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Modeling of Pollutants Removal in Subsurface Vertical Flow and Horizontal Flow Constructed Wetlands

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Abstract: Reject water is a by-product of every municipal and agro-industrial wastewater treatment plant (WWTP) applying sewage sludge stabilization. It is usually returned without pre-treatment to the biological part of WWTP, having a negative impact on the nitrogen removal process. The current models of pollutants removal in constructed wetlands concern municipal and industrial wastewater, whereas there is no such model for reject water. In the presented study, the results of treatment of reject water from dairy WWTP in subsurface vertical flow (SS VF) and subsurface horizontal flow (SS HF) beds were presented. During a one-year research period, SS VF bed reached 50.7% efficiency of TN removal and 73.8% of NH_4^+-N, while SS HF bed effectiveness was at 41.4% and 62.0%, respectively. In the case of BOD_5 (biochemical oxygen demand), COD (chemical oxygen demand), NH_4^+-N, and TN (total nitrogen), the P-k-C* model was applied. Multi-model nonlinear segmented regression analysis was performed. Final mathematical models with estimates of parameters determining the treatment effectiveness were obtained. Treatment efficiency increased up to the specific temperature, then it was constant. The results obtained in this work suggest that it may be possible to describe pollutant removal behavior using simplified models. In the case of TP (total phosphorus) removal, distribution tests along with a t-test were performed. All models predict better treatment efficiency in SS VF bed, except for TP.

Keywords: P-k-C* model; subsurface vertical flow constructed wetlands; subsurface horizontal flow constructed wetlands; reject water; treatment modeling; dairy wastewater treatment plant; anaerobic sewage sludge digestion

1. Introduction

In Europe, the biggest municipal and agro-industrial wastewater treatment plants (WWTPs) utilize anaerobic sewage sludge digestion with biogas production [1]. The possibility of heat and electric energy production from biogas is an advantage of such sewage sludge treatment, which decreases the cost of waste treatment significantly.

One of the major problems connected with anaerobic digestion chamber exploitation is reject water, which is generated in the process of dewatering stabilized sewage sludge. It is usually returned without pre-treatment to the biological part of a WWTP, having a negative impact on the treatment process and causing problems with reaching high effectiveness of nitrogen removal [2–4]. Reject water from anaerobic sewage sludge digestion is characterized by a high concentration of nitrogen in the form of ammonium nitrogen (NH_4^+-N) and irregular flow because of periodically working devices for sewage sludge dewatering [5–8]. The problem of reject water treatment in a conventional sludge activated system is caused by low biodegrability, indicated by proportions of easily biodegradable organic matter expressed by biochemical oxygen demand (BOD_5) to total organic matter expressed by chemical oxygen demand (COD).

Constructed wetlands are considered environmentally friendly technology [9–11]. They have been used worldwide for the treatment of municipal and industrial wastewater, as well as reject water from anaerobic sewage sludge digestion in municipal WWTPs and landfill leachate, which is also characterized by high NH_4^+-N concentration [12–15]. Reject water generated during anaerobic digestion in dairy WWTPs differs from that from municipal WWTPs. There is a lack of experiments concerning constructed wetlands treatment of reject water generated during anaerobic digestion in dairy WWTPs [16].

The aim of the research was to define parameters, with their expected values, of the mathematical models describing the pollutants removal in subsurface vertical flow (SS-VF) and subsurface horizontal flow (SS-HF) constructed wetlands treating reject water from anaerobic sludge digestion in dairy WWTPs. The P-k-C* model was applied [12,17,18]. The issue of modeling the functioning of constructed wetlands is one of the most important challenges in that field [19,20]. The current models of pollutant removal concern the municipal and industrial wastewater treatment, whereas there is no such model for reject water treatment, so the necessity for such research goals was confirmed.

2. Materials and Methods

2.1. Study Sites

The research was conducted over a twelve-month period using a pilot-scale plant built in the biggest dairy plant in Poland, which Person Equivalent (PE) is up to 500,000, located in Wysokie Mazowieckie in the Podlaskie province. The average air temperature is 6.8 °C and precipitation is 562 mm year^{-1}.

A problem with high concentration of nutrients in the reject water was observed after changing aerobic sewage sludge stabilization to anaerobic. After sewage sludge dewatering with centrifuge, reject water was returned to the main sewage line without separate treatment. In comparison with reject water obtained during aerobic sewage sludge stabilization, the concentration of ammonium nitrogen increased significantly [2].

The research installation (N 52.54.20-E 22.299.20) was designed by the authors using earlier experience with reject water treatment. Its main elements are SS VF and SS HF beds with 5 m^2 surface area and 0.8 m height (Figure 1). The beds were used in parallel. In addition, the installation includes a sedimentation and retention tank, outflow, and sampling points (I, II, III). Figure 2 presents the cross section of SS VF and SS HF beds.

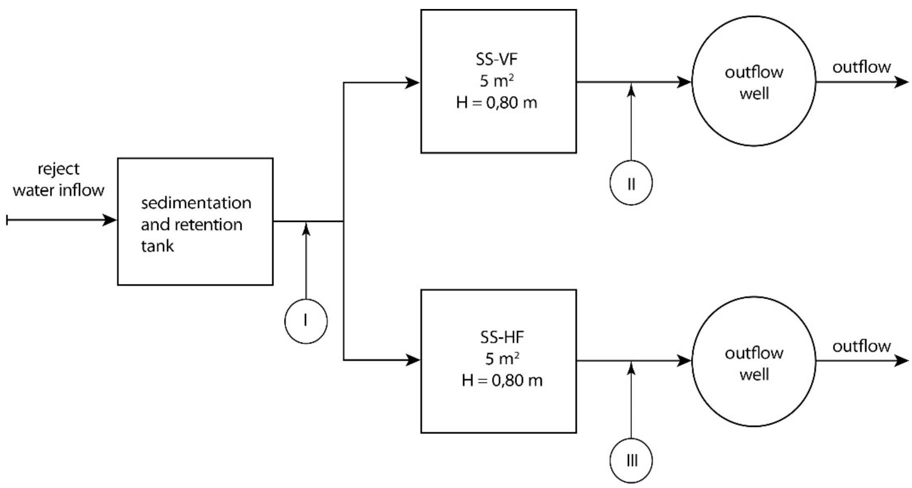

Figure 1. Scheme of research installation, with sampling points (I, II, III).

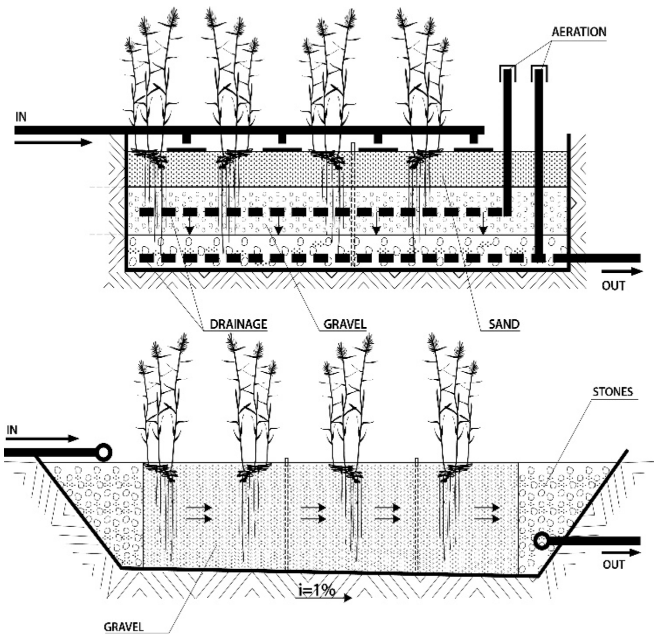

Figure 2. Cross section of SS VF (subsurface vertical flow) and HF (subsurface horizontal flow beds) beds.

