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

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Abstract:

The design of water treatment plants requires simultaneous analysis of technical, economic and environmental aspects, identified by multiple conflicting objectives. We demonstrated the advantages of an interactive multiobjective optimization (MOO) method over a posteriori methods in an unexplored field, namely the design of a biological treatment plant for drinking water production, that tackles the process drawbacks, contrarily to what happens in a traditional volumetric-load-driven design procedure. Specifically, we consider a groundwater denitrification biofilter, simulated by the Activated Sludge Model modified with two-stage denitrification kinetics. Three objectives were defined (nitrate removal efficiency, drawbacks on produced water, investment and management costs) and the interactive method NIMBUS applied to identify the best-suited design without any a priori evaluation, as for volumetric-load-driven design procedures. When compared to an evolutionary MOO algorithm, the interactive solution process was faster, more understandable and user-friendly and supported the decision maker well in identifying the most preferred solution (main design/operating parameters) to be implemented. Approach strength has been proved through both sensitivity analysis and positive experimental validation through a pilot scale biofilter operated for three months. In synthesis, without any "a priori" evaluation based on practical experience, the MOO design approach allowed obtaining a preferred Pareto optimal design, characterized by volumetric loading in the range 0.85-2.54 $kg_N\ m^{-3}\ d^{-1}$ (EBCTs: 5-15 min), a carbon dosage of 0.5-0.8 $g_{C,dos}/g_{C,stoich}$, with SRTs in the range 4-27 d.

Keywords: Water treatment; Interactive method; NIMBUS method; IND-NIMBUS; Decision support; Pareto optimality.

1. Introduction

Designing a water treatment plant involves selecting the best process and defining the optimal configuration to guarantee the required removal efficiency at the lowest global cost, standing for economic and environmental costs at the same time. An effective plant design should manage multiple aspects, such as water quality, available treatment technologies, operational constraints, construction challenges, regulatory requirements, consumer/environmental concerns and economic feasibility

(Crittenden et al., 2012). Even when considering only the last aspect, one must balance between treatment efficacy, investment and operational costs for treatment unit realization and running, and both investment and operational costs for additional treatment units that could be required as a consequence of specific drawbacks of the selected process (such as water pH variation, biomass leakage, biological or chemical reactions by-products in the treated effluent) (AWWA and ASCE, 2005). Experience-based design approaches, such as volumetric-load-driven procedures for biological treatment processes, do not optimize design cases from both investment and management points of view, as they are based on average behavior considerations (Rivas et al., 2008).

Thus, designing a water treatment plant represents a complex decision problem involving multiple conflicting criteria (Crittenden et al., 2012). Indeed, decision support tools and methods (Hakanen et al., 2013; Hamouda et al., 2009; Hartikainen et al., 2015; Maier et al., 2014; Nicklow et al., 2010; Poch et al., 2004; Reed et al., 2013) are needed to support the choice of the best design highlighting the trade-offs involved. Despite the broad literature on applying decision support tools for water management problems (Brown et al., 2015), tools to support treatment unit design are limited and most references focus on water resource systems and wastewater treatment applications. However, drinking water treatment design has specificities that do not allow the direct application of results from the wastewater field. For instance, adopting a biological process for drinking water treatment requires the careful evaluation of typical process drawbacks that are usually neglected in the wastewater field, such as by-products of (incomplete) biological reactions or biomass leakage in treated water.

An example of applying a biological process for drinking water production is contamination of the water supply source by nitrate. Pollution by nitrate endangers public health (Dahab and Lee, 1988; Kapoor and Viraraghavan, 1997; Soares, 2000; Shrimali and Singh, 2001; Magram, 2010, Sharma and Sobti, 2012) and regulation limits have been imposed by US Environmental Protection Agency (US EPA) and the European Commission: in Italy, the regulatory limit is set at 50 mg/L for nitrate and 0.5 mg/L for nitrite in drinking water. Nitrate removal is attainable through different treatment processes, as catalysis, reverse osmosis, ion exchange, and heterotrophic denitrification (Kapoor and Viraraghavan, 1997; Soares, 2000; Shrimali and Singh, 2001). Among the full-scale applied alternatives, biodenitrification is the most cost-effective, as operational costs are minimized by the

absence of any brine to be disposed of or treated (Soares, 2000; Shrimali and Singh, 2001; Aslan, 2008). Biodenitrification exploits the bacteria ability to reduce nitrates to gaseous nitrogen in an anoxic environment, given the presence of biodegradable organic carbon which ensures anoxic conditions and acts as the electron donor (Rittman and McCarty, 2001). The main weakness of biodenitrification applied to drinking water production is the sensitivity to variations in feeding and operating conditions, which can affect the overall process performance, enhancing the accumulation of nitrite. Besides, two aspects have to be considered in the risk of bacterial regrowth in distribution networks, which are usually neglected in case of wastewater: the release of biomass and the presence of readily biodegradable organic carbon in the effluent from the biological reactor. These two represent further parameters to be considered in process evaluation.

Tools that support optimization of multiple conflicting objectives belong to the field of multiobjective optimization (MOO). MOO methods optimize mathematical models involving multiple conflicting objective functions and allow the identification of so-called Pareto optimal solutions, reflecting the trade-offs among the conflicting objectives. A solution is called Pareto optimal if none of the objective function values can be improved without impairing at least one of the other objectives. When considering two solutions P_1 and P_2, P_1 is said to be dominated by P_2 if P_2 is better than P_1 with respect to at least one objective and not worse than P_1 with respect to all other objectives. Thus, a Pareto optimal solution is not dominated by any other solution.

