

Multivariate methods in modelling the removal of nitrogen and phosphorus in deep-bed two-media down-flow sand filters

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ABSTRACT

Multiple linear regression (MLR) was applied to study the operation and efficiency of simultaneous denitrification with sodium acetate (NaAc) as a carbon and energy source and the chemical precipitation of phosphate with iron salts in a pilot sand filter. Different loads were simulated with the addition of primary settled wastewater (PW) containing suspended solids (SS) and biological SS from the aeration tank. SS from PW partly passed the filter. Biological SS was caught by the filter. The validation of the model showed that phosphorus concentration and chemical oxygen demand reduction were easier to predict correctly than the time of operation of the filter. The reduction of total phosphorus differed between 0.01 and 0.31 mg P/L, and the nitrate concentration in the filtrate differed between 0 and 5.2 mg N/L when comparing the experimental values with the model. Experiments showed filtrate concentrations of 0–0.12 mg PO₄-P/L, 3.0–8.8 mg NO₃-N/L, and 1.9–34 mg SS/L. The addition of NaAc gave a small increase in the reduction of PO₄-P probably because of the assimilation of PO₄-P into the denitrifying sludge. The dosage of Fe gave a small decrease in the reduction of NO₃-N probably because of PO₄-P limitation for denitrifying bacteria, as Fe acts as a precipitation agent upon PO₄-P.

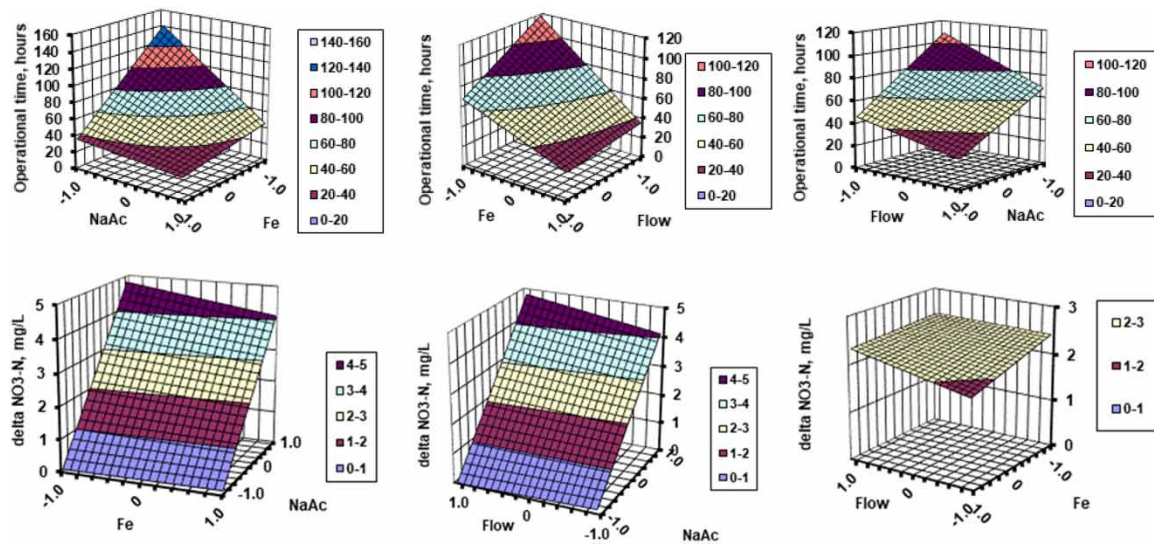
Key words: MLR, multiple linear regression, nutrient removal, operational time, sand filter, validation

HIGHLIGHTS

- Multiple linear regression (MLR) was applied to study processes in sand filters.
- Different loadings and removal of suspended solids, organic material, and phosphorus were evaluated.
- The models were found to well describe the responses of operational cases.
- Suspended solids were removed more efficiently from secondary than primary settled wastewater.
- The dosage of iron salt decreased the reduction of NO₃-N due to PO₄-P limitation.

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GRAPHICAL ABSTRACT



INTRODUCTION

New demands for the concentration of nitrogen, phosphorus, and BOD₇ in the effluent were expected for the Henriksdal wastewater treatment plant (WWTP) in Stockholm, Sweden, which is the largest WWTP in Europe that is excavated into rock. The capacity and removal efficiency were increased by building three new biological lines and a sand filter step as a final treatment step. In order to facilitate the design and operation of the full-scale sand filters, results from the present study were used. The pilot tests were performed at the Henriksdal WWTP, and one central aim was to study simultaneous suspended solids separation, denitrification, and phosphorus precipitation. The multivariate method multiple linear regression (MLR) with a full factorial design was used to overcome the problems with interpreting the results from the many values resulting from the study.

Previously, Healy *et al.* (2007) studied the removal of total suspended solids (TSS), total chemical oxygen demand (COD), filtered COD (COD_f), total nitrogen, nitrate nitrogen (NO₃-N), ammonium nitrogen (NH₄-N), and total Kjeldahl nitrogen (TKN) in a pilot-scale sand filter. However, the filter had a low hydraulic load. Sperlich *et al.* (2013) studied a pilot-scale dual filter with anthracite and sand at a hydraulic load of 6 m/h. The removal of TSS, NO₃-N, nitrite nitrogen (NO₂-N), total phosphorus, and COD were calculated. Altmann *et al.* (2015) studied the removal of phosphorus, TSS, and micropollutants in a pilot-scale dual media deep-bed filter with anthracite and sand at a hydraulic load of 7.5 m/h. Powdered activated carbon and ferric chloride were added in the study. Huang *et al.* (2011) used MLR in a two-factor designed model without quadratic or interaction terms in calculate the model response. Katayama & Imori (2014) studied selection criteria for identifying relevant predictors in high-dimensional multivariate linear regression analysis, which included high exponents of power for the factors of the variables.

In the present study, the reduction of NO₃-N, PO₄-P, total P, total P minus PO₄-P, TSS (mentioned as SS), and COD_f were investigated in a pilot-scale deep-bed two-media down-flow filter. The concentrations in the filtrate of these substances except COD_f were also studied. Hydraulic loads of 5 and 10 m/h were investigated.

METHODS

Pilot plant filter

A pilot plant filter was studied at the Henriksdal WWTP in Stockholm. It was operated with wastewater from the full-scale plant. The concentration of phosphate is often high in the effluent from the secondary sedimentation tanks in the Henriksdal plant. Precipitation of phosphate with iron salts in the filters has been introduced. In the pilot plant study, precipitation with iron was, therefore, of great interest. Even more, strict nitrogen demands were expected later. Denitrification in the filter with dosage of a carbon source was also studied.

The pilot plant study was performed in a stainless steel filter unit with a filter bed height of 1.5 m, which is the same height as in the full-scale filters, a filter diameter of Ø0.4 m, and a total height of 3.0 m. Eight parallel pilot

plant filters were constructed for the evaluation. Filter 7, in which the denitrification study was done, contained a 0.3 m sand layer with a grain size of $\text{Ø}0.8\text{--}1.2$ mm and above that 1.2 m ceramic grains, with a grain size of $\text{Ø}2.5\text{--}4.0$ mm (mentioned as expanded clay in Figure 1). The filters functioned as deep-bed two-media down-flow filters. At the Henriksdal WWTP, 60 full-scale filters have been installed, each with a length of 10 m and a width of 6 m.

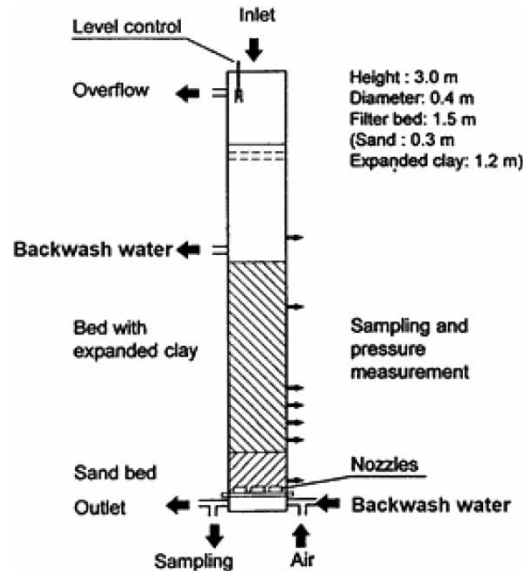


Figure 1 | Pilot plant filter at the Henriksdal WWTP. Expanded clay refers to the ceramic grains in the filter bed.

Sampling and analytical methods

The time taken to reach a high-pressure drop in the filter was measured by a conductive level indicator. The signal from this indicator was recorded. The end of the filter cycle was detected by the level meter as a level of around 2.75 m. When the filter bed was clogged, the water level in the filter starts to increase until finally the water level reached the overflow pipe and the filter was no longer able to filtrate all the influent. Composite samples from the influent and the filtrate were taken in all experiments. The sampling points are marked with Inlet and Sampling as shown in Figure 1. SS were analysed by a gravimetric method according to the Swedish standard methods SS028112-3 and SS-EN 872 but with glass microfibre filters of grade GF/C (pore size $1.2\ \mu\text{m}$, Whatman). When soluble fractions were analysed, the samples were first filtered with glass microfibre filters of grade MGC (pore size $1.2\ \mu\text{m}$, Munktell). The concentrations of nitrate, phosphate, total phosphorus, and filtered COD (COD_f) in the influent and the filtrate were analysed using cuvette tests (LCK339, LCK349, LCK348, and LCK114, respectively, Dr Lange, Germany), and the cuvettes were measured using a filter photometer LP 2W (Dr Lange, Germany). The pressure levels along the bed height of the filter were recorded manually at six different locations in the filter bed and one location below the water surface during the pilot study (Figure 1). These are marked with small arrows on the right side of the filter. Values from the pressure meters are not presented in this paper.

Operational conditions

Filtration of secondary settled wastewater from the Henriksdal WWTP was studied in the pilot plant filter. The experiments were conducted with hydraulic loads of 5 and 10 m/h. In order to achieve denitrification in the filter, sodium acetate (NaAc) was dosed as a carbon source in selected experiments. The dosage was mainly around $71\text{--}72\ \text{g NaAc}\cdot 3\text{H}_2\text{O}/\text{m}^3$. In order to simulate occasions with a high hydraulic load to the plant, 19% of primary settled wastewater and 81% of secondary settled wastewater were mixed in the study, giving $23\text{--}37\ \text{mg SS/L}$ in the influent to the filter. In the full-scale plant, part of the primary settled wastewater (PW) bypasses the biological treatment at high flows and is led to the filters directly. Sludge escape was simulated by mixing secondary settled wastewater with a small flow of activated sludge from the corresponding aeration tank. This resulted most often in around $60\text{--}71\ \text{mg SS/L}$ in the influent to the filter.

