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WASTEWATER TREATMENT IN SUBMERGED AERATED BIOFILTER UNDER CONDITION OF HIGH AMMONIUM CONCENTRATION

OCZYSZCZANIE ŚCIEKÓW W ZANURZONYM, NAPOWIETRZANYM ZŁOŻU BIOLOGICZNYM W WARUNKACH WYSOKICH STEŻEŃ AZOTU AMONOWEGO

Abstract: Treatment of wastewater produced in Service Areas (SA) located on expressways and highways is a worldwide problem because of increasing amount of roads and specific composition of those kinds of sewage. Insufficient removal of pollutants from wastewater discharged into surface water may cause serious environmental problems. In the present study efficiency of treatment of wastewater with high ammonium concentration in biological membrane system used on SA was investigated. Concentrations of ammonia nitrogen in wastewater flowing into bioreactor in none of the tested objects did not fall below 99.0 mg of N-NH₄·dm⁻³. Because of high ammonium content in sewage and high pH reaching value about 9, it is almost impossible to create favorable conditions for microorganisms that run purification processes resulting in low efficiency of phosphorus and nitrogen removal (reduction of biogenic compounds did not exceed 15%). Treatment of wastewater consisting mainly of urine with commonly used biological membrane technology has to be widely tested to perform suitable quality of discharged wastewater, to provide safety of surface water environment.

Keywords: biological membrane system, ammonia nitrogen, wastewater treatment

Introduction

The expanding worldwide problem of decreasing water resources [1] with parameters suitable for water intended for drinking and food purposes, requires the use of ever newer and more effective solutions for the protection of water resources. There are many legal conditions and technological solutions applied on a global scale, which are intended to prevent excessive pollution of surface water by treated sewage discharged to it. There is no doubt that urban wastewater from entire cities or agglomerations can pose a serious threat to the environment in case of insufficient treatment and discharge to surface water. Similarly, deterioration of water quality may be caused by improperly designed or faulty selected local treatment systems, used for the treatment of domestic sewage [2].

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Constantly growing network of roads makes it necessary to create places with sanitary facilities for travelers. Service Areas (SA) are separated in a lane of expressways and highways and are equipped with parking, sanitary infrastructure and lighting. The most common type of SA are those belonging to the first category in which equipment includes lighting, parking and recreational facilities, sanitary facilities, and rarely - small catering facilities. The constantly growing number of such facilities and associated with their activities the formation of wastewater require action leading to their purification and disposal. The most commonly used solution is to equip the SA in cesspools used for periodic detention of wastewater which are then exported into the collective wastewater treatment plant. Due to the large unevenness in traffic, and therefore - high irregularity of sewage inflow there is a risk of underestimating the size of the required cesspool. Another drawback is the high operating costs associated with the need for frequent sewage disposal, often for a very long distance. Not without significance is also the composition and nature of the wastewater held in the cesspools which, due to the ongoing processes of anaerobic digestion, is a heavy burden for collective wastewater treatment systems and can adversely affect the processes they run. Disadvantages of cesspools used as a technological solution appropriate for SA mean that there is a need to look for other solutions that can effectively replace them. The choice of technology used for wastewater treatment should be based on tests of physico-chemical (qualitative) composition of sewage and on information of real amount of wastewater produced in this facility. In the scientific literature exists a concept of a "typical composition of domestic wastewater", on the basis of which purification technology is selected. This way of technology selection is very vague and does not incorporate the true nature of produced wastewater which may differ significantly from the composition of wastewater recognized as "similar", which generates the formation of numerous problems in the operation phase [3]. Suggesting the literature data on concentrations and loads of pollutants in wastewater is still a common mistake when solution for sewage treatment is selected. Knowledge of the values of parameters such as BOD₅, COD and TOC inform about the content of organic matter in wastewater [4]. It is also important to determine the content of total suspended solids, phosphorus and nitrogen, which allows to assess the nature of sewage and choose the best method of treatment.