A MOO problem can be formulated as (Miettinen, 1999):

$$minimize\{z_1(\underline{x}), \dots, z_j(\underline{x}), \dots, z_m(\underline{x})\} \tag{1}$$

subject to

$$\underline{x} = (x_1, \dots, x_i, \dots, x_n) \in S, \tag{2}$$

where

$$S = \{\underline{x} \in \mathbb{R}^n | \ (lb_i \leq x_i \leq ub_i, \ i = 1, \dots, n) \ and \ (p_k(\underline{x}) = 0, \ k = 1, \dots, eq) \ and \ (q_h(\underline{x}) \leq 0, h = 1, \dots ieq)\}. \tag{3}$$

The real-valued objective functions $z_j: S \rightarrow \mathbb{R}$ are simultaneously optimized in the feasible region $S \subset \mathbb{R}^n$, which is a set in the design space defined by the lower (lb_i) and upper (ub_i) bounds for the *n* design

variables x_i as well as equality (p_k) and inequality (q_h) constraints. By mapping a design variable vector with the objective functions, objective vectors in the *m*-dimensional objective space are obtained. In what follows, we use the terms solution and design as synonyms. The whole set of Pareto optimal solutions is referred to as a Pareto front. Because the objective functions in MOO problems are typically conflicting, there exist several different Pareto optimal solutions and additional preference information from a human decision maker (DM) is needed to identify the most preferred solution.

Many MOO methods have been developed to solve complex design optimization problems and the choice depends on the characteristics of the problem to be solved (Miettinen, 1999; Miettinen et al., 2008; Miettinen and Hakanen, 2017); such as the number and the form of objective functions, design variables and the type of preference information available from the DM (e.g., desired values of objectives or acceptable trade-offs).

MOO methods can be classified on how they involve the DM's preferences (Miettinen, 1999): before running the optimization algorithm ("a priori" methods), after having found a set of Pareto optimal solutions ("a posteriori" methods) or during an iterative optimization process where the DM's preferences are used to find more preferred solutions (interactive methods). Brown et al. (2015) reported a dramatic increase in the use of a posteriori methods in water systems in recent years. In particular, so-called evolutionary multiobjective optimization (EMO) algorithms have been assessed in water resource management considering algorithmic performance evaluation and identifying future research challenges (Maier et al., 2014; Reed et al., 2013). In the above-mentioned papers, decision making and interactive approaches are briefly mentioned as future challenges (e.g. support through visualization). Additionally, Brown et al. (2015) summarized the short history of water resource systems and highlighted the use of progressive articulation of preference information in MOO (Singh et al., 2008) and indicated it as an emerging research area. An example of an interactive approach is the Modelling to Generate Alternatives (MGA) method and related approaches (Brill et al., 1990; Zechman and Ranjithan, 2007) that have reached main stream in environmental decision making with a nice connection between interactive MOO and sensitivity analysis (Reed et al., 2013). In the MGA method, a small amount of solutions is given to the DM, who can provide additional information not modeled or quantified. In interactive MOO methods, such information is assumed to be the preferences of the

DM. Brill et al. (1990), showed that “human-machine decision-making system will perform better when the human is presented with a few different alternatives than when presented with a homogeneous set of alternatives, as might result from sensitivity analysis”. Similarly, interactive MOO methods have been seen to perform better than generating a large set of Pareto optimal solutions for the DM (Miettinen et al., 2008). However, literature of drinking water treatment design rarely focuses on applying interactive methods or do not consider Pareto optimality of the obtained results (Monarchi et al., 1973). This paper is focused on the optimization of the design of a biological treatment plant for drinking water production that tackles the process drawbacks, contrarily to what happens in a traditional volumetric-load-driven design procedure. In detail, a groundwater biodenitrification filter is considered as a case study for nitrate removal, taking into account two important drawbacks: effluent quality, requiring specific monitoring and post-treatments, and investment and management costs. Indeed, as for the former aspect, products of incomplete biological reactions and biomass leakage in treated water cannot be neglected in a biofilter design for drinking water treatment. To find an optimal design to our case study by active guidance of the DM, we applied the interactive NIMBUS method (Miettinen and Mäkelä, 2006), implemented in the IND-NIMBUS software (Miettinen, 2006). The reason for using IND-NIMBUS is that it is one of the few available implementations of interactive MOO methods and it has been successfully applied to practical problems in various domains (Hakanen et al., 2013; Hartikainen et al., 2015; Ojalehto et al., 2014; Steponavičė et al., 2014). In addition, we compared the efficacy of IND-NIMBUS and an EMO algorithm NSGA-II (Deb et al., 2002) with its implementation NGPM (Lin, 2011) to see the effect of incorporating DM’s preferences into the optimization process and could identify several benefits. The biological process was modelled modifying the Activated Sludge Model 1 (ASM1) (Henze et al., 2000) to take into account nitrite formation, which is usually neglected in biological processes for wastewater treatment. A pilot scale biofilter, managed for about three months, gave data for the validation of the optimization outputs. This work explores the application of interactive MOO methods as supporting tools for process design in a novel field: drinking water treatment. This application lead to new insights on the integration of MOO approaches with conventional process design which can be extended to similar problems, highlighting advantages and helping the choice of the most appropriate decision support tool.

2. Material and Methods

2.1 Design problem definition

The design of the denitrification biofilter has been tailored to remove nitrate from groundwater pumped by Milan city supply wells, characterized by the simultaneous presence of organic pollutants, mainly volatile organic compounds and pesticides. Table 1 reports the main characteristics of groundwater observed during a 70 days monitoring period (three analyses per week).