Use of multivariate methods in evaluating the function of the filter

Multivariate methods were applied to study the efficiency of filter operation. The influence of different processes and operational parameters on 12 different responses was studied using a full factorial design to evaluate empirical relationships with MLR. Models were calculated for three different operational cases. First, the case with only secondary settled wastewater in the influent to the filter was evaluated for two different flows. In the second case, 19% of the volume of PW was added to the filter inlet to simulate the conditions with high flows through the plant when part of PW bypasses the biological treatment and is led directly to the filters. In the third case, SS from the aeration tank outlet was added to the filter inlet to simulate sludge escape. In case 1, the three variables were the dosage of Fe or not, the dosage of NaAc or not, and the filter flow (Q). In case 2, the flow is replaced by the addition of PW or not. Finally, in case 3, the flow is replaced by the addition of SS or not. All values for the variables were normalized before the calculations and were stated within the range of -1 to $+1$. In Table 1, the values representing -1 and $+1$ are presented. The dosage of Fe and NaAc is represented by the amount of the added substances. The surface loading of 5.0 or 10.0 m/h represents a lower or higher flow through the filters. In cases 2 and 3, the surface loading was 10.0 m/h.

Table 1 | The values of the variables for the three different cases

	Fe dosed (g/m ³)		NaAc·3H ₂ O dosed (g/m ³)		Flow (m/h)		PW added		SS added	
	-1	+1	-1	+1	-1	+1	-1	+1	-1	+1
Case 1	0	1.89	0	71.5	5	10				
Case 2	0	1.89/2.84	0	71.5			No	Yes		
Case 3	0	1.89/2.84	0	71.5					No	Yes

1.89 g Fe/m³ was dosed when FeCl₃ was added and 2.84 g Fe/m³ was dosed when FeSO₄ was added in cases 1, 2, and 3. An earlier study showed no significant difference between FeSO₄ and FeCl₃ as a precipitation agent (Jonsson 1998). The set point for the carbon source dosage was 71.6 g NaAc·3H₂O/m³. The actual dosage became 71.5 g NaAc·3H₂O/m³ as an average, which was very close to the set point of 71.6 g/m³. The variables, on which the calculations were based, are presented in Table 2 for cases 1, 2, and 3.

Table 2 | Variables on which the models were based ($- = -1$ and $+ = +1$)

Exp	Variables, case 1			Variables, case 2			Variables, case 3		
	Fe	NaAc	Q	Fe	NaAc	PW	Fe	NaAc	SS
1	-	-	-	-	-	-	-	-	-
2	+	-	-	+	-	-	+	-	-
3	-	+	-	-	+	-	-	+	-
4	+	+	-	+	+	-	+	+	-
5	-	-	+	-	-	+	-	-	+
6	+	-	+	+	-	+	+	-	+
7	-	+	+	-	+	+	-	+	+
8	+	+	+	+	+	+	+	+	+

Exp = Experiment (number).

Interaction terms but not quadratic terms were included in the model. The empirical models of MLR for the calculated responses, \hat{y} , in cases 1, 2, and 3, respectively, have the formulas according to Equations (1)–(3).

$$\hat{y} = b_0 + b_1 \cdot x_{\text{Fe}} + b_2 \cdot x_{\text{NaAc}} + b_3 \cdot x_{\text{Q}} + b_{12} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} + b_{13} \cdot x_{\text{Fe}} \cdot x_{\text{Q}} + b_{23} \cdot x_{\text{NaAc}} \cdot x_{\text{Q}} + b_{123} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} \cdot x_{\text{Q}} \quad (1)$$

$$\hat{y} = b_0 + b_1 \cdot x_{\text{Fe}} + b_2 \cdot x_{\text{NaAc}} + b_3 \cdot x_{\text{PW}} + b_{12} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} + b_{13} \cdot x_{\text{Fe}} \cdot x_{\text{PW}} + b_{23} \cdot x_{\text{NaAc}} \cdot x_{\text{PW}} + b_{123} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} \cdot x_{\text{PW}} \quad (2)$$

$$\hat{y} = b_0 + b_1 \cdot x_{\text{Fe}} + b_2 \cdot x_{\text{NaAc}} + b_3 \cdot x_{\text{SS}} + b_{12} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} + b_{13} \cdot x_{\text{Fe}} \cdot x_{\text{SS}} + b_{23} \cdot x_{\text{NaAc}} \cdot x_{\text{SS}} + b_{123} \cdot x_{\text{Fe}} \cdot x_{\text{NaAc}} \cdot x_{\text{SS}} \quad (3)$$

Quadratic terms, e.g. $b_{11} \cdot x_{Fe} \cdot x_{Fe}$, were not included. They give maxima or minima inside the response surface diagram, but these terms have no meaning when the variables are discrete. The dosage of Fe or no dosage of Fe and the dosage of NaAc or no dosage of NaAc are examples of discrete variables.

$$y = \hat{y} + \varepsilon \quad (4)$$

y in Equation (4) is the actual response and ε contains the neglected terms mentioned above, terms with higher exponents of power in the Taylor's equation for approximations, and the experimental error.

High absolute values for the coefficients b_i , b_{ij} , and b_{ijk} give high positive or negative influence on the response, \hat{y} , depending on the sign before b_i , b_{ij} , and b_{ijk} . The desire is, of course, that the responses of the reduced concentrations and the time of operation before clogging are high and that the concentrations in the filtrate are low. The results are presented in three-dimensional response surface diagrams for some selected responses based on the response models, i.e. the equations for \hat{y} in Equations (1)–(3). In MLR, the b_i , b_{ij} , and b_{ijk} coefficients are calculated from matrix multiplications according to $(X'X)^{-1} \cdot X'Y = b$, where X , Y , and b , with the concentrations of reduced $\text{NO}_3\text{-N}$ in case 1 as an example for Y , are

$$\begin{array}{cccccccc}
 X_0 & X_1 & X_2 & X_3 & x_1x_2 & x_1x_3 & x_2x_3 & x_1x_2x_3 \\
 \begin{bmatrix}
 +1 & -1 & -1 & -1 & +1 & +1 & +1 & -1 \\
 +1 & +1 & -1 & -1 & -1 & -1 & +1 & +1 \\
 +1 & -1 & +1 & -1 & -1 & +1 & -1 & +1 \\
 +1 & +1 & +1 & -1 & +1 & -1 & -1 & -1 \\
 +1 & -1 & -1 & +1 & +1 & -1 & -1 & +1 \\
 +1 & +1 & -1 & +1 & -1 & +1 & -1 & -1 \\
 +1 & -1 & +1 & +1 & -1 & -1 & +1 & -1 \\
 +1 & +1 & +1 & +1 & +1 & +1 & +1 & +1
 \end{bmatrix}
 & Y = \begin{bmatrix}
 y_1 \\
 y_2 \\
 y_3 \\
 y_4 \\
 y_5 \\
 y_6 \\
 y_7 \\
 y_8
 \end{bmatrix} = \begin{bmatrix}
 0.1 \\
 0.2 \\
 4.7 \\
 3.5 \\
 0 \\
 0 \\
 4.9 \\
 4.7
 \end{bmatrix} & b = \begin{bmatrix}
 b_0 \\
 b_1 \\
 b_2 \\
 b_3 \\
 b_{12} \\
 b_{13} \\
 b_{23} \\
 b_{123}
 \end{bmatrix}
 \end{array}$$

RESULTS AND DISCUSSION

The real responses from the experiments, y , on which the calculations were based, are presented in Table 3 for case 1, Table 4 for case 2, and Table 5 for case 3. The responses are operational time from the start of a cycle to clogging of the filter, the concentrations in the filtrate, and the concentrations reduced in the filter in mg/L, both from analyses of composite samples, where Δ represents the reduction of the substance.

The empirical relationships for different \hat{y} , which are calculated by MLR, are shown in Table 6 for case 1, in Table 7 for case 2, and in Table 8 for case 3, where operational time is the time of operation from the start to the clogging of the filter. Models with high accuracy are, however, impossible to create because of varying conditions of the wastewater in different experiments with a variety of components in the influent wastewater, different concentrations of substances, and different ion strengths. For case 1, b_1 is also called b_{Fe} , $b_2 = b_{NaAc}$, $b_3 = b_Q$, $b_{12} = b_{Fe NaAc}$, $b_{13} = b_{Fe Q}$, $b_{23} = b_{NaAc Q}$, b_{123} is also called $b_{Fe NaAc Q}$, and b_0 is a constant. For case 2, the b values are named in a similar way but Q is replaced by PW, and for case 3, Q is replaced by SS.

In the example, $(X'X)^{-1} \cdot X'Y = b$ is $1/8 \cdot X'Y = b$. For the reduction of $\text{NO}_3\text{-N}$ in case 1, b_1 is $1/8 \cdot (-y_1 + y_2 - y_3 + y_4 - y_5 + y_6 - y_7 + y_8) = -1.3/8 = -0.1625$ and b_2 is $1/8 \cdot (-y_1 - y_2 + y_3 + y_4 - y_5 - y_6 + y_7 + y_8) = +17.5/8 = +2.1875$, which also could be seen in Table 6. Y is presented in Table 3. The other coefficients, b_i , b_{ij} , and b_{ijk} , are calculated in the same way. In this example, the dosage of Fe had a small negative effect on the reduction of $\text{NO}_3\text{-N}$ in the filter, and the dosage of NaAc had, as expected, a high positive effect on the reduction of $\text{NO}_3\text{-N}$ in the filter. $b_Q = +0.1375$ according to Table 6, which corresponds to the effect of the magnitude of the flow through the filter. This value is low enough, indicating that only a small increase in the $\text{NO}_3\text{-N}$ reduction will result from a higher flow. This is also confirmed in the response surface diagrams (Figure 2). The coefficients of interaction terms are also small and these gave little influence on the reduction of $\text{NO}_3\text{-N}$ in the filter.

