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The efficiency and technological reliability of biogenic compounds removal during long-term operation of a one-stage subsurface horizontal flow constructed wetland

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ABSTRACT

The paper presents the results of a study of the efficiency and technological reliability of total nitrogen and total phosphorus removal during long term operation of a one-stage constructed wetland system with subsurface horizontal wastewater flow. The flow rate of the wastewater treatment plant was 1.2 m^3/d during the research period. Physical and chemical analyses of raw wastewater and treated effluent were carried out in the years 1997–2010 (14 years). During this study period, 56 series of analyses were performed and 112 wastewater samples were collected. It was observed that the average efficiency of total nitrogen and total phosphorus removal amounted to 51% and 73%, respectively. The technological reliability, determined by means of the Weibull reliability method, was 45% for total nitrogen removal and 48% in the case of total phosphorus removal. Total nitrogen and total phosphorus concentrations in the treated wastewater did not exceed the permissible values in 39.3% and 37.5% of cases, respectively. A negative correlation was found between the sorption capacity of the soil-plant filter for the removal of total phosphorus and the time of plant operation. The efficiency of total nitrogen removal was stable over time.

1. Introduction

In Poland, almost 38% of the population lives in rural areas [1]. Almost half of the villages located in rural communes are characterized by a scattered development pattern, i.e. the distance between buildings is greater than 100 m. Such an arrangement of farmsteads is problematic as far as the construction of a central wastewater disposal and treatment system is concerned [2]. Due to economic reasons, the only solution for the implementation of a proper wastewater management programme in rural areas is to conduct treatment in household treatment plants [3–5]. At present, there are about 202 800 household treatment plants in Poland, and about 22,000 new facilities are built each year [6]. The technological solutions for household treatment plants include: 1 – systems with infiltration drainage, 2 – sand filters, 3 – treatment plants with activated sludge, 4 – treatment plants with a trickling filter, 5 – hybrid systems (activated sludge + trickling filter), and 6 – wetland systems (one-stage and hybrid) [7]. The possibility of employing a given wastewater treatment technology in a given location is governed by numerous factors, the most important of which include the size of the property, type of receiver of treated wastewater, and legal requirements pertaining to the quality of treated wastewater [8]. Along with the increasing number of household treatment plants, the need to control their efficiency and reliability grows as well.

A survey carried out in 2011 in south-eastern Poland shows that the most common solution used in household sewage treatment plants were systems with infiltration drainage (71%). Systems with activated sludge constituted 15.7%, systems with biological reservoir – 11.6%, hybrid systems (active sediment with biological reservoir) – 1.6%, and

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constructed wetland systems – only 0.1% [9]. According to the studies of Orlik and Jóźwiakowski [10], the effects of removing contaminants in systems consisting of precipitator and infiltration drainage are small and in the case of removal of general suspensions do not exceed 40%, and in the case of BOD_5 and COD, TN and TP they do not exceed 38%. It has also been shown that these systems provide only mechanical wastewater treatment and thus contribute to the degradation of groundwater quality [11]. Significantly better results in the removal of contaminants (at the level of 70–80%) can be achieved in domestic sewage treatment plants with activated sludge and biological deposit, or in hybrid systems, where these two technologies are combined [12–14]. However, a significant drawback of domestic sewage treatment plants with activated sludge and biological reservoir is the high sensitivity to unevenness of the amount and composition of incoming sewage. The activated sludge treatment plants are also not resistant to periodical shortages of electrical energy (pump and blower interruptions).

Small volumes of wastewater can be treated in wetland systems, both one-stage as well as hybrid ones [15–18]. The efficiency of pollution removal in these systems has been the focus of numerous works [3,4,15,19–23]. Increasing attention is drawn to the reliability of wetland systems, especially with respect to the removal of the biogenic compounds nitrogen and phosphorus [24–31]. However, there are few studies pertaining to the technological reliability and efficiency of biogenic compounds removal over the course of a long-term operation of wetland systems [32–34].

Legal regulations currently in force in Poland and many other countries worldwide stipulate that wastewater treatment plants should be monitored for the concentration of biogenic indicators, such as nitrogen and phosphorus, only when the wastewater is discharged to standing waters or their direct tributaries. However, taking into account the increasing number of small wetland systems, discharging of biogenic compounds with treated wastewater into flowing waters is becoming an important issue [35–39]. The specific nature of the treatment plants considered, as well as the possibility of efficient removal of biogenic compounds are also important [40].

The purpose of the present study was to analyze the technological reliability and efficiency of removal of biogenic compounds (total nitrogen and total phosphorus) over a period of 14 years in a one-stage horizontal subsurface flow constructed wetland (HSFCW) planted with the basket willow *Salix viminalis* L.

2. Material and methods

2.1. Characteristics of the experimental facility

The monitored facility was located in the town of Jastków, in south-eastern Poland (51°18′N, 22°26′E). Since 1994 it had been used for the treatment of regular domestic wastewater (kitchen, toilet, shower, washing machine etc). The flow rate of the treatment plant was 1.2 m^3/d during the study period. The average hydraulic retention time (HRT) was 49.6 days and hydraulic loading rate (HLR) was 0.006 $m^3/m^2/d$. The facility consisted of two-chambers preliminary sedimentation tank of 13.7 m^3 of active volume and singe bed with horizontal flow of 1.2 m deep and with a surface area of 186 m^2 and a 1% slope. To protect the grand water against the contamination 1 mm HDPE (High Density Polyethylene) foil lining was used for sealing the bed. The bed was filled with a medium sand with a top layer of humus in which the willow *Salix viminalis* L. was planted with density 4 seedlings per 1 m^2 (Fig. 1). The willow was harvested and removed from the bed in each March during the monitoring period. Effluent from the plant was diverted to a pond with a surface area of 1190 m^2 [41].

In 2008, at the end of the research period, clogging of the tested system was observed, which was manifested by the accumulation of wastewater on the surface of the bed. Due to this operational problems which was not possible to overcome the owner took the decision to close this facility in 2011, after 17 years of its operation.

2.2. Analytical methods

Analyses of the quality of raw wastewater and treated effluent were carried out for the data collected in the years 1997–2010. During this study period, 56 sampling events were performed and 112 wastewater samples were collected to analyze the following parameters: temperature of wastewater, concentration of dissolved oxygen, pH, nitrate and nitrite nitrogen, ammonia nitrogen, total nitrogen and total phosphorus. TSS (Total Suspended Solids), BOD_5 (Biochemical Oxygen Demand) and COD (Chemical Oxygen Demand) were also investigated in this treatment plant but the results of this study were presented in a previous paper [41].