Wastewater generated at the SA are referred to as domestic sewage, but their composition, due to the nature of SA activity, significantly deviates from them. On first category Service Areas which are not equipped with a small catering facilities, sewage generated in the vast majority consist of urine. It consists of urea, inorganic ions, natural organic metabolites, trace amounts of antibiotics and other organic compounds resulting from the synthesis [5]. In typical domestic wastewater urine is the smallest percentage (approx. 1%), however, introduces the biggest load of nutrients - 80-90% of nitrogen and up to 50-70% of phosphorus [6-8]. In the wastewater, consisting mainly of urine, the percentage of each of nutrients may be much higher. Urea during storage of urine in a tank (for example, septic tank) undergoes ammonification - biological degradation by urease, a catalyst for the hydrolysis of urea to ammonia and carbon dioxide [7]. As a result of hydrolysis and formation of ammonia, pH of wastewater increases and may reach value of about 9 [9]. Flushing the toilet with a tap water causes enrichment of a mixture of calcium and magnesium [5], which with its periodic detention creates the right conditions for the precipitation of struvite crystals (a process initiated by hydrolysis of the urea, and limited by the content of magnesium) [10]. Increase of the pH leads to reduction in the wastewater the concentration of calcium, magnesium and phosphorus by precipitation of carbonates

and phosphates, as well as loss of nitrogen due to ammonia volatilization [6, 7]. After exceeding the concentration of $150 \text{ mg NH}_3 \cdot \text{dm}^{-3}$, nitrification process is inhibited. High pH value as well as high concentration of ammonium cause sterile conditions in the bioreactor and inhibits growth of biomass used in biological treatment systems. The bacteria involved in the nitrification process, i.e. *Nitrosomonas* and *Nitrobacter* are very sensitive to pH changes and the presence of poisonous substances in the wastewater. Thiourea is considered as the most toxic substance for those strains of bacteria [11].

By their specific nature, it is difficult to qualify the wastewater from Service Areas for the so-called "typical domestic sewage". This is mainly urine mixed with water, a small amount of detergent, and the blackwater. Studies have shown that from the human body is emitted about 20% of phosphorus, and the ratio N : P in produced domestic wastewater is in range from 3 : 1 to 5 : 1 [12]. Due to the high concentration of nitrogen, insufficient cleaning of this type of wastewater can have serious environmental consequences for surface water, but also in the longer term - coastal and marine ecosystems [13]. The most common biogenic compound easily getting into the groundwater is nitrogen in the nitrate form [14]. Excessive phosphorus load discharged into the environment, may reduce the usefulness of water to be used for food purposes, and also as a limiting factor, it can intensify the primary production in surface water [15, 16]. For this reason it is important to select technology corresponding to the type of produced wastewater.

So far, treatment technology aimed to eliminate problems arising during the operation of septic tanks is a technology based on microbial activity of activated sludge or biofilm. Extremely important parameters which should be taken into account when selecting treatment technologies based on living organisms is content of easily digestible organic matter in the effluent relative to the amount of nitrogen. C : N ratio can dramatically affect the physical properties of the forming activated sludge in the system [17] and also effectiveness of wastewater treatment. Easily digestible organic matter and nitrogen contained in the wastewater are the basic building materials of cells of microorganisms responsible for cleaning processes. They are also a source of necessary energy. Systems using activated sludge are typically operated in conditions of limited amounts of carbon. C : N ratio in urban wastewater ranges from 20 : 1 to 25 : 1, but in a typical household sewage amount of carbon is even more limited [18]. Effective removal of organic matter from wastewater and reducing the amount of nitrogen to the level eliminating the risk of eutrophication in the receiver at a low C : N ratio is not possible without the dosage of the external carbon [19]. Proper C : N ratio in wastewater is necessary for nitrification process and growth of nitrifying bacteria, but also aids the process of denitrification in the anoxic zone [20]. For the proper conditions for denitrification it is necessary to provide amounts of carbon and nitrogen in the ratio of 5 : 1. Lack of enough amount of organic matter may result in a technological problems such as low efficiency of contaminants removal and long time of activated sludge maturation [21].

