



Article Influence of the Hybrid Sewage Treatment Plant's Exploitation on Its Operation Effectiveness in Rural Areas

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Abstract: The article evaluates the effectiveness of the removal of organic pollutants—nitrogen and phosphorus—from household sewage in a hybrid bioreactor with a submerged fixed bed. The experiment was carried out in two exploitation variants that were both conducted in a laboratory model of the hybrid bioreactor: (I) cycles of 120 min of aeration and 60 min of no aeration with a constant sewage dosage, and (II) cycles 60 min of aeration and 60 min of no aeration, with a periodic sewage dosage in the no-aeration phase. The experiment was carried out on real sewage primarily treated in a septic tank. The amount of pollution removal was calculated and compared with the mandatory standards according to Polish law. Moreover, the susceptibility of the sewage to the biological treatment, nitrification, and denitrification activity was determined. The research shows a higher effectiveness for the 60/60 model in comparison to the 120/60 model. High operation efficiency was observed regarding the removal of organic pollution and nitrate nitrogen. The tested structure showed very low nitrification activity combined with intense denitrification. These processes were observed in the 60/60 variant. The structure was often overloaded with the nitrate nitrogen, which was considered to be the nitrification process inhibitor. It was suggested that phosphorus was also removed by the denitrifying bacteria.

Keywords: household sewage; hybrid bioreactor; nitrification; denitrification

1. Introduction

A significant number of rural settlements in Poland, as well as small urban centers, still lack the satisfactory sewage management regulations, contrary to their common and widespread water supply systems [1]. The observed dynamic development of the sewerage systems and sewage treatment infrastructure in the rural areas is in most cases due to the European Union (EU) directives and funding. Regardless of the significant increase of the sewerage system's length in the rural areas, the needs are only partially fulfilled. The local sanitation system is definitely more important; it drains the sewage into the septic tanks. Despite the dynamic development of the sewerage system in the villages, it accounts for only 19% of the whole water supply system. The disproportion between the water supply systems and the sanitation systems causes serious threats to the sanitary conditions and the environment in these areas. The characteristics of the rural settlement, which consists of dispersed buildings, should impose on designers and investors the usage of effective, reliable, and relatively cheap technologies to treat the individual households' sewage [2]. With such technologies, it is possible for the Polish villages to attain the needed ecological goals, which are environment-friendly, as in the case of collective sewerage system and local sewage treatment plant building plans, and are very often placed in the indefinite future. In Polish conditions, small amounts of sewage treatment

technologies that use the filtering drainages, sand filters, or hydrobotanic beds are the most frequent ones. These systems also are environmentally-friendly and promote the sustainable development of rural areas. Currently, systems with small amounts of sewage treatment in artificial conditions, using biological beds or the active sludge technologies, are becoming more and more popular.

In practice, the classic systems of the household sewage treatment plants with the active sludge are "miniatures" of the large systems. However, regarding the different sewage characteristics in the areas with the dispersed buildings, contrary to the areas within the collective sewerage system, very often exploitation problems occur in such small systems, which is related to the interruptions in the aeration due to the lack of electricity supply or the instability of active sludge biogenesis development related to a high heterogeneity of sewage supply.

Systems with microorganisms' biofilm are considered to be much more beneficial in comparison to the traditional systems, where the biomass is suspended in the bioreactor [3–5]. These systems are characterized by the stable sewage treatment process, a long period of biomass retention, and a much higher resistance to the toxic substances and sudden changes in the external conditions. Moreover, bioreactors with the biomass attached to the filling material are much smaller than the classic systems with the suspended microorganisms; therefore, they are easier to use in small sewage treatment plants.

In the cases with existing mini-sewage treatment plants that operate on the active sludge technology, it is possible to increase their operation stability, which influences their reliability, by introducing flexible or stable microorganisms immobilized on the artificial carriers [6,7]. Such hybrid systems are the combination of suspended and immobilized biomass, and are more resistant to the quantitative and qualitative changes of sewage than the classic active sludge; they also lack some of the biological beds' defects [8,9]. The introduction of plastic blocks into the classic bioreactor with the active sludge may increase the effectiveness of organic matter removal from the sewage without the need to expand the treatment plant. Both the ecology and exploitation of such a procedure is arguable, and it is much cheaper than adding new objects to the system [10]. However, exploitation bioreactors require considerable costs for the aeration of a chamber. Thus, analysis solutions that decrease the exploitation costs of the bioreactor are necessary; they permit removal pollutions such as other technologies. Such solutions can include periodic aeration or a sewage dosage bioreactor. Vertical flow filters with filling in the form of polyurethane foam can be such an example [11,12]. Tomei et al. [13] observed very high organic removal (99% after just 5 h of treatment), and the effective biodegradation of the organic fraction of the wastewater (>90% at the end of the test) was observed during an experiment with the use of a hybrid system consisting of a biological reactor containing spiral-coiled polymeric tubing through which the mixed sewage was pumped. A hybrid DMBR-IVCW system, which combines a dynamic membrane bioreactor (DMBR) unit and an integrated vertical-flow constructed wetland (IVCW) unit, was applied by Kong et al. [14] to treat domestic sewage. Zhu et al. [15] used a hybrid membrane bioreactor (HMBR) for ship domestic sewage treatment. The degradation of organics in an aeration tank during this experiment follows the first-order reaction.