As for the design problem, an upflow submerged biofilter (influent flow rate of 0.6 m^3/h) was considered, with the dosage of sodium acetate as organic carbon source to support biomass growth, with stoichiometric carbon requirements of 0.70 g_C/g_{DO}, 1.69 $g_C/g_{NO3\text{-}N}$ and 1.27 $g_C/g_{NO2\text{-}N}$ (C: total organic carbon, DO: dissolved oxygen, NO3-N: nitrate, NO2-N: nitrite), as recommended by Henze et al. (2008). Analyses were performed according to Standard Methods (APHA, 2012).

Table 1. Main characteristics of groundwater to be treated in a biofilter. Active biomass, described as suspended mg_{COD}/L (COD: Chemical Oxygen Demand), is calculated based on VSS (Volatile Suspended Solids) measures considering biomass molecule as $C_5H_7O_2N$ according to bioenergetic evaluation by Henze et al. (2008).

	Symbol	Unit	Number of data	Mean	Standard Deviation	Minimum	Maximum
Temperature	T	°C	31	17.4	1.08	14.6	18.8
pH	pH	-	31	7.3	0.12	7.0	7.5
Dissolved Oxygen (DO)	$S_{o,in}$	mg_{DO}/L	29	6.2	0.55	5.0	7.2
Nitrate	$S_{NO3,in}$	$mg_{NO3\text{-}N}/L$	30	8.9	0.65	7.7	10.0
Nitrite	$S_{NO2,in}$	$mg_{NO2\text{-}N}/L$	24	0.01	0.025	0.00	0.09
Total Organic Carbon (TOC)	$S_{S,in}$	mg_C/L	4	< 1	-	-	-
Active biomass	$X_{BH,in}$	mg_{COD}/L	19	0.81	0.090	0.00	0.28

The design optimization was performed through an iterative procedure involving the biofilter simulation model (Section 2.2), the optimizer (Section 2.3) and two data exchanger tools (Supporting Information S1). After system initialization (Section 2.4), MOO methods were applied. By systematically varying the design variable values, the optimizers generated different designs whose performance was evaluated

by the simulation model in terms of the objective and constraint function values, finally, leading to a Pareto optimal design.

A pilot scale groundwater denitrification biofilter was built for the validation of MOO outputs: a biological upflow submerged filter (Biofor®, Degrémont; V = 0.2 m^3) with expanded clay as biomass support (Biolite®, Degrémont; d_{50} = 3.5 mm). The flow rate was maintained constant at 0.6 m^3/h, implying an upflow rate of 8.3 m/h. In addition to organic carbon dosage, phosphorus (P) was dosed setting the phosphorus to nitrogen (N) ratio at 0.05 g_P/g_N, to consider both aerobic and anoxic metabolism requirements (Tang et al., 2011). Thus, the validation of the optimization results was done through data collected during tracer tests (Supporting Information S2) and during a stationary phase of the biological processes that lasted about three months.

2.2 Biofilter simulation model

The modeling of the denitrification biofilter was performed through AQUASIM 2.0 - EAWAG (Reichert, 2008). Biofilter hydraulics was modeled through a series of eight Continuously Stirred Tank Reactors (CSTRs), which permits easy simulation of a Plug Flow Reactor with a low to moderate axial dispersion (d = 0.05 - 0.07) (Tchobanoglous et al., 2014). Each CSTR had the same constant volume (V_{cstr}) 12.5% of the reactor volume (V_r). A fraction (Q_{rec}) of the influent flow rate was assumed to be recirculated to simulate biomass retention in each compartment and calculated from V_r and sludge retention time (SRT) as:

$$Q_{rec} = \frac{V_r}{SRT}. \quad (4)$$

Biological processes occurring in the reactor were modeled through a modification of ASM1. They are biological growth in aerobic and anoxic conditions (both on nitric and nitrous nitrogen), biomass decay and organic compounds hydrolysis, as reported in Supporting Information S3 together with stoichiometric coefficients, process rates and parameters.

The ASM1 process rates were modified with 2-step denitrification kinetics from Magrì and Flotats (2008) and Kornaros and Lyberatos (1998), describing respectively a denitrifying biomass with a nitrite reduction rate considerably greater than the nitrate reduction rate and a nitrite accumulating biomass (nitrite reduction kinetic inhibited by nitrate presence). The modified model was formulated to predict

incomplete denitrification both in terms of nitrate residuals with complete degradation of nitrite and nitrite accumulation. Once having set influent water inflow (Q) and characteristics (Table 1), design and operating parameters, the model calculates the state variable values of the biological system at the stationary phase. The state variables were the concentration of dissolved nitrate (S_{NO3}), nitrite (S_{NO2}), oxygen (S_O) and organic substrate (S_S) as well as the content of active biomass (X_{BH}), particulate non-biodegradable products arising from biomass decay (X_P) and slowly biodegradable substrate arising from particulate decay (X_S).

2.3 Multiobjective design optimization problem

The aim of the optimization of the biofilter design was minimizing nitrate concentration in the effluent, investment and maintenance costs and effluent quality, in terms of carbon and nitrite concentration, in order to contain post-treatment requirements. To formulate this problem, three design variables (DVs) and three objective functions (OBJs) were identified.

The design variables represent design and operating parameters most significantly affecting the design performances:

- DV_1: reactor volume V_r (ranging between 50 L and 1200 L),
- DV_2: readily biodegradable substrate being externally added C_{dos} (dosed carbon, as COD, ranging between 0 mg_{COD}/L and 200 mg_{COD}/L),
- DV_3: sludge retention time SRT (ranging between 1 d and 100 d).

The objective functions are:

- OBJ_1: nitrate removal efficiency, as the residual concentration in the effluent (NO_3-$N_{,out}$),
- OBJ_2: total cost of the plant considering both investment and operating costs (*Costs*),
- OBJ_3: quality of the biofilter effluent, both in terms of organic carbon and nitrite concentration (COD_{out}).