In the validation of the models, a comparison of the model responses, \hat{y} , with the values of analyses, y , from experiments, not included in the calculation of the models, was done (Table 9). The fit is not perfect but within the range of what could be expected. For example, in comparison 1, \hat{y} for ΔCODf is 32 mg/L compared to 33 mg/L for y . In comparison 4, both \hat{y} and y are 0.26 mg/L for $\Delta\text{PO}_4\text{-P}$. For the time of operation, however,

Table 3 | Variables and responses, in mg/L, on which the models were based, for case 1

Exp	Variables			Responses					
	Fe	NaAc	Q	Operational time	SS	Total P	PO ₄ -P	Total P – PO ₄ -P	NO ₃ -N
1	–	–	–	175.2	4.6	0.23	0.10	0.13	8.8
2	+	–	–	41.8	3.3	0.07	0	0.07	8.2
3	–	+	–	61.4	4.7	0.14	0.03	0.11	3.0
4	+	+	–	30.8	3.3	0.02	0	0.02	4.9
5	–	–	+	104.9	1.9	0.17	0.11	0.06	8.5
6	+	–	+	31.3	5.1	0.14	0.02	0.12	8.5
7	–	+	+	28.3	5.6	0.24	0.12	0.12	4.1
8	+	+	+	22.4	6.4	0.13	0	0.13	5.4

Exp	Responses					
	ΔSS	Δtotal P	ΔPO ₄ -P	Δ(total P – PO ₄ -P)	ΔNO ₃ -N	ΔCODf
1	3.5	0.13	0.07	0.06	0.1	0
2	–0.6	0.21	0.17	0.04	0.2	0
3	1.9	0.18	0.10	0.07	4.7	32
4	–0.1	0.24	0.16	0.08	3.5	0
5	2.3	0.02	0.01	0.01	0	0
6	–0.1	0.28	0.26	0.03	0	0
7	0.3	0.19	0.16	0.03	4.9	32
8	7.1	0.16	0.15	0.01	4.7	37

Table 4 | Variables and responses, in mg/L, on which the models were based, for case 2

Exp	Variables			Responses					
	Fe	NaAc	PW	Operational time	SS	Total P	PO ₄ -P	Total P – PO ₄ -P	NO ₃ -N
1	–	–	–	104.9	1.9	0.17	0.11	0.06	8.5
2	+	–	–	31.3	5.1	0.14	0.02	0.12	8.5
3	–	+	–	28.3	5.6	0.24	0.12	0.12	4.1
4	+	+	–	22.4	6.4	0.13	0	0.13	5.4
5	–	–	+	18.2	14	0.70	0.06	0.64	5.8
6	+	–	+	18.4	34	0.60	0.05	0.55	5.8
7	–	+	+	14.1	27	0.67	0.04	0.63	4.3
8	+	+	+	13.7	26	0.62	0.02	0.60	3.5

Exp	Responses					
	ΔSS	Δtotal P	ΔPO ₄ -P	Δ(total P – PO ₄ -P)	ΔNO ₃ -N	ΔCODf
1	2.3	0.02	0.01	0.01	0	0
2	–0.1	0.28	0.26	0.03	0	0
3	0.3	0.19	0.16	0.03	4.9	32
4	7.1	0.16	0.15	0.01	4.7	37
5	13.6	0.36	0.02	0.34	0	0
6	3.0	0.54	0.04	0.50	0	0
7	3.0	0.09	–0.02	0.11	4.3	24
8	1.0	0.26	0.01	0.25	6.0	32

Table 5 | Variables and responses, in mg/L, on which the models were based, for case 3

Exp	Variables			Responses					
	Fe	NaAc	SS	Operational time	SS	Total P	PO ₄ -P	Total P – PO ₄ -P	NO ₃ -N
1	–	–	–	104.9	1.9	0.17	0.11	0.06	8.5
2	+	–	–	31.3	5.1	0.14	0.02	0.12	8.5
3	–	+	–	28.3	5.6	0.24	0.12	0.12	4.1
4	+	+	–	22.4	6.4	0.13	0	0.13	5.4
5	–	–	+	10.1	4.9	0.20	0.05	0.15	8.5
6	+	–	+	4.3	3.2	0.10	0.02	0.08	8.5
7	–	+	+	6.9	8.0	0.09	0	0.09	8.4
8	+	+	+	5.2	6.0	0.05	0	0.05	8.2

Exp	Responses					
	ΔSS	Δtotal P	ΔPO ₄ -P	Δ(total – PO ₄ -P)	ΔNO ₃ -N	ΔCOD _f
1	2.3	0.02	0.01	0.01	0	0
2	–0.1	0.28	0.26	0.03	0	0
3	0.3	0.19	0.16	0.03	4.9	32
4	7.1	0.16	0.15	0.01	4.7	37
5	57	1.84	0.03	1.81	0	0
6	68	2.38	0.24	2.14	0	0
7	52	1.69	0.14	1.55	0.8	32
8	36	1.80	0.10	1.70	2.0	24

Table 6 | Model equations, \hat{y} , for different responses from MLR, case 1

Operational time =	$62.0125 - 30.4375 \cdot \text{Fe} - 26.2875 \cdot \text{NaAc} - 15.2875 \cdot \text{Q} + 21.3125 \cdot \text{Fe} \cdot \text{NaAc} + 10.5625 \cdot \text{Fe} \cdot \text{Q} + 4.9125 \cdot \text{NaAc} \cdot \text{Q} - 4.3875 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
SS =	$4.3625 + 0.1625 \cdot \text{Fe} + 0.6375 \cdot \text{NaAc} + 0.3875 \cdot \text{Q} - 0.3125 \cdot \text{Fe} \cdot \text{NaAc} + 0.8375 \cdot \text{Fe} \cdot \text{Q} + 0.6125 \cdot \text{NaAc} \cdot \text{Q} - 0.2875 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
Total P =	$0.1425 - 0.0525 \cdot \text{Fe} - 0.01 \cdot \text{NaAc} + 0.0275 \cdot \text{Q} - 0.005 \cdot \text{Fe} \cdot \text{NaAc} + 0.0175 \cdot \text{Fe} \cdot \text{Q} + 0.025 \cdot \text{NaAc} \cdot \text{Q} - 0.015 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
PO ₄ -P =	$0.0475 - 0.0425 \cdot \text{Fe} - 0.01 \cdot \text{NaAc} + 0.015 \cdot \text{Q} + 0.005 \cdot \text{Fe} \cdot \text{NaAc} - 0.01 \cdot \text{Fe} \cdot \text{Q} + 0.0075 \cdot \text{NaAc} \cdot \text{Q} - 0.0125 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
Total P – PO ₄ -P =	$0.095 - 0.01 \cdot \text{Fe} + 0 \cdot \text{NaAc} + 0.0125 \cdot \text{Q} - 0.01 \cdot \text{Fe} \cdot \text{NaAc} + 0.0275 \cdot \text{Fe} \cdot \text{Q} + 0.0175 \cdot \text{NaAc} \cdot \text{Q} - 0.0025 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
NO ₃ -N =	$6.425 + 0.325 \cdot \text{Fe} - 2.075 \cdot \text{NaAc} + 0.2 \cdot \text{Q} + 0.475 \cdot \text{Fe} \cdot \text{NaAc} + 0 \cdot \text{Fe} \cdot \text{Q} + 0.2 \cdot \text{NaAc} \cdot \text{Q} - 0.15 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
ΔSS =	$1.7875 - 0.2125 \cdot \text{Fe} + 0.5125 \cdot \text{NaAc} + 0.6125 \cdot \text{Q} + 1.4125 \cdot \text{Fe} \cdot \text{NaAc} + 1.3125 \cdot \text{Fe} \cdot \text{Q} + 0.7875 \cdot \text{NaAc} \cdot \text{Q} + 0.8875 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
Δtot P =	$0.1763 + 0.0463 \cdot \text{Fe} + 0.0163 \cdot \text{NaAc} - 0.0138 \cdot \text{Q} - 0.0388 \cdot \text{Fe} \cdot \text{NaAc} + 0.0113 \cdot \text{Fe} \cdot \text{Q} - 0.0038 \cdot \text{NaAc} \cdot \text{Q} - 0.03375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
ΔPO ₄ -P =	$0.135 + 0.05 \cdot \text{Fe} + 0.0075 \cdot \text{NaAc} + 0.01 \cdot \text{Q} - 0.0375 \cdot \text{Fe} \cdot \text{NaAc} + 0.01 \cdot \text{Fe} \cdot \text{Q} + 0.0025 \cdot \text{NaAc} \cdot \text{Q} - 0.0275 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
Δ(total P – PO ₄ -P) =	$0.0413 - 0.0013 \cdot \text{Fe} + 0.0063 \cdot \text{NaAc} - 0.0213 \cdot \text{Q} - 0.0013 \cdot \text{Fe} \cdot \text{NaAc} + 0.0013 \cdot \text{Fe} \cdot \text{Q} - 0.0063 \cdot \text{NaAc} \cdot \text{Q} - 0.00875 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
ΔNO ₃ -N =	$2.2625 - 0.1625 \cdot \text{Fe} + 2.1875 \cdot \text{NaAc} + 0.1375 \cdot \text{Q} - 0.1875 \cdot \text{Fe} \cdot \text{NaAc} + 0.1125 \cdot \text{Fe} \cdot \text{Q} + 0.2125 \cdot \text{NaAc} \cdot \text{Q} + 0.1375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$
ΔCOD _f =	$12.625 - 3.375 \cdot \text{Fe} + 12.625 \cdot \text{NaAc} + 4.625 \cdot \text{Q} - 3.375 \cdot \text{Fe} \cdot \text{NaAc} + 4.625 \cdot \text{Fe} \cdot \text{Q} + 4.625 \cdot \text{NaAc} \cdot \text{Q} + 4.625 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{Q}$

(Fe = X_{Fe}, NaAc = X_{NaAc}, Q = X_Q).