The technology of hybrid reactors belongs to the biological methods of wastewater disposal. It uses the process of the natural immobilization of biomass, which consists of the formation of a biological membrane on the surface of carriers, and in consequence increases the efficiency of wastewater treatment. [16]. In addition, this technology eliminates the common problem of clogging deposits. Moreover, regarding the activated sludge method, it was shown that immobilized biomass on supports in technological systems solves the problem of sludge swelling and allows resignation from the recirculation of activated sludge [17,18]. In the biogenesis of hybrid reactors, regarding activated sludge, there is no development of filamentous bacteria [19]. According to Chan et al. [20], hybrid reactor technology, in contrast to the conventional activated sludge system, is distinguished by better oxygen permeability, a shorter sewage retention time, higher charges of organic compounds, a higher degree of nitrification, and a larger contact area with wastewater. Sindhi and Shah [21] outlined the disadvantages of hybrid reactors, including the limited possibility of process control and the lower popularity of this technology. Hybrid reactor technology brings a number of technological possibilities.

An important advantage of this system is the possibility of accepting a large pollutant load. The process of the natural immobilization of biomass on supports, and thus the prolonged age of microorganisms, independent of the hydraulic time of sewage retention, allows for highly effective nitrification [22].

The main premise behind conducting the research was that in Poland, technologies based on activated sludge or biological beds for removing pollution from a single building are widely used. However, these systems work with low efficiency due to insufficient reductions in nutrients as well as organic compounds. In addition, these systems are very sensitive to changes in sewage inflow. However, both activated sludge and biological biofilms have their advantages, so it was decided to combine both solutions and investigate the hybrid system. To intensify the work of simultaneous denitrification/nitrification, it was decided that the reactor would work under changing conditions (anoxic/oxygen), which in turn could increase the reduction of nutrients. Such cyclic aeration may in effect be less costly for the operator compared to aerated solutions with recruiters, which was another reason to undertake such research.

The aim of the research is to evaluate the influence of various exploitation conditions (changeable aeration versus the lack of aeration, constant versus periodic sewage dosage) on the effectiveness of organic and nutrient removal as well as on the efficiency of the elementary processes in a hybrid bioreactor with a submerged fixed bed.

2. Materials and Methods

2.1. The Research Station

The tests were carried out on a laboratory model of the hybrid bioreactor. The model is made of a cuboid container with a built-in secondary settlement tank. The container is divided into two parts: the first is the aeration tank, and the second is the settlement tank. The container walls are made of metal, except for one that is made of a plexiglass slab. The aeration tank dimensions are as follows: length—700 mm; width—300 mm; height—700 mm; thus, the total volume is $V_{tot} = 147 \text{ dm}^3$. The secondary settlement tank's total volume is $V_{tott} = 58.5 \text{ dm}^3$, as shown in Figure 1. The plastic bed block was placed inside the aeration tank, which was submerged in the sewage. The block's dimensions are: length—400 mm, width—300 mm, and height—300 mm. The specific surface of the carriers was 150 m²·m⁻³. Sewage was evenly distributed in the aeration tank due to the installed overflow. The treated sewage was drained from the secondary settlement tank with a sawtooth overflow weir. The inflow and outflow of the sewage from the bioreactor were placed in the way, which enabled the piston flow. The bioreactor's content was subsurface aerated with the disc diffuser covered with PTFE membrane produced by Stamford Scientific International Inc. (Poughkeepsie, NY, USA). The air was pumped into the system with a HIBFLOW HP-60 compressor. The rotameter was placed between the compressor and the diffuser to enable the regulation of the supplied air amount. The excessive sludge was drained outside of the settlement tank by the peristaltic pump type PER-R0601. Part of the sludge from the secondary tank was recirculated into the aeration tank by the same type of peristaltic pump.



Figure 1. Model of the hybrid bioreactor.