The abnormal conditions for nitrification process in specific wastewater may be also affected by high concentration of ammonia (NH_3) and free nitrous acid (HNO_2), whose presence is due to the high pH, extreme temperatures and high concentrations of ammonia nitrogen (N-NH_4^+), short age of activated sludge as well as too little or too much of the available organic matter. Waste water containing low concentrations of ammonia nitrogen creates better conditions for the growth of nitrifying bacteria showing differential sensitivity to concentrations of individual substrates in wastewater. The toxic effects of ammonia on microorganisms is largely dependent on the pH [22]. Proper growth of bacteria

leading the process of ammonia oxidation takes place at a pH of the sewage in the range of 5.8-8.5, and for the occurrence of the oxidation of nitrite is necessary to maintain the pH at 6.5-8.5 [23]. To ensure in the wastewater suitable conditions for the growth of bacteria responsible for nitrification, is also necessary to ensure wastewater oxygenation level of 1.5-2.0 g O₂·m⁻³.

The aim of the study was to evaluate the operating conditions of sewage treatment plants operating in a biological membrane technology under low organic matter load together with high pH values and high concentrations of ammonia nitrogen, as well as to determine the effect of wastewater quality and the operating conditions on the effectiveness of biogenic compounds removal.

Materials and methods

Wastewater from sewage treatment plants operating on the basis of the biofilm and activated sludge technology, located on the Service Areas (SA) built by the expressway were analyzed. Four facilities in which purification processes occur first in the primary settlement stage (septic tank), from where after the mechanical treatment by sedimentation and flotation wastewater are directed to the bioreactor were analyzed. The bioreactor is a tank with a capacity of 6 m³, in which the central part is a submerged fixed bed, aerated from the bottom by the diffuser system. The tank is equipped with a refining chamber in which occurs the separation of excess sludge before outflow of sewage into the receiver (dry ditch). The size of each of the wastewater treatment plants was determined as 50 RLM, on the basis of a predetermined amount of sewage discharged per day. For the project purpose also information about the qualitative composition of "typical domestic sewage" was used.

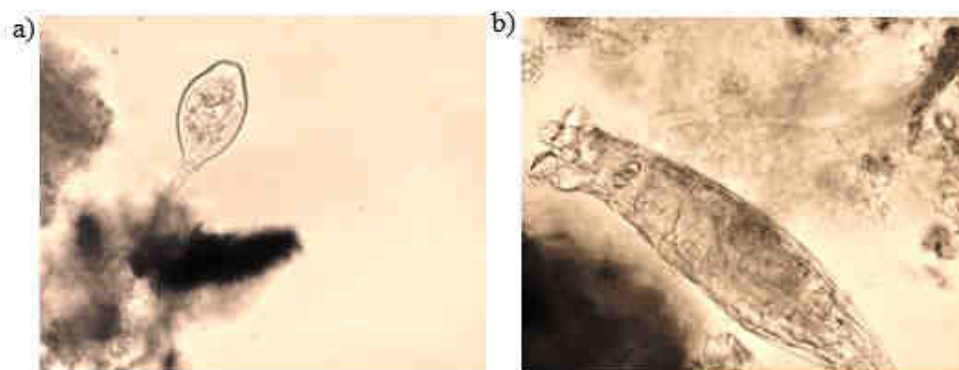


Fig. 1. Organisms in activated sludge: a) settled ciliates, b) representative of rotifers - photo A. Bawiec

In the primary settling tank occurs decrease in the levels of total suspended solids, COD and BOD₅ by sedimentation of particles with a density greater than the density of water at the bottom, thereby forming the layer of sewage sludge. The sewage sludge undergo the process of anaerobic decomposition, resulting in an initial stabilization. A compound having a density less than the density of water being carried on a surface in the form of scum. For the system used on SA, it is recommended to remove accumulated

sludge at its height exceeding 0.8 m and the thickness of scum exceeding 25 cm, with a frequency of at least 4 times a year.