A detailed description of their definition is reported in Supporting Information S4.

Functions OBJ_1 to OBJ_3 to be minimized are respectively expressed as nitric nitrogen concentration in the effluent:

$$OBJ_1 = NO_3 - N_{out}\left[{}^{mg_N}/_{L}\right] = S_{NO3}, \quad (5)$$

as the sum of costs of reactor realization, reagent consumption for carbon supply, energy consumption for backwashing and sludge disposal:

$$OBJ_2 = Costs[€]$$
$$= 0.8\frac{€}{L} \cdot V_R + 10yr$$
$$\cdot \left(C_{dos} \cdot 27.92\frac{€ \cdot L}{yr \cdot mg} + \frac{1}{t_{cycle}} \cdot 160.3\frac{€ \cdot h}{yr} + \frac{\Delta M_{TOT,bw}}{t_{cycle}} \cdot 1.314 \right. \quad (6)$$
$$\left. \cdot 10^{-3}\frac{€ \cdot h}{yr \cdot mg} \right)$$

and as the sum of organic carbon and nitrous nitrogen concentrations in the effluent as COD:

$$OBJ_3 = COD_{out}\left[{}^{mg_{COD}}/_{L}\right] = S_S + S_{NO2} \cdot 1.71\frac{mg_{COD}}{mg_N}. \quad (7)$$

The symbols used have already been introduced, except for $\Delta M_{tot,bw}$, in (6) which is sludge removal for a single backwashing and t_{cycle} which is the treatment cycle duration, both calculated from the definition of SRT and the backwashing procedure reported by Richard (Richard, 1989) on real scale Biofor(R) through equations detailed in Supporting Information S4.

Constraints were set on state variables influencing the biological system and the OBJ values to consider two conditions that cannot be violated, but that are not already stated in the simulation model:

- $CONSTR_1$: maximum acceptable particulate matter accumulation:

$$CONSTR_1: \overline{X_{TOT}} \leq 6750\ \frac{mg_{COD}}{L} \quad (8)$$

- $CONSTR_2$: maximum acceptable value for nitric nitrogen concentration in the effluent:

$$CONSTR_2: S_{NO3} \leq 8.5\ {}^{mg_N}/_{L}. \quad (9)$$

The first constraint limits particulate matter concentration under the value corresponding to 75% of reactor voids filled by particulate matter; while the second constraint assures a nitric nitrogen concentration lower than 75% of the regulation limit on drinking water.

It has to be noted that no constraint has been set on nitrous nitrogen and organic carbon concentrations in the effluent, even though a regulation limit does exist especially for nitrous nitrogen. This is because no drinking water treatment plant would supply the effluent of a biodenitrification unit as it is: post-treatments have to be provided, at least for safety reasons in case of malfunctioning. Thus, nitrite and

organic carbon concentrations in the biofilter effluent have to be minimized rather than keeping below regulation limits to minimize costs of the required post-treatments. Consequently, this evaluation is integrated in the optimization problem in OBJ_3 instead of adding specific constraints.

2.4 Multiobjective optimization methods

To solve the design problem, the interactive MOO method NIMBUS was applied and its performance compared to an EMO algorithm. Typically, interactive methods calculate few Pareto optimal solutions based on the preference information specified by a human DM and iteratively support the DM in finding the most preferred solution. The synchronous NIMBUS method (Miettinen and Mäkelä, 2006) was applied, implemented in the IND-NIMBUS software framework (Miettinen, 2006; Ojalehto et al., 2014).

NIMBUS starts by computing the ranges of the OBJ values in the Pareto front and an initial Pareto optimal solution, which is shown to the DM. If the DM is satisfied, the most preferred solution has been found. Otherwise, the DM is asked to express preferences on how the solution should be changed to get a more preferred solution. This is done through a classification of the OBJs, indicating whether their values at the current Pareto optimal solution:

1. should be improved as much as possible,
2. should be improved up to a given bound,
3. are acceptable as they are,
4. are allowed to impair till a given bound, or
5. may change freely at the moment.

Because of Pareto optimality, if some OBJ is improved, some others should be allowed to impair. The classification information of the DM is considered when new Pareto optimal solutions are computed (up to four in the synchronous NIMBUS method). These solutions are then shown to the DM who can select the most preferred of them and either stop or classify again. This iteration continues until the DM is satisfied and is convinced that better solutions do not exist. The interaction is enabled through a graphical user interface of IND-NIMBUS (see an example screenshot in Supporting Information S5).

Pareto optimal solutions are computed by converting the MOO problem with the preference information into single objective optimization sub-problems, which are solved with appropriate optimizers. In the

present work, they were solved with the differential evolution optimizer (Storn and Price, 1997) (with 6000 maximum iterations allowed, with maximum 400 generations, crossover rate of 0.9, and F factor of 0.8).

Contrary to interactive methods, EMO algorithms operate with a set of solutions (called a population) whose evolution through different iterations (called generations) selects nondominated solutions providing an approximation of the Pareto front after a number of generations. No expression of preferences is required from the DM up to the final choice of the most preferred solution from the final nondominated set. As an EMO algorithm, the commonly used NSGA-II algorithm (Deb et al., 2002) and its Matlab implementation NGPM (Lin, 2011) were used. In NSGA-II, a population of solutions is initialized randomly. Then, for each generation, the population evolves by using genetic operators (selection, crossover and mutation) that include randomness to obtain a new population that is both closer to the Pareto front and as diverse as possible. After the maximum number of generations, the output is a set of nondominated solutions that is used to approximate the Pareto front (the parameters for NGPM were: population size 100 points with 100 generations, crossover rate 1.2 and mutation probability 0.5).