2.2. Research Procedures

The research analyzed a real sewage sampled from the septic tank of the single-family household. The transported sewage was dosed into the bioreactor using the peristaltic pump. The aeration tank was inoculate activated sludge from the RetroFAST hybrid sewage treatment plant's treated domestic wastewater. The experiment was carried out in two variants in order to simulate bioreactor's performance in different exploitation conditions: I—constant sewage dosage 24 h a day, with changeable aeration cycles—120 min with aeration/60 min with no aeration; II—sewage dosage during the periods with no aeration, and various aeration cycles—60 min with aeration/60 min with no aeration. The average quantity of air pressed to the bioreactor was 55 dm³ h^{-1} . After taking the samples from the raw sewage and the treated sewage, physicochemical analyses of the following pollution indexes were performed: temperature, total suspended solids, BOD₅, COD_{Cr}, ammonium, nitrite and nitrate nitrogen, and total phosphorus. The sewage temperature was measured using a digital thermometer, and total suspended solids were measured with the gravimetric method as follows: BOD₅ according to the standard PN-EN 1899–1:2002 and PN-EN 1899–2:2002; COD_{Cr} according to the standard PN-ISO 15705:2005; ammonium, nitrite and nitrate nitrogen using the photocolorimetric method according to PN-ISO 7150–1:2002, PN-87/C-04576.07 and PN-EN 26777:1999; and total phosphorus was measured using the photocolorimetric method according to the standard PN-EN 1189-2000.

Once a day, the following parameters were monitored during the process in the aeration tank: sewage temperature, dissolved oxygen concentration, oxygenation, and pH reaction. The evaluations were made with the pH/oximeter CPX type equipped with the proper measurement probes: –COG-1 type oxygen and a PEPS-1electrode. Nitrification and denitrification efficiency was evaluated on the basis of the Carrera et al. [23] formulas:

$$L(N-NH_4) = \frac{(N-NH_4)_i}{M \cdot HRT}$$
(1)

$$r_{N} = \frac{(N-NH_{4})_{i} - (N-NH_{4})_{O}}{M \cdot HRT}$$

$$\tag{2}$$

$$\mathbf{r}_{\mathrm{D}} = \frac{(\mathrm{N-NO}_{\mathrm{x}})_{\mathrm{i}} - (\mathrm{N-NO}_{\mathrm{x}})_{\mathrm{O}}}{\mathrm{M} \cdot \mathrm{HRT}}$$
(3)

where:

L(N-NH₄)—bioreactor's loading with the ammonium nitrogen, gN-NH₄·gsmo⁻¹·d⁻¹;

M—biomass concentration in the bioreactor, $gsmo \cdot m^{-3}$;

HRT—hydraulic retention time, d;

 r_N —nitrification rate, gN-NH₄·gsmo⁻¹·d⁻¹;

 r_D —denitrification rate, gN-NO_x·gsmo⁻¹·d⁻¹;

 $(N-NH_4)_i$ —ammonium nitrogen concentration in the bioreactor's inflow, gN-NH₄·m⁻³;

 $(N-NH_4)_0$ —ammonium nitrogen concentration in the bioreactor's outflow, gN-NH₄·m⁻³;

 $(N-NO_x)_i$ —oxidized nitrogen concentration in the bioreactor's inflow, gN-NO_x·m⁻³;

 $(N-NO_x)_o$ —oxidized nitrogen concentration in the bioreactor's outflow, gN-NO_x·m⁻³;

3. Results

Figure 2 presents how BOD₅ has been changing in the raw and treated sewage during the experiment. High BOD₅ variation is characteristic of the raw sewage. It results from the raw sewage samples that were treated in this experiment being taken from a septic tank that was a part of the sewage drainage and treatment system for a single household. The high variability of the pollutant's amount is typical of the sewage from the single households in the rural parts of Poland. The average BOD₅ value in the raw sewage during the experiment was $289.0 \pm 120.3 \text{ mgO}_2 \cdot \text{dm}^{-3}$. The average BOD₅ values in the raw sewage were similar in both experiment variants: average BOD₅ for variant I was 284.1, and the average BOD₅ for II was 295.6 mgO₂·dm⁻³. In the case of sewage treated in the beginning of the process and after the conditions changed, the BOD₅ value changed, which was caused on one hand by the bioreactor's accustomation and the creation of the biomass suspended and immobilized on the bed, and on the other hand by the existing biomass' acclimatization to the variable conditions in the bioreactor. The acclimatization period was definitely shorter after the conditions changed than in the beginning of the experiment. This results from the biomass having already been formed and developed during the change of the experiment's variant, so the adaptation process to the changeable conditions was much shorter than in the situation when there was an insufficient amount of microorganisms in the bioreactor to mineralize the organic matter. The relatively short adaptation period is also the result of biofilm creation, which is much more resistant to the sudden conditions' changes than the suspended biomass. During the experiment, except for the mentioned periods of the accustomation and acclimatization of biomass, the BOD₅ values in the bioreactor's outflow were slightly changeable. The average BOD₅ value in the treated sewage in the variant I was $38.9 \pm 19.9 \text{ mgO}_2 \cdot \text{dm}^{-3}$, and in variant II it was $24.0 \pm 18.1 \text{ mgO}_2 \cdot \text{dm}^{-3}$. The average BOD₅ reduction during variant I equaled 86.3%, and in II, it equaled 91.9%. Such results confirm that the replacement of the constant sewage dosage into the bioreactor with the periodic dosage and shortening of the aeration period from 120 to 60 min caused the improvement of BOD₅ removal from the sewage. According to the mandatory Polish legislation about the quality of sewage drained into the collector, the tested change of the bioreactor's exploitation conditions regarding the average BOD₅ values fulfilled the quality standards for the treated sewage from the treatment plants of 15,000 equivalent people (EP), and in the case of the reduction, it was larger than 100,000 EP. In the case of variant I, the treatment plant met the standards for smaller objects: up to 2000 EP.