The SS VF bed was built of three layers of stones 0.2 m (fraction 16–60 mm), gravel 0.4 m (fraction 2–16 mm), and sand 0.2 m (fraction 0.5–2 mm). Two pipes (diameter 50 mm) with perforation 2 mm were used for passive aeration. The SS-HF bed was built of gravel (2–16 mm) and stones (16–60 mm). The gravel hydraulic conductivity was 4×10^{-2} ms^{-1}. Both beds were planted with reeds (*Phragmites australis*). Reject water was taken directly from centrifuge which dewatered sludge after anaerobic digestion in a dairy WWTP.

2.2. Sampling and Analytical Procedures

The study was carried out between April 2015 and March 2016. Both beds were supplied from sedimentation and retention tanks with the same amount of reject water, which allowed comparison of these beds' effectiveness. The daily hydraulic load was 0.1 m^3 m^{-2} d^{-1} in both SS VF and SS HF beds. Hydraulic retention time (HRL) for SS HF bed was approximately 8 days. Samples were collected three times a month (influent to SS-VF and SS-HF and effluents from both beds). The air temperature during the research period varied from −11 to 26 °C, while reject water temperature varied from 4 °C to 20 °C.

The basic physical and chemical analyses were performed: BOD_5, COD, total organic carbon (TOC), total suspended solids (TSS), total Kjeldahl nitrogen (TKN), ammonium nitrogen (NH_4^+-N), nitrate nitrogen (V) (NO_3-N), nitrite nitrogen (III) (NO_2-N), total phosphorus (TP), dissolved oxygen, and alkalinity. TN value was calculated as a sum of TKN, NO_3–N, and NO_2^--N. To evaluate biodegrability of reject water, BOD_5/COD and BOD_5/TN ratios were determined. Determinations were conducted in a certified laboratory in accordance with the procedures set out in the Regulation of the Environmental Protection Minister [21] from November 18, 2014, and in accordance with the American Public Health Association (2005) [22].

2.3. Modeling of Pollutants Removal

Statistical analysis of NH_4^+-N and TN, as well as BOD_5; COD removal was performed, based on the P-k-C* model [12]:

$$\frac{C_{out} - C^*}{C_{in} - C^*} = \left(1 + \frac{k(T)}{qP}\right)^{-P} \tag{1}$$

where C_{out}—output concentration (g/m^3), C_{in}—input concentration (g/m^3), C^*—background concentration (g/m^3), $k(T)$—chemical reaction coefficient (temperature dependent), q—hydraulic load (m/d), P—number of tanks in series.

Model (1) can be further simplified if some assumptions are made. The apparent removal efficiency η_{app} can be defined as:

$$\eta_{app} = 1 - \frac{C_{out} - C^*}{C_{in} - C^*} \tag{2}$$

This apparent efficiency model's real efficiency η in limit $C^* \to 0$. If limit $P \to \infty$ is taken, the right side of Equation (1) simplifies:

$$\lim_{P\to\infty}\left(1 + \frac{k(T)}{qP}\right)^{-P} = \exp(-k(T)/q) \tag{3}$$

Chemical reaction coefficient k is dependent on reject water temperature, using direct or modified first order Arrhenius dependency:

$$k_d(T) = K_{20}\theta^{T-20} \equiv \exp(\ln K_{20} + [T - 20]\ln\theta) \tag{4}$$

$$k_m(T) = K_{20}\theta^{T-20}\theta_m^{\min(T-T_k,0)} \equiv \exp(\ln K_{20} + [T - 20]\ln\theta + [\min(T - T_k, 0)]\ln\theta_m) \tag{5}$$

where k_d, k_m—direct and modified chemical reaction coefficients; K_{20}—direct or modified chemical reaction coefficients normalized for 20 °C; θ—temperature coefficient; θ_m—modifier for temperature coefficient for temperatures less than specific temperature T_k, where trend changes; $\min(\cdot,\cdot)$—minimum function of 2 arguments.

Modified first order dependency allows for an additional change of the reaction coefficient below an estimated specific temperature. It was introduced to better fit modeled data, while not changing the basic rule of first order linear dependency on a logarithmic scale.

By using all available combinations of previously described equations, 16 models with different parameters subsets were estimated. The final parameter subset selection was performed using methodology given by Burnham & Anderson [23]. A detailed description of fitted models, their selection and fitting procedure are presented in Appendix A.

It is worth noting that a full analysis of a P-k-C* model is difficult and can lead to unexpected results [24]. In most of the cases, plug flow models can be too simple to describe the dynamics of particular pollutant removal. Sensitivity analysis of fitted models in literature [25] may suggest that some parameters, like P, may be insensitive and set to values obtained from earlier studies. There are some earlier results suggesting that temperature dependency can change [16].

3. Results and Discussion

3.1. Treatment Efficiency and Load Removal

Table 1 presents the characteristics of reject water before and after treatment with SS VF and SS HF beds during research period, while Table 2 shows removed pollutants load.

The BOD_5/COD and BOD_5/TN ratios give information about biodegradability [26]. Its value can be used to assess the reject water susceptibility for high efficiency of biological treatment in conventional systems (e.g., activated sludge method). An average BOD_5/COD ratio was 0.59, while BOD_5/TN ratio 0.43. Low BOD_5/COD ratio pointed out low degradability of the organic compounds. Reject water from dairy WWTP with anaerobic sewage sludge digestion was discovered to have a higher BOD_5/COD ratio than municipal WWTPs. In municipal, it ranges from 0.25 to 0.32 for reject water from the WWTP in Gdansk [27], and 0.2 for reject water from the WWTP in Minworth [8]. The WWTP in Gdansk records BOD_5/TN ratios at the level of 0.37 up to 0.54. In this case, the conventional path of nitrogen removal cannot be applied as the content of easily biodegradable organics is insufficient [28,29]. A very high concentration of ammonium nitrogen should be viewed as the main reason for this.

Table 1. The characteristics of reject water before and after treatment in SS VF and SS HF beds (38 research series, 114 samples).

	Before Treatment (mg L^{-1})	After Treatment (mg L^{-1})	
Parameter		**SS-VF**	**SS-HF**
BOD_5	120.53 ± 16.97	21.55 ± 8.27	31.18 ± 10.04
COD	201.92 ± 21.48	40.82 ± 12.14	49.79 ± 12.44
TOC	53.18 ± 9.91	10.18 ± 4.52	18.08 ± 5.58
TSS	141.01 ± 18.12	20.60 ± 4.84	26.88 ± 4.61
TN	276.66 ± 42.71	134.29 ± 19.44	158.87 ± 20.29
NH_4^+-N	195.53 ± 38.32	50.71 ± 16.60	73.29 ± 20.53
NO_3^--N	0.69 ± 0.28	12.68 ± 4.76	3.31 ± 1.33
NO_2^--N	0.17 ± 0.07	0.13 ± 0.05	0.63 ± 0.20
TP	22.17 ± 3.54	16.43 ± 2.00	13.95 ± 2.54

Mean ± standard deviation.

Table 2. Removed pollutant loads (mean values).