Samples were collected four times a year, once each season: in February (winter), May (spring), August (summer), and November (autumn). The temperature of wastewater, concentration of dissolved oxygen and pH were determined using a Multi 340i handheld meter from WTW. Nitrate and nitrite nitrogen were determined using a Slandi LF 300 photometer, and ammonium nitrogen was determined using a PC Spectro spectrophotometer manufactured by AQUALYTIC. Total nitrogen was determined using the AQUALYTIC PC Spectro spectrophotometer, after oxidation of the samples at 100 °C in a WTW CR4200 thermo reactor. Total phosphorus was determined using an MPM 2010 spectrophotometer manufactured by WTW, after oxidation of the samples at 120 °C in a WTW CR4200 thermo reactor. Sampling, sample transportation, processing and analysis were done according to the relevant Polish Norms PN-74/C-04620/00 [43] and EN 25667-2 [44] which are compatible with APHA [45,46].

Biogenic compounds in samples of wastewater were analyzed according to the following methods which are recommended by the ordinance of the Minister of Environment in Poland [47]:

- Ammonium nitrogen – spectrophotometric method according to: PN-ISO 7150-1 [48],
- Nitrate nitrogen – spectrophotometric method according to: PN-C-04576-08 [49],
- Nitrite nitrogen – spectrophotometric method according to: PN-EN 26,777 [50],
- Total nitrogen – spectrophotometric method according to: PB-NL-FCH-11 [51],
- Total phosphorus – spectrophotometric method according to: PN-EN ISO 6878 [52].

2.3. Statistical analysis

The analysis of the obtained results was carried out in four stages. The first stage involved determination of the frequency of occurrence of characteristic total nitrogen and total phosphorus concentrations in the influent and effluent wastewater. The guidelines provided by Jóźwiak and Podgórski [53] were used in order to determine the size and range of classes.

$$k \leqslant 5\log n \quad (1)$$

where k – number of classes ($5 < k < 20$), n – abundance of the sample.

On the basis of formula (1), 5 classes with a span of 20 mg/l and 5 classes for total phosphorus with a span of 15 mg/l have been determined for effluent from total nitrogen. In the effluent treated for total nitrogen, 5 classes with a span of 10 mg/l were established and 5 classes with a span of 2.5 mg/l were established for total phosphorus (Figs. 2 and 3).

The classes for the particular pollutant indicators were selected so that the frequency distribution was as detailed as possible, without affecting the clarity of the structure of the statistical set.

In the second stage of the analysis, regarding the results obtained on the basis of the mean concentrations of the analyzed pollutant indicators in the influent (C_d) and the effluent (C_o) collected from the

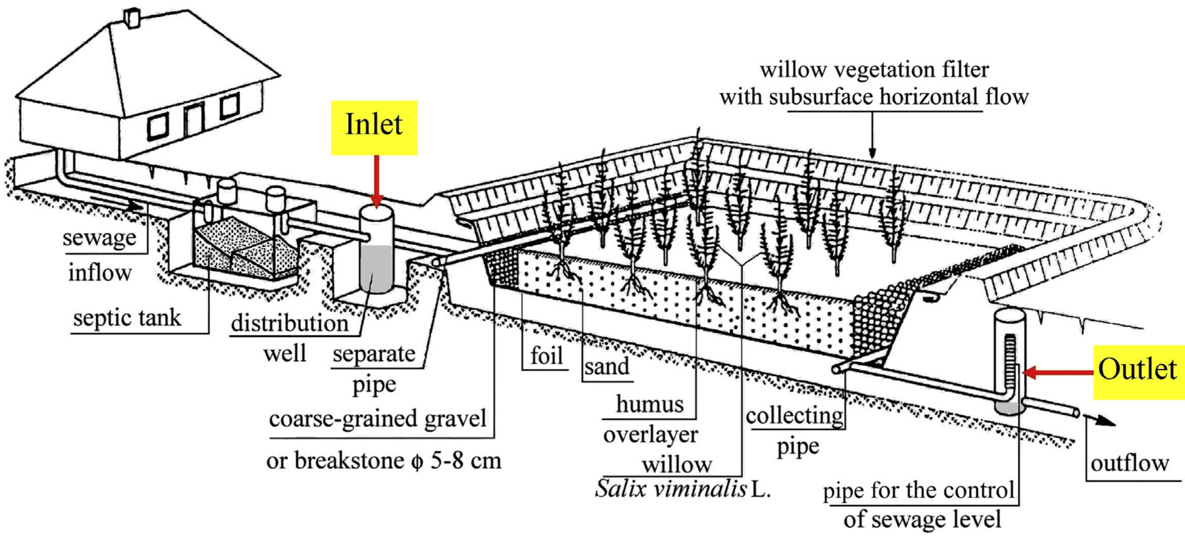


Fig. 1. Technological scheme of a one-stage constructed wetland with subsurface horizontal flow (based on a design by Drupka et al. [42] and Jóźwiakowski et al. [41]).

constructed wetland, the efficiency of nitrogen and phosphorus removal was calculated according to the formula:

$$D = 100 \cdot [1 - C_o/C_d], \% \quad (2)$$

In the third stage, the efficiency of nitrogen and phosphorus removal was determined using the Weibull reliability theory. The assumed permissible values of the analyzed parameters represent the Polish standards for the concentrations of total nitrogen (30 mg/l) and total phosphorus (5 mg/l) in sewage discharged from treatment plants of up to 2000 PE (population equivalents) into lakes and their tributaries.

The operation reliability of the wastewater treatment plant was determined with the Weibull method. The Weibull distribution is a useful general probability distribution, which can also be used to determine the reliability of operation of a wastewater treatment plant [26,31,41]. The Weibull distribution is characterized by the probability density function (3) with the parameters b, c, and θ:

$$f(x) = \frac{c}{b}\cdot\left(\frac{x-\theta}{b}\right)^{(c-1)}\cdot e^{-\left(\frac{x-\theta}{b}\right)^{c}} \quad (3)$$

where x is a variable defining the concentration of a given contamination parameter in treated wastewater, b – scale parameter, c – shape parameter, θ – location parameter.

It is assumed that $\theta < x$, $b > 0$, $c > 0$.

The Weibull distribution parameters were estimated by the maximum likelihood method. The goodness-of-fit of the Weibull distribution to the empirical data was evaluated with the Hollander–Proschan test. The results were analyzed using STATISTICA 8 software.

In the fourth stage of the analysis, the correlation relationship pertaining to the influence of the operation time on the efficiency of total phosphorus and total nitrogen removal was determined. The correlation analysis was based on the Pearson correlation coefficient, where the independent working time of the object (horizontal axis) was an independent variable, and the dependent variable was the dependent concentration of total nitrogen and the concentration of total phosphorus in treated wastewater (vertical axis).