COD changes during the experiment are similar to BOD_5 , as shown in Figure 2. The average COD value in the raw sewage in the whole tested period was $651.49 \pm 213.79 \text{ mgO}_2 \cdot \text{dm}^{-3}$, whereas in variant I, it was $706.49 \pm 221.43 \text{ mgO}_2 \cdot \text{dm}^{-3}$, and in variant II, it was $568.89 \pm 189.10 \text{ mgO}_2 \cdot \text{dm}^{-3}$. In this case, the reduction of COD values in the raw sewage observed in the second variant might have caused the higher reduction of this index, regarding the smaller COD loading of the sludge. The average COD value in the bioreactor's outflow in variant I was $219.9 \pm 131.2 \text{ mgO}_2 \cdot \text{dm}^{-3}$, which caused the 68.9% reduction, whereas in variant II, it was $87.5 \pm 30.4 \text{ mgO}_2 \cdot \text{dm}^{-3}$, with a reduction of 84.6%. In the case of variant I, the average COD value in the treated sewage and the reduction exceeded the admissible level stated in the regulation cited above for all of the treatment

plant's sizes, and variant II met all of the standards for all of the treatment plants' sizes. In the case of the COD value, better bioreactor's operation could be observed in variant II.



Figure 2. Changes of BOD₅, COD_{Cr}, and total suspended solids values during the experiment.

Similarly, in case of the total suspended solids' value, its high changeability in the raw sewage can be seen, as well as a definitely higher treatment capacity in variant II, as shown in Figure 2. Regarding the raw sewage, the total suspended solid's concentration in variant I was $284.8 \pm 144.4 \text{ mg} \cdot \text{dm}^{-3}$, and in variant II, it was $206.5 \pm 58.2 \text{ mg} \cdot \text{dm}^{-3}$. Due to the system's accustomation, a high variability of total suspended solids' concentration can be noticed during variant I. In the beginning of variant II, due to the changes in exploitation as well as in the other bioreactor's conditions, more than a $100.0 \text{ mg} \cdot \text{dm}^{-3}$ increase of the total suspended solids concentration was observed in the outflow, which decreased after seven days to below $50.0 \text{ mg} \cdot \text{dm}^{-3}$. The average concentration of this indicator for the outflow in variant I was $89.5 \pm 59.1 \text{ mg} \cdot \text{dm}^{-3}$, while for variant II it was significantly lower, and was $39.8 \pm 35.8 \text{ mg} \cdot \text{dm}^{-3}$. The total suspended solids' reduction in variant I equaled 68.6%, and for II, it equaled 80.1%. In case of variant I, the total suspended solids' concentration in the outflow exceeded the admissible standards according to the Polish regulations for all of the treatment plants' sizes, and in case of variant II, the requirements were met for the smallest objects: up to 2000 RLM.

Nitrogen compounds' transformations are greatly influenced by exploitation conditions. Regarding the ammonium nitrogen in the raw sewage, as shown in Figure 3, the average concentration for variant I was $67.0 \pm 18.1 \text{ mg} \cdot \text{dm}^{-3}$, and for variant II, it was $63.2 \pm 16.9 \text{ mg} \cdot \text{dm}^{-3}$. Therefore, it can be assumed that during the experiment, the concentration of this nitrogen form stood steady on a similar level. However, important differences occurred in the treated sewage. The average ammonium nitrogen concentration in the raw sewage in variant I was $78.9 \pm 19.0 \text{ mg} \cdot \text{dm}^{-3}$, and in variant II, it was $46.9 \pm 12.2 \text{ mg} \cdot \text{dm}^{-3}$, which caused reductions of 17.8% (concentration increase in the outflow compared with the inflow) and 25.8%, respectively.

For variant I, the ammonium nitrogen concentration in the outflow was lower than in the inflow, regardless of the much higher oxygen concentration than is necessary for the nitrification process (the average oxygen concentration in the bioreactor in variant I was $3.5 \text{ mgO}_2 \cdot \text{dm}^{-3}$). Similarly to the previously discussed pollution indicators, such a state is influenced by the lack of specialized microorganisms that are capable of the nitrification process. The situation changes in variant II, where this indicator is reduced. During variant II, the average oxygen concentration in the bioreactor was $1.8 \text{ mgO}_2 \cdot \text{dm}^{-3}$. It is important that not only the change of the exploitation conditions influenced

the transformations of the nitrogen compounds. Regarding the nitrifiers needing old sludge age, when the concentration of the biomass (suspended as well as immobilized on the filling) increased over time, the effectiveness of ammonium nitrogen removal from the sewage also increased. This influenced the nitrification process, which is presented in Figure 4.