-	Removed Load (g m^{-2} d^{-1})	
Parameter	**SS-VF**	**SS-HF**
BOD_5	9.90	8.93
COD	16.11	15.21
TOC	4.30	3.51
TSS	12.00	11.41
TN	14.24	11.78
NH_4^+-N	14.48	12.22
TP	0.57	0.82

The average calculated treatment efficiency in SS VF bed was: BOD_5 82.12%, COD 79.79%, TOC 80.85%, TSS 85.30%, TN 51.46%, NH_4^+-N 74.06%, and TP 25.42%. The average calculated treatment efficiency in SS HF bed was: BOD_5 74.13%, COD 75.34%, TOC 66.01%, TSS 89.90%, TN 42.58%, NH_4^+-N 62.52%, and TP 37.20%. After the treatment, the average ratios of BOD_5/COD were 0.53 for SS VF bed and 0.63 for SS HF bed, and BOD_5/TN were 0.16 and 0.20, respectively.

Through the analysis of the effect of organic matter removal (g m^{-2} d^{-1}) expressed by BOD_5, COD, and TOC (Table 2), similar values to household sewage treatment with constructed wetlands were achieved [10]. In the case of TN and NH_4^+-N, higher efficiency was observed mainly due to high concentration of nitrogen in reject water before treatment in SS VF and SS HF beds. The TP removal effect was similar to the observed in the case of household and municipal sewage treatment.

3.2. Modeling of Pollutants Removal

Statistical analysis of NH_4^+-N and TN, as well as BOD_5, COD removal, presented in Appendix B, revealed existence of 3 group models, each with stable core parameters, which dominate the variability of logarithmic likelihood and residuals distribution. The most-preferred parameter set was T_k, θ_m, K_{20}—a model without temperature dependency after T_k, with Akaike weight greater than 0.94 in all but other cases except in one (TN removal in SS HF bed). Residual analysis suggests also T_k, θ_m, K_{20} model selection in this case. Selected models are presented graphically in Figure 3, using efficiency scale.

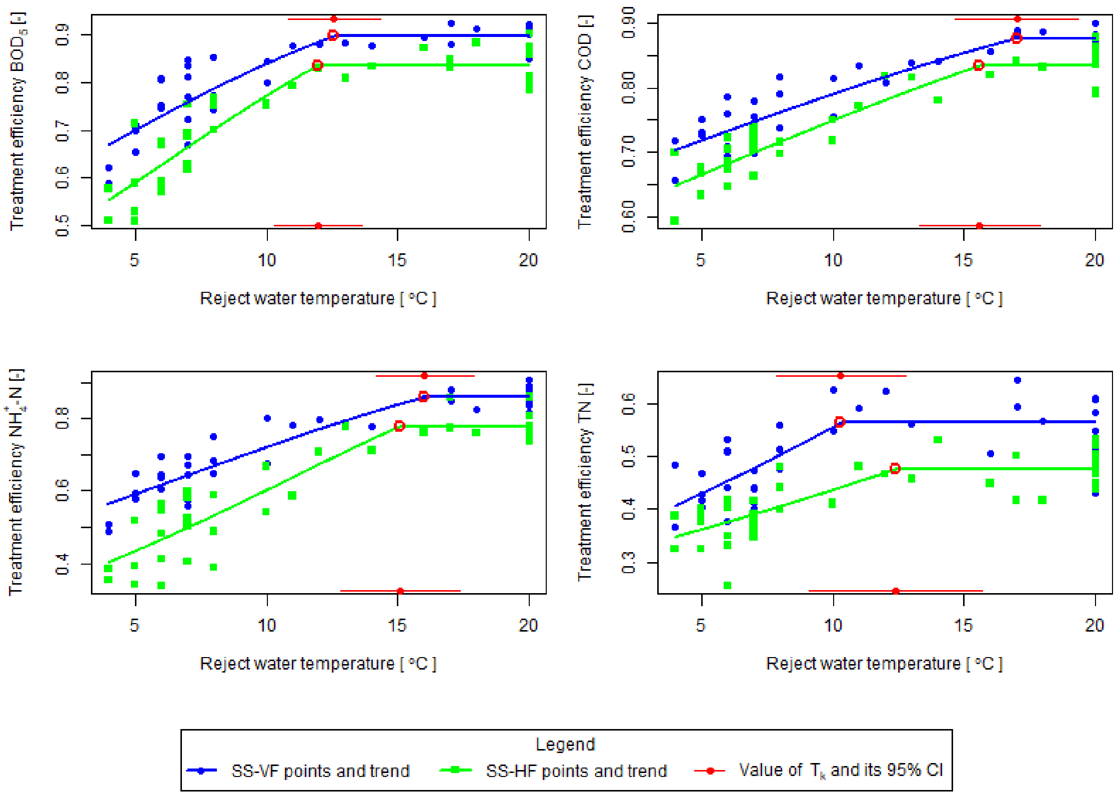

Figure 3. Selected model fit.

In the cases of BOD_5, COD, TN, and $NH_4{}^+$-N, the treatment efficiency increased until a specific temperature T_k was reached, and then it was constant. Results obtained in this work suggest that it may be possible to effectively describe pollutant removal behavior in a consistent way using simplified models. Any additional parameter, with assumption of proper overall fit, will have a large confidence interval, suggesting overall insensitivity. Simplicity of obtained models does not interfere with their statistical validity.

Created models show that simple ratios obtained over the whole period of the research study are insufficient to properly describe behavior of beds, and a more dedicated approach, like P-k-C* modeling, is necessary. Only ratios in stable periods of bed work (after specific temperature T_k) are sufficient to describe it.

Values of specific temperature T_k were marked in place of trend change and on the upper and lower section of figures along with 95% confidence intervals (CI). Parameter estimates of the fitted models, along with their 95% confidence intervals, and residual sum of squares were presented in Table 3.

Figure 4 presents standard a box-whisker plot of TP removal efficiency and reject water temperature during the whole research period. Mean values are described as red dots inside each box, and horizontal line represents median. Dashed lines inside efficiency vs. temperature plot represent mean values.

The calculated treatment efficiency of TP removal was on average 25.42% (SS VF) and 37.20% (SS HF). The efficiency of TP removal in SS VF bed was stable and higher than in SS HF bed. Phosphorous removal occurs mainly in the processes of chemical precipitation and sorption, which do not depend on temperature [12]. A distribution analysis of TP treatment efficiency revealed no significant deviation from normality (Shapiro-Wilk normality test, for SS HF: test statistics 0.952, p-value = 0.1, for SS VF: test statistics 0.96, p-value = 0.19). An F-test revealed no statistically significant difference between the variances (F test statistics 1.74, numerator and denominator degrees of freedom—37, p-value = 0.095). T-test revealed statistically significant differences between mean treatment efficiency of TP (t statistics 9.52, 74 degrees of freedom, p-value $< 2 \times 10^{-14}$). For all performed tests, significance level was set to $\alpha = 0.05$.

Table 3. Parameters of selected models for BOD_5, COD, NH_4^+-N and TN.