3. Results

3.1. Pollutant concentrations in influent and effluent from constructed wetland

Total nitrogen and total phosphorus concentrations in the influent of the analyzed wastewater treatment plant, examined over the 14 year period, in a majority of cases corresponded to the average levels described in the literature, which meant the wastewater could be classified as household sewage [31,54]. The characteristic values of total nitrogen, total phosphorus and other indicators in the influent and effluent wastewater are presented in Table 1.

The pH of inflowing wastewater fluctuated, almost imperceptibly, between 7.03 and 8.10. By contrast, the temperature of the influent during the investigated period varied appreciably over a range from 0.5 to 22.0 °C. The concentration of dissolved oxygen in the wastewater inflowing to the treatment plant was very low, ranging from 0.08 to

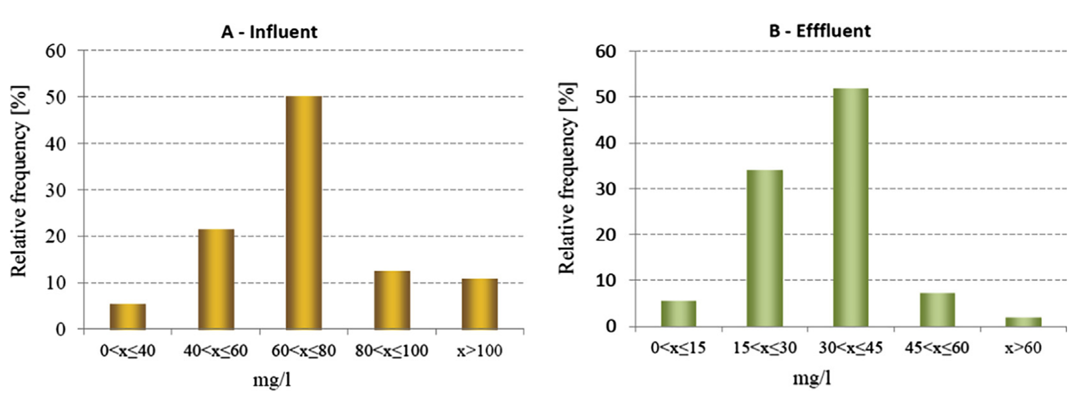


Fig. 2. Histogram of total nitrogen concentrations in the influent (A) and effluent (B).

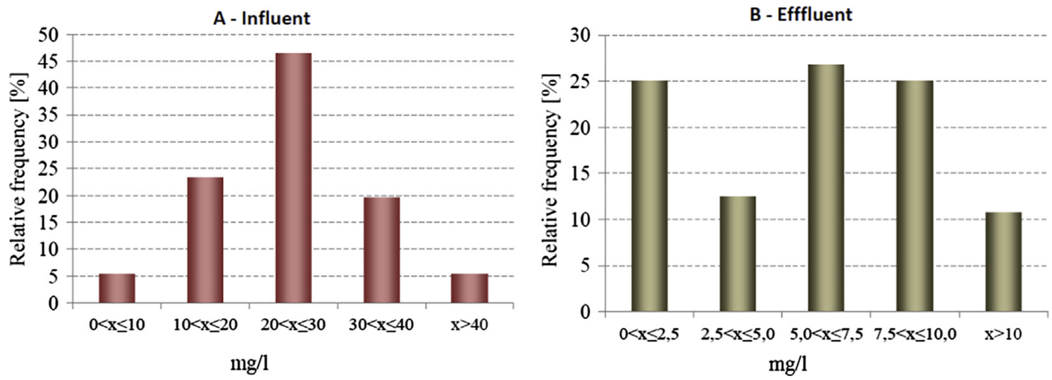


Fig. 3. Histogram of total phosphorus concentrations in the influent (A) and effluent (B).

Table 1
Statistics characterizing the concentrations of different indicators at the inflow and outflow of the investigated HSFCW system during long-term operation (14 years).

Parameters		Statistics indicators					
		Average	Median	Min	Max	SD	Cv
Temperature of wastewater [°C]	In	11.7	12.6	0.5	22.0	6.1	0.52
	Out	10.7	11.7	0.0	21.4	6.4	0.60
Dissolved oxygen [mgO_2/l]	In	0.50	0.30	0.08	3.21	0.60	1.21
	Out	2.10	1.65	0.20	8.10	1.40	0.69
pH	In	7.49	7.48	7.03	8.10	0.3	0.03
	Out	7.05	7.10	6.18	7.68	0.3	0.05
BOD_5[a] [mg O_2/l]	In	163.2	161.0	62.0	309.0	54.1	0.41
	Out	21.7	20.0	6.0	61.0	12.7	0.71
COD[a] [mg O_2/l]	In	329.8	338.0	101.0	580.0	97.2	0.34
	Out	57.8	52.0	9.0	134.0	29.0	0.60
TSS[a] [mg/l]	In	89.9	72.5	10.9	239.0	56.8	0.62
	Out	29.7	29.0	4.2	57.0	15.2	0.51
Total nitrogen [mg/l]	In	71.5	70.4	37.1	137.0	21.5	0.30
	Out	32.9	33.0	9.0	92.0	12.8	0.39
Ammonium nitrogen [mg/l]	In	55.6	55.6	20.2	88.0	15.6	0.28
	Out	23.8	24.5	3.9	52.6	9.1	0.38
Nitrate nitrogen [mg/l]	In	0.40	0.16	0.11	2.09	0.5	1.36
	Out	3.00	0.98	0.01	22.6	4.5	1.50
Nitrite nitrogen [mg/l]	In	0.10	0.127	0.006	0.820	0.10	0.98
	Out	0.20	0.071	0.006	1.428	0.20	1.48
Total phosphorus [mg/l]	In	24.8	25.2	5.2	42.8	8.8	0.36
	Out	6.1	6.2	0.1	17.9	3.7	0.61

Notation: In – Inflow; Out – Outflow; SD – Standard deviation; Cv – Coefficient of variation.
[a] From Jóźwiakowski et al. [41].

3.21 mg/l, with an average of 0.5 mg/l. High concentrations of ammonium nitrogen (20.2–88.0 mg/l) and low levels of nitrate nitrogen (0.11–2.09 mg/l) and nitrite nitrogen (0.006–0.820 mg/l) in wastewater inflowing to the treatment plant were also obtained throughout the study period (Table 1). In another paper, it was shown that the average concentration of total suspended solids in the wastewater inflowing to the treatment plant was 90 mg/l and the BOD_5 and COD values were 163 and 330 ml/l, respectively [41].

The average concentration of total nitrogen in the influent wastewater amounted to 71.5 mg/l, whereas the median equaled 70.4 mg/l. Incidentally, this indicator reached the maximum and minimum values of 137.0 and 37.1 mg/l, respectively, but exhibited a low variation of $C_v = 0.30$.