In the bioreactor takes place the decomposition of pollutants discharged with mechanically purified sewage in the way of biological processes. By maintaining a high oxygen transfer in the reactor tank, the right conditions for the growth of microorganisms for aerobic processes of organic matter decomposition and nitrification are provided. In addition to bacteria forming the activated sludge, numerous protozoa (including floating and settled ciliates Fig. 1a), rotifers (Fig. 1b) and nematodes (Fig. 2), which support the processes of purification are settled in it. Support for the activated sludge is a centrally located plastic grate, aerated from the bottom via fine bubbles diffusers operating continuously (Fig. 3).



Fig. 2. Nematode in activated sludge in biological reactor - photo A. Bawiec



Fig. 3. Grate made of plastic with biological membrane - photo A. Bawiec

Physicochemical tests included determining the concentration of nitrogen and phosphorus as well as BOD₅ in the wastewater taken from primary settling tank (inflow) and from the refining chamber in bioreactor (outflow). Analyzes were made in Laboratory of Water and Wastewater in Department of Environmental Engineering and Geodesy of University of Life Sciences in Wrocław according to the methods specified in standards. In addition, to perform microscopic analysis of activated sludge collected from a bioreactor an inverted microscope Delta Optical IB-100, with the 40x magnification was used.

Results and discussion

Samples of wastewater flowing in and flowing out of wastewater treatment plant operated on the SA facilities, collected in 2014 were analyzed. Sewage discharged into the bioreactors operated in the analyzed objects have a different composition from the “typical domestic wastewater”. Distinguishing feature of wastewater generated there from wastewater discharged from households is a significant predominance of urine over other sewage components, due to the lack of catering facilities, showers and laundry facilities. Research of wastewater quality showed dominance of ammonium over other forms of nitrogen and relatively low organic matter content expressed in BOD₅ in the range of 6.7-16.6 mg O₂·dm⁻³. Due to such an unusual composition, determined on the basis of the value of BOD₅ and concentrations of total nitrogen C : N ratio was consecutively for each object 1 : 17; 1 : 35; 1 : 14; 1 : 13. Such distribution of organic matter in relation to the nitrogen content does not provide optimal conditions for nitrification processes that should occur in the bioreactor. Concentrations of total nitrogen in the analyzed samples of raw and treated wastewater collected from Service Areas every two months is showed on Figure 4a-b.

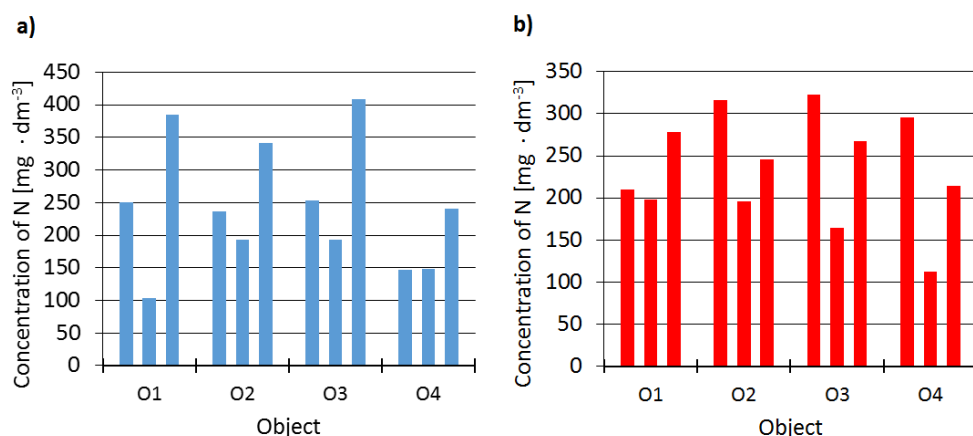


Fig. 4. Concentration of total nitrogen in a) raw and b) treated wastewater samples collected in June, August, October 2014, in four analyzed objects (O1, O2, O3, O4)

The data presented in Figure 4 show the concentration of total nitrogen in raw sewage flowing into the settling tanks in three consecutive sampling periods (June, August, October 2014), in four analyzed objects (O1, O2, O3, O4). Concentrations of total nitrogen in raw sewage did not fall below 100 mg N·dm⁻³. The lowest concentration in all objects were

recorded in August, while the highest in the samples collected in October, which may be related to the thermal conditions prevailing in the area. The highest concentration of total nitrogen occurred in October in the third facility and exceeded $400 \text{ mg N} \cdot \text{dm}^{-3}$.