Horizontal Flow Bed SS-HF				Vertical Flow Bed SS-VF			
BOD_5							
Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI	Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI
T_k	11.959	10.302	13.616	T_k	12.580	10.836	14.323
θ_m	1.106	1.073	1.140	θ_m	1.087	1.061	1.113
K_{20}	0.180	0.170	0.190	K_{20}	0.225	0.213	0.238
RSS	1.974	-		RSS	2.327	-	
COD							
Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI	Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI
T_k	15.578	13.293	17.864	T_k	17.000	14.656	19.344
θ_m	1.048	1.035	01.06	θ_m	1.043	1.033	1.052
K_{20}	0.179	0.172	0.187	K_{20}	0.209	0.200	0.218
RSS	0.831	-		RSS	0.884	-	
NH_4^+-N							
Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI	Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI
T_k	15,087	12,82	17,355	T_k	16.007	14.153	17.862
θ_m	1.101	1.073	1.131	θ_m	1.074	1.058	1.089
K_{20}	0.151	0.139	0.164	K_{20}	0.195	0.184	0.207
RSS	2.800	-		RSS	1.748	-	
TN							
Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI	Parameter	Estimate/Value	Lower 95% CI	Upper 95% CI
T_k	12.401	9.132	15.671	T_k	10.312	7.860	12.763
θ_m	1.051	1.024	1.078	θ_m	1.077	1.033	1.122
K_{20}	0.065	0.060	0.069	K_{20}	0.083	0.077	0.089
RSS	1.026	-		RSS	1.629	-	

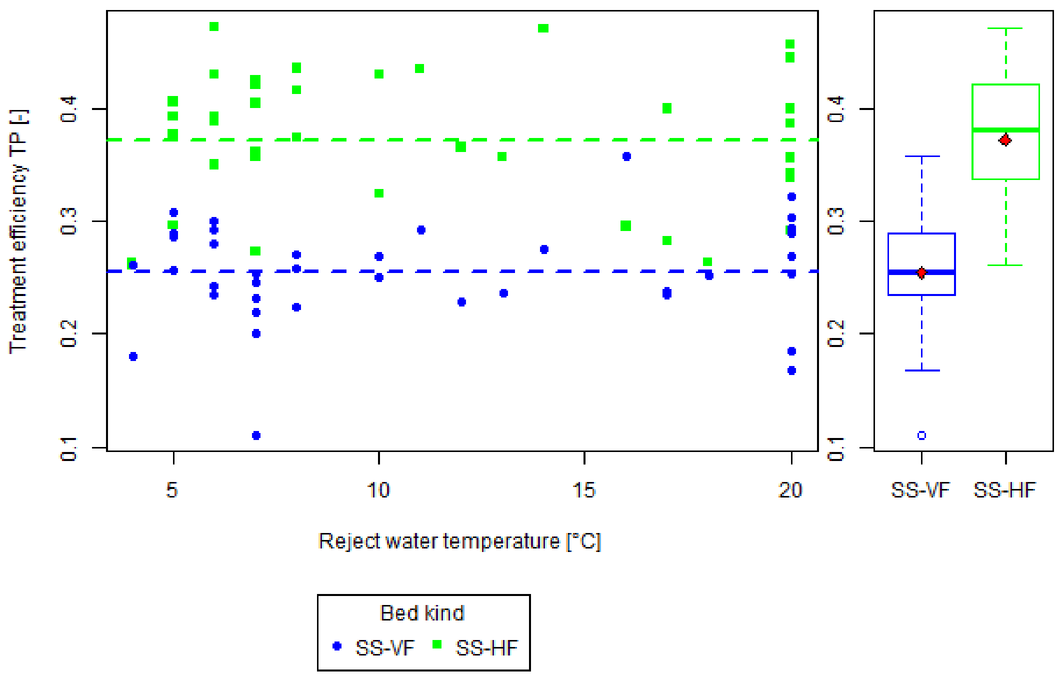

Figure 4. TP removal efficiency and reject water temperature during the research period.

The mathematical models found practical implementation. An application, which allows determining the effectiveness of reject water treatment in SS VF and SS HF constructed wetlands depending on the reject water temperature, was developed (Figure 5).

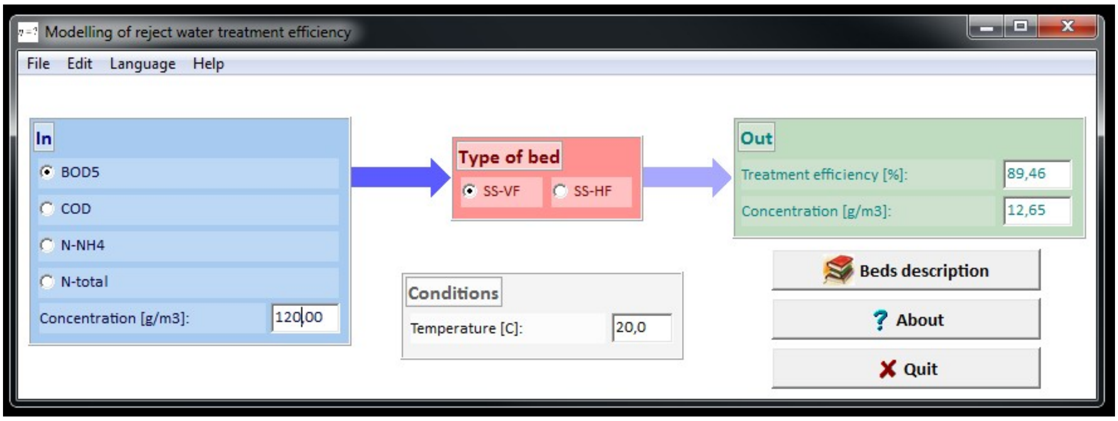

Figure 5. An application "Modeling of reject water treatment efficiency".

4. Conclusions

The study showed a high efficiency of SS VF and SS HF beds for removing main pollutants from the reject water generated during anaerobic sewage sludge digestion in dairy WWTP. Results for SS VF and SS HF, respectively, were: COD 79.79% and 75.34%, for BOD_5 82.12% and 74.13%, for TN 51.46% and 42.58%, for NH_4^+-N 74.06% and 62.52%, for TP 25.42% and 37.20%. A higher efficiency of main organic pollutants removal was observed in the case SS VF bed during the whole research. The efficiency of TP removal was stable and higher for the SS HF bed.

The results have been used for modeling the work of SS VF and SS HF beds for reject water treatment. Models for determining the effectiveness of treatment depending on the temperature and type of beds were presented. P-k-C* model was sufficient to describe the observed variability of constructed wetland behavior. In the cases of BOD_5, COD, TN, and NH_4^+-N, the treatment efficiency increased to the specific temperature, then it was constant. Results obtained in this work suggest that it may be possible to effectively describe pollutant removal behavior using simplified models.

All models predict better treatment efficiency in SS VF bed, except for TP. In the case of TP removal, distribution tests along with a t-test were performed. The created models show that simple ratios obtained over the whole period of study are insufficient to properly describe behavior of beds. A more dedicated approach, like P-k-C* modeling, is necessary. Only ratios in stable periods of bed work (after specific temperature) are sufficient to describe it.

An application in the form of software for modeling the efficiency of reject water treatment from sewage sludge digestion in dairy WWTPs was prepared on the basis of the results from the research.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A. Statistical Modeling

The 16 models obtained from (1) were constructed by taking limit of certain variables simplifying the expression:

C^* (allowed to vary or set to 0)

P (allowed to vary or taken limit resulting in (3) dependency)

θ (allowed to vary or set to 1)
θ_m and related T_k (if θ_m is present, then (5) form was used, otherwise (4)).

All models were estimated using non-linear least squares procedure with a variant of Levenberg-Marquardt's algorithm for optimization [30]. It is worth noting that both sides of (1) are always constrained to a (0–1) interval. Residuals from modeling this dependency will be therefore non-homoscedastic. Final model estimation procedure reweighted them by term proportional to [31]:

$$s_i \sim \frac{1}{\hat{r}_i(1-\hat{r}_i)} \tag{A1}$$

where $\hat{r}_i$—estimated and right-hand side of (1), s_i—residual correction for i-th observation.