In the treated wastewater, the mean total nitrogen concentration was 32.9 mg/l. The median was comparable at 33.0 mg/l. The extreme concentrations of total nitrogen equaled 92.0 and 9.0 mg/l. The coefficient of variation of this parameter in the effluent was $C_v = 0.39$.

As far as total phosphorus was concerned, its average concentration in the influent wastewater was 24.8 mg/l, with a median of 25.2 mg/l. The maximum concentration reached 42.8 mg/l, and the minimum was 5.2 mg/l. The coefficient of variation of total phosphorus in the influent was slightly higher than in the case of total nitrogen and amounted to $C_v = 0.36$.

In the treated wastewater, the mean concentration of total phosphorus equaled 6.1 mg/l. The median value was almost the same at 6.2 mg/l. A high variability of total phosphorus ($C_v = 0.61$) was observed in the effluent samples. The extreme concentration values of total phosphorus in the treated wastewater ranged from 0.1 to 17.9 mg/l.

In the next part of this manuscript, the frequency of occurrence of total nitrogen and total phosphorus concentrations in the wastewater during treatment in analyzed object is discussed.

Based on formula (1) for the analysis of total nitrogen concentrations in the influent wastewater, five concentration ranges with a span of 20 mg/l were adopted. Fig. 2 shows that in 50% of cases, the concentrations of total nitrogen ranged from 60 to 80 mg/l. The range from 40 to 60 mg/l corresponded to 21.4% of cases. Therefore, in 71.4% of cases, total nitrogen concentrations in the influent wastewater ranged from 40 to 80 mg/l. In the course of the monitoring, it was observed that in 5.4% of cases, total nitrogen concentrations in the influent were lower than 40.0 mg/l, while in 10.7% of cases, the concentrations were very high and exceeded 100 mg/l.

For total phosphorus analysis in influent wastewater, five ranges spanning 10 mg/l each were assumed. In 46.4% of cases, total phosphorus concentrations in the influent wastewater were in the range from 20 to 30 mg/l. The ranges from 10 to 20 mg/l and 30 to 40 mg/l corresponded to 23.2% and 19.6% of cases, respectively. In 5.4% of cases, total phosphorus concentrations were below 10 mg/l, while in 5.4% of cases the concentrations exceeded 40 mg/l. The characteristic distribution of the particular total phosphorus concentrations in the influent wastewater is presented in Fig. 3.

For the analysis of characteristic total nitrogen concentrations in the treated wastewater, five classes with a span of 15 mg/l were assumed. The limit values of the class ranges were set so that the upper limit value of one range was equal to 30 mg/l, which corresponded to the maximum permissible concentration of total nitrogen in treated wastewater according to the assumed criteria. Total nitrogen concentrations of up to 15 mg/l and up to 30 mg/l were found in 15 out of the 56 conducted analyses, adding up to 39.3% of cases. In the remaining cases, i.e. almost 61%, the concentrations exceeded the limit value. Most often (approximately 52% of cases), the concentrations of total nitrogen in the treated wastewater ranged from 30 to 45 mg/l. In five out of the 56 analyzed samples of treated wastewater, the total nitrogen concentration exceeded 45 mg/l, which corresponded to almost 9% of

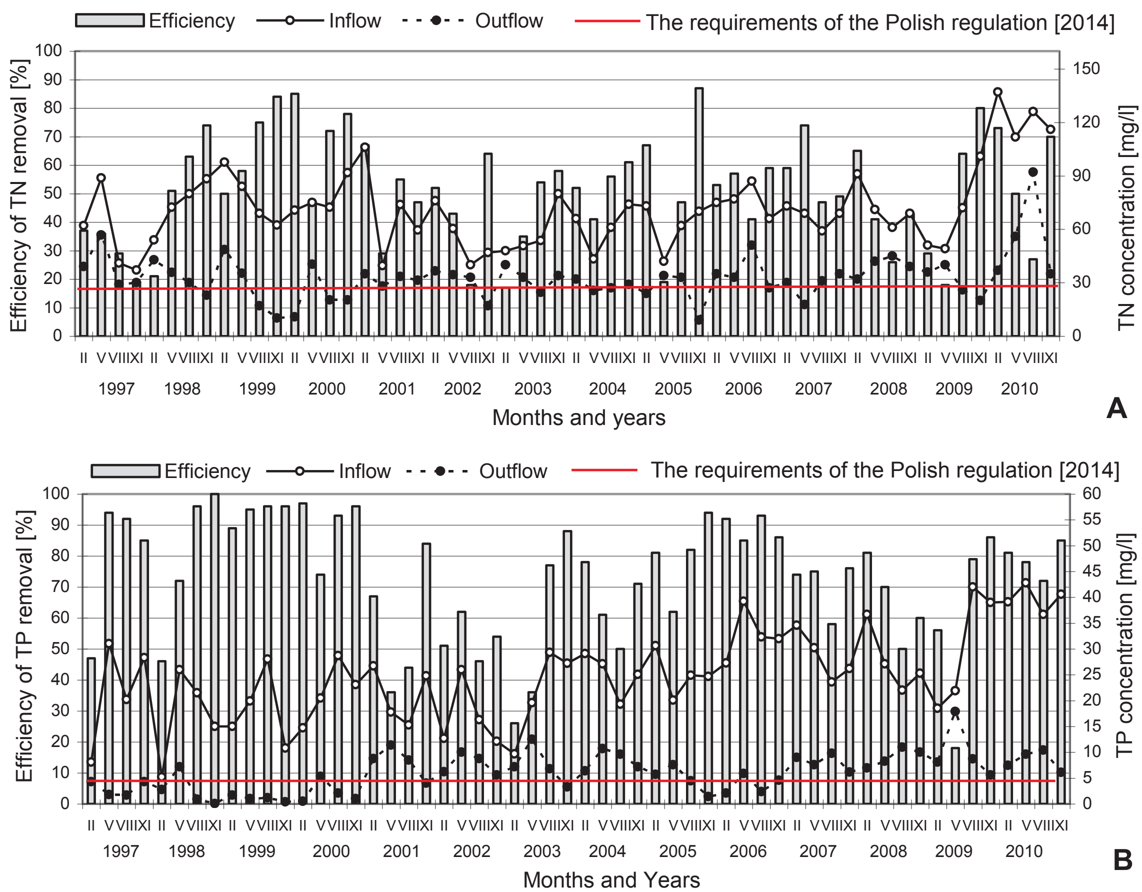


Fig. 4. Efficiency of removal of biogenic compounds and their concentration in the influent and effluent during long-term operation of the one-stage constructed wetland system (A: TN – total nitrogen; B: TP – total phosphorus).

cases. Detailed distribution data for characteristic total nitrogen concentrations in treated wastewater are presented in Fig. 2.