The graph presented in Figure 4b shows the concentration of total nitrogen in treated wastewater collected from the clarification chambers of bioreactors. The research shows that in June 2014 the concentration of the tested parameter increased as a result of treatment in three of the four test objects. In August, an increase in the concentration of total nitrogen at the outlet of the bioreactor were observed in the objects O1 and O2, while in October in each of the studied Serving Areas concentration in treated sewage were lower than in raw wastewater. Lack of reduction of total nitrogen in wastewater passing through the bioreactor may be due to high concentrations of ammonium that shows inhibitory effect on the activity of microorganisms running treatment processes under aerobic conditions. The increase of nitrogen concentration at the outlet may be the result of dying of organisms forming biomass and which are flowing out of the system with treated wastewater.

Concentrations of ammonia nitrogen in the analyzed samples of raw and treated wastewater are presented on Figure 5a-b.

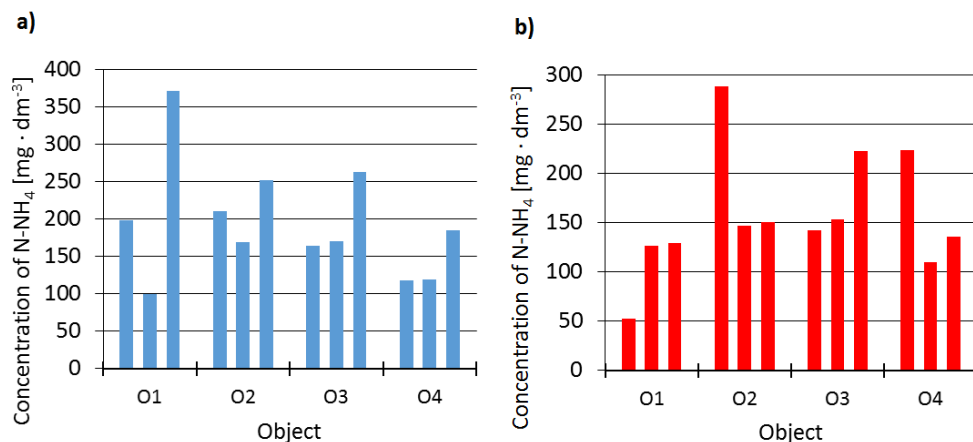


Fig. 5. Concentration of ammonia nitrogen in a) raw and b) treated wastewater samples collected in June, August, October 2014, in four analyzed objects (O1, O2, O3, O4)

The concentration of ammonia nitrogen in raw sewage flowing into the settlers used as the first stage of treatment, in none of the tested objects did not fall below $99.0 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$. The highest concentration of ammonia nitrogen was recorded in October on the object O1 and it amounted to $370.68 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$. The lowest concentrations of this form of nitrogen were observed in the outflow from the object O4 (from 117.44 to $185.15 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$). In each of the analyzed objects, the highest concentration of N-NH_4 reported in October. The wide variation in the size of the concentration of this form of nitrogen in individual SAs stems from the huge inequality of sewage inflow and diverse participation of urine in the sewage flowing into the settling tanks. The qualitative composition of wastewater is affected not only by the number of passengers, but also by kind of means of transport they use for travel (in the case of

emptying toilets from buses, the concentration of ammonia nitrogen in wastewater influent can be several times larger than when operating only passengers travelling by cars).