Model parameter selection was performed using methodology from [23]. After all 16 models were built, they were weighted using Akaike weight sw_i, which is a measure of a model fit to the data, normalized to (0–1) interval, relative to the best model from the set of analyzed ones. The Akaike weight was calculated from differences Δ_i of second order [32] AIC [33]. Models with very similar logarithmic likelihood and residual distribution, containing only additional parameters, were removed from weighting procedure [23]. Top weighted models, forming 0.90–0.95 of summary Akaike weights, were retained for further analysis. Residuals structure of the remaining parameter sets was also checked, to further reject oversimplified ones using an approach based on [34]. Remaining parameter subsets were presented along with their basic statistics. Parameter T_k, if present, was presented graphically in a similar way to [35].

Model calculation was performed using R version 3.5.1 [36]. The nlfb procedure from nlsr package [37] was used for optimization of models. Second order AIC was calculated using AICc procedure from AICcmodavg package [38].

Appendix B. Result of Statistical Modeling

Tables A1–A4 present fitted models grouped by almost identical values of logarithmic likelihood and residuals structure. Analysis revealed three preferred groups of models, each with stable core parameters, which dominate the variability of logarithmic likelihood and residuals distribution. For the simplest models from each group, containing the core parameters, Akaike weight was calculated. Any other model, with addition of one or more parameters, does not change its fit significantly.

Analysis results consistently supported choice of model with parameter subset T_k, θ_m, K_{20}—a model with no temperature dependency after T_k. In almost all cases, Akaike weight for this preferred model is greater than 0.95, with suggests strong support. The second one is θ, K_{20}—a model with non-changing temperature dependency. Model θ, K_{20} for total nitrogen for horizontal flow bed had approximately w_i equal to 0.16, suggesting some support for further analysis. The third model contains only K_{20} variable, and obviously does not depend on temperature. It has almost no support, and was not chosen for residual analysis.

Residual analysis, presented in Figures A1–A4, support selection for first preferred model in all cases; there is signal of non-linearity in models θ, K_{20}, even a slight one for total nitrogen in horizontal flow bed. Residuals analysis suggests that models with T_k, θ_m, K_{20} parameters also have properties allowing for further analysis of those models parameters—normal distribution, homoscedasticity and no visible autocorrelation—which were also performed.

Table A1. Fitted models for BOD_5 with their statistics.

Horizontal Flow Bed SS-HF							Vertical Flow Bed SS-VF						
Model	**Variables**	**k**	**logLik**	**AICc**	**Δ_i**	**w_i**	**Model**	**Variables**	**k**	**logLik**	**AICc**	**Δ_i**	**w_i**
1	T_k, θ_m, K_{20}	4	2.272	4.668	0.000	>0.999	1	T_k, θ_m, K_{20}	4	−0.855	10.923	0.000	>0.996
2	P, T_k, θ_m, K_{20}	5	2.485	6.905	2.237		2	$\theta, T_k, \theta_m, K_{20}$	5	−0.260	12.394	1.471	
3	$\theta, T_k, \theta_m, K_{20}$	5	2.280	7.315	2.648		3	P, T_k, θ_m, K_{20}	5	−0.606	13.088	2.164	
4	$C^*, T_k, \theta_m, K_{20}$	5	2.272	7.330	2.663		4	$C^*, T_k, \theta_m, K_{20}$	5	−0.855	13.586	2.663	
5	$\theta, P, T_k, \theta_m, K_{20}$	6	2.485	9.739	5.072		5	$\theta, P, T_k, \theta_m, K_{20}$	6	−0.031	14.772	3.849	
6	$C^*, P, T_k, \theta_m, K_{20}$	6	2.485	9.740	5.072		6	$C^*, \theta, T_k, \theta_m, K_{20}$	6	−0.260	15.229	4.306	
7	$C^*, \theta, T_k, \theta_m, K_{20}$	6	2.280	10.150	5.483		7	$C^*, P, T_k, \theta_m, K_{20}$	6	−0.606	15.922	4.999	
8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	2.485	12.763	8.096		8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	−0.031	17.796	6.873	
9	θ, K_{20}	3	−8.793	24.293	19.625	5.476×10^{-5}	9	θ, K_{20}	3	−7.783	11.349	11.349	3.421×10^{-3}
10	θ, P, K_{20}	4	−7.897	25.007	20.339		10	θ, P, K_{20}	4	−6.763	22.738	11.815	
11	C^*, θ, K_{20}	4	−8.793	26.799	22.131		11	C^*, θ, K_{20}	4	−7.783	24.778	13.855	
12	C^*, θ, P, K_{20}	5	−7.897	27.670	23.002		12	C^*, θ, P, K_{20}	5	−6.763	25.401	14.478	
13	K_{20}	2	−31.510	67.364	62.696	5.430×10^{-14}	13	K_{20}	2	−32.052	68.446	57.523	3.218×10^{-13}
14	C^*, K_{20}	3	−31.510	69.727	65.059		14	C^*, K_{20}	3	−32.052	59.886	70.809	
15	P, K_{20}	3	−31.510	69.727	65.059		15	P, K_{20}	3	−32.052	59.886	70.809	
16	C^*, P, K_{20}	4	−31.510	72.233	67.565		16	C^*, P, K_{20}	4	−32.052	62.392	73.316	

Table A2. Fitted models for COD with their statistics.

Horizontal Flow Bed SS-HF							Vertical Flow Bed SS-VF						
Model	**Variables**	**k**	**logLik**	**AICc**	**Δ_i**	**w_i**	**Model**	**Variables**	**k**	**logLik**	**AICc**	**Δ_i**	**w_i**
1	T_k, θ_m, K_{20}	4	18.702	−28.193	0.000	>0.965	1	T_k, θ_m, K_{20}	4	17.528	−25.844	0.000	>0.970
2	P, T_k, θ_m, K_{20}	5	18.823	−25.772	2.421		2	$\theta, T_k, \theta_m, K_{20}$	5	18.001	−24.127	1.716	
3	$\theta, T_k, \theta_m, K_{20}$	5	18.711	−25.547	2.645		3	P, T_k, θ_m, K_{20}	5	17.591	−23.307	2.537	
4	$C^*, T_k, \theta_m, K_{20}$	5	18.702	−25.530	2.663		4	$C^*, T_k, \theta_m, K_{20}$	5	17.528	−23.181	2.663	
5	$\theta, P, T_k, \theta_m, K_{20}$	6	18.832	−22.955	5.238		5	$\theta, P, T_k, \theta_m, K_{20}$	6	18.002	−21.295	4.549	
6	$C^*, P, T_k, \theta_m, K_{20}$	6	18.823	−22.937	5.256		6	$C^*, \theta, T_k, \theta_m, K_{20}$	6	18.001	−21.293	4.551	
7	$C^*, \theta, T_k, \theta_m, K_{20}$	6	18.711	−22.713	5.480		7	$C^*, P, T_k, \theta_m, K_{20}$	6	17.591	−20.472	5.372	
8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	18.832	−19.931	8.261		8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	18.002	−18.271	7.573	
9	θ, K_{20}	3	14.117	−21.528	6.664	3.449×10^{-2}	9	θ, K_{20}	3	12.794	−18.883	6.961	2.986×10^{-2}
10	θ, P, K_{20}	4	14.821	−20.430	7.762		10	θ, P, K_{20}	4	13.497	−17.782	8.062	
11	C^*, θ, K_{20}	4	14.117	−19.022	9.170		11	C^*, θ, K_{20}	4	12.794	−16.376	9.468	
12	C^*, θ, P, K_{20}	5	14.821	−17.768	10.425		12	C^*, θ, P, K_{20}	5	13.497	−15.119	10.725	
13	K_{20}	2	−18.244	40.831	69.023	9.922×10^{-16}	13	K_{20}	2	−20.165	44.672	70.516	4.726×10^{-16}
14	C^*, K_{20}	3	−18.244	43.194	71.386		14	C^*, K_{20}	3	−20.165	47.035	72.879	
15	P, K_{20}	3	−18.244	43.194	71.386		15	P, K_{20}	3	−20.165	47.035	72.879	
16	C^*, P, K_{20}	4	−18.244	45.700	73.892		16	C^*, P, K_{20}	4	−20.165	49.542	75.385	

Table A3. Fitted models for NH_4^+-N with their statistics.