The analysis of characteristic total phosphorus concentrations in the effluent was also conducted for five class ranges with a span of 2.5 mg/l. Similarly to the previous case, a limit value was assumed while determining the class ranges, corresponding to the permissible concentration of total phosphorus in treated wastewater, which equals 5 mg/l. The concentrations of total phosphorus up to 2.5 and up to 5 mg/l were found in 21 cases, i.e. 27% of samples. In 29 out of the 56 cases, the phosphorus concentration in treated wastewater ranged from 5 to 7.5 mg/l and from 7.5 to 10 mg/l. This corresponded to almost 52% of cases. In six cases (10.7%), total phosphorus concentrations were higher than 10 mg/l. Detailed distribution data for characteristic total phosphorus concentrations in treated wastewater are given in Fig. 3.

3.2. *Efficiency of biogenic compounds removal*

Fig. 4 presents the results of analyses of the composition of the influent and effluent wastewater as well as the efficiency of biogenic compounds removal in the one-stage constructed wetland system over the many years of its operation. Table 2 shows the statistical characteristics of the efficiency of removal of biogenic compounds.

3.2.1. *Total nitrogen and ammonium nitrogen*

According to Brix [55] pollutants removal (mainly nitrogen) in constructed wetland systems are primarily related to the effect of a biological membrane which is being created during the flow of sewage through the soil. Plants play an auxiliary role in the process of treatment. Oxygen zones are formed around the plant roots, while areas with poor oxygenation occur at the remaining parts of the bed.

Table 2
Statistical characteristics of the efficiency of biogenic compounds removal during long-term operation of the one-stage constructed wetland system.

Parameters	Statistics				
	Average	Min.	Max.	Standard deviation	Coefficient of variation
Total nitrogen removal [%]	51.3	16.7	87.1	18.9	0.37
Ammonium nitrogen removal [%]	53.7	−7.4	92.3	21.3	0.40
Total phosphorus removal [%]	72.7	18.3	99.6	19.9	0.27

Microfilms of different bacteria develops on the soil particles and on the plant rots and rhizomes, species composition is adapted to the variable trophic conditions. The microorganisms decompose organic matter in aerobic and anaerobic processes. The plant roots and rhizomes maintain the filtration properties of the bed by loosening its internal structure [56]. The developed system of rhizomes and roots of the below-ground parts of the plants ensures their intensive growth. The major removal mechanism of nitrogen in CWs is nitrification and denitrification. Volatilisation, adsorption and plant uptake play a much less important role in nitrogen removal [57,58].

It was observed that the efficiency of removal of total nitrogen and ammonium nitrogen during long-term operation of the investigated system was diversified. Total nitrogen was removed with an efficiency ranging from 16.7% to 87.1%, whereas the average elimination rate reached 51.3% (Table 2). The literature data indicate that the removal efficiency of total nitrogen in one-stage constructed wetland systems is low and usually ranges between 40 and 43% [16,19]. An exceptionally high total nitrogen elimination efficiency (76%) was obtained in Portugal in a one-stage constructed wetland system with reed located in the Capinha region [21]. Mayo and Bigambo [59] showed, in model studies, that the efficiency of total nitrogen removal in wetland systems may reach up to 48.9%. According to these authors, the denitrification process enables the removal of 29.9% of nitrogen, an additional 10.2% of nitrogen is taken up by plants, and 8.2% undergoes sedimentation. Vymazal [19] claimed that it is impossible to achieve a total nitrogen removal exceeding 50% in one-stage constructed wetland systems with subsurface horizontal flow which are used to treat of domestic wastewater due to the lack of adequate conditions for nitrification and mainly because of the lack of a quick and evenly distributed supply of oxygen to the cells of the microorganisms involved in the biological decomposition of pollutants. In this present study, it was shown that during the long-term operation, the analyzed system provided an average efficiency of ammonium nitrogen removal of only in 53.7% and 51.3% respectively (Table 2). The results of study presented in this work confirmed the previous experience of Vymazal [19].

On the basis of data given by Jóźwiakowski et. al. [41] as regards to BOD_5 and total nitrogen concentration in effluents, it was found that the concentration of total nitrogen in effluent discharged from the monitored facility is dependent on two parameters. In the first case, it was found that the concentration of total nitrogen in effluent depends on the concentration of this parameter in inflowing effluent, while in the second case it was found that the concentration of total nitrogen in outgoing wastewater is influenced by the ratio of BOD_5/TN in incoming wastewater.

In both cases the correlations determined by the coefficient of correlation $R_{x,y}$ according to the scale of Stanisz [60] are at the average level and amount to $R_{x,y} = 0.38$ and $R_{x,y} = -0.32$ respectively. The significance of the calculated correlation coefficients was investigated by *t*-Student's test at the level of significance $\alpha = 0.05$. In both cases, the statistical significance of the tested correlations was found. Since both factors may simultaneously affect the concentration of total nitrogen in the effluent, a nomogram (Fig. 5) described in the equation $TN_{outflow} = 339917 + 0.1406 \times TN_{inflow} - 4.4771 \times BOD_5/TN$ has been developed. On the nomogram on the "x" axis, there is the first independent variable, i. e. the range of total nitrogen concentration in incoming wastewater, on the "y" axis, the second independent variable, i. e. the range of BOD_5/TN ratio, whereas on the "z" axis there is a dependent variable, i. e. the concentration of total nitrogen in treated wastewater. The first of the described dependencies could be explained by the fact that nitrification and denitrification processes occurring in the reservoir were not resistant to high dynamics of changes in nitrogen compounds concentration in incoming effluents. The microorganisms inhabiting the deposit did not multiply quickly enough when sewage inflows contain high concentrations of total nitrogen and therefore a significant part of ammonium nitrogen was not distributed in the deposit in the nitrification process and was discharged with treated sewage. Explaining the second relationship, i. e. the effect of BOD_5/TN on the efficiency of nitrogen removal of nitrogen compounds in the bed, it could be stated that the higher the content of organic matter expressed by BOD_5, which was the nourishment for the micro-organisms responsible for the denitrification process, the higher the intensity of this process and thus the nitrogen concentration in the effluent from the plant was lower.