2.5 Sensitivity analysis

A sensitivity analysis was performed to test the robustness of the solutions and generalize the efficacy of the MOO method in solving the design problem: case-study specific parameters and the adopted biofilter simulation model were changed and the obtained MOO solutions were compared.

First, the variability of the optimization results was assessed due to the variation of four parameters, with considered values in parentheses:

- nitric nitrogen concentration in raw water (NO_3-N_{in}, tested values: 4.5 mg_N/L, 20 mg_N/L, 30 mg_N/L);
- influent flow rate (Q, tested values: 0.3 m^3/h, 1.2 m^3/h, 1.8 m^3/h);
- biomass concentration in backwashing water ($X_{TOT,bw}$, tested values: 100 mg_{COD}/L, 400 mg_{COD}/L, 600 mg_{COD}/L);
- dispersion in the biofilter simulated through a different number of CSTRs in series (*n*, tested values: 4 and 6).

Then, the effect of simulation model variation was studied considering two different simulation models presented in Kornaros and Lyberatos (1998) and Magrí and Flotats (2008), characterized by process rates and parameters described in Supporting Information S6.

3. Results and Discussion

A preliminary assessment of the MOO problem was performed through a feasible region investigation by generating 3000 random points between the upper and lower bounds of the DVs and simulating the system for each point. The feasibility of the points was checked by evaluating the corresponding constraint function values and the ratio of feasible points was calculated to identify which constraint was limiting. Results are summarized in Supporting Information S7. This analysis permits to highlight the conflicts among OBJs: while a reduction of COD_{out} (OBJ_3) and costs (OBJ_2) can be achieved concurrently, both OBJs conflict with NO_3-N_{out} concentrations (OBJ_3). The conflict is especially critical in case of desired OBJ_3 values below 0.2 mg_N/L NO_3-N_{out}. The DM's role, hence, is to find the preferred balance of such conflicting objectives.

3.1 Multiobjective optimization through IND-NIMBUS

The optimization applying IND-NIMBUS was started by evaluating the ranges of the OBJs (so-called nadir and ideal objective vectors) (Miettinen, 1999) and a first Pareto optimal solution, reported in Table 2 (iteration 1). Then, an iterative solution process guided by the DM's preference information (as classification, see Section 2.4), was conducted resulting in Pareto optimal solutions reported in Table 2 (iterations 2-4). All the solutions generated are shown in Figure 1.

The ideal and the nadir objective vectors showed that solutions do exist that are able to perform complete NO_3-N removal (ideal $OBJ_1 = 0$) or to assure almost no worsening of effluent quality (ideal $OBJ_3 = 0$), but not simultaneously.

The first Pareto optimal solution (solution 1) allows an 87% nitrate nitrogen removal with 7202 euro costs (12003 $euro \cdot m^{-3} \cdot h$) and COD effluent concentration of about 1.16 mg_{COD}/L. Higher nitrogen removal efficiencies are reported in the literature for full scale drinking water biodenitrification units (Richard, 1989), and an efficiency higher than 90% is reported for pilot scale units (Matějů et al., 1992). Furthermore, the low level of COD_{out} limits post-treatment requirements but it can correspond to a dramatic contribution of nitrous nitrogen considering the low value of dosed carbon (49% of

stoichiometric requirements). Thus, further optimization was needed and the DM started providing preference information to find more preferable solutions.

Table 2. Pareto optimal solutions generated by IND-NIMBUS and DM's preference information at different iterations. The most preferred Pareto optimal solution is highlighted in grey.

NIMBUS iteration	Pareto optimal solution	DM's Preference Information					DV1	DV2	DV3	OBJ1	OBJ2	OBJ3
		MOO Starting Point	OBJ that should improve	OBJ acceptable as it is	OBJ allowed to impair	OBJ that may change freely	V_r	C_{dos}	SRT	NO_3-N_{out}	Costs	COD_{out}
							[L]	[mg_{COD}/L]	[d]	[mg_N/L]	[euros]	[mg_{COD}/L]
1	**Nadir**	-	-	-	-	-	-	-	-	8.89	56,100	158
	Ideal	-	-	-	-	-	-	-	-	0	41.3	0.2
	1	-	-	-	-	-	83	25.4	26.8	1.13293	7,202	1.16173
2	**2**	1	OBJ3 (as much as possible)	OBJ1	-	OBJ2	52	31.4	4.1	0.71954	8,921	1.12766
	3						151	31.3	4.1	0.73631	8,961	1.12750
	4						73	31.3	4.1	0.72356	8,909	1.12768
3	**5**	4	OBJ1 (as much as possible)	-	-	OBJ2 OBJ3	101	42.8	7.5	0.00019	12,169	4.07301
	6						107	42.9	7.6	0.00017	12,214	4.17777
	7						117	39.0	10.2	0.00117	11,115	2.56437
	8						89	27.9	26.1	0.40238	7,898	1.27773
4	**9**	1	OBJ1 (up to 0.89 mg_N/L)	-	OBJ2 (up to 10000€) OBJ3 (up to 2 mg_{COD}/L)	-	57	31.3	4.1	0.75324	8,875	1.27357

To demonstrate the DM's role in the solution process, we discuss here some particular aspects. At the second iteration, the DM accepted the OBJ_1 value as it is, asking to improve as much as possible OBJ_3 and letting OBJ_2 change freely. Three Pareto optimal solutions were computed with a negligible difference in performances. A little improvement was obtained in OBJ_3 (2.9% reduction of COD_{out} value) thanks to a higher carbon dosage (61% of stoichiometric requirements), that implied an improvement in OBJ_1 (36% reduction). As the dosed carbon was still lower than stoichiometric requirements, the risk of incomplete denitrification still existed and the nitrous nitrogen contribution to COD_{out} could be relevant. Thus, the DM started iteration 3 from solution 4. After the preferences had been expressed, four more Pareto optimal solutions were computed, with almost complete NO_3 removal, higher costs and higher values of COD_{out}. The results suggested to the DM that a 90% NO_3 removal

(0.89 mg_N/L NO_3-N_{out}), with costs around 10,000 euros and COD_{out} concentration around 2 mg_{COD}/L could be a good trade-off.