Horizontal Flow Bed SS-HF							Vertical Flow Bed SS-VF						
Model	**Variables**	**k**	**logLik**	**AICc**	Δ_i	w_i	**Model**	**Variables**	**k**	**logLik**	**AICc**	Δ_i	w_i
1	T_k, θ_m, K_{20}	4	−4.371	17.954	0.000	>0.980	1	T_k, θ_m, K_{20}	4	4.583	0.046	0.000	>0.940
2	$\theta, T_k, \theta_m, K_{20}$	5	−4.342	20.559	2.605		2	$\theta, T_k, \theta_m, K_{20}$	5	5.060	1.754	1.708	
3	P, T_k, θ_m, K_{20}	5	−4.370	20.615	2.662		3	P, T_k, θ_m, K_{20}	5	4.883	2.109	2.063	
4	$C^*, T_k, \theta_m, K_{20}$	5	−4.371	20.617	2.663		4	$C^*, T_k, \theta_m, K_{20}$	5	4.583	2.709	2.663	
5	$C^*, \theta, T_k, \theta_m, K_{20}$	6	−4.342	23.394	5.440		5	$\theta, P, T_k, \theta_m, K_{20}$	6	5.117	4.476	4.429	
6	$C^*, P, T_k, \theta_m, K_{20}$	6	−4.370	23.450	5.497		6	$C^*, \theta, T_k, \theta_m, K_{20}$	6	5.060	4.589	4.542	
7	$\theta, P, T_k, \theta_m, K_{20}$	6	−4.638	23.986	6.032		7	$C^*, P, T_k, \theta_m, K_{20}$	6	4.883	4.944	4.897	
8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	−4.638	27.010	9.056		8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	5.117	7.499	7.453	
9	θ, K_{20}	3	−9.506	25.719	7.765	2.018×10^{-2}	9	$\theta,$	3	0.566	5.574	5.527	5.932×10^{-2}
10	θ, P, K_{20}	4	−8.692	26.597	8.643		10	θ, P, K_{20}	4	1.709	5.793	5.747	
11	C^*, θ, K_{20}	4	−9.506	28.225	10.271		11	C^*, θ, K_{20}	4	0.566	8.080	8.034	
12	C^*, θ, P, K_{20}	5	−8.692	29.260	11.306		12	C^*, θ, P, K_{20}	5	1.709	8.456	8.410	
13	K_{20}	2	−37.860	80.062	62.108	3.195×10^{-14}	13	K_{20}	2	−33.102	70.547	70.500	4.618×10^{-16}
14	C^*, K_{20}	3	−37.860	82.425	64.471		14	C^*, K_{20}	3	−33.102	72.910	72.863	
15	P, K_{20}	3	−37.860	82.425	64.471		15	P, K_{20}	3	−33.102	72.910	72.863	
16	$C^*, P,$	4	−37.860	84.931	66.978		16	C^*, P, K_{20}	4	−33.102	75.416	75.370	

Table A4. Fitted models for TN with their statistics.

Horizontal Flow Bed SS-HF							Vertical Flow Bed SS-VF						
Model	**Variables**	**k**	**logLik**	**AICc**	Δ_i	w_i	**Model**	**Variables**	**k**	**logLik**	**AICc**	Δ_i	w_i
1	T_k, θ_m, K_{20}	4	14.709	−20.206	0.000	>0.838	1	T_k, θ_m, K_{20}	4	5.926	−2.640	0.000	>0.992
2	$\theta, T_k, \theta_m, K_{20}$	5	14.744	−17.613	2.593		2	$\theta, T_k, \theta_m, K_{20}$	5	6.384	−0.893	1.747	
3	$C^*, T_k, \theta_m, K_{20}$	5	14.709	−17.543	2.663		3	$C^*, T_k, \theta_m, K_{20}$	5	5.926	0.023	2.663	
4	P, T_k, θ_m, K_{20}	5	14.709	−17.543	2.663		4	P, T_k, θ_m, K_{20}	5	5.926	0.023	2.663	
5	$C^*, \theta, T_k, \theta_m, K_{20}$	6	14.744	−14.779	5.428		5	$C^*, \theta, T_k, \theta_m, K_{20}$	6	6.384	1.942	4.582	
6	$\theta, T_k, \theta_m, K_{20}$	6	14.732	−14.755	5.451		6	$\theta, P, T_k, \theta_m, K_{20}$	6	6.384	1.942	4.582	
7	$C^*, P, T_k, \theta_m, K_{20}$	6	14.709	−14.709	5.498		7	$C^*, P, T_k, \theta_m, K_{20}$	6	5.926	2.858	5.498	
8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	14.732	−11.731	8.475		8	$C^*, \theta, P, T_k, \theta_m, K_{20}$	7	6.384	4.966	7.605	
9	θ, K_{20}	3	11.810	−16.915	3.291	>0.161	9	θ, K_{20}	3	−0.182	7.071	9.710	7.729×10^{-3}
10	θ, P, K_{20}	4	11.887	−14.562	5.644		10	θ, P, K_{20}	4	−0.084	9.381	12.020	
11	C^*, θ, K_{20}	4	11.810	−14.409	5.798		11	C^*, θ, K_{20}	4	−0.182	9.577	12.216	
12	C^*, θ, P, K_{20}	5	11.887	−11.899	8.307		12	C^*, θ, P, K_{20}	5	−0.084	12.044	14.683	
13	K_{20}	2	−2.405	9.154	29.360	3.532×10^{-7}	13	K_{20}	2	−9.124	22.590	25.229	3.297×10^{-6}
14	C^*, K_{20}	3	−2.405	11.517	31.723		14	C^*, K_{20}	3	−9.124	24.953	27.592	
15	P, K_{20}	3	−2.405	11.517	31.723		15	P, K_{20}	3	−9.124	24.953	27.592	
16	C^*, P, K_{20}	4	−2.405	14.023	34.229		16	C^*, P, K_{20}	4	−9.124	27.459	30.099	