The graph shown in Figure 5b shows the concentrations of ammonia nitrogen in the wastewater flowing out of the bioreactor, which is the second stage of wastewater treatment. In the treated wastewater on the objects O2 and O4, the concentration of the nitrogen forms were greatest in June 2014 and in both bioreactors it exceeded $220 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$. The lowest levels were recorded in the same month on the object O1 - $52.47 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$. In the remaining months, the concentration of ammonia nitrogen in each of operated bioreactors range of $100\text{--}150 \text{ mg N-NH}_4 \cdot \text{dm}^{-3}$. High concentrations of ammonia nitrogen in the outlet of the plant are the result of very high concentrations of this compound in the inlet. Because of the high pH of the effluent flowing into the plant, high concentration of N-NH_4 and the lack of required concentration of organic matter, in the bioreactor there are no favorable conditions to the growth of nitrifying bacteria, therefore, the nitrification of ammonium nitrogen which goes into the form of nitrate nitrogen, does not occur with the expected yield.

The graphs in Figures 6a-b show the concentration of nitrate nitrogen in raw and purified sewage from SAs (O1, O2, O3 and O4).

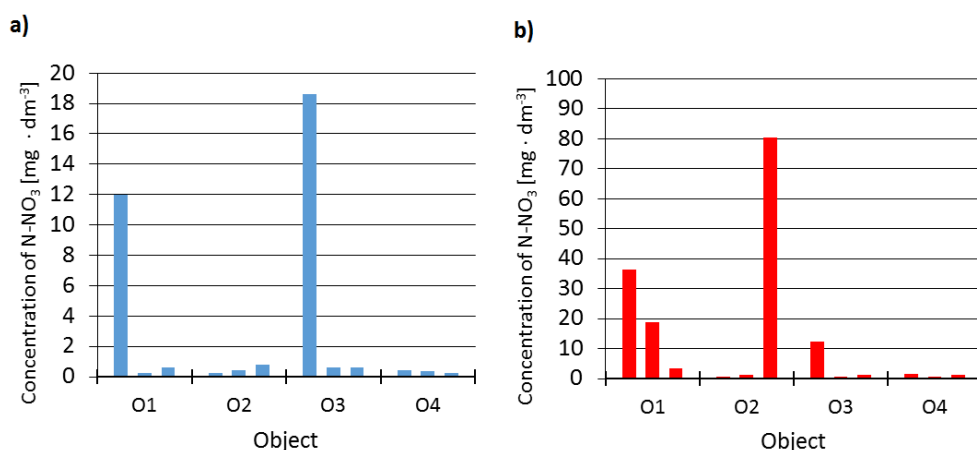


Fig. 6. Concentration of nitrate nitrogen in a) raw and b) treated wastewater samples collected in June, August, October 2014, in four analyzed objects (O1, O2, O3, O4)

The data presented in the graph illustrating the concentration of nitrate nitrogen at the inlet to the treatment plant shows that in most cases the concentration of this form of nitrogen were close to zero, and only in two cases (object O1 and O3, June 2014), showed values in excess of $10 \text{ mg N-NO}_3 \cdot \text{dm}^{-3}$. Such a small share of $\text{NO}_3\text{-N}$ relative to N-NH_4 is connected with the nature of waste water resulting from operations carried out on the SA.

At the outlet of the reactor increase in the concentration of nitrate nitrogen in the wastewater was observed, with the exception of the object O3 in August 2014, where a slight decline occurred. The increase in the concentration of nitrate nitrogen on the outlet may indicate the presence of ammonia nitrogen oxidation when favorable conditions for this process were ensured (periodic changes in pH and ammonia concentrations in

conditions of continuous aeration of wastewater may promote the growth of nitrifying bacteria).

Except from nitrogen, the component of wastewater which has huge impact on quality of receiving water is phosphorus. Its concentration limits the occurrence of eutrophication in surface water, what largely determines the possibility of their use as a drinking water and for industrial purposes. The concentrations of phosphorus in the wastewater treated in the plants operated on Service Areas are presented on graphs in Figures 7a-b.

