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Potential of Interactive Multiobjective Optimization in Supporting the Design of a Groundwater Biotenitrification Process

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SUPPLEMENTARY MATERIAL

Text S1. Conceptual representation of the design optimization problem

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

Figure S1. Flowchart representing the design optimization problem constituting units

Text S2. Hydraulic characterization of the biofilter

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.

Text S3. Biofilter simulation model

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

Text S4. Optimization problem formulation

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

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

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

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

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.

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

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.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Text S5. User interface of IND-NIMBUS

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

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

Text S6. Simulation models considered in the sensitivity analysis

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

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

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 Lyberatos, 1998).

Name	Process	Expression
r₁	Aerobic Growth	$\hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_O}{S_O + k_{O,H}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₂	Anoxic growth on nitrate	$\eta_{g1} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO3}}{S_{NO3} + k_{NO3}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₃	Anoxic growth on nitrite	$\eta_{g2} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO2}}{S_{NO2} + k_{NO2}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot \left(\frac{k_{I,NO3}}{k_{I,NO3} + S_{NO3}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₄	Biomass decay	$b_H \cdot X_{BH} \cdot \vartheta^{T-20}$
r₅	Hydrolysis of entrapped organics	$k_h \cdot \frac{X_S}{K_X \cdot X_{BH} + X_S} \cdot \left[\left(\frac{S_O}{S_O + k_{O,H}} \right) + \eta_h \cdot \left(\frac{k_{O,H}}{S_O + k_{O,H}} \right) \cdot \left(\frac{S_{NO3} + S_{NO2}}{S_{NO3} + S_{NO2} + k_{NO}} \right) \right] \cdot X_{BH}$

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, 2008).

Name	Process	Expression
r₁	Aerobic Growth	$\hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_O}{S_O + k_{O,H}} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₂	Anoxic growth on nitrate	$\eta_{g1} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO3}}{S_{NO3} + k_{NO3}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₃	Anoxic growth on nitrite	$\eta_{g2} \cdot \hat{\mu}_H \cdot \left(\frac{S_S}{S_S + k_S} \right) \cdot \left(\frac{S_{NO2}}{S_{NO2} + k_{NO2}} \right) \cdot \left(\frac{k_O}{k_O + S_O} \right) \cdot X_{BH} \cdot \vartheta^{T-20}$
r₄	Biomass decay	$b_H \cdot X_{BH} \cdot \vartheta^{T-20}$
r₅	Hydrolysis of entrapped organics	$k_h \cdot \frac{X_S}{K_X \cdot X_{BH} + X_S} \cdot \left[\left(\frac{S_O}{S_O + k_{O,H}} \right) + \eta_h \cdot \left(\frac{k_{O,H}}{S_O + k_{O,H}} \right) \cdot \left(\frac{S_{NO3} + S_{NO2}}{S_{NO3} + S_{NO2} + k_{NO}} \right) \right] \cdot X_{BH}$

Table S3 Model parameters and assumed values for the model by Kornaros and Lyberatos (Kornaros and 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 _C OD	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

Text S7. Investigation of the feasible region

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

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

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

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

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

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

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

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.

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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:

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1. problem initialization;
 2. optimization looking for a similar value of OBJ_1 , the best value for OBJ_3 and letting OBJ_2 change freely;
 3. from the solution with lower OBJ_1 value, optimization looking for the best OBJ_1 value, letting both OBJ_2 and OBJ_3 change freely;
 4. optimization to an acceptable level of OBJ_2 and OBJ_3 letting OBJ_1 change freely;
 5. optimization to an acceptable level of OBJ_1 , OBJ_2 and OBJ_3 ;
 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 SI.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.

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

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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