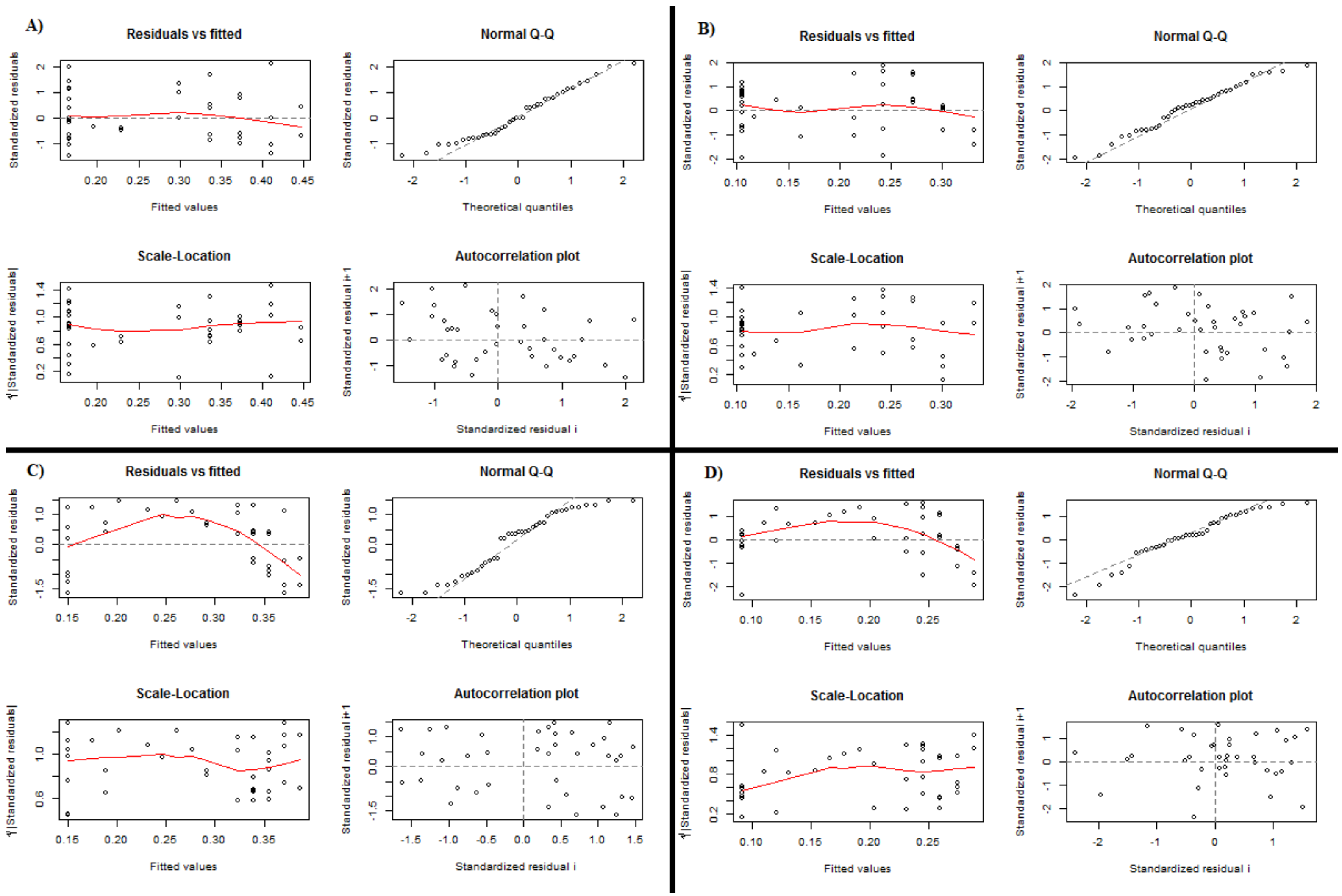

Figure A1. Diagnostic for BOD_5 models. (**A**) Horizontal bed, model T_k, θ_m, K_{20}; (**B**) vertical bed, model T_k, θ_m, K_{20}; (**C**) horizontal bed, model θ, K_{20}; (**D**) vertical bed, model θ, K_{20}.

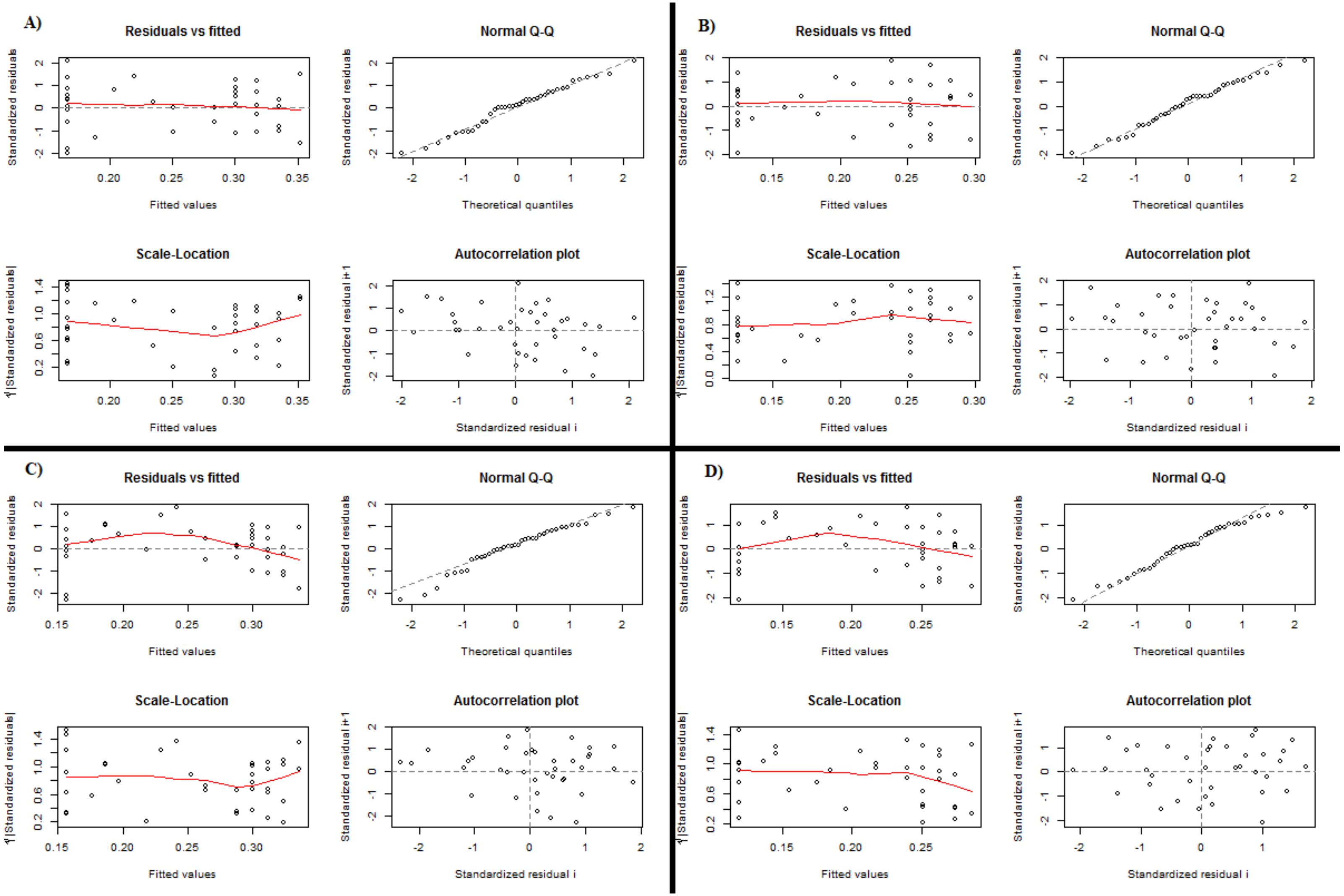

Figure A2. Diagnostic for COD models. (**A**) Horizontal bed, model T_k, θ_m, K_{20}; (**B**) vertical bed, model T_k, θ_m, K_{20}; (**C**) horizontal bed, model θ, K_{20}; (**D**) vertical bed, model θ, K_{20}.