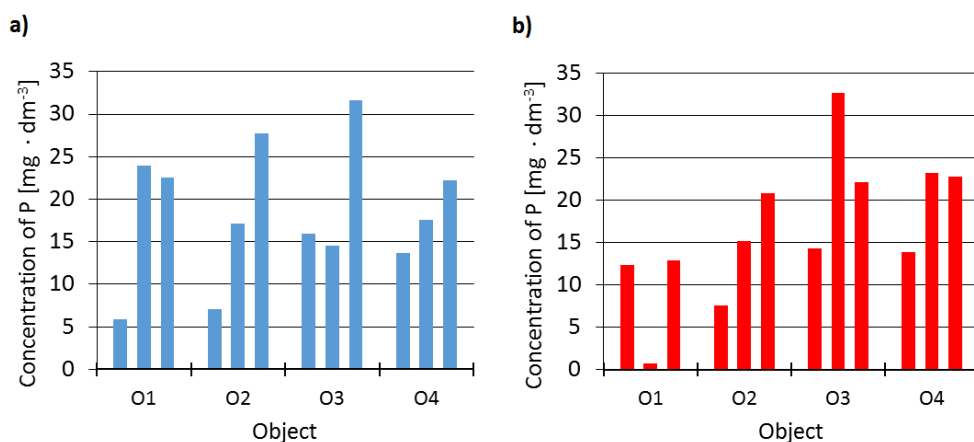


Fig. 7. Concentration of total phosphorus in a) raw and b) treated wastewater samples collected in June, August, October 2014, in four analyzed objects (O1, O2, O3, O4)

The concentration of total phosphorus in raw sewage flowing into settling tanks in sewage treatment plants on SAs ranged from 5.92 to 31.66 mg P·dm⁻³. This wide range of encountered concentrations is the result of very high variability of the quality of raw wastewater, which composition largely depends on the number of travelers using the SA. The highest concentrations of phosphorus in raw sewage were recorded in three locations - O2, O3 and O4 in October, and the lowest in June, in the wastewater from the O1, O2 and O4.

In the analyzed samples taken at intervals of two months, there was no clear trend in changes in the concentration of total phosphorus in the wastewater as a result of their treatment. In the object O1 and O2 in June an increase in the concentration of total phosphorus took place, while in the other two sampling dates - decline. However in the object O3 in June a decrease of the concentration, and the increase in August and October was observed. In the facility O4 higher concentrations of P on the outlet from the treatment plant than in the inlet were reported.

During the research, obtained removal efficiency of nitrogen and phosphorus from wastewater does not exceed 15% (Fig. 8). The problem with removal of nitrogen from wastewater characterized by a high concentration of ammonia nitrogen is caused by a sterilizing effect of high pH and the toxic effect of ammonia being a result of hydrolysis, to microorganisms colonizing the biofilm. With continuously maintaining a high level of urea in wastewater and low concentrations of organic matter expressed in BOD₅, creating favorable conditions for the growth of nitrifying bacteria of the first and the second phase is

almost impossible. Another factor worsening the living conditions of microorganisms is the use of cleaning agents and disinfectants based on chlorine, which used in large quantities can cause poisoning of biofilm and finally death of the biofilters organisms. Removal of phosphorus from wastewater in bioreactors on the SA does not occur with high efficiency due to the unfavorable composition of the wastewater and continuous aeration (biological phosphorus removal processes occur while ensuring alternating aerobic and anaerobic conditions).