Figure 3. Changes of ammonium nitrogen, nitrites, nitrates, and total phosphorus concentration during the experiment.



Figure 4. Nitrification rate and sludge loading with ammonium nitrogen during the experiment.

During system's accustomation, a lack of nitrification was observed, which is confirmed by the negative values of the nitrification rate. After the accustomation period, when an appropriate amount of nitrifiers' biomass was produced, the nitrification rate was increasing. During variant II, in which the reduction of ammonium nitrogen was observed, the average nitrification rate was $0.08 \pm 0.09 \text{ mgN-NH}_4 \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$. If the sludge loading with ammonium nitrogen is much higher than the nitrification rate, N-NH₄ removal is less efficient, and the ammonium form cumulates in the bioreactor. It seems that the loading with ammonium nitrogen and the hydraulic retention time

didn't play any significant role in the transformation of this nitrogen form, because in both variants, the amounts were similar, as shown in Table 1. However, the reduction of ammonium nitrogen was observed with the average hydraulic retention rate lower than 7 d. It is possible that the nitrification rate was influenced by the sewage temperature in the bioreactor, as shown in Figure 5.



Table 1. Sludge loading with ammonium nitrogen and hydraulic retention time in the bioreactor in the analyzed variants.

Figure 5. Relationship of the nitrification rate and temperature in the bioreactor.

This indicates the growth of the process' rate with the increase of the temperature, which is a common regularity. This presented relationship is statistically important at $\alpha = 0.05$.

From the beginning of the experiment until the end of variant I, the tendency of nitrate nitrogen reduction is observed, as shown in Figure 3. It is correlated with a lack of nitrification in this period. The average nitrate nitrogen concentration in the raw sewage during variant I was 1.7 ± 0.8 mgN-NO₃·dm⁻³, and in the treated sewage, it was 0.77 ± 0.3 mgN-NO₃·dm⁻³. In this case, the N-NO₃ reduction was most likely caused by heterotrophic bacteria, which are able to develop much more quickly than the chemoautotrophic nitrifiers. The average reduction of this nitrogen form was 54.7%. Significant nitrate (III) reduction can be observed in variant II of the process, where the average concentration in the outflow was 0.01 mgN-NO₃·dm⁻³ against the inflow concentration of 1.4 mgN-NO₃·dm⁻³, which means that the reduction was 99.3%. This is the example of intense denitrification that occurs together with nitrification. Periodic feeding of the bioreactor with the sewage (during the non-aeration phase) causes the anoxic conditions in the active sludge flocs and in the biofilm, mostly in its deeper parts, which enables the denitrification of N-NO₃ in the biofilm or the flocs' peripheries, which is the result of the nitrification process.

The average rate of the denitrification process in the variant I rate was $3.47 \pm 5.13 \text{ gN-NO}_x \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$, and in variant II, it was $7.56 \pm 5.71 \text{ gN-NO}_x \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$. Periodic feeding of the bioreactor with the sewage and the shorter aeration time, which caused the decrease of oxygen concentration in the bioreactor, caused the increase of the denitrification rate. The high denitrification rate and therefore the high reduction of the oxygenated form of

nitrogen resulted from the treated sewage being the source of easily decomposable substrates for the heterotrophic microbes. Figure 6 shows that when the COD/BOD₅ ratio didn't exceed 2.6, the denitrification rate was between 1.9–14.1 gN-NO_x·gsmo⁻¹·d⁻¹. Alongside an increase of hardly decomposable COD (a higher COD/BOD₅ ratio), the denitrification rate decreased, and when the COD/BOD₅ ratio was 6, this process stopped.



Figure 6. Relationship between the COD/BOD₅ ratio in the raw sewage and the nitrification rate.

The average concentration of nitrite nitrogen in the raw sewage during variants I and II was $0.05 \pm 0.04 \text{ mgN-NO}_2 \cdot \text{dm}^{-3}$. Due to the temporary N-NO₂ form, nitrite nitrogen concentration is fluctuating during the whole process. In the treated sewage during variant I, the average concentration of N-NO₂ is $0.041 \pm 0.038 \text{ mgN-NO}_2 \cdot \text{dm}^{-3}$. A significant increase of N-NO₂ concentration is observed in variant II of the process, when the average concentration was $0.32 \pm 0.28 \text{ mgN-NO}_2 \cdot \text{dm}^{-3}$. The change of exploitation conditions (decrease of oxygen concentration in the bioreactor) is believed to inhibit the second part of the nitrification process, which causes the observed accumulation of this indicator in the treated sewage.