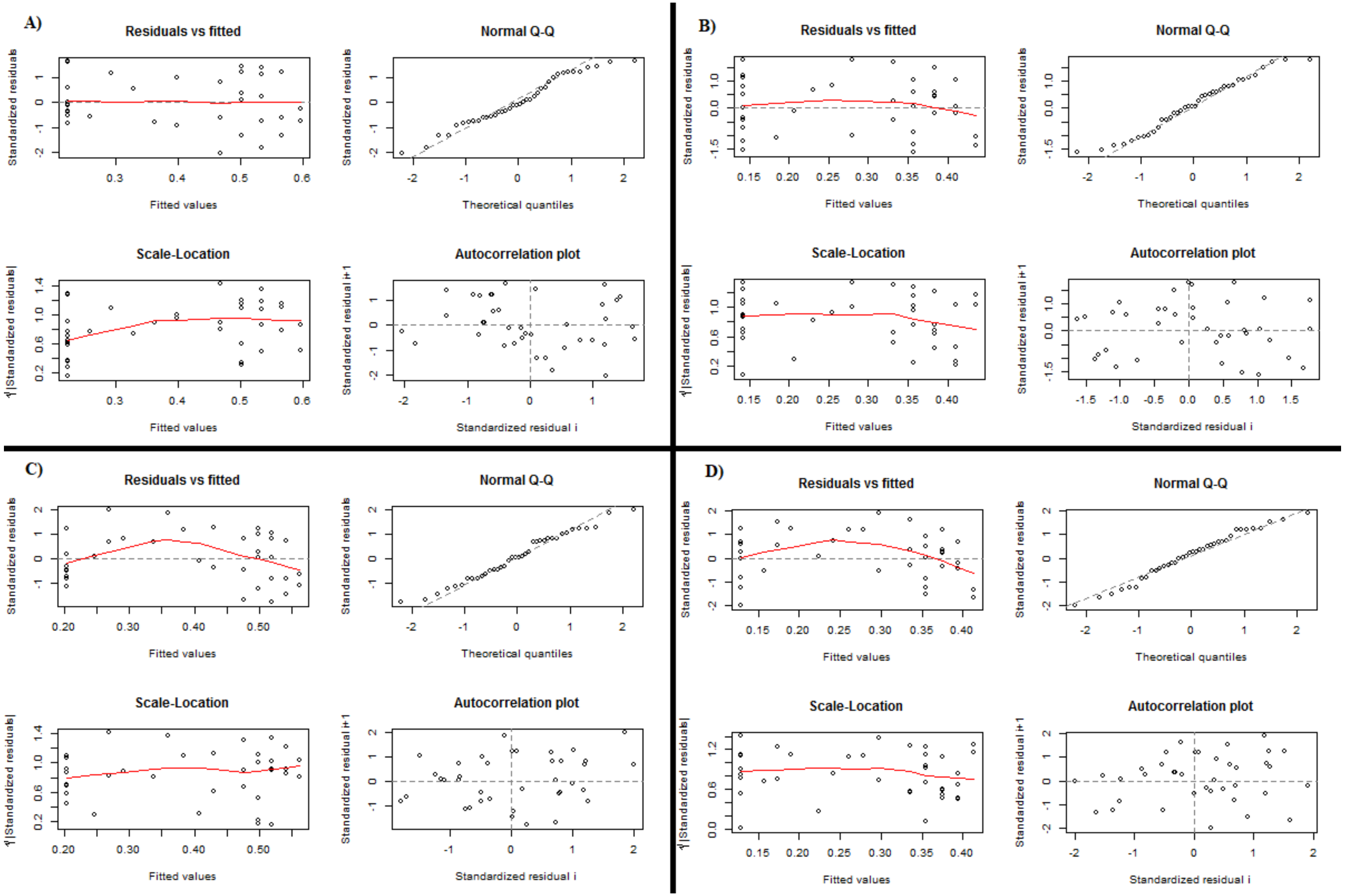

Figure A3. Diagnostic for NH_4^+-N models. (**A**) Horizontal bed, model T_k, θ_m, K_{20}; (**B**) vertical bed, model T_k, θ_m, K_{20}; (**C**) horizontal bed, model θ, K_{20}; (**D**) vertical bed, model θ, K_{20}.

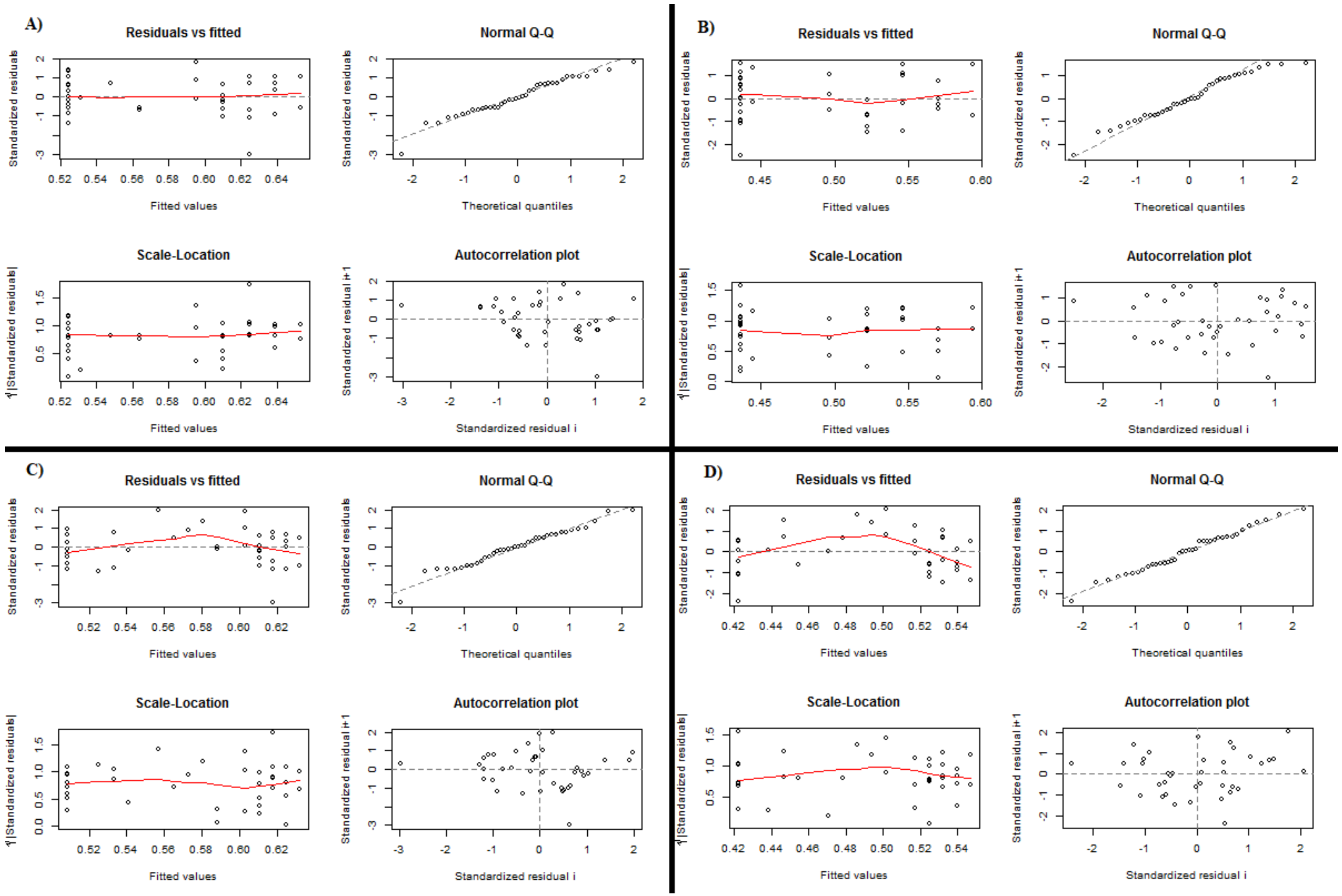

Figure A4. Diagnostic for TN models. (**A**) Horizontal bed, model T_k, θ_m, K_{20}; (**B**) vertical bed, model T_k, θ_m, K_{20}; (**C**) horizontal bed, model θ, K_{20}; (**D**) vertical bed, model θ, K_{20}.

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