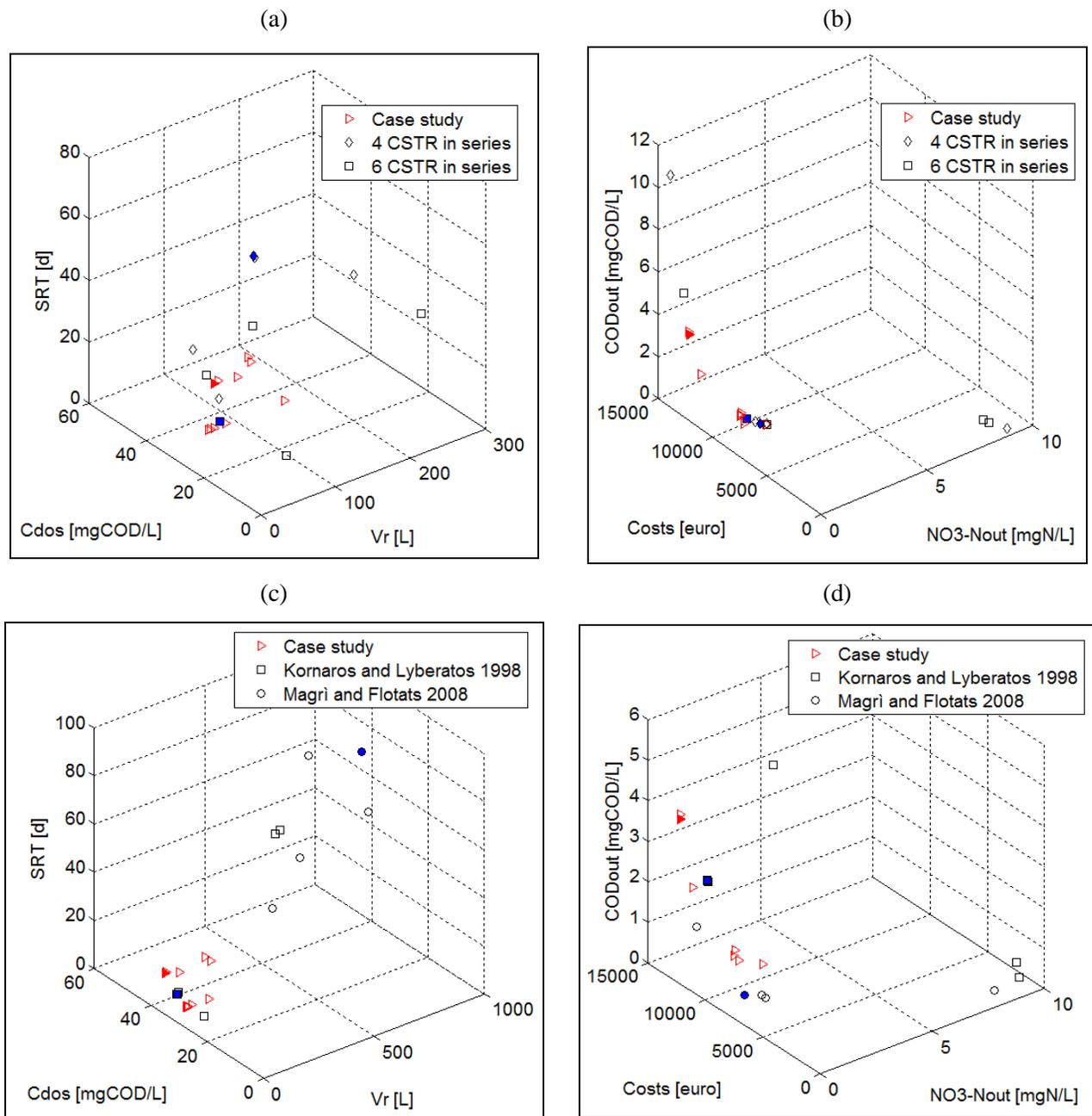


Figure S7. Pareto optimal solutions generated by IND-NIMBUS by varying the number of CSTRs considered in the biofilter hydraulic model (a,b) and the simulation model (c,d): solutions represented with case study results in the design (a, c) and objectives spaces (b, d). Most preferred solutions have been represented as filled points.

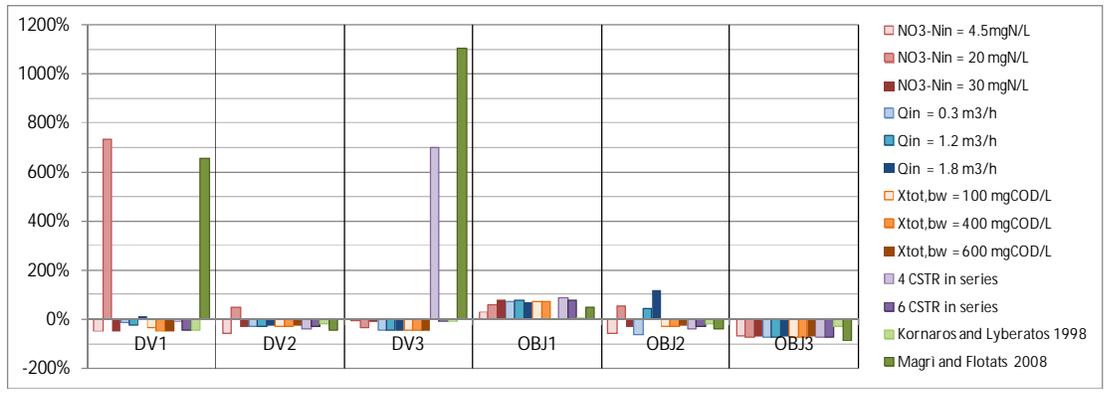


Figure SI.8. Variation of design variables and objective functions values of most preferred design found through IND-NIMBUS sensitivity analysis.

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