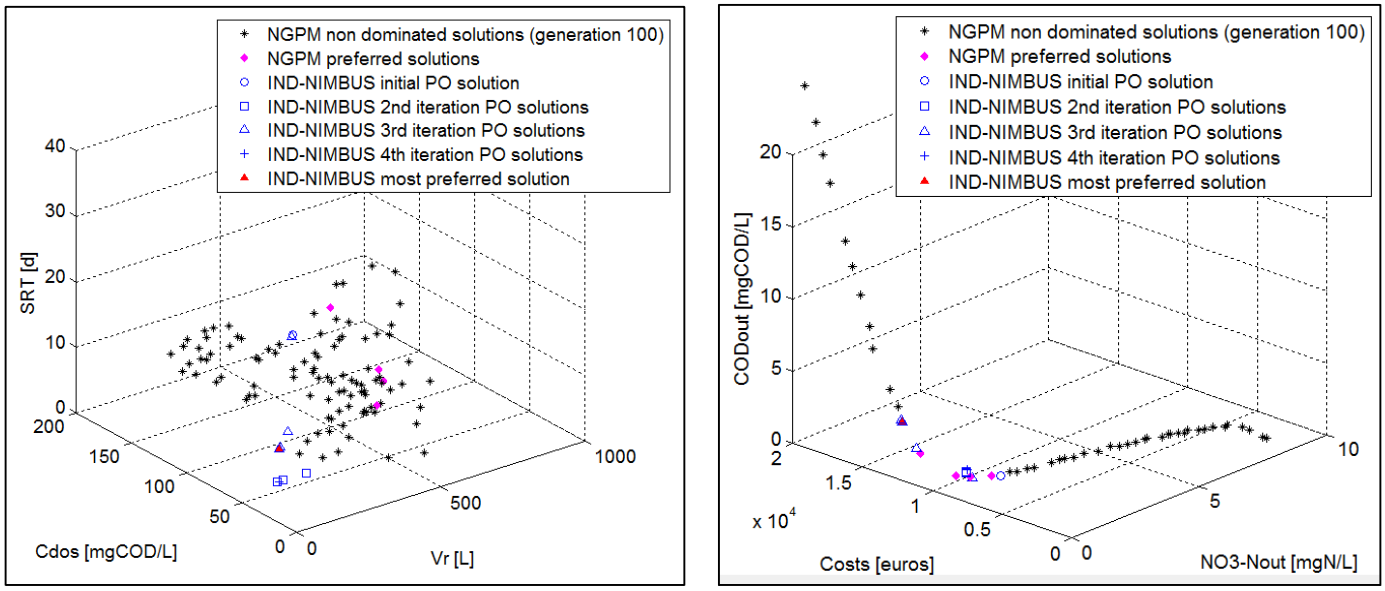

Figure 1. Nondominated solutions found with NGPM (generation 100) and Pareto optimal solutions found with IND-NIMBUS in the design space (a) and in the objective space (b).

The obtained Pareto optimal solutions were few and thus easy to compare, but carrying relevant information supporting the choice of the most preferred design and insight about the feasibility of the preferences expressed.

Indeed, some trade-offs clearly appeared to the DM. First of all, the iterations guided by the DM's preference information rapidly reached solutions assuring almost complete nitrate removal (solutions 2 - 9) at acceptable costs, but also that the lowest OBJ_3 value for Pareto optimal solutions is 1.13 mg_{COD}/L. It also showed that Pareto optimal solutions can be obtained with a dosed carbon lower than the stoichiometric requirements, since the simulation model takes into account the organic carbon available from biomass decay (endogenous organic carbon). However, supplying carbon under the stoichiometric level does not assure the complete removal of nitrous nitrogen, which can represent an important fraction of the OBJ_3 value. Therefore, the iterative comparison of solutions showed to the DM a critical issue: obtained values for OBJ_3 are difficult to be fully evaluated: the minimum value of OBJ_3

corresponded to an acceptable concentration of residual carbon (0.42 mg_C/L), in case of complete denitrification, but it corresponded also to an unacceptable value of NO_2-N (0.66 mg_N/L), in case of incomplete denitrification. Thus, the iterative process revealed a difficulty in identifying the acceptable level of impairment for OBJ_3, as it aggregates the concentrations of NO_2-N and TOC in the effluent water. The results suggested that further studies should be performed, with the implementation of a fourth OBJ (splitting OBJ_3 in two), which offers a more informative optimization model.

Considering what has been outlined so far, solution 5 represented the most preferred design to be implemented. Compared to other Pareto optimal solutions, it involves a higher COD_{out} value. However, it assures a high removal of nitric nitrogen (>90%, NO_3-N_{out} < 0.89 mg_N/L) with a SRT value that implies good biomass retention, good endogenous carbon and carbon dosage equal to 83% of the stoichiometric requirements that, together, are more likely to involve complete denitrification with a high TOC contribution and almost zero NO_2-N contribution to the COD_{out} value (accepted to be around 4 mg_{COD}/L). Overall, the interactive solution process enabled the DM to learn about the interdependencies of the objectives and the feasibility of expressed preferences and get convinced of the goodness of the solution found.