The reduction of total phosphorus from the sewage was insignificant during the whole experiment. In variant I, the total phosphorus concentration in the raw sewage was $18.4 \pm 4.9 \text{ mgP}_{og} \cdot \text{dm}^{-3}$, and in variant II, it was 19.8 mgPog·dm⁻³. Therefore, it can be stated that in the whole tested period, the concentration of this indicator was constant in time, as shown in Figure 3. For the treated sewage in variant I, the reduction of this index could not be observed except for in the very beginning and at the end of the experiment (average concentration was $20.1 \pm 9.3 \text{ mgP}_{\text{og}} \cdot \text{dm}^{-3}$). This can be explained by the lack of developed biomass suspended in the bioreactor at the beginning of the experiment, which meant that microorganisms didn't consume the phosphorus from the sewage. The improvement of this situation could be noticed during variant II. Except for one case, during this period, the total phosphorus concentration in the bioreactor was decreasing. Its average concentration was $16.2 \pm 6.8 \text{ mgP}_{og} \cdot \text{dm}^{-3}$, so the reduction level was 18.2%. Phosphorus was consumed by the suspended bacteria in the amount that allowed them to cover the basic needs; however, the excessive intake of this biogenic compound was not observed. This was most probably due to the lack of an anaerobic zone in the system, which caused a lack of PAO bacteria. It may also be assumed that since a high denitrification ratio was observed in variant II, simultaneous phosphorus intake could occur as the result of denitrifying dephosphatation, as shown in Figure 7.



Figure 7. Relationship of denitrification ratio and total phosphorus concentration in the treated sewage.

The relationship between denitrification ratio $r_{denitrification}$ and total phosphorus concentration in the treated sewage is the evidence. The calculated correlation coefficient r = 0.62 was statistically important at $\alpha = 0.05$ level. The result of dephosphatation denitrification, apart from the high N-NO₃ reduction, was the phosphorus removal from the sewage. This process was more intense during variant II of the experiment, which had no aeration.

The influence of the BOD_5/P_{gen} ratio in the raw sewage on the total phosphorus concentration in the bioreactor's outflow was also noticed. The lowest concentration of this indicator in the treated sewage was observed when $BOD_5/P_{gen} > 23$, as shown in Figure 8. When the BOD_5/P_{gen} ratio was lower, the lack of an easily decomposed substrate used by the Poli-P bacteria in the dephosphatation process was observed.



Figure 8. Relationship of the total phosphorus concentration in the treated sewage and the BOD_5/P_{tot} ratio in the raw sewage.

4. Discussion

The quantity and quality of sewage is highly determined by the rural households' characteristic. Tenants who did not have access to the sewage treatment and drainage systems are accustomed to saving water. This is the reason for the concentration of higher pollutants in a smaller amount of water. Moreover, the water usage trends in the households causes the high heterogeneity of the sewage drainage, which exposes the treatment systems to the instable performance. In case of the systems operating on classic active sludge, which has become a more and more popular trend in Poland for small amounts of sewage treatment, severe exploitation problems may occur, such as sludge rinsing during significant hydraulic frictions, which in consequence influences the treated sewage quality. A high irregularity of the pollutants' concentration in the household sewage is also shown in the research carried out by Wasik and Chmielowski [24], Kaczor et al. [25], Bugajski et al. [26], Nowak et al. [27], Chmielowski et al. [28]. These authors, while testing the quality of sewage drained from the rural areas, noticed significant fluctuations in the BOD₅ values, COD, and/or total nitrogen. The analyses of the new sewage treatment methods show that the introduction of the biomass immobilized on the carriers as the submerged or mobile bed into the classic bioreactor with the active sludge increases the effectiveness of pollutant removal from the sewage. In the analyzed system, the obtained organic pollutants' removal depended on the variant and ranged from 68.9% to 91.9%: the removal of total suspended solids ranged from 68.6% to 80.1%, the removal of ammonium nitrogen ranged from 17.8% to 25.8%, the removal of N-NO₃ ranged from 54.7% to 99.3%. However, the removal of total phosphorus was maintained at about 18.2%. The presented results indicate the high effectiveness of the organic matter and N-NO₃ removal, and significantly better effectiveness was observed in the system that operated in the sequence of 60 min of no aeration/60 min of aeration with the periodic sewage dosage in the no-aeration phase. This situation is probably because heterotrophic bacteria that was capable of organic matter removal and denitrification dominated in the tested system, whereas the denitrification was the aerobic process. The average loading of the bioreactor with the COD loading was 2.43 gCOD·gsmo⁻¹·d⁻¹ and 0.63 kgCOD·m⁻³·d⁻¹, respectively. The average organic matter removal rate was 0.46 kgCOD·m^{-3·d⁻¹}, which equaled 1.19 gCOD·gsmo^{-1·d⁻¹ in variant I,} and 0.49 kgCOD·m⁻³·d⁻¹ and 1.80 gCOD·gsmo⁻¹·d⁻¹ in variant II. The same results described other researchers. Guo et al. [29] obtained similar results concerning the removal of sewage pollutants in a hybrid bioreactor filled with a bed made of the silk fibers. The results of the research showed a 90% reduction of COD and a 50%-80% reduction of total nitrogen. The cited researchers showed, similarly to the authors of the present paper, that the introduction of the raw sewage into the bioreactor during aeration decreases the effectiveness of nitrogen removal in comparison to the situation when the bioreactor is fed during the anoxic phase. Low denitrification effectiveness is caused by the lack of organic carbon, which acts as the electron donor for the denitrifiers, because the aerobic microorganisms oxidize a lot of the available carbon source.