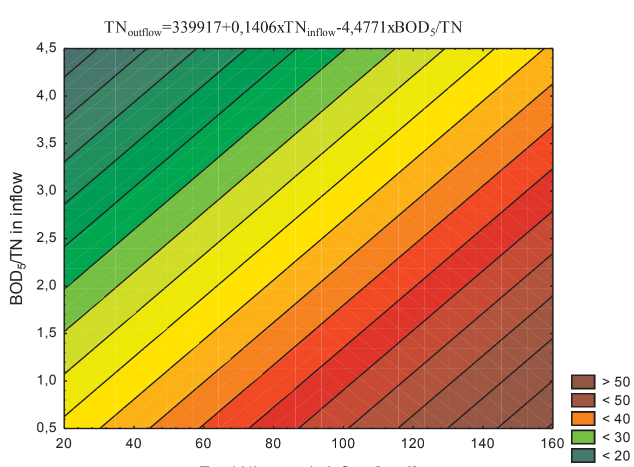


Fig. 5. Nomogram for forecasting the concentration of total nitrogen in treated wastewater according to the concentration of total nitrogen and the ratio BOD_5/N_{og} in inflowing wastewater.

In order to create suitable conditions for nitrification and improve the elimination of ammonia nitrogen in constructed wetland systems, aeration of wastewater may be employed. Jamieson et al. [61] observed an increase in the removal efficiency of NH_4-N from 50.5% to 93.3% after aeration of wastewater was carried out in a subsurface system with horizontal flow. Another study [62] showed that application of an unconventional method of oxygenation of wastewater inflowing to a sand filter provided better ammonium nitrogen removal from domestic wastewater. Jóźwiakowski et al. [62] showed that use of a 0.1% hydrogen peroxide solution can increase the efficiency of removing ammonium nitrogen from wastewater from 31 to 81%. This study indicates that oxygenation of wastewater with hydrogen peroxide can be applied only for the optimization of the nitrification process in wastewater treatment plants.

3.2.2. *Total phosphorus*

The results of the measurements showed that total phosphorus was removed with an efficiency from 18.3% to 99.6%, whereas the average elimination rate reached 72.7% (Table 2). Previous results obtained in one-stage constructed wetland systems indicate that the average efficiency of total phosphorus removal in such objects usually amounts to 32–50% [16,19]. Brix et al. [63] demonstrated that a high efficiency of phosphorus removal in constructed wetland systems could be achieved by supplementing the substrate with high sorption capacity materials. For example, an exceptionally high total phosphorus elimination (99%) was observed in the Longdao River, in Beijing (China), in which the system was filled with a mixture of materials characterized by a high sorption capacity (including gravel with iron, dolomite, and limestone) [64]. Phosphorus sorption is conducive to de-acidification and alkalization of wastewater [65].

3.3. *Technological reliability of biogenic compounds removal*

The reliability of the considered wetland system was determined using the Weibull reliability method [26,31,41]. A hypothesis that the Weibull distribution could be used to approximate the empirical data was verified for the estimated parameters of distribution. The probability values p for the two indicators showed that the empirical data could be described with the Weibull distribution, i.e. assuming that the null hypothesis was true. The goodness-of-fit of the Weibull distribution to the estimated parameters was tested using the Hollander-Proschan test, the results of which are presented in Table 3.

Fig. 6 shows the horizontal axis of total nitrogen (A) and total phosphorus (B) in treated wastewater. Vertical axis shows the reliability given in % on a scale from 0 to 100. The graph shows the distribution with a 5% confidence interval. The graph shows the function dependence of the variable y on the independent variable x.

The technological reliability of the analyzed wetland system regarding total nitrogen removal to the required limit concentration of 30 mg/l was 45%, which is presented in the model devised on the basis of the Weibull distribution, shown in Fig. 6A. This means that permissible concentrations of this element in the effluent wastewater could be observed on almost 201 days. According to the guidelines proposed

Table 3
Results of the estimation of the Weibull distribution parameters together with the measures of goodness of fit to empirical data.

Index	Distribution parameters			Hollander-Proschan test	
	b	c	θ	Test value	p[a]
Total nitrogen	36.882	2.599	0.555	0.00503	0.9959
Total phosphorus	6.676	1.539	0.000	−0.56025	0.5753

Notation: b – scale parameter, c – shape parameter, θ – location parameter.
[a] p – probability value; if $p \le 0.05$, then the null hypothesis that the empirical data can be described with the Weibull distribution must be rejected. If $p \le 0.05$, the zero hypothesis is rejected, which is: empirical data can be described by Weibull distribution. In both cases, p is greater than 0.05.

by Andraka and Dzienis [66], a treatment plant characterized by a PE (Person Equivalent) lower than 2000 should operate with a reliability of at least 97.3%, and a producer's risk of α = 0.05. In other words, such a treatment plant is allowed to be inoperable for a maximum of 9 days per year. In the plant investigated in the present study, this number was considerably exceeded: when the number of days during which the total nitrogen concentration in the treated wastewater exceeded the limit value was compared to the number of days in which the exceeded value had no negative impact on the assessment of plant operation, it was found that the concentrations of this parameter in the effluent were excessive on as many as 192 days a year.

The technological reliability of removal of phosphorus compounds to the maximum concentration of 5 mg/l was 48%, as shown in the model devised on the basis of the Weibull distribution in Fig. 6B. The total phosphorus removal efficiency in the analyzed system indicates that the limit value of this parameter can be exceeded on almost 190 days of a year. In line with the afore-mentioned guidelines, excessive total phosphorus concentrations negatively impact the assessment of the system's phosphorus removal efficiency 181 days a year. Because phosphorus removal occurs mainly as a result of adsorption on the surface of soil grains in the filter, an attempt was made to show the dependency of the sorption capacity of a system on its operation time.