Finally, considering the calculation time, 8 hours were needed for the problem initialization but only 2 hours were necessary for each optimization. This calculation time was still regarded as acceptable but it would have also been possible to employ an approach directed to computationally expensive problems as e.g. in Steponavičė et al. (2014).

Despite the calculation time, the application of IND-NIMBUS was intuitive and user-friendly from the DM's point of view, since she was not asked to understand the working principles of the optimization algorithm. The limited number of solutions to compare helped the DM in the choice of the best suited one. Lastly, the iterative procedure gave a deeper insight on relationships among objectives, increasing the ability and the confidence in choosing the optimal design to be implemented, without neglecting potentially interesting portions of the Pareto front.

3.2 Multiobjective optimization through NGPM

The NGPM software, where no preferences are considered, was also applied to study the effect of including DM's preferences into the optimization. The NGPM optimization took 10.35 hours and

generated 100 nondominated points with an increasing ratio of feasible points for each population (reaching 100% at generation 3). As it is typical of EMO methods, Pareto optimality of solutions is not assured and it is only known that they are nondominated. In this sense, the population obtained at generation 100 represented an approximation of the Pareto front. Smallest and largest values for each objective in the final population are reported in Table 3.

Table 3. Main descriptive statistics of nondominated solutions produced by NGPM (generation 100).

	DV_1	**DV_2**	**DV_3**	**OBJ_1**	**OBJ_2**	**OBJ_3**
	V_r	**C_{dos}**	**SRT**	**NO_3-N_{out}**	**Costs**	**COD_{out}**
	[L]	**[mg_{COD}/L]**	**[d]**	**[mg_N/L]**	**[euro]**	**[mg_{COD}/L]**
Minimum	75	5	5	$2 \cdot 10^{-6}$	1,619	0.3
Maximum	984	200	34	8.50	56,409	158

When considering the aim of the optimization, a single Pareto optimal design to be implemented had to be found among the 100 nondominated solutions obtained and therefore a choice by the DM was needed based on her experience. In fact, it should be stressed that EMO methods do not give any additional information to help the DM's choice. The NGPM optimization gave indications on the most suitable range of values for V_r and SRT design variables (Table 3), but no suggestion could be deduced about C_{dos}, which involves a wide range of design performances. The choice could be made by visually analyzing the objective space in a 3D plot, because the problem had only three objectives (this is no more possible if the number of OBJs is higher). Thus, in this case, the most preferred solution could be sought among solutions with NO_3-N_{out} lower than 1 mg_N/L and COD_{out} lower than 5 mg_{COD}/L. This corresponded to the four preferred solutions that are shown in Figure 1, characterized by V_r, SRT and C_{dos} respectively equal to 231 - 444 L, 11 - 28 d and 26 - 37 mg_{COD}/L (corresponding to C_{dos}/C_{stech} equal to 50-70%). The carbon dosage of these solutions is too low since with a carbon dosage lower than stoichiometric requirements, the risk of incomplete denitrification increases as C_{dos}/C_{stech} ratio decreases. However, it appeared to be complicated to find a proper solution among the NGPM solutions with a lower COD_{out}, which is obtained through a higher dosage of carbon and acceptable values of the other two objectives.

3.3 Discussion and validation of optimization results

The presented results showed that MOO is a valuable tool for drinking water treatment design optimization, with strengths and weaknesses depending on the applied method. When comparing solutions found with the two optimization methods, one must keep in mind that NGPM solutions are only nondominated while IND-NIMBUS solutions are assured to be Pareto optimal (if the single objective optimizer guarantees optimality). Then, a comparison can be done by checking if the IND-NIMBUS solutions belong to the region of design and objective spaces where NGPM solutions are concentrated. Considering results reported in Tables 2 and 3, all IND-NIMBUS solutions have reactor volume values on a different range compared to NGPM and the same can be said about SRT values of solutions 1 and 9. This indicates that Pareto optimal solutions found with IND-NIMBUS do not overlap with nondominated solutions found with NGPM. Thus, NGPM did not find solutions that the DM found with IND-NIMBUS, as visualized in Figure 1 in both design and objective spaces.

General considerations on biofilter optimal design can be drawn by normalizing IND-NIMBUS results: Pareto optimal solutions resulted to be characterized by volumetric loading in the range 0.85-2.54 $kg_N\ m^{-3}\ d^{-1}$ (with EBCTs in the range 5-15 min) and a carbon dosage of 0.49-0.83 $g_{C,dos}/g_{C,stoich}$, with SRTs in the range 4-27 d. Pareto optimal solutions in these ranges are expected to involve a nitric nitrogen removal efficiency of 87-100%, with 809-1573 €/$g_{N,removed}$ and 0.04±0.097 $g_{COD,out}/g_{COD,in,}$ with the COD content in the effluent being made up both by residual carbon and nitrite.

Considering the performances of the pilot scale biofilter built for the validation of the MOO results (data not shown), it has to be reported that applying the suggested carbon dosage for volumetric loadings and EBCTs led to lower nitric nitrogen efficiencies (40-75%) with average COD_{out} equal to 4.8 mg_{COD}/L, but with peaks of nitrous nitrogen up to 5.4 mg_N/L. On the contrary, higher efficiencies (55-100%) were obtained for volumetric loadings and EBCTs in the proposed ranges but with a carbon dosage of 1.05-1.34 $g_{C,dos}/g_{C,stoich}$ with average COD_{out} equal to 7.4 mg_{COD}/L but composed by residual organic carbon for 87% on average. These observations confirm the need of splitting the third objective in two, for a better optimization of nitrous nitrogen concentration in the effluent as a function of the dosed carbon.