Helmer and Kunst [30] and Helmer et al. [31] tested the possibility of the removal of nitrogen compounds using the rotating biological contractor. The authors found that the concentration of nitrogen compounds decreased with an oxygen concentration of $1.0 \text{ mgO}_2 \cdot \text{dm}^{-3}$ and without the external organic carbon source. Ammonium nitrogen was transformed into gas nitrogen in autotrophic conditions. About 40% of inorganic and organic nitrogen was found to be transformed into the final gas products. Therefore, it was suggested that heterotrophic microorganisms are capable of nitrification and denitrification at the same time in aerobic conditions. Microbiological analyses showed that *Thiosphaera pantotropha* and *Nitrosomonas* sp. were able to nitrify/denitrify simultaneously. Similar results were obtained by Menoud et al. [32] in the analyses of sewage nitrogen transformations in the bioreactor with SIPORAXTM blocks. It was ascertained that as the result of the oxygen concentration gradient, nitrifiers develop in the external parts of the carrier, whereas denitrifiers develop in the internal parts. The performed research showed that the maximum capacity of the simultaneous nitrification was $0.61/0.83 \text{ kgN}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$, and the oxygen concentration was above $1.0 \text{ mgO}_2 \cdot \text{dm}^{-3}$. Rodgers [33], while testing the system with the plastic bed periodically submerged

in the bioreactor and a constant sewage dosage, gained an organic matter removal rate as high as $3.8 \text{ kgCOD} \cdot \text{m}^{-3} \cdot \text{d}^{-1}$ with a 92% efficiency of COD reduction.

Research regarding a moving bed sequencing batch reactor has also been conducted by other authors. Cao et al. [34] analyzed the effect of dissolved oxygen concentration on oxygen diffusion and the bacterial community structure in a moving bed sequencing batch reactor. Sytek-Szmeichel et al. [35] tested the efficiency of wastewater treatment in MBSBBR systems in specified technological conditions with a sequence MBSBBR bioreactor. Dulkadiroglu et al. [36] modeled nitrate concentrations in a moving bed sequencing batch biofilm reactor using an artificial neural network technique. In the work Gilbert et al. [37], the low temperature partial nitritation/anammox in a moving bed biofilm reactor treating low strength wastewater was evaluated. Koupaie et al. [38] evaluated an integrated anaerobic/aerobic fixed-bed sequencing batch biofilm reactor for the decolorization and biodegradation of azo dye Acid Red 18, where the comparison of using two types of packing media was carried out. Bassin et al. [39] focused on effect of different operational conditions on biofilm development, nitrification, and nitrifying microbial population in moving-bed biofilm reactors. The research carried out by Lim et al. [40] concerned the enhancement of nitrogen removal in a moving bed sequencing batch reactor with intermittent aeration during an aerobic REACT period. Persson et al. [41] studied the structure and composition of biofilm communities in a moving bed biofilm reactor for nitritation/anammox at low temperatures. Jaroszyński et al. [42] analyzed the impact of free ammonia on anammox rates in a moving bed biofilm reactor. The studies carried out by Zekker et al. [43] focused on the effects of anammox enrichment from reject water on blank biofilm carriers and carriers containing nitrifying biomass on the operation of two moving bed biofilm reactors.

In the nitrifying beds loaded with a high amount of ammonium nitrogen, the nitrification rate ranged between 0.18–0.35 gN-NH₄·gsmo⁻¹·d⁻¹ [44,45], and in the one found by Helmer et al. [31] in the rotating disk filters, the nitrification rate remained on the level of 7.2 mgN-NH₄·gsmo⁻¹·h⁻¹ with an oxygen concentration of 1.0 mgO₂·dm⁻³. The presented analyses show the nitrification rate at the level of 0.08 gN-NH₄·gsmo⁻¹·d⁻¹.

The increase of the denitrification rate in variant II of the experiment could have been caused by the periodic sewage dosage during the no-aeration phase. Such a type of sewage dosage, especially with a high concentration of dissolved COD fraction, significantly increased the denitrification process [46].