Table 7 | Model equations, \hat{y} , for different responses from MLR, case 2

Operational time =	$31.4125 - 9.9625 \cdot \text{Fe} - 11.7875 \cdot \text{NaAc} - 15.3125 \cdot \text{PW} + 8.3875 \cdot \text{Fe} \cdot \text{NaAc} + 9.9125 \cdot \text{Fe} \cdot \text{PW} + 9.5875 \cdot \text{NaAc} \cdot \text{PW} - 8.5375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
SS =	$15.05 + 2.825 \cdot \text{Fe} + 1.2 \cdot \text{NaAc} + 10.3 \cdot \text{PW} - 2.875 \cdot \text{Fe} \cdot \text{NaAc} + 1.825 \cdot \text{Fe} \cdot \text{PW} - 0.05 \cdot \text{NaAc} \cdot \text{PW} - 2.275 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
Total P =	$0.40875 - 0.03625 \cdot \text{Fe} + 0.00625 \cdot \text{NaAc} + 0.23875 \cdot \text{PW} - 0.00375 \cdot \text{Fe} \cdot \text{NaAc} - 0.00125 \cdot \text{Fe} \cdot \text{PW} - 0.00875 \cdot \text{NaAc} \cdot \text{PW} + 0.01625 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
PO ₄ -P =	$0.0525 - 0.03 \cdot \text{Fe} - 0.0075 \cdot \text{NaAc} - 0.01 \cdot \text{PW} - 0.005 \cdot \text{Fe} \cdot \text{NaAc} + 0.0225 \cdot \text{Fe} \cdot \text{PW} - 0.005 \cdot \text{NaAc} \cdot \text{PW} + 0.0025 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
Total P-PO ₄ -P =	$0.35625 - 0.00625 \cdot \text{Fe} + 0.01375 \cdot \text{NaAc} + 0.24875 \cdot \text{PW} + 0.00125 \cdot \text{Fe} \cdot \text{NaAc} - 0.02375 \cdot \text{Fe} \cdot \text{PW} - 0.00375 \cdot \text{NaAc} \cdot \text{PW} + 0.01375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
NO ₃ -N =	$5.7375 + 0.0625 \cdot \text{Fe} - 1.4125 \cdot \text{NaAc} - 0.8875 \cdot \text{PW} + 0.0625 \cdot \text{Fe} \cdot \text{NaAc} - 0.2625 \cdot \text{Fe} \cdot \text{PW} + 0.4625 \cdot \text{NaAc} \cdot \text{PW} - 0.2625 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
ΔSS =	$3.775 - 1.025 \cdot \text{Fe} - 0.925 \cdot \text{NaAc} + 1.375 \cdot \text{PW} + 2.225 \cdot \text{Fe} \cdot \text{NaAc} - 2.125 \cdot \text{Fe} \cdot \text{PW} - 2.225 \cdot \text{NaAc} \cdot \text{PW} - 0.075 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
Δtotal P =	$0.2375 + 0.0725 \cdot \text{Fe} - 0.0625 \cdot \text{NaAc} + 0.075 \cdot \text{PW} - 0.0375 \cdot \text{Fe} \cdot \text{NaAc} + 0.015 \cdot \text{Fe} \cdot \text{PW} - 0.075 \cdot \text{NaAc} \cdot \text{PW} + 0.035 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
ΔPO ₄ -P =	$0.07875 + 0.03625 \cdot \text{Fe} - 0.00375 \cdot \text{NaAc} - 0.06625 \cdot \text{PW} - 0.03125 \cdot \text{Fe} \cdot \text{NaAc} - 0.02375 \cdot \text{Fe} \cdot \text{PW} - 0.01375 \cdot \text{NaAc} \cdot \text{PW} + 0.03375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
Δ(total P - PO ₄ -P) =	$0.16 + 0.0375 \cdot \text{Fe} - 0.06 \cdot \text{NaAc} + 0.14 \cdot \text{PW} - 0.0075 \cdot \text{Fe} \cdot \text{NaAc} + 0.0375 \cdot \text{Fe} \cdot \text{PW} - 0.06 \cdot \text{NaAc} \cdot \text{PW} + 0.0025 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
ΔNO ₃ -N =	$2.4875 + 0.1875 \cdot \text{Fe} + 2.4875 \cdot \text{NaAc} + 0.0875 \cdot \text{PW} + 0.1875 \cdot \text{Fe} \cdot \text{NaAc} + 0.2375 \cdot \text{Fe} \cdot \text{PW} + 0.0875 \cdot \text{NaAc} \cdot \text{PW} + 0.2375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$
ΔCOD _f =	$15.625 + 1.625 \cdot \text{Fe} + 15.625 \cdot \text{NaAc} - 1.625 \cdot \text{PW} + 1.625 \cdot \text{Fe} \cdot \text{NaAc} + 0.375 \cdot \text{Fe} \cdot \text{PW} - 1.625 \cdot \text{NaAc} \cdot \text{PW} + 0.375 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{PW}$

(Fe = x_{Fe} , NaAc = x_{NaAc} , PW = x_{PW}).**Table 8** | Model equations, \hat{y} , for different responses from MLR, case 3

Operational time =	$26.675 - 10.875 \cdot \text{Fe} - 10.975 \cdot \text{NaAc} - 20.05 \cdot \text{SS} + 8.975 \cdot \text{Fe} \cdot \text{NaAc} + 9 \cdot \text{Fe} \cdot \text{SS} + 10.4 \cdot \text{NaAc} \cdot \text{SS} - 7.95 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
SS =	$5.1375 + 0.0375 \cdot \text{Fe} + 1.3625 \cdot \text{NaAc} + 0.3875 \cdot \text{SS} - 0.3375 \cdot \text{Fe} \cdot \text{NaAc} - 0.9625 \cdot \text{Fe} \cdot \text{SS} + 0.1125 \cdot \text{NaAc} \cdot \text{SS} + 0.2625 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
Total P =	$0.14 - 0.035 \cdot \text{Fe} - 0.0125 \cdot \text{NaAc} - 0.03 \cdot \text{SS} - 0.0025 \cdot \text{Fe} \cdot \text{NaAc} + 0.0 \cdot \text{Fe} \cdot \text{SS} - 0.0275 \cdot \text{NaAc} \cdot \text{SS} + 0.0175 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
PO ₄ -P =	$0.04 - 0.03 \cdot \text{Fe} - 0.01 \cdot \text{NaAc} - 0.0225 \cdot \text{SS} + 0.0 \cdot \text{Fe} \cdot \text{NaAc} + 0.0225 \cdot \text{Fe} \cdot \text{SS} - 0.0075 \cdot \text{NaAc} \cdot \text{SS} + 0.0075 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
Total P-PO ₄ -P =	$0.1 - 0.005 \cdot \text{Fe} - 0.0025 \cdot \text{NaAc} - 0.0075 \cdot \text{SS} - 0.0025 \cdot \text{Fe} \cdot \text{NaAc} - 0.0225 \cdot \text{Fe} \cdot \text{SS} - 0.02 \cdot \text{NaAc} \cdot \text{SS} + 0.01 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
NO ₃ -N =	$7.5125 + 0.1375 \cdot \text{Fe} - 0.9875 \cdot \text{NaAc} + 0.8875 \cdot \text{SS} + 0.1375 \cdot \text{Fe} \cdot \text{NaAc} - 0.1875 \cdot \text{Fe} \cdot \text{SS} + 0.8875 \cdot \text{NaAc} \cdot \text{SS} - 0.1875 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
ΔSS =	$27.825 - 0.05 \cdot \text{Fe} - 3.975 \cdot \text{NaAc} + 25.425 \cdot \text{SS} - 2.25 \cdot \text{Fe} \cdot \text{NaAc} - 1.15 \cdot \text{Fe} \cdot \text{SS} - 5.275 \cdot \text{NaAc} \cdot \text{SS} - 4.55 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
Δtotal P =	$1.045 + 0.11 \cdot \text{Fe} - 0.085 \cdot \text{NaAc} + 0.8825 \cdot \text{SS} - 0.09 \cdot \text{Fe} \cdot \text{NaAc} + 0.0525 \cdot \text{Fe} \cdot \text{SS} - 0.0975 \cdot \text{NaAc} \cdot \text{SS} - 0.0175 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
ΔPO ₄ -P =	$0.13625 + 0.05125 \cdot \text{Fe} + 0.00125 \cdot \text{NaAc} - 0.00875 \cdot \text{SS} - 0.06375 \cdot \text{Fe} \cdot \text{NaAc} - 0.00875 \cdot \text{Fe} \cdot \text{SS} - 0.00875 \cdot \text{NaAc} \cdot \text{SS} + 0.00125 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
Δ(total P - PO ₄ -P) =	$0.91 + 0.06 \cdot \text{Fe} - 0.0875 \cdot \text{NaAc} + 0.89 \cdot \text{SS} - 0.0275 \cdot \text{Fe} \cdot \text{NaAc} + 0.06 \cdot \text{Fe} \cdot \text{SS} - 0.0875 \cdot \text{NaAc} \cdot \text{SS} - 0.0175 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
ΔNO ₃ -N =	$1.55 + 0.125 \cdot \text{Fe} + 1.55 \cdot \text{NaAc} - 0.85 \cdot \text{SS} + 0.125 \cdot \text{Fe} \cdot \text{NaAc} + 0.175 \cdot \text{Fe} \cdot \text{SS} - 0.85 \cdot \text{NaAc} \cdot \text{SS} + 0.175 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$
ΔCOD _f =	$15.625 - 0.375 \cdot \text{Fe} + 15.625 \cdot \text{NaAc} - 1.625 \cdot \text{SS} - 0.375 \cdot \text{Fe} \cdot \text{NaAc} - 1.625 \cdot \text{Fe} \cdot \text{SS} - 1.625 \cdot \text{NaAc} \cdot \text{SS} - 1.625 \cdot \text{Fe} \cdot \text{NaAc} \cdot \text{SS}$

(Fe = x_{Fe} , NaAc = x_{NaAc} , SS = x_{SS}).

