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INFLUENCE OF COMPOSITION OF RAW WASTEWATER ON REMOVAL OF NITROGEN COMPOUNDS IN MULTISTAGE TREATMENT WETLANDS

Influence of composition of raw wastewater on the efficiency of organic matter and nitrogen removal in multistage treatment wetlands has been examined. Commonly used indicators, like COD/BOD_5 and BOD_5/TN ratio, determining wastewater susceptibility to biodegradation in conventional treatment technologies were assessed. It was confirmed that multistage treatment wetland may be used for treatment of wastewater with high loads of organic pollutants and unfavorable composition for a biological treatment expressed by the ratio of COD/BOD_5 and BOD_5/TN equal to 2.7 and 0.7, respectively.

1. INTRODUCTION

Nitrification and denitrification are widely accepted as main processes responsible for removal of nitrogen compounds from wastewater in both, conventional and treatment wetland plants [1, 2]. However, nitrification requires aerobic conditions while denitrification occurs in anaerobic environment with dissolved oxygen (DO) below 0.5 mg O_2/dm^3. Denitrification is also sensitive for various carbon sources [2, 3]. Thus, the availability of easy degradable source of carbon expressed as BOD_5 in total organic matter content (COD) in wastewater becomes important for biological nutrient removal (BNR). The most common way for indicating the wastewater susceptibility to biodegradation is based on the ratio of COD/BOD_5. According to Miksch and Sikora [3] when COD/BOD_5 is below 2, wastewater is easily degradable and the efficiency of removal of organic matter is over 90%. The necessity of low carbon-to-nitrogen (expressed, e.g. by BOD_5/TN) ratio is another widely accepted concept for activated sludge or attached growth technologies. Thus effective removal of nitrogen (over 90%) is

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possible only when BOD_5/TN is over 4.0 [3]. To fulfill the requirements of BNR manipulation of aeration and external carbon sources is a common practice in case of high-tech solutions, while in treatment wetland (TW) technology such practices are not usual. In TW technology, vertical subsurface flow (VSSF) beds with intermittent loadings are considered more aerated and thus predestined for oxidation of ammonium nitrogen, while horizontal subsurface flow (HSSF) beds are considered to be more dedicated to denitrification due to limited oxygen conditions [4]. Moreover, the natural property of TW is accumulation of organic matter and the accumulated organics is considered internal carbon source which could be used during denitrification. In this way, wastewater, depending on its origin, can have various compositions and, as a consequence, different biodegradability under treatment processes [1]. Liu et al. [5] proved that VSSF beds loaded with higher rates of COD/TN, like 5 or even 10, in wastewater indicated higher rates of nitrogen removal in comparison to low loaded ones. In the case of VSSF bed with intermittent aerations and COD/TN in wastewater equal to 10, the removal efficiency of TN was 73.7%, while for COD/TN ratio equal of 2.5 it was only 31.9%. It has been suggested that the level of easily degradable organic matter and nitrogen (measured by TKN) to ensure effective nitrification should be like BOD_5/TKN below 1 in treatment wetlands [1]. The analysis of COD/BOD_5 and BOD_5/TN (or BOD_5/TKN) ratios can provide useful information as far as: (i) the rate of biodegradability of the studied wastewater (ii) nitrification capacity of the reactor basing on the quantification of the degradable dissolved organic matter, which is fundamental for effective removal of ammonium nitrogen in the conventional nitrification/denitrification process.

The objective of the paper was to evaluate the influence of raw wastewater composition expressed by COD/BOD_5 and BOD_5/TN ratios on the efficiency of organics and nitrogen removal in multistage treatment wetlands (MTWs). The research has been carried out for the MTWs treating domestic wastewater (DW) and reject water from the digested sludge dewatering (RW). The quality of wastewater and reject water was different, allowing the evaluation of the effect of composition on the efficiency of nitrogen removal.

2. EXPERIMENTAL

Treatment wetlands and experimental design. The investigation was carried out in two local MTWs and the pilot MTW for reject water (RW) from digested sludge dewatering located in the conventional WWTP Wschód. The full-scale MTWs were situated in the villages of Wiklino and Darżlubie in Northern Poland. After treatment in a three chamber septic tank (with two days hydraulic retention time), the wastewater was pumped into the MTWs responsible for biological treatment. MTW in

Wiklino consisted of two HSSF beds (HSSF I and HSSF II), one before and the other following the VSSF bed. The detailed characteristics of MTWs are presented in Table 1.