When comparing the applied optimization methods, the reported results proved that NGPM can be applied to get an approximation of the Pareto front, keeping in mind that the distance from the real Pareto front is unknown and decreases only by increasing the number of generations. Thus, it can be applied if the purpose of the optimization is the estimation of possible trade-offs. However:

- If the purpose is finding the most preferred Pareto optimal solution as the design to be implemented, NGPM is not able to identify it and further optimizations or analyses are needed.
- NGPM results can give some indications to help the DM's choice of the most preferred solution, but an unsupported choice has to be made mainly based on the DM's experience.
- The evaluation of the NGPM results has to be done by plotting them in the design and objective spaces. However, this is possible only for up to three dimensions and in higher dimensions other visualizations are needed (Miettinen, 2014).

On the contrary, IND-NIMBUS has not been intended for studying the whole Pareto front but to help the DM in finding her/his preferred Pareto optimal solution. IND-NIMBUS proved to be well suited for the design problem considered:

- It allowed obtaining Pareto optimal solutions, whose evaluation was supported by an easy visualization of the results. This enables also the consideration of a higher number of objectives.
- It allowed performing multiple optimization iterations that increased DM's knowledge on the MOO problem in the areas of interest. It also showed the best and the worst design performances in terms of defined objectives and revealed the need for a better MOO problem formulation (i.e., by splitting the third objective into two distinct ones for a better evaluation of required post-treatments).
- The DM's choice was effectively supported to find the most preferred design to be implemented and the DM was not overloaded by having to study too much information at a time.

3.4 Sensitivity analysis results

A sensitivity analysis was performed evaluating the variation of the feasible region, NGPM and IND-NIMBUS results and detailed outputs are available in Supporting Information S8.

468 The feasible region analysis appeared to be affected mainly by nitric nitrogen concentration in the
469 influent: increasing values of NO_3-N_{in} decreased the number of feasible solutions, because of an
470 increasing number of points violating constraint 2 (adequate nitrate removal).
471 As for IND-NIMBUS, the sensitivity analysis was conducted performing five steps of the interactive
472 method to identify the most preferred design, for each different MOO problem. The obtained results
473 revealed that the greatest variations of DVs and OBJ values for the most preferred design, were
474 associated with NO_3-N_{in} variation (+736% DV_1 value), and the application of 4-CSTRs model (+698%
475 DV_3 value) and Magrì and Flotats (2008) model (+654% DV_1 value and 1105% DV_3 value).
476 As for NGPM, the highest variations were observed varying influent flow rate that led to reactor volume
477 variation up to +255%, involving a cost variation of +208%. However, the nondominated solutions
478 were also significantly affected by NO_3-N_{in}: increasing values increased both the reactor volume (up to
479 +95%) and the dosed carbon (up to 198%) leading to designs with higher costs (up to 196%) but also
480 higher residual concentrations in the effluent of NO_3-N (up to +56%) and COD (up to +189%). Lower
481 but remarkable variations were observed increasing hydraulic dispersion (e.g. for lower number of
482 CSTRs in series) and applying the simulation model of Magrì and Flotats (2008): the former involved
483 higher values of SRT (up to +126%), while the latter identified designs with higher reactor volumes (up
484 to +174%) and higher values of NO_3-N_{out} (up to +63%).
485 The presented results allowed generalizing the efficacy of the proposed MOO approach indicating that
486 NO_3-N_{in} is the most important parameter affecting the optimization results, together with influent flow
487 rate, hydraulic dispersion in the reactor and the considered simulation model.

488 **4. Conclusions**

489 Reported results show that it is beneficial to formulate a denitrification biofilter design problem as a
490 multiobjective optimization problem. We demonstrated that applying an interactive MOO method
491 offers advantages over a widely used a posteriori evolutionary algorithm and, thus, it is necessary to
492 choose the correct MOO method to find the most preferred Pareto optimal design to be implemented.
493 Considering the specific design problem of a pilot scale biofilter for groundwater biodenitrification,
494 results showed that:

- The interactive method was the most suitable one, effectively supporting the design process up to the identification of the final, most preferred, Pareto optimal design.
- General considerations on biodenitrification designs can be drawn by normalizing IND-NIMBUS results: Pareto optimal designs resulted to be characterized by volumetric loading in the range 0.85-2.54 $kg_N\ m^{-3}\ d^{-1}$ (with EBCTs in the range 5-15 min) and a carbon dosage of 0.49-0.83 $g_{C,dos}/g_{C,stoich}$, with SRTs in the range 4-27 d. These values are expected to involve a nitric nitrogen removal efficiency of 87-100% with 809-1573 $€/g_{N,removed}$ and 0.044±0.097 $g_{COD,out}/g_{COD,in}$ and the COD content in the effluent made up by residual carbon and nitrite.
- Observations on a pilot scale biofilter validated the identified Pareto optimal designs. However, they also confirmed the need of splitting the third objective in two, for a better optimization of nitrous nitrogen concentration in the effluent as a function of the dosed carbon.

In general, the presented MOO design approach with the aid of the IND-NIMBUS software allowed obtaining a Pareto optimal design without any "a priori" evaluation based on practical experience, as in the case of volumetric-load-driven design procedures. In order to narrow and improve the range of the Pareto optimal designs, further studies should be done adopting a four-objective model by splitting the "effluent water quality" objective in two, considering nitrite and carbon concentrations separately. In that case, the efficiency of the EMO software NPGM applied would suffer further but IND-NIMBUS would still be applicable.

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