3.4. Influence of operation time on the concentration of biogenic compounds in the effluent

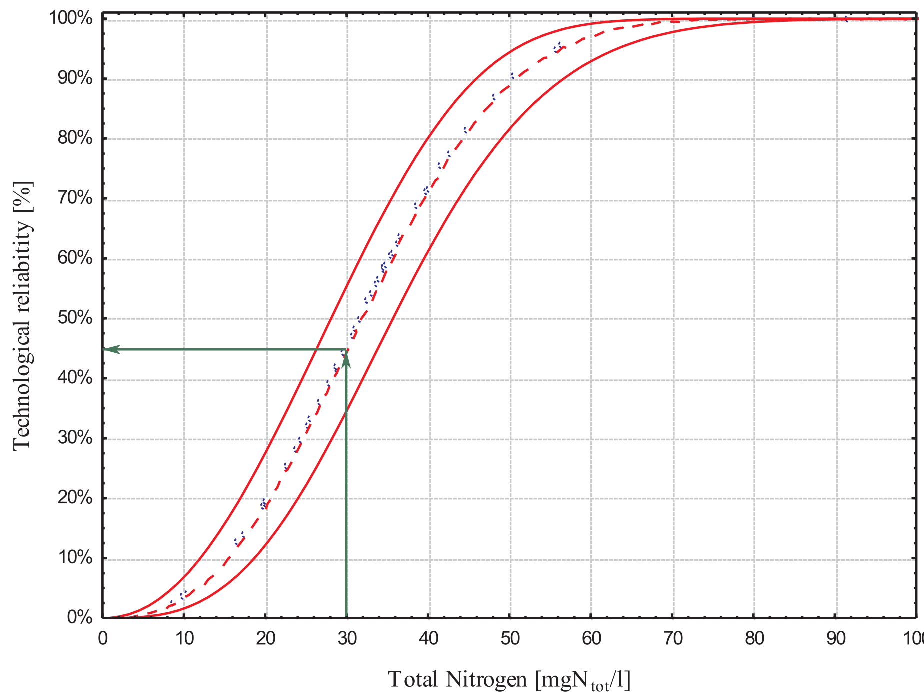
An analysis of the effect of the operation time of the facility under study on the concentration of total nitrogen in treated wastewater was conducted as well (Fig. 7A). The calculated correlation between total nitrogen in effluent wastewater and the days of operation of the treatment plant was $r_{xy} = 0.22$. This correlation is weak. Additionally, if the incidental value – amounting to 92.0 mg/l – is considered an outlier and excluded, the correlation drops down to $r_{xy} = 0.10$, which, according to Stanisz [60] is a very weak correlation. The regression model, which is described with an R^2 equation, only explains 4.6% of the observed dependency. Thus, it can be stated that there is no relationship between total nitrogen in treated wastewater and the operation time of the treatment plant. A similar finding was reported by Pierzgalski et al. [67], who investigated a pilot-scale vertical subsurface flow plant and observed a very high removal efficiency of N-compounds from the very beginning of plant operation which could not be attributed to microbial activity (nitrification/denitrification). The author suggested that in the first phase of operation, adsorption processes were responsible for nitrogen reduction in wastewater, with biological processes gaining advantage as operation time increased and the microbial community became larger. Also Wojciechowska et al. [30] confirmed the importance of adsorption processes for the removal of N-compounds in the startup period of constructed wetland operation.

The correlation between the operation time of the constructed wetland system (independent variable) and the concentration of total phosphorus in the treated wastewater (dependent variable) was $r_{xy} = 0.55$. According to Stanisz [60], such a level of correlation is considered high on the assumed scale. Therefore, there is a strict dependency between these variables, i.e. the sorption capacity of a constructed wetland system decreases in time. In the analyzed case, the correlation was statistically significant, reaching $\alpha = 0.05$, and the test value amounted to $t = 0.000013$. On the basis of the equation describing the regression line presented in Fig. 7B, it can be stated that total phosphorus in treated wastewater increased by 0.0014 mg/l along with the operation time over the consecutive days. Thus, in each consecutive year of system operation, the total phosphorus concentration in treated wastewater increased by 0.51 mg/l. Taking into account the analyzed 14-year system operation period, total phosphorus in the effluent wastewater increased by about 7.15 mg/l in relation to the initial operation period. The results indicate that in the course of long-term operation of the considered object, the phosphorus removal efficiency dropped as a result of the decreasing sorption capacity of the material filling the filter. A situation like this has been drawn attention to by Vymazal [68].

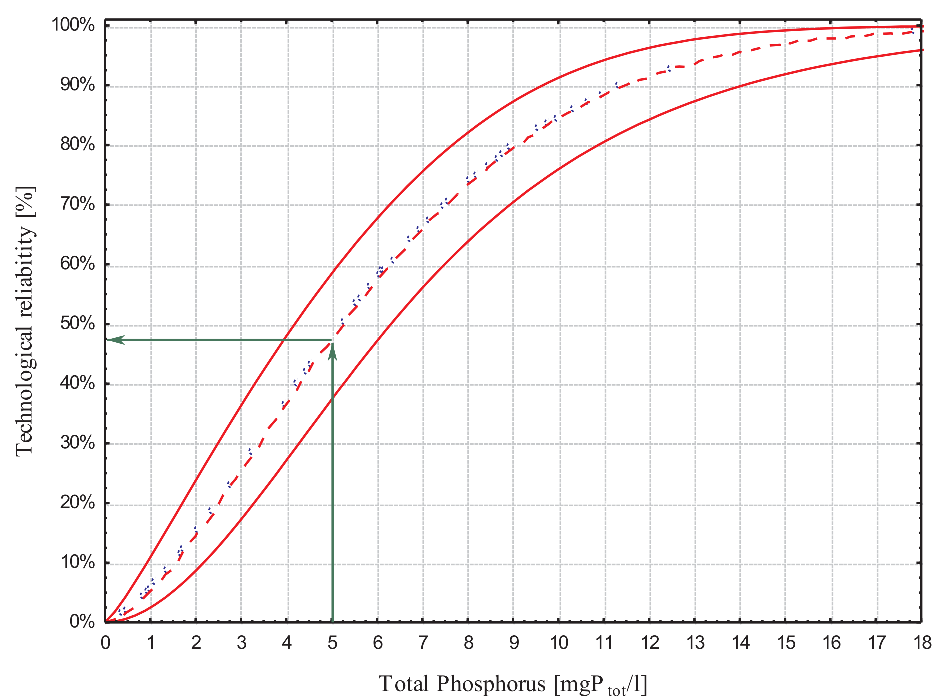
4. Discussion

The results of the 14-year long research on the reliability and efficiency of removal of biogenic compounds in a one-stage wetland system with horizontal flow show that the performance of the system was not satisfactory. Treatment plants of this type, as indicated by numerous authors, are highly efficient in eliminating organic matter expressed as BOD_5 and COD [16,19,21,41]. However, in the case of biogenic compounds, which include nitrogen and phosphorus, the general efficiency of such facilities is much lower. Also, small treatment plants rarely achieve an efficiency comparable to larger objects. There are many reasons for such a state of affairs, but the most important ones include simple design and a lack of constant control over the treatment processes. However, taking into account the economic arguments resulting from the guidelines found in WFD/2000/60 UE [69] and in The Council Directive 91/271/EEC [70] concerning urban wastewater treatment, which preclude the construction of sewerage and collection systems of wastewater treatment plants in non-urbanized zones, the best possible solutions pertaining to the neutralization of wastewater in the place of its origin should be sought and tested. Undoubtedly, constructed wetland systems are one such solution. They blend well with the environment and efficiently removal organic matter and total suspended solids. In order to improve the efficiency and reliability of neutralization of nitrogen compounds, hybrid systems should be designed – incorporating both vertical and horizontal flow of wastewater. Previous studies show that hybrid constructed wetland systems exhibit a much greater reliability as well as efficiency of nitrogen and phosphorus removal [3,4,19,20,22,30,31]. The efficiency of phosphorus removal can be improved by applying P-sorption substrate amendments in a constructed wetland or their use as the last, separate stage of treatment in P-filters filled with carbonate-siliceous rocks [71,72].