Table 1

Characteristics of studied multistage treatment wetlands (MTWs)

Plant	Flow [m^3/d] (pe)	Configuration	Substratum [mm]	Contact time [d]	Area [m^2]	Depth [m]	Hydraulic load [mm/d]	Unit area [m^2/pe]
Wiklino	20.5 (220)	HSSF I	2–6	12.3	1050	0.6	19.5	4.7
		VSSF	2–6	–	312	0.4	65.7	1.4
		HSSF II	2–6	6.3	540	0.6	38.02	2.4
Darżlubie	56.7 (750)	HSSF I	2–6	5.1	1200	0.6	47.3	1.6
		Filter K	2–6	–	400	–	141.2	0.6
		HSSF II	2–6	2.1	500	0.6	113.4	0.7
		VSSF	2–6	–	500	1.0	113.4	0.7
		HSSF III	2–6	4.2	1000	0.6	56.7	1.2
Pilot Wschód	0.24 (5)	VSSF I	2–8	–	7.5	0.6	3.2	1.5
		VSSF II	2–8	–	5.0	0.6	4.8	1.0
		HSSF	2–8	4.1	3.9	0.6	28.5	0.78

The MTW in Darżlubie consisted of five stages: three HSSF beds, one VSSF bed (placed as a third stage of the treatment) and the cascade filter (filter K) placed after the first HSSF bed.

The pilot MTW Wschód consisted of two settling tanks (each having the volume of 1 m^3) in the mechanical stage and three subsurface flow beds (two VSSF working in a series and followed by HSSF) in the biological stage. After each stage of treatment, 1 m^3 tank was placed with the pump for collecting and ensuring intermittent feeding of equal single doses of RW for each stage (about 0.11 m^3 twice a day).

Sample collection and analysis. Composite samples (mixed for 6 h) were taken from influent and final effluent, after each stage of treatment following the hydraulic retention time in each stage of treatment (e.g. for Wiklino influent at the beginning (0 day), effluents from HSSF I and influent to VSSF after 12.3 days, effluent from HSSF II after 18.6 days, cf. Table 1). The contact time for VSSF beds, measured with a tracer, equaled 20–22 min in all investigated filters.

Chemical analyses were performed according to standard methods [6, 7]. Concentrations of the following pollutants were measured: organics (BOD_5 and COD) and nitrogen: total Kjeldahl nitrogen (TKN), ammonia nitrogen and nitrates, as well as nitrites (their sum gives total nitrogen TN).

The temperature of wastewater, as well as dissolved oxygen (DO) were measured at the sampling points onsite,while samples were collected using the measuring probe (WTW Multi 340i/SET).

Removal efficiency η, %, was calculated as:

$$\eta = \frac{L_{\text{inf}} - L_{\text{eff}}}{L_{\text{inf}}} \times 100$$

where: L_{inf}, g/d is pollutants' load in influent, L_{eff}, g/d – pollutants' load in effluent after subsequent stages of treatment in the constructed wetland.

Mass removal rate *MRR*, g/(m^2·d), was calculated from the following equation:

$$MRR = \frac{C_{\text{inf}} Q_{\text{inf}} - C_{\text{eff}} Q_{\text{eff}}}{A}$$

where: A, m^2, is the area of TW, Q_{inf} and Q_{eff}, m^3/d – average flow rates of influent and effluent, respectively, C_{inf} and C_{eff}, mg/(dm^3·s) – average concentrations of influent and effluent pollutants, respectively.

3. RESULTS

3.1. QUALITY OF INFLUENT AND EFFLUENT IN THE ANALYZED MTWs

The analyzed wastewaters differ significantly, even those of the same kind like DW from local MTWs in Wiklino and Darżlubie. The inlet concentrations of pollutants of the analyzed DW were much higher in comparison to the literature values for domestic wastewater discharged to TWs in Europe [9–12]. However they were consistent with other results of examinations carried out in Poland and could be considered typical of Polish conditions [13–15]. There are many reasons of such a phenomenon: (1) separate sewer system, (2) smaller water consumption per p.e. than recommended in the national guideline for designing, (3) finally low efficiency of pollutants removal in the mechanical part (Table 2). The average concentrations of TN discharged to the MTW in Wiklino and Darżlubie did not differ significantly, being 130.4 mg/dm^3 and 120.8 mg/dm^3, respectively (Table 2). In the effluent, the dominant form of nitrogen was ammonium, but its share in concentration of TN was different: up to 90% for wastewater in Darżlubie and only 77% for wastewater in Wiklino (Table 3).