The high level of nitrates (III) reduction in the sewage, which was observed in the presented research regardless of the oxygen dissolved in the bioreactor, suggests the presence of microorganisms that are capable of denitrification in the aerobic conditions. This phenomenon is often observed in the systems with biomass immobilized on the carrier (e.g., circular, moving, membrane, and fluidal beds) or in the so-called granulated active sludge in the periodically operating systems [47–57]. This is caused by the occurrence of the oxygen concentration gradients in the biological membrane or in the granulated active sludge. The occurrence of external aerobic layers together with the anoxic conditions in the deeper membrane layers is possible in this situation [58,59]. Podedworna and Żubrowska-Sudoł [60] in the research of the moving-bed sequencing batch biofilm reactor, MBSBBR, obtained a similar reduction of organic pollution with the loading between 0.227–0.684 kgCOD·m⁻³·d⁻¹. The quoted authors gained nearly 100% nitrification of the ammonium nitrogen with the sludge age of 1–2 d, which resulted from the nitrifiers' immobilization on the bed. However, the high denitrification efficiency resulted from the increase of the bioreactor loading up to 0.528–0.687 kgCOD·m⁻³·d⁻¹.

In the discussed experiment, the increase in the phosphorus removal effectiveness was observed together with the increase of the BOD_5/P ratio. A similar phenomenon was observed by Vaboliene et al. [61] in the experiment in the active sludge bioreactor with the simultaneous nitrification/denitrification option. The lowest total phosphorus concentration was found with the BOD_7/P ratio higher than 15. Although a relationship between the total phosphorus concentration in the outflow and the denitrification rate was observed, the denitrification caused only partial phosphorus removal from the sewage. In the anaerobic conditions, the efficacy of the energy production from the denitrifying dephosphatation amounted to about 40% in relation to the energy produced

in the aerobic conditions. The denitrifying dephosphatation could have been limited in the tested system due to the low N-NO₃ concentration, which is the electron acceptor for the Poli-P bacteria [62]. Mishima et al. [63], in their research of a flow bioreactor with the anoxic, anaerobic, and aerobic zones filled with the hygroscopic gel, found an increased phosphates intake during the denitrification process. A similar observation was found in the presented research.

It seems that similarly to ammonium nitrogen, phosphorus was removed mainly due to the heterotrophic bacteria intake. Partly, this was caused by a low suspended biomass' concentration in the bioreactor, which enables the possibility of the increased phosphorus intake from the sewage. A relatively small amount of the suspended biomass in the bioreactor was observed, which was $86.8 \text{ mg} \cdot \text{dm}^{-3}$ on average. Similar observations were found by Hamoda and Al-Ghusain [64], where in the tested bioreactor with the submerged bed (the ceramic vertical elements), the suspended biomass constituted about 5% of the total biomass in the bioreactor, which corresponded with the 120 mg $\cdot \text{dm}^{-3}$ concentration in the beginning to 25 mg $\cdot \text{dm}^{-3}$ at the end of the bioreactor. The main suspension mass was attached to the bed.

5. Conclusions

The work presents the effectiveness of the removal of organic pollutants, nitrogen and phosphorus, from household sewage in a hybrid bioreactor with a submerged fixed bed. The experiment was carried out in a laboratory model of the hybrid bioreactor in two exploitation variants: variant I-120 min of aeration/60 min of no aeration and a constant sewage dosage, and variant II-60 min of aeration/60 min of no aeration with a periodic sewage dosage in the no-aeration phase. The experiment was carried out on real sewage primarily treated in the septic tank. On the basis of the carried out research, it can be concluded that the tested hybrid system provided high organic pollutants' and nitrate nitrogen reduction from the sewage. Regarding the removal of organic pollutants and nutrients, the variant with 60 min of aeration and 60 min of no-aeration phases and a periodic sewage dosage during the no-aeration phase seemed to be more favorable. Such exploitation increases the denitrification process. In the tested system, a low nitrification rate combined with high denitrification was observed. This situation was observed mostly in variant II. That the system was often overloaded with ammonium nitrogen caused the inhibition of the nitrification process. Ammonium nitrogen as well as total phosphorus were mainly removed due to the intake of heterotrophic microorganisms. The denitrifying dephosphatation process could have had an influence on the total phosphorus removal, which is confirmed by the statistically important relationship between the total phosphorus concentration in the treated sewage and the denitrification rate. Generally, on the basis of the performed study, the following conclusions can be formed:

- (1) There was a higher stable amount of pollutants removal from the sewage in the case with biofilm creation. The biofilm is much more resistant to sudden changes in the conditions than the suspended biomass.
- (2) In variant II (60 min of aeration and 60 min of no-aeration phases, and a periodic sewage dosage during the no-aeration phase), in comparison to variant I (constant sewage dosage 24 h a day, changeable aeration cycles—120 min with aeration/60 min with no aeration) there was a greater removal of organic pollutants and nutrient. The mean removal of organic compounds was equal to 86.3% for BOD₅ and 68.9% for COD in variant I, but for variant II, these rates were 91.9% and 80.1% for BOD₅ and COD, respectively. Ammonium nitrogen was observed to increase the concentration in the outflow during variant I by 17.8%, and by 25.8% for variant II.
- (3) The rate of nitrification during variant II was