In the light of the results obtained in our study, the current guidelines pertaining to the maximum total nitrogen and phosphorus concentrations in treated wastewater discharged from small treatment plants seem confusing. On the one hand – in line with the Polish law – when the effluent is discharged to watercourses, biogenic compounds cannot be accounted for in the evaluation of treatment plant operation. On the other hand, when the treated wastewater is discharged to standing waters, the regulations are the same as in the case of collective treatment plants with more than 2000 PE [47]. According to this regulation, the permissible total nitrogen and phosphorus concentrations in the effluent discharged to standing waters should be less than 30 mg/l and 5 mg/l, respectively. The authors of this work share the opinion that when a large number of small treatment plants operate in a given country, the appropriate regulations pertaining to the removal of nitrogen and phosphorus should be valid for all treatment plants, regardless of their size or the method of sewage discharge. In order to protect water resources against eutrophication, solutions which enable



A



B

Fig. 6. Results of the Weibull reliability analysis for total nitrogen (A) and total phosphorus (B) in the effluent. Notation: dashed red line – reliability function, continuous red line – confidence intervals, green arrows – probability of achieving the total nitrogen limit in the effluent.

a high removal efficiency of biogenic compounds, e.g. hybrid constructed wetland systems, should be recommended [3,4,19,20,22]. Non-conventional methods which aim at improving nitrogen and phosphorus removal in constructed wetland systems can be recommended as well [62,72] instead of the methods used in classic wastewater treatment plants, which are mostly based on activated sludge [73].

On the basis of the experiences of Mucha et al. [34] and Jóźwiakowski et al. [41] it can be said that one-stage constructed wetlands could ensure stable removal of organic matter (OM) and total suspended solids (TSS) in long-term operation in Polish conditions. The quality of effluent for OM and TSS fulfil Polish requirements.

5. Conclusions

It was found that the average total nitrogen and total phosphorus removal efficiencies over the long-term operation (14 years) of the considered plant amounted to 51% and 73%, respectively. The

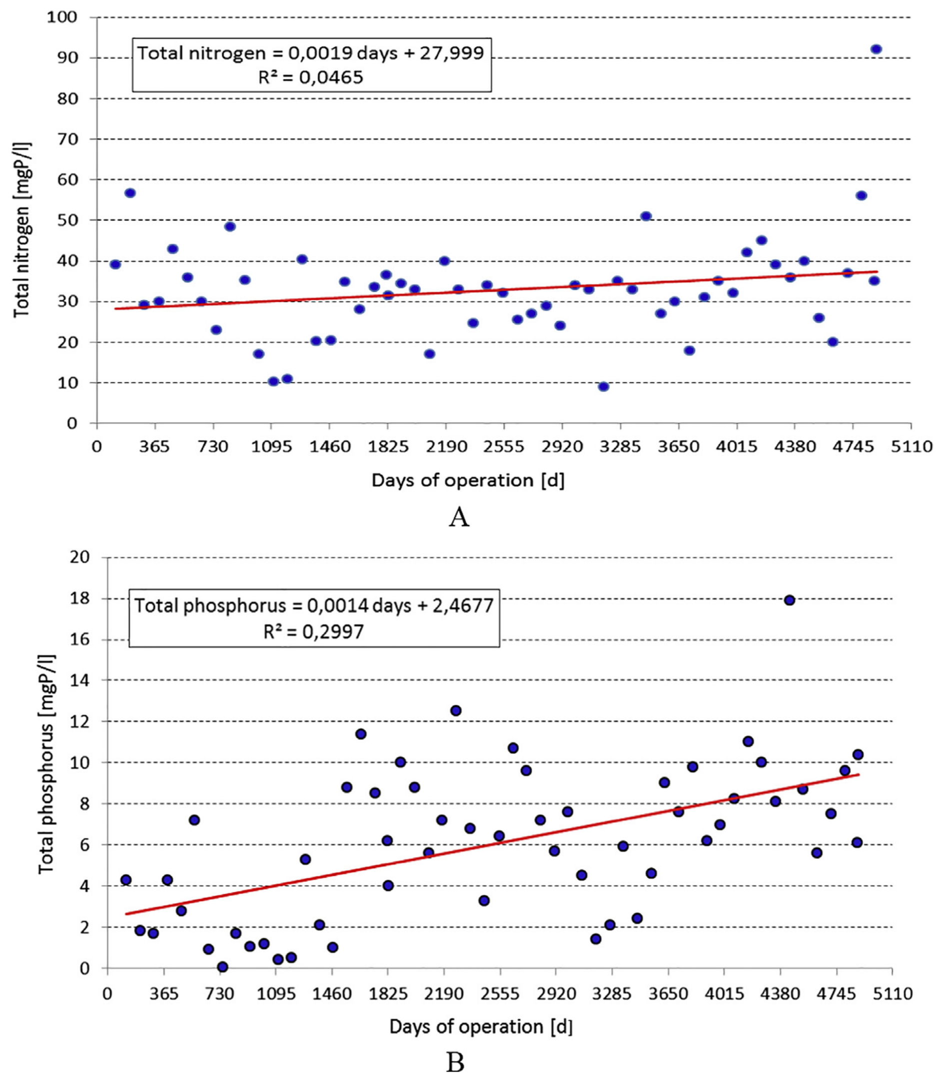


Fig. 7. Concentration of total nitrogen (A) and total phosphorus (B) in the effluent as a function of days of treatment plant operation.

technological reliability, determined by means of the Weibull reliability method, was 45% for total nitrogen removal and 48% in the case of total phosphorus removal. The total nitrogen concentration in treated wastewater did not exceed the limit value in 39.3% of cases, and total phosphorus – in 37.5% of cases. Therefore, the one-stage constructed wetland system did not ensure satisfactory efficiency of biogenic compounds removal during its long-term operation. The high efficiency of total nitrogen removal in one-stage constructed wetland systems with subsurface horizontal flow is impossible to achieve due to the lack of adequate conditions for nitrification. Mainly because of the lack of a quick and evenly distributed supply of oxygen to the cells of the microorganisms involved in the biological decomposition of pollutants. In the case of phosphorus, the effectiveness of its removal is high in the initial period of operation of such treatment plants. But along with the working time, the efficiency of phosphorus removal decreases and in consequence the concentration of phosphorus in effluent wastewater is increasing, which could be explained as a result of the decreasing sorption capacity of the material filling the bed.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.seppur.2018.03.058.

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