Reject water (RW) discharged to the pilot MTW Wschód was characterized by the typical composition as for the liquors generated during mechanical dewatering of the digested sludge [16]. Furthermore, the characteristic of the RW was similar to that of landfill leachate from the mature municipal damping site [17–20]. The share of ammonium nitrogen in TN was 90% in the RW fed to the first tank. Due to the processes occurring in the second tank, the composition of pollutants changed and the share of NH_4^+-N in TN increased to 94.4%.

Table 2

Characteristics of influent wastewater in the analyzed MTWs

Parameter	Wiklino, *n* = 16	Darżlubie, *n* = 12	Wschód, *n* = 20
	Mean±σ min–max		
TSS, mg/dm^3	364.0±55.7 267.4–456.1	504.9±25.9 478.2–562.8	408.7±70.3 320.6–729.8
VSS, mg/dm^3	266.9±38.9 177–301.7	322±23.9 279.9–357.9	365±45.2 262.5–460.3
TN, mg/dm^3	130.4±9.2 118.4–148.0	120.8±4.3 114.3–128.9	788.1±170.9 710.4–1789.2
NH_4^+-N, mg/dm^3	99.5±10.5 92.0–125.4	108.0±3.7 99.5–113.5	744.2±183.3 509–1006.6
Org-N, mg/dm^3	30.6±10.07 18.3–49.9	12.4±2.3 9.9–15.5	46.7±23.9 18.4–88.8
COD, mg/dm^3	714.6±110.7 508.8–932.5	843.8±40.7 791.4–901.5	1022.7±93.5 880.0–1260.0
BOD_5, mg/dm^3	382.1±72.0 280.6–500.7	368.7±16.0 340.2–390.5	378.9±87.0 270.8–569.0
Temperature of wastewater, °C	17.3±0.6 16.4–18.3	12.0±5.0 5.0–18.8	16.4±3.2 9.1–20.6
DO, mg/dm^3	0.2±0.2 0.0–0.9	0.7±0.2 0.4–1.1	0.6±0.2 0.4–0.9
COD/BOD5	1.9	2.4	2.7
BOD_5/TN	3.4	3.1	0.5

In both influents to the local MTWs, the values of the rate of BOD_5/TN were similar but far too low when compared with the recommended values. Although both MTWs were fed only with domestic wastewater (from rural settlements) without supply of industrial waste, their composition indicates that they are not easily susceptible to the conventional biological methods of treatment. The unfavorable composition may result from the higher concentration of pollutants in wastewater which is the effect of reduced water consumption. As a consequence of the reduced amount of water consumption, the treatment time in the septic tank is prolonged, which initiates the starting processes of anaerobic decomposition of wastewater. In this way, a part of organic matter can be decomposed but nitrogen concentration remains unchanged. This could lead to illusory increasing of concentration of total nitrogen in comparison to the concentration of organic matter. In the case of RW, the values of both COD/BOD_5 and BOD_5/TN were lower in comparison to the recommended values indicating that this wastewater is hardly susceptible to the conventional biological degradation.

Table 3

Characteristics of effluent wastewater in the analyzed MTWs

Parameter	Wiklino, n = 16	Darżlubie, n = 12	Wschód, n = 20
	Mean±σ min–max		
TSS, mg/dm^3	24.2±4.8 18.4–32.3	44.4±4.2 40.5–45.7	23.6±6.1 13.3–35.2
VSS, mg/dm^3	15.1±7.9 7.8–17.4	35.7±9.9 25.8–42.1	16.3±3.7 14.8–17.8
TN, mg/dm^3	22.8±3.5 17.9–28.6	10.0±0.7 9.3–11.2	124.2±51.9 45.6–188
NH_4^+-N, mg/dm^3	4.2±1.7 0.4–6.5	4.0±0.2 3.5–4.4	96.0±38.7 30.2–154.6
Org-N, mg/dm^3	4.0±0.6 2.8–5.1	3.8±0.4 3.2–4.7	23.0±16.2 83–66.9
COD, mg/dm^3	84.5±15.7 62.0–114.5	71.3±3.1 66.7–76.3	263.9±38.9 183.6–340
BOD_5, mg/dm^3	15.8±2.7 10.7–20.4	30.4±1.7 27.9–32.5	21.0±4.7 10.6–29.4
Temperature of wastewater, °C	11.9±5.3 5.2–19.0	12.9±5.6 4.7–20.1	16.5±3.2 9.1–20.3
DO, mg/dm^3	2.1±0.6 1.3–3.2	1.9±0.2 1.7–2.2	2.2±0.2 1.9–2.6
COD/BOD5	5.3	2.3	12.6
BOD_5/TN	0.7	3.0	0.2