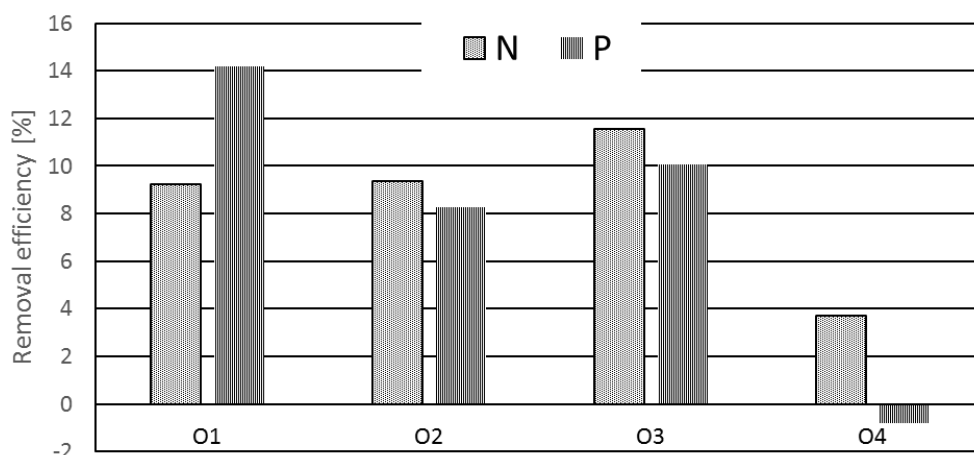


Fig. 8. Efficiency of nitrogen and phosphorus removal in wastewater treatment plants located on Service Areas

The graph shown on Figure 8 presents average efficiency of nitrogen and phosphorus removal in wastewater treatment plants working in attached growth processes technology located on Service Areas (O1, O2, O3, O4). In the analyzed period, total nitrogen removal efficiency did not exceed 12% ($\eta = 11.55\%$ in wastewater treatment plant in O3) while the lowest efficiency was achieved on the object O4 - 3.7%. The highest reduction of total phosphorus was achieved in the treatment plant in object O1 and it amounted to 14.2%. The lowest total phosphorus removal efficiency was observed as in the case of nitrogen in the wastewater treatment facility in O4, where increase of its concentration in treated sewage comparing to raw wastewater occurred. It may be linked to the leaching of dead fragments of biofilm into the outflow or with intensive use of the surfactants rich in phosphate compounds.

Conclusions

Wastewater treatment in Service Areas due to the specific nature of the produced wastewater (sewage consist mainly of urine, lack of catering facilities), is a difficult task, and the method of effective treatment has not yet been developed. Failure to meet the requirements of the water permit is not the result of negligence of the exploiters, but the result of the erroneous assumptions of the quality of sewage flowing into the system, based on the literature data on the quality of the "typical domestic sewage". Obtaining low efficiency of nitrogen removal from wastewater is related to the lack of environmental

conditions for the growth of nitrifying bacteria. High concentrations of ammonia nitrogen leads to the formation of ammonia which is toxic for nitrifying microorganisms and the pH rises beyond the level of bacteria tolerance. An additional factor is the use of strong sterilizing disinfectants for cleaning toilets that discharged with sewage into the settling tank and bioreactor sterilizes wastewater itself. Delivery of active chlorine which is the main active ingredient in disinfectants to the settling tank eliminates microorganisms and higher organisms inhabiting the activated sludge flocs and biofilm. Also, cleaners containing surfactants, discharged in excess to the treatment plant, adversely affect the condition of the biofilm.

The solution that could improve the operation of sewage treatment plants in the SA is the separation of urine from the remaining wastewater stream, which would be possible in the case of men's lavatories (assembly urinals and sewage disposal tank). Such a solution would reduce the concentration of urea flowing into treatment system, reducing the amount of ammonia formed in wastewater. In addition, urine collected in the reservoir due to its sterility could be used for the fertilizing purposes [24]. Another action that could support the work of treatment is to eliminate the use of chlorine-based cleaners, to the use of cleaners based on organic acids (citric acid), which are biodegradable agents and less harmful for environment.

Research carried out in June, September and October 2014 in wastewater treatment plants located in Service Areas showed that treatment of wastewater with high concentrations of ammonium and high pH causes operational problems and obtaining high efficiency of contaminants removal is not possible. There is a need to continue research, verify obtained results and compile technology for treatment domestic wastewater produced on Service Areas because it is problem which occurs worldwide in all types on treatment plants using biofilm and activated sludge technology for treatment of sewage with high ammonium concentration.

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