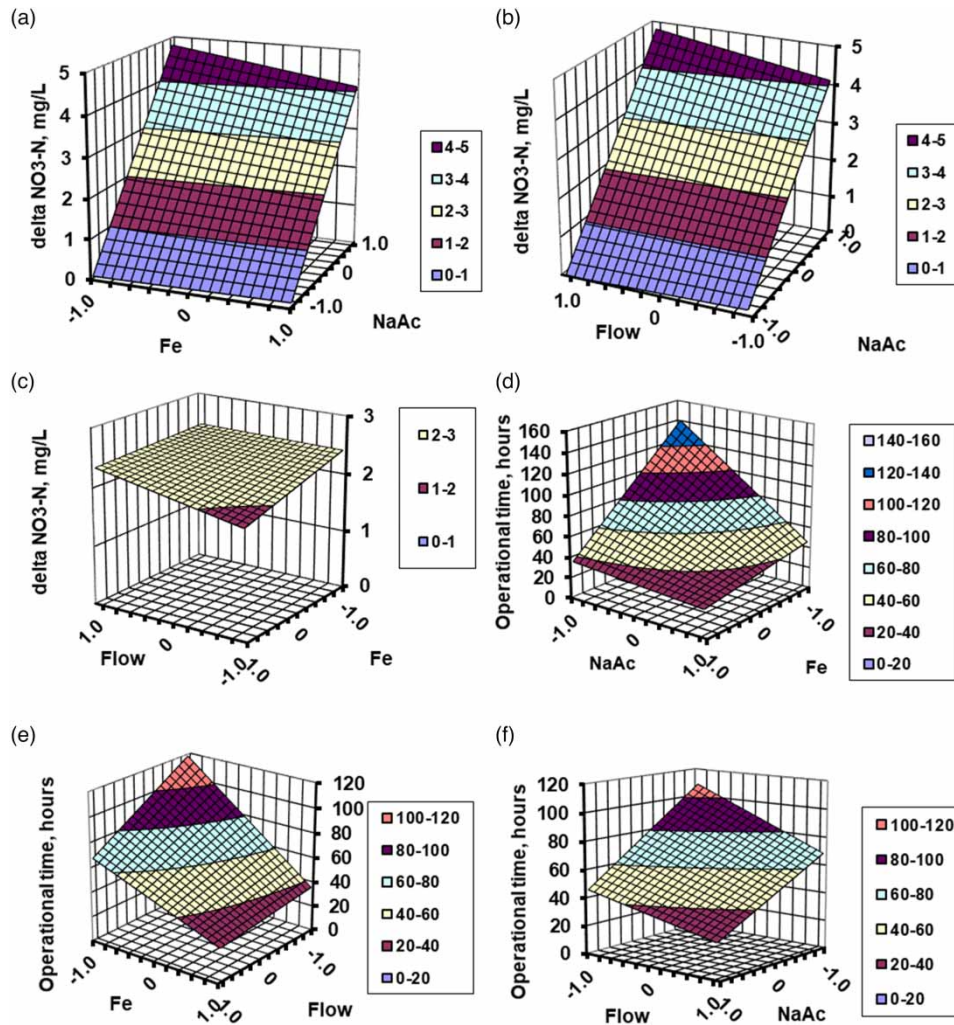


Figure 2 | Reduction of NO₃-N in the filter in mg N/L and time of operation in hours for different variables for case 1.

the fit is less precise. The difference between \hat{y} and y is significant for most of the examples. For examples 1, 2, and 3, the difference is only little more than 10 h, which is interpreted as not too large, but in other cases, the difference is higher. Furthermore \hat{y}_5 is 4.3 h for the time of operation. This was the case with the addition of SS, which often had times of operation of 4–10 h. \hat{y}_5 of 4.3 h and y_5 of 8.9 h both lay within this range, but y_5 was, however, twice as long as \hat{y}_5 (Table 9).

The model response values, \hat{y} , corresponded totally with the experimental values from which the models were calculated, y , for variable values -1 or $+1$, i.e. the lowest and highest values of x in the studied interval. From that fact, the result of the experiments was given in Tables 3–5. No maxima and minima could be found between the values -1 and $+1$ of the variables as the quadratic terms were absent in the equations of \hat{y} . In Figures 2–4, response surface diagrams are presented, showing examples of the responses, \hat{y} , calculated in the study. These diagrams are very illuminating showing clearly the results from the MLR calculations. The equations of the models for all responses are presented in Tables 6–8. The response surface diagrams are represented as functions of the variables in three combinations, i.e. the dosage of Fe and NaAc, the dosage of Fe and Q, PW, or SS, and the dosage of NaAc and Q, PW, or SS, respectively, with the third variable not shown in the diagrams chosen to zero, i.e. in the middle of the interval between -1 and $+1$. Note that the maximum values of the y -axis in the three diagrams can be different. In three different cases, 12 responses were calculated from three variables. In most calculations, a variable gave either a positive or a negative response regardless of the other two variables, but sometimes an increase changed to a decrease, and vice versa, if one or both of the other variables were changed. This was shown in the response surface diagram as a ‘saddle effect’, e.g. at no dosage of Fe; both low flow and no dosage of NaAc favour the SS reduction, but when Fe was dosed, both high flow and NaAc dosage gave a higher

Table 9 | Validation of the models

Response	\hat{y}_1	y_1	\hat{y}_2	y_2	\hat{y}_3	y_3
Time, h	28.3	40.2	25.2	36.2	41.8	53.1
SS	5.6	5.0	6.0	4.7	3.3	8.5
Total P	0.24	0.17	0.18	0.16	0.07	0.13
PO ₄ -P	0.12	0.05	0.06	0.02	0	0.02
Total P – PO ₄ -P	0.12	0.13	0.13	0.14	0.07	0.11
NO ₃ -N	4.1	5.2	4.8	10.0	8.2	8.5
Δ SS	0.3	0	3.8	1.1	0	0
Δ total P	0.19	0.18	0.17	0.15	0.21	0.24
Δ PO ₄ -P	0.16	0.19	0.15	0.15	0.17	0.22
Δ (total P – PO ₄ -P)	0.03	0	0.02	0	0.04	0.02
Δ NO ₃ -N	4.9	4.7	4.8	0	0.2	0
Δ CODf	32	33	35	32	0	0
Response	\hat{y}_4	y_4	\hat{y}_5	y_5	\hat{y}_6	y_6
Time, h	31.3	48.3	4.3	8.9	18.4	9.5
SS	5.1	7.1	3.2	7.0	34	16
Total P	0.14	0.23	0.10	0.29	0.60	0.26
PO ₄ -P	0.02	0.06	0.02	0.02	0.05	0.01
Total P – PO ₄ -P	0.12	0.22	0.08	0.27	0.55	0.25
NO ₃ -N	8.5	8.5	8.5	8.5	5.8	5.8
Δ SS	0	0	68.1	57.8	3.0	7.0
Δ total P	0.28	0.20	2.38	2.07	0.54	0.67
Δ PO ₄ -P	0.26	0.26	0.24	0.24	0.04	0.15
Δ (total P – PO ₄ -P)	0.03	0	2.14	1.85	0.50	0.52
Δ NO ₃ -N	0	0	0	0	0	0
Δ CODf	0	0	0	0	0	0

The model responses, \hat{y} , were compared with responses, y , from experiments not included in the calculation of the models, in mg/L. Time in hours.

reduction of SS. This might be a result of relatively high values of $b_{Fe NaAc}$ and $b_{Fe Q}$ compared with the values for b_{Fe} , b_{NaAc} , and b_Q . In Figure 4, a ‘saddle’ can be seen for the concentration of SS in the filtrate, in case 3, as a function of Fe dosage and SS addition.

The reduction of NO₃-N in the filter is one of the most interesting responses. A biofilm with microorganisms was growing on the surface of the ceramic grains in the filter bed. In this biofilm, denitrification, i.e. reduction of nitrate, occurs if there is enough carbon source in the influent to the filter. Both an external carbon source, e.g. sodium acetate, and an internal carbon source, e.g. PW, to a much lower extent, gave denitrification. A long-time of operation before clogging is essential for the performance of the filter. At short operational times, the filters are waiting inactive in a queue for backwashing while the other filters become overloaded. When part of PW containing high concentrations of phosphorus enters the filters after bypassing the biological treatment, the load of phosphorus to the filters increases, and the concentration of PO₄-P in the filtrate is thereby of great interest. When there is a sludge escape in the secondary sedimentation tanks, high concentrations of biological SS are entering the filters. If the load of SS is too high for the filter, the SS concentration in the filtrate might increase because of filter break-through. This value is thereby of great interest. More probable is that the filter clogs, giving short times of operation. These six responses were, therefore, chosen as examples to be presented in the response surface diagrams. El Mouhri *et al.* (2021) studied tannery wastewater, although not with MLR, showing response surface diagrams to illustrate the results. Casella (2019) worked with the combination of MLR and multiple mass probability. In MLR, several variables were used, but quadratic and interaction terms as parameters for the variables were excluded. Leverenz *et al.* (2009) studied the parameters influent COD concentration, dosing frequency in an intermittently dosed sand filter, time of operation, i.e. clogging, and the hydraulic loading rate to assess the

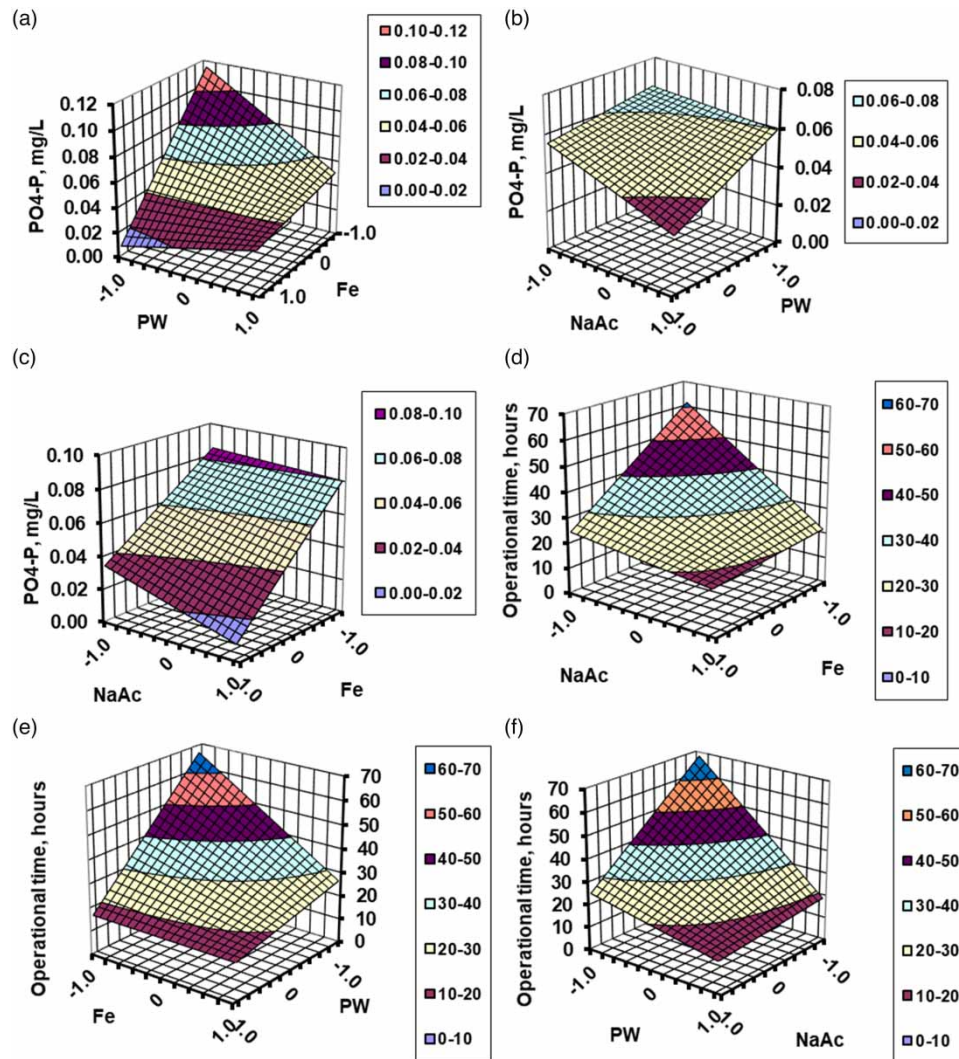


Figure 3 | PO₄-P in the filtrate in mg P/L and time of operation in hours for different variables for case 2.

importance of the parameters on the growth of heterotrophic bacteria by using MLR analysis. The resulting equation contained multiplied variables with exponents of power different from 1. In the Supplementary Material, the remaining response surface diagrams from our study are presented in Figures S1, S2, and S3. The concentrations of the substances in the influent to the filters are shown in Supplementary Tables S1, S2, and S3.

MLR was used to calculate empirical relationships with equations predicting different responses, \hat{y} , from the variables, x . Interaction terms were included but not quadratic terms. The exponents of power were 1. The results from the modelling were evaluated by comparing the real analysed results, y , with the calculated values of the responses, \hat{y} , when x_i is -1 or $+1$ (Tables 3–5) and the coefficients of the variables, b_i , in the model equations (Tables 6–8). Interaction terms also affected the responses, but a specific variable was more difficult to differentiate in these terms. Most illuminating in interpreting the results was to study the three-dimensional response surface diagrams (Figures 2–4).

Relations between the flow rate and surface loading

The flow rate had a relatively large impact on the time of operation but generally had a minor influence on the concentration of the substances reduced in the filter or on the concentration of the substances in the filtrate except for $\Delta(\text{total P minus PO}_4\text{-P})$ where a lower flow gave an obvious higher reduction with $b_Q = -0.0213$ in case 1. Ncube *et al.* (2016) studied SS removal in a quadruple media filter at hydraulic rates between 5 and 60 m/h, compared to 5 and 10 m/h in our study. They found that higher rates have less impact on the

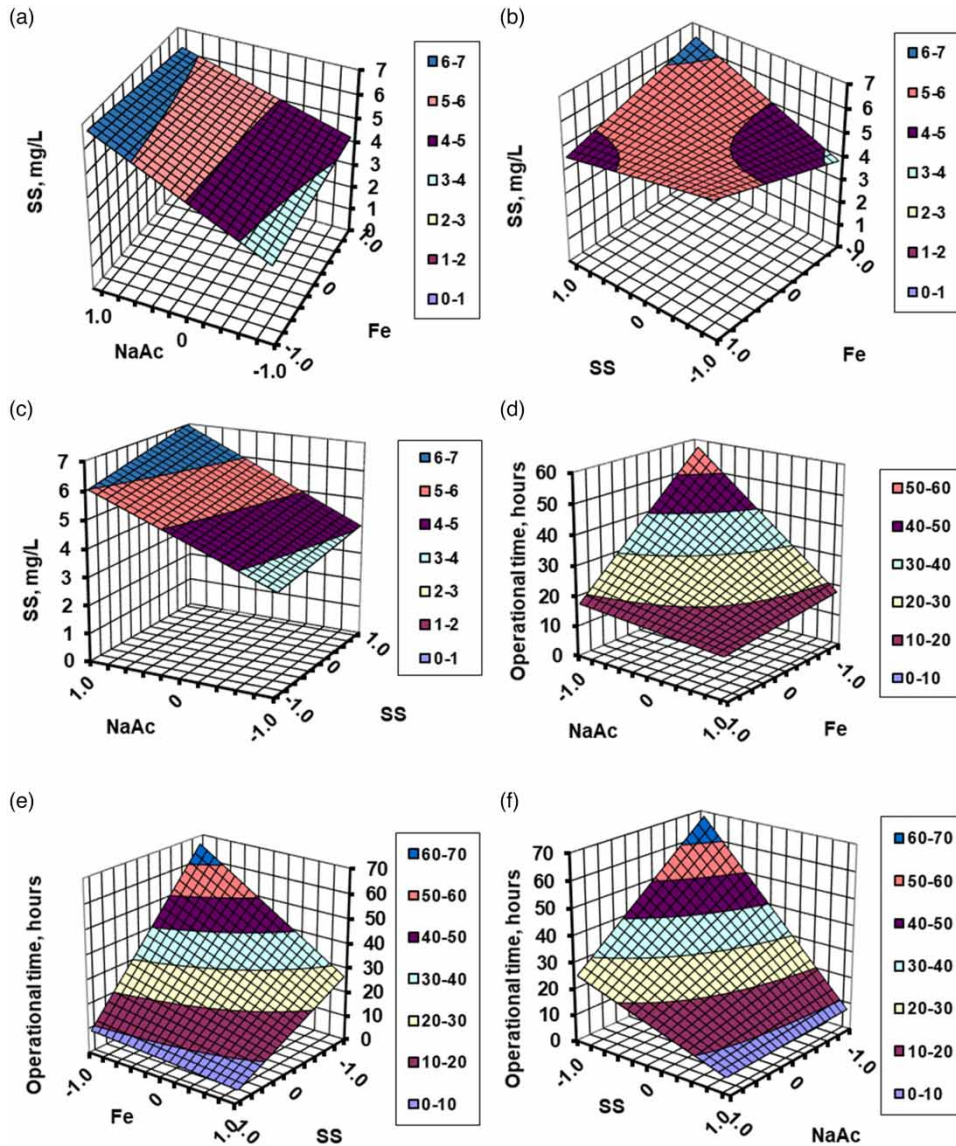


Figure 4 | SS in the filtrate in mg SS/L and time of operation in hours for different variables for case 3.

performance of the multimedia filter than for a single media filter. SS could be transported deeper into the filter and be stored there, probably also to some extent on the surface of every media layer, ensuring that the head loss was distributed to all media layers. In our study, the head loss could be distributed between two layers. The time of operation before clogging, however, was close to reciprocally proportional to the flow when the concentration of SS in the influent and other conditions were the same. Else, it was logarithmically proportional to the flow. Higher concentrations of SS in the influent gave a possibility for the filter to remove more SS in the filter, but also a very small tendency of filter break-through might increase the concentration of SS in the filtrate. High surface loadings in the filter were not considered to be a problem for the concentration of the substances in the filtrate, but high flows also containing a high influent concentration of SS could give very short times of operation causing problems in the WWTP.

Removal of suspended solids

When extra SS were added to the filter, by the addition of primary settled wastewater, there were more SS that could be separated in the filter by filtration. The concentration of SS in the filtrate, however, increased to a very small extent by 0.7 mg SS/L or 16%. NaAc dosage resulted in the production of SS in the filter during

denitrification. Part of this SS might pass through the filter due to smaller flocs increasing the concentration of SS in the filtrate.

The concentration of SS in the filtrate with the addition of PW was 14–34 mg SS/L (influent 28–37 mg SS/L) and 3.2–8.0 mg SS/L with the addition of SS (influent 42–71 mg SS/L), compared with 1.9–6.4 mg SS/L without addition (influent 4.2–13.5 mg SS/L). This suggests that SS in PW, with a smaller floc size, is more difficult to separate in the filter than SS with a larger floc size originating from the aeration tank. This is even more obvious when the results from Δ SS in the filter gave values of 1.0–13.6 mg/L with PW addition and 36–68 mg/L with SS addition compared with no addition, which gave 0.0–7.1 mg SS/L. This shows that the filter removes more flocs from SS than from PW. The influent contained, however, a higher SS concentration when SS were added compared to PW. The low values from wastewater without PW or SS addition were a result of a low influent concentration of SS not giving the filter enough possibility to remove and accumulate high amounts of SS from the influent to the filter.

The somewhat higher Δ SS at higher hydraulic velocities ($b_Q = +0.6125$ in case 1) was probably a result of higher amounts of SS in the influent to the filter, either with the same SS concentration in the influent and a higher flow or, as an alternative, a higher flow that induces sludge escape from the secondary sedimentation tanks giving higher concentrations of SS in the influent to the filter. The latter was not seen here. Both alternatives give a higher amount of SS that could be separated in the filter. PW addition gave an increased SS concentration in the filtrate, but SS addition gave SS concentrations in the filtrate not much different from these analysed in the filtrate from secondary settled wastewater without any addition to the influent of the filter. This suggests a small filter break-through for the smaller PW flocs but not for the larger SS flocs.

The concentration of SS in the filtrate increases, however to a small extent, when NaAc is dosed compared to no NaAc dosage, which might be SS originating from denitrification in the filter. For the concentration of SS in the filtrate in case 1, $b_Q = +0.3875$, in case 2 $b_{PW} = +10.3$, and in case 3 $b_{SS} = +0.3875$. This shows that a higher hydraulic load and the addition of biological SS had a small increase of SS in the filtrate, but the increase was much larger when PW was added. For Δ SS in case 1, $b_Q = +0.6125$, in case 2 $b_{PW} = +1.375$, and in case 3 $b_{SS} = +25.425$. An increase in the reduction of SS occurred in all three cases, but the largest increase was detected for case 3 with SS addition. The conclusion is that the filter removes large amounts of biological SS, but it is suspended solids originating from PW that pass the filter.

Several studies of the removal of SS by wastewater filtration in down-flow filters have been reported. The tests cover a wide range of SS in the filter feed but also the applied hydraulic load (Table 10). Kobler & Boller (1997) compared SS particle removal in different types of filters. Deep-bed filters had the highest removal efficiency for total SS. No filter break-through was found for SS particles $>1 \mu\text{m}$ in size for one plant, while another plant had no particle break-through for SS particles $>10 \mu\text{m}$. Shandalov *et al.* (1997) studied filtration in a pilot plant deep-bed one-media sand filter with the grain size of $\text{Ø}1.4\text{--}2.0 \text{ mm}$ giving 90% reduction of SS. They also made a mathematical model from the result of the SS analyses using numerical solutions. Kaminski *et al.* (1997) performed a laboratory study in a sand filter with a grain size of $\text{Ø}1.1 \text{ mm}$ where the SS reduction became 47–75% depending on the hydraulic loads of 20 and 5 m/h, respectively. Sauter *et al.* (2021) studied filtration in a pilot plant deep-bed dual media sand filter with the addition of FeCl_3 at hydraulic loads of 5–10 m/h and inlet concentrations of $7.0 \pm 4.2 \text{ mg SS/L}$. The lower sand bed had a height of 0.6 m and the upper filter bed consisted of 1.2 m of either anthracite or granular activated carbon (GAC), which gave a concentration of $<1 \text{ mg SS/L}$ in the filtrate. Altmann *et al.* (2015) performed a study in a pilot-scale deep-bed dual media sand filter at an influent of 7.5 m/h and FeCl_3 dosage. The filter bed had a sand layer of $\text{Ø}0.7\text{--}1.25$ and an upper anthracite layer of $\text{Ø}1.4\text{--}2.5 \text{ mm}$. The concentration of SS in the influent was 3.8–5.1 mg SS/L and that in the filtrate was 0.1–0.4 mg SS/L. Sperlich *et al.* (2013) studied a pilot-scale deep-bed dual media sand filter with 0.6 m sand and a 1.2 m upper anthracite layer at a hydraulic load of 6 m/h and influent concentrations of 1.8–30 mg SS/L. The concentration of SS in the filtrate became $\leq 2 \text{ mg SS/L}$ for all investigated influent SS concentrations. Ben Aim *et al.* (1997) performed a laboratory study in a filter filled with 0.09 m polystyrene latex particles. The influent load was 5 m/h and had a concentration of 10 mg SS/L. The reduction of SS became approximately 65%. Healy *et al.* (2007) studied a pilot-scale denitrifying sand filter at a very low hydraulic loading rate of 0.0004 m/h and a concentration of 370 mg SS/L in the influent. The reduction of both SS and COD was $>99\%$. Jeong *et al.* (2019) performed a laboratory-scale study in one-media sand filter at hydraulic loads of 7.8–54.6 m/h. The concentration of SS was reduced from 18.5 to 5.7 mg SS/L in the filter.

Table 10 | Suspended solids separation in down-flow sand filters

Upper layer grain size (mm)	Upper layer bed height (m)	Lower layer grain size (mm)	Lower layer bed height (m)	Hydraulic load (m/h)	Suspended solids in filter feed (mg/L)	Suspended solids in filtrate (mg/L)	Reference
2.5–4.0	1.2	0.8–1.2	0.3	5	2.7–8.1	3.3–4.7	Present study
				10	4.2–13.5	1.9–6.4	
				10	27–37	14–34	
				10	42–71	3.2–8.0	
One media		1.4–2.0	1.15	14.9	38	3.8	Shandalov <i>et al.</i> (1997)
One media		1.1	0.20	5	10–60	2.5–15	Kaminski <i>et al.</i> (1997)
				10	10–60	5.3–32	
1.4–2.5	1.2	0.7–1.25	0.6	5–10	7.0	< 1	Sauter <i>et al.</i> (2021)
1.4–2.4	1.2						
1.4–2.5	1.2	0.7–1.25	0.6	7.5	3.8–5.1	0.1–0.4	Altmann <i>et al.</i> (2015)
2.5–4.0	1.2	0.71–1.25	0.6	6	1.8–30	≤ 2	Sperlich <i>et al.</i> (2013)
1.4–2.5	1.2						
One media		0.450	0.09	5	10	6.5	Ben Aim <i>et al.</i> (1997)
0.45	0.25	0.11	0.30	0.00042	370	<3.7	Healy <i>et al.</i> (2007)
					90	≈0	
One media		0.521–1.28	0.4	7.8–54.6	18.5	5.7	Jeong <i>et al.</i> (2019)

Pilot scale and laboratory scale.

Denitrification

As expected, NaAc acted as an energy and carbon source for the heterotrophic bacteria responsible for denitrification and thereby improved the denitrification simultaneously as $\text{PO}_4\text{-P}$ was assimilated into the denitrifying sludge while both $\Delta\text{NO}_3\text{-N}$ and ΔCODf increased. For $\Delta\text{NO}_3\text{-N}$ in case 1, $b_{\text{Fe}} = -0.1625$, $b_{\text{NaAc}} = +2.1875$, and $b_{\text{Q}} = +0.1375$, showing dominance of the NaAc dosage. It also showed that the Fe dosage decreased $\Delta\text{NO}_3\text{-N}$, which probably was a result of the precipitation of $\text{PO}_4\text{-P}$ by Fe. This might give a rate-limiting shortage of $\text{PO}_4\text{-P}$ needed for the assimilation into the denitrifying sludge. Dalahmeh *et al.* (2014) also mentioned the assimilation of phosphorus into the microbial biomass in addition, in their case, to adsorption. The influence of the flow was small as a higher flow only increased $\Delta\text{NO}_3\text{-N}$ to a very small extent.

Primary settled wastewater, PW, also acted as a carbon source containing organic material although to a much lower degree than NaAc, due to the slow degradation rate of most of the organic material in the PW. For $\Delta\text{NO}_3\text{-N}$ in case 2, $b_{\text{NaAc}} = +2.4875$ and $b_{\text{PW}} = +0.0875$. The reduction of $\text{NO}_3\text{-N}$ and soluble COD was mainly zero without the dosage of NaAc, showing the very dominating influence as, for the bacteria, an easily degradable carbon source. For $\text{NO}_3\text{-N}$ concentration in the filtrate in case 2, $b_{\text{NaAc}} = -1.4125$ and $b_{\text{PW}} = -0.8875$. The added biological particles in case 3 analysed as suspended solids, SS, did not act as a carbon source and were not increasing $\Delta\text{NO}_3\text{-N}$ or ΔCODf . For $\Delta\text{NO}_3\text{-N}$ in case 3, $b_{\text{NaAc}} = +1.55$, $b_{\text{SS}} = -0.85$, and $b_{\text{Fe}} = +0.125$. For ΔCODf in case 3, $b_{\text{NaAc}} = +15.625$, $b_{\text{SS}} = -1.625$, and $b_{\text{Fe}} = -0.375$. As can be seen from b_{Fe} , the dosage of Fe increased to a small extent $\Delta\text{NO}_3\text{-N}$ but decreased ΔCODf .

Operational time between backwashings

The time of operation between backwashings is of vital importance for the performance of the filter. In case 1, $b_{\text{Fe}} = -30.4375$, $b_{\text{NaAc}} = -26.2875$, and $b_{\text{Q}} = -15.2875$, showing that the dosage of iron decreased the operational time most, followed by the dosage of NaAc, and last by increasing the flow through the filter. In case 2, $b_{\text{Fe}} = -9.9625$, $b_{\text{NaAc}} = -11.7875$, and $b_{\text{PW}} = -15.3125$, showing that the time of operation was mostly influenced by the addition of PW giving a decrease in operational time followed by the dosage of NaAc and the dosage of Fe, which gave the smallest shortening of the operational time. In case 3, $b_{\text{Fe}} = -10.875$, $b_{\text{NaAc}} = -10.975$, and $b_{\text{SS}} = -20.05$. This shows that the dosage of Fe and NaAc gave approximately the same decrease

in the time of operation, while the addition of SS decreased the time twice as much. The time of operation in the study had a maximum value of 175 h when no dosage of Fe or NaAc was done while the surface loading was kept to 5 m/h. Al-Jadhai (2003) performed a study with sand of $\text{Ø}2.0\text{--}3.36$ mm, which gave a time of operation of 100 h at 8 m/h and 53 h at 18 m/h. In our study, the highest value was 104.9 h at 10 m/h. When Fe was dosed, the operational time decreased to 31.3 h, and when NaAc was dosed, the operational time decreased to 28.3 h. Boller (1984) reports a time of operation of 20 h at 7.4 m/h and 30 h at 10 m/h when Fe was dosed giving an effluent concentration of 0.1–0.2 mg total P/L. The operational time of 30 h is close to 31.3 h in this study. Simultaneous Fe and NaAc dosage in our study shortened the time of operation to 22.4 h. When PW was added, including a dosage of Fe, the longest time of operation was 18.4 h. With a dosage of NaAc, the time of operation decreased to 14.1 h, and with simultaneous Fe and NaAc dosage, it was 13.7 h. When extra SS were added, the operational time decreased to 10.1 h compared to 18.2 h with only PW addition, which shows that SS addition decreased the time of operation much more than PW addition. This is probably a result of the higher concentration of SS in the influent when SS were added compared to PW.

Simultaneous SS addition and Fe dosage gave the shortest operational time of only 4.3 h between backwashings. Radtke & Sturm (2007) performed a study in two-media down-flow filters and found, for the filter with grains 1.2 m $\text{Ø}1.4\text{--}2.5$ mm plus 0.6 m $\text{Ø}0.71\text{--}1.25$ mm at 10 m/h and an influent with 50 mg SS/L, a time of operation of 5.5 h, and with 90 mg SS/L a time of operation less than 2 h. In a filter with grains 1.2 m $\text{Ø}2.0\text{--}4.0$ mm plus 0.6 m $\text{Ø}1.0\text{--}1.6$ mm at 15 m/h and an influent with 50 mg SS/L, the time of operation became 7.0 h. A filtration velocity of 10 m/h with 90 mg SS/L gave a time of operation of 6.0 h but with higher concentrations of SS in the filtrate. This is close to 4.3, 5.2, 6.9, and 10.1 h in our study with high concentrations of SS in the influent. Moran *et al.* (1993a) report a time of operation to filter break-through of 46 h at 6.5 m/h, and 29 and 46 h at 19.8 m/h in a deep-bed one-media down-flow filter. Moran *et al.* (1993b) report at 13.3 m/h a time of operation to filter break-through in two deep-bed two-media down-flow filters of 46.5 and 20.0 h at the influent concentration of 6.3 and 24.2 mg SS/L, respectively.

In a laboratory deep-bed two-media down-flow filter with a hydraulic load of 12.6 m/h studied by Tchobanoglous (1970), a time of operation of 19 h was detected with 22 mg SS/L in the influent and 48 h with 14 mg SS/L. Williams *et al.* (2007) studied tertiary filtration in a pilot-scale two-media rapid deep filter bed with coagulated secondary wastewater as an influent at different loading rates. The time of operation, defined as a head loss of 3.4 m corresponding to the terminal head loss, i.e. clogging, in the full-scale filter, became 22 h at a load of 12.2 m/h, 17 h at 15.3 m/h, nearly 14 h at 18.3 m/h, 11 h at 21.4 m/h, and 10.5 h at 24.4 m/h at a coagulant dose of 12 mg/L. 22 h at 12.2 m/h can be compared with 31.3 h at 10 m/h when ferric chloride was dosed, 28.3 h when NaAc was dosed, and 22.4 h when Fe and NaAc were dosed simultaneously in our study. Al-Saedi *et al.* (2019) studied an up-flow sand filter for microbial clogging. Ethanol dosage to the influent wastewater gave a time of operation of 36% and sucrose dosage gave 66% time of operation compared to only wastewater to the influent. In our study, 35% of the time of operation remained at 5 m/h and 27% at 10 m/h when NaAc was dosed, which is close to 36% with ethanol dosage found by Al-Saedi *et al.* (2019).

Removal of phosphorous compounds

When Fe was dosed, $\Delta\text{PO}_4\text{-P}$ increased, which was expected, as Fe acted as a precipitation agent for $\text{PO}_4\text{-P}$. When NaAc was dosed, $\Delta\text{PO}_4\text{-P}$ also increased, although to a very small extent, probably because of the $\text{PO}_4\text{-P}$ assimilation during denitrification. For $\Delta\text{PO}_4\text{-P}$ in case 1, $b_{\text{Fe}} = +0.05$, $b_{\text{NaAc}} = +0.0075$, and $b_{\text{Q}} = +0.01$, showing a dominant influence from the Fe dosage. The flow had a small impact on $\Delta\text{PO}_4\text{-P}$. PW contains organic compounds which some contain phosphorus. The PW acts as a weak carbon source in the filter, and during denitrification, organic material in the PW breaks down leaving among others free phosphorus as $\text{PO}_4\text{-P}$ increasing the concentration of $\text{PO}_4\text{-P}$ in the filter. This $\text{PO}_4\text{-P}$ can be assimilated into the sludge produced during denitrification or be precipitated when Fe was dosed. When Fe was dosed, $\Delta\text{PO}_4\text{-P}$ was found to be 0.15/0.26 mg/L compared to 0.01/0.04 mg/L by the addition of PW, which is very low, and 0.10/0.24 mg/L by the addition of SS. If only NaAc was dosed, $\Delta\text{PO}_4\text{-P}$ was 0.16 mg/L and that was higher than with PW addition, 0.02 mg/L (0.00 mg/L for simultaneous PW and NaAc addition), and higher than with SS addition, 0.03 mg/L (0.14 mg/L for simultaneous SS and NaAc addition). For $\Delta\text{PO}_4\text{-P}$ in case 2, $b_{\text{Fe}} = +0.03625$, $b_{\text{NaAc}} = -0.00375$, and $b_{\text{PW}} = -0.06625$. For $\Delta\text{PO}_4\text{-P}$ in case 3, $b_{\text{Fe}} = +0.05125$, $b_{\text{NaAc}} = +0.00125$, and $b_{\text{SS}} = -0.00875$.

The concentration of $\text{PO}_4\text{-P}$ in the filtrate was low when Fe was dosed giving values of 0.00/0.02 mg/L for wastewater without addition, 0.02/0.05 mg/L with PW addition, and 0.00/0.02 mg/L with SS addition. Even

without Fe dosage, the concentration of $\text{PO}_4\text{-P}$ was low in the filtrate when PW or SS were added. For $\text{PO}_4\text{-P}$ in the filtrate in case 1, $b_{\text{Fe}} = -0.0425$, $b_{\text{NaAc}} = -0.01$, and $b_{\text{Q}} = +0.015$. For $\text{PO}_4\text{-P}$ in the filtrate in case 2, $b_{\text{Fe}} = -0.03$, $b_{\text{NaAc}} = -0.0075$, and $b_{\text{PW}} = -0.01$. For $\text{PO}_4\text{-P}$ in the filtrate in case 3, $b_{\text{Fe}} = -0.03$, $b_{\text{NaAc}} = -0.01$, and $b_{\text{SS}} = -0.0225$. In all cases, Fe and NaAc decreased the concentration of $\text{PO}_4\text{-P}$ in the filtrate. Both PW and SS addition also decreased the concentration of $\text{PO}_4\text{-P}$ but an increase in the flow increased instead of $\text{PO}_4\text{-P}$ in the filtrate.

SS bound phosphorus, total P minus $\text{PO}_4\text{-P}$, was used to predict the behaviour of SS in the filter as it was approximately proportional to SS. Earlier problems with the SS analysis combined with a very safe analysis of phosphorus made it tempting to use the total P minus $\text{PO}_4\text{-P}$ analyses when evaluating the filter. When SS were added, the concentration of SS in the influent was most often 60–71 mg SS/L, and with PW addition 23–37 mg SS/L. This can be interpreted as that influent SS gives a higher possibility of reduction of SS and thereby of total P and total P minus $\text{PO}_4\text{-P}$ than influent PW containing less SS.

PW contains dissolved organic phosphorous compounds that were not caught by the filter. Some PW also contains suspended bound phosphorus that was caught in the filter increasing both the reduction and the filtrate concentration of total P minus $\text{PO}_4\text{-P}$. When SS contained suspended bound phosphorus, SS addition also increased $\Delta(\text{total P minus } \text{PO}_4\text{-P})$, but the concentration in the filtrate was still low. With simultaneous Fe dosage and PW addition, the concentration of total P in the filtrate was 0.60/0.62 mg/L compared to 0.13/0.14 mg/L without PW addition and 0.05/0.10 mg/L with SS addition, all with simultaneous Fe dosage. It is obvious that PW increases the concentration of total P and thereby total P minus $\text{PO}_4\text{-P}$ in the filtrate considerably, while SS addition does not have that effect probably because of some filter break-through by smaller flocs from PW. For the concentration of total P minus $\text{PO}_4\text{-P}$ in the filtrate in case 2, $b_{\text{PW}} = +0.24875$, and in case 3, $b_{\text{SS}} = -0.0075$. For the concentration of total P in the filtrate in case 2, $b_{\text{PW}} = +0.23875$, and in case 3, $b_{\text{SS}} = -0.03$. This points to a large increase in the concentration when PW was added, but a decrease when SS were added.

Both $\Delta\text{total P}$ and $\Delta(\text{total P minus } \text{PO}_4\text{-P})$ were highly increased by the addition of SS probably also because there was more P as suspended bound P entering the filter that could be reduced when SS were added. $\Delta\text{total P}$ showed an increased value both for Fe dosage, which was expected, and for PW addition. $b_{\text{Fe}} = +0.0725$, $b_{\text{NaAc}} = -0.0625$, and $b_{\text{PW}} = +0.075$ for case 2. With Fe dosage, $\Delta\text{total P}$ became 0.16/0.28 mg/L, with PW addition 0.26/0.54 mg/L, and with SS addition 1.80/2.38 mg/L. $\Delta(\text{total P minus } \text{PO}_4\text{-P})$, also with Fe dosage, became 0.01/0.03 mg/L, with PW addition 0.25/0.50 mg/L, and with SS addition 1.70/2.14 mg/L. This suggests that SS might contain more suspended solids bound phosphorus than PW, but this is contradicted by the analysis, showing that during the period of the study as an average the solid phase of PW contained 3.1% total P minus $\text{PO}_4\text{-P}$. The solid phase of the secondary sedimented water, which probably consists of biological SS, contained an average 3.2% total P minus $\text{PO}_4\text{-P}$ during the same period. $\Delta(\text{total P minus } \text{PO}_4\text{-P})$ at SS addition was higher than at PW addition, but this can be explained by the higher values of ΔSS when biological SS was added compared to PW. The smaller flocs of PW might not be caught by the filter. Part of the bound phosphorus in PW might be dissolved by passing through the filter. The different concentrations of SS in added PW and added SS must also be considered. When only Fe was dosed, most of the $\Delta\text{total P}$ existed as $\text{PO}_4\text{-P}$, which was expected, but when PW or SS were added, most of the reduction was in the SS form as total P minus $\text{PO}_4\text{-P}$. All these results can explain the different values for $\Delta\text{total P}$ and $\Delta(\text{total P minus } \text{PO}_4\text{-P})$ mentioned above.

CONCLUSIONS

MLR was a useful tool to identify relations in a dual media down-flow sand filter between different types of loads, precipitation of phosphate, removal of total phosphorous, suspended solids, and organic material, and denitrification. In the models, interaction terms were included but not quadratic terms.

The time of operation before clogging was close to reciprocally proportional to the flow at the same concentration of SS in the influent and under otherwise similar conditions. Else, the operational time between backwashings was logarithmically proportional to the flow.

Smaller flocs of SS originating from primary settled wastewater were not separated in the filter giving high concentrations in the filtrate of SS and associated nutrients. SS originating from the aeration tank were removed by the filter.

The flow rate had generally a minor influence on the removal and the concentrations of substances in the filtrate except for the reduction of total P minus PO₄-P where a lower flow gave a higher reduction.

PW partly contains dissolved organic phosphorous compounds that pass through the filter and give higher concentrations of phosphorus in the filtrate.

As expected, the removal of NO₃-N and soluble COD was highly increased when NaAc was dosed as well as the dosage of Fe decreased the concentration of PO₄-P in the filtrate.

Primary settled wastewater, PW, containing organic material acted as a carbon source in the denitrification, although to a much lower degree than NaAc. By dosing NaAc, the removal of PO₄-P increased both from the dosage of Fe and, to a small degree, from the assimilation of PO₄-P into the denitrifying sludge.

The dosage of Fe slightly decreases the reduction of NO₃-N probably because of a lack of PO₄-P for the assimilation into the denitrifying sludge since Fe acts as a precipitation agent upon PO₄-P.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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