During the study, in both local MTWs stable and effective removal of pollutants was provided. Concentrations of characteristic pollutants in the effluent wastewater varied in a very small range from the values which did not exceed the limit values for treated wastewater from units below 2000 PE according to the current regulation [6]. The effluent from Darżlubie still had low COD/BOD_5 rate in comparison with COD/BOD_5 rate for the effluent in Wiklino.

The treated RW showed very low levels of TSS and organic matter expressed as BOD_5. These indicators meet the requirements of the above mentioned current regulation [6]. However, characteristic values for RW, COD and TKN, exceeded the limits. The concentration of organic matter expressed as COD changed over a wide range values from 183.6 to 340.0 mg/dm^3. In the treated RW, nitrogen occurred mainly in the form of ammonium nitrogen with the concentration varying in a wide range from 99.6 to 198.6 mg/dm^3.

3.2. EFFICIENCY OF REMOVAL OF POLLUTANT

Despite unfavorable composition of the analyzed wastewaters, the MTWs ensure very high efficiency of pollutant removal (Fig. 1).

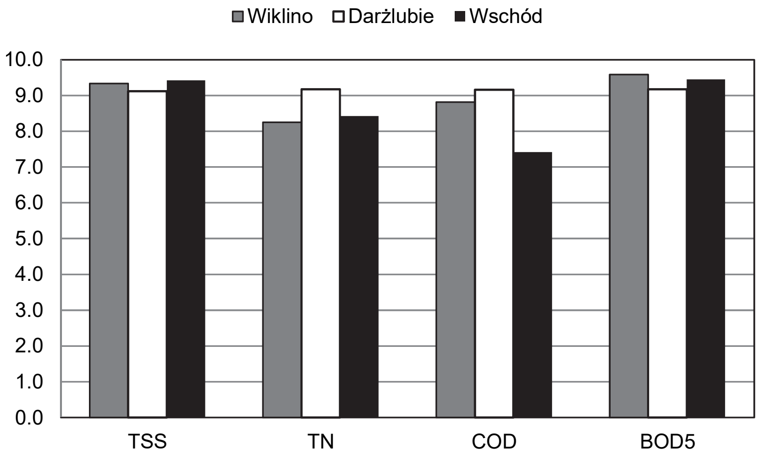
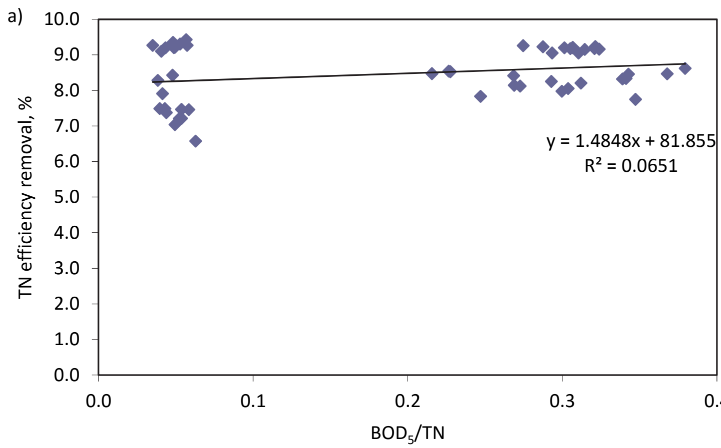


Fig. 1. Average removal efficiency of selected pollutants in the analyzed MTWs

The local MTWs ensured the organic removal over 90% and nitrogen removal over 80% in the monitoring period. The pilot plant Wschód showed very good efficiency of easily degradable organic (BOD_5) and TN reduction while it was lower in the total organics measured as COD. The achieved results are consistent with the data provided by the literature and prove that MTW are very efficient in removal of both, organics and nitrogen [10, 11, 20]. Moreover, the results suggest that in the systems composed of at least three beds with different– horizontal and vertical flow, the efficient treatment of wastewater of unfavorable compositions expressed by COD/BOD_5 and BOD_5/TN is possible (Fig. 2).

The analysis of the achieved results for TN removal efficiency depending on BOD_5/TN ratio showed two ranges of rates (0.4–0.6 and 2.0–3.8) for which similar removal efficiency of nitrogen was received (Fig. 2a). The first was the range of values for RW with total nitrogen removal efficiency from 70.0 to 94.0% and the other with the similar range of removal efficiency was observed for domestic wastewater (65.0–94.0%). The dependence of TN removal efficiency on COD/BOD_5 ratio showed no significant differences in the analyzed rates from 1.0 up to 3.8 (Fig. 2b). Moreover, the achieved results indicated the lack of simple linear regression between total nitrogen removal efficiency and wastewater composition expressed by BOD_5/TN and COD/BOD_5 in raw wastewater.



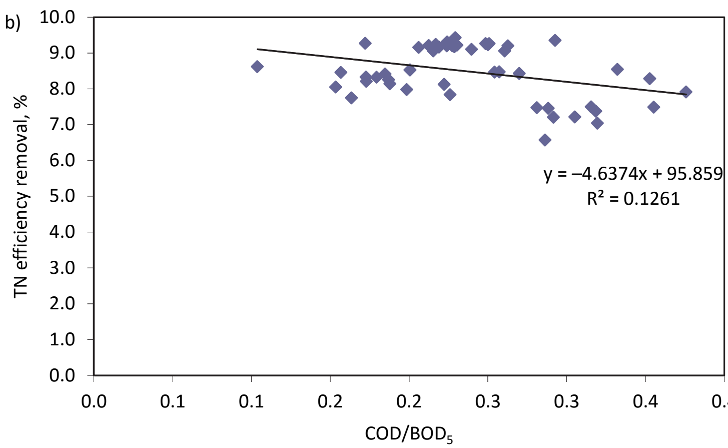


Fig. 2. Dependence of the total nitrogen (TN) removal efficiency on:
a) BOD_5/TN, b) COD/BOD_5 in raw wastewater

3.3. MASS REMOVAL RATE VIA TN LOAD

The applied simple linear regression model (LRM) confirmed a significant linear relationship between the mass removal rate (*MRR*) of TN and the TN load in raw wastewater. A very high coefficient of determination $R^2 = 0.99$ for the model: TN $MRR = f$(TN load) allowed the determination of one common relationship of the analyzed MTWs, the local plants, as well as the pilot facility (Fig. 3).

The high degree of fitting the results obtained describing the removal of 99% of TN in the analyzed MTWs allows the use of the above relationship to predict the rate of TN removal in the load from 1 to 2.0 g/(m^2·d) for wastewater treatment plants (domestic wastewater), and from 10 to 25 g/(m^2·d) for other wastewater treatment facilities with high pollutant loads. The results are fully consistent with the literature reports where the advisable load of TN does not exceed 25 g/(m^2·d) [1, 4, 22].

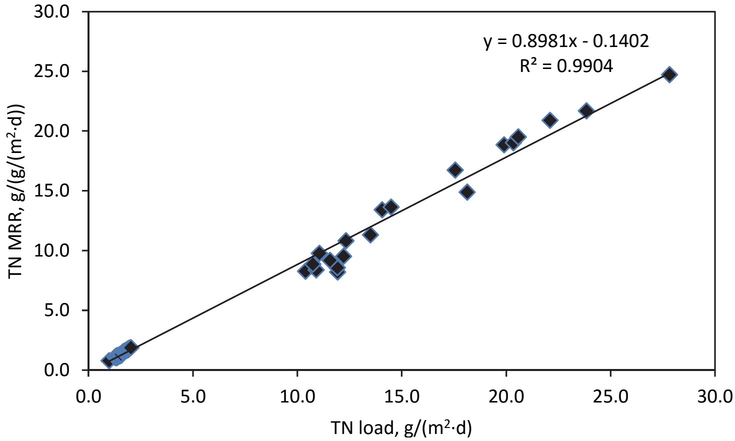


Fig. 3. Dependence of the TN mass removal rate on the TN load

3.4. DISCUSSION

In MTWs, both vertical (VSSF) and horizontal (HSSF) flow beds are incorporated to enhance the conditions suitable for aerobic and anoxic and even anaerobic decay [1, 4, 8, 21]. The entire system of MTW works properly when certain requirements are fulfilled. In the recent papers, the key factors supporting proper maintenance of TWs have been discussed. Among others, the load of hydraulic, organics and nitrogen influence the TWs operation, their influence on clogging and pollutants removal have been described [1, 18]. In the case of the MTWs analyzed in this work, all above mentioned parameters fulfill the recommended conditions that leads to effective removal of both nitrogen and carbonaceous compounds. Moreover, the working conditions of these facilities (Table 1) could be in considered as "perfect" and classified as the low loaded system in the nomenclature commonly used in high-tech solutions. The *MRR* of total nitrogen in the analyzed MTWs is directly proportional to the load of nitrogen in raw wastewater and there was no influence observed of improper composition of raw wastewater on efficiency removal of TN. Many factors determine good performance of MTWs under investigation, and the work undertaken only partially reveals the problem. It could be indicated that (i) not only the concentration of the pollutants in raw wastewater but also their composition is very important for unit processes leading to the efficient TN removal in MTW, (ii) MTW creates favorable conditions that could ensure efficient removal of TN independently of their initial concentrations, e.g. in unconventional processes [1, 23, 24]. Since in TWs a variety of physical and biochemical processes occur leading to successful TN removal, thus, the degree of dispersion of pollutants could play an important role, as well [1, 21, 24]. In contrast to the conventional treatment with activated sludge, in TW sedimentation, filtration and adsorption mechanisms of pollutant removal are very important [1, 2, 4, 21, 24]. The organic matter in the form of particular as well as colloidal fraction (even hardly degradable) is trapped inside the bed. Then, due to long residence time of wastewater in MTW it is slowly degraded, producing smaller molecules and consequently it could become an internal source of easily available carbon for denitrification, similarly to dead tissues of plants, as well as microorganisms [1]. This explanation could be especially true in this case since in the analyzed raw wastewater, the TSS concentration was very high and TSS consisted mainly of organic solids (VSS) (Table 2).

The MTWs provide a variety of conditions and thus could favor different mechanisms of nitrogen removal, among others such as short-circuiting the classical nitrification-denitrification pathway. Ammonium oxidizers have more versatile metabolism than it was previously assumed; aerobic nitrifies and ANAMMOX bacteria may be natural partners in many oxygen-limited environments. They even coexist with heterotrophic bacteria more easily than nitrifies, since heterotrophic bacteria consume oxygen for the decomposition of the organic matter, favoring in this way planctomycete--like bacteria over nitrifiers [21, 23]. In 2007, Dang and Sun confirmed completely autotrophic nitrogen-removal over nitrite (CANON) deammonification, responsible for nitrogen loss in the pilot studies with landfill leachate [23]. The results of their studies proved that TWs can achieve treatment of high ammonia and low organics content wastewater [24]. Furthermore, the authors observed that the ANAMMOX process was still working even when wastewater temperature decreased below 20 °C. These observations were partly proven also by Gajewska [25]. The carried out nitrogen mass balance for HSSF bed in Wiklino was characteristic of the existence of a combination of CANON and ANAMMOX processes. The compatibility of the stoichiometry equation with real parameters (input and output) was at a satisfactory level of 89.0%. These processes of nitrogen removal, often so-called unconventional, could be one of the reasons of lack of dependence of the nitrogen removal and the organic matter [23–25].

4. CONCLUSIONS

Multistage treatment wetland may be used for treatment of wastewater with high loads of organic pollutants and unfavorable composition expressed by the ratio of COD/BOD_5 and BOD_5/TN equal to 2.7 and 0.7, respectively.

Limited usefulness of commonly used indicators, like COD/BOD_5 and BOD_5/TN ratio in conventional treatment technologies to determine wastewater susceptibility to biodegradation during their treatment in multistage treatment wetland was confirmed.

Very effective removal of both organic matter and nitrogen, as well as the lack of significant correlation between organic matter and TN removal suggested the presence of alternative pathways of removal of nitrogen compounds with reduced available carbon demands or a great variety of unit processes, more complex in comparison with high-tech solutions which finally lead to the decomposition of even hardly degradable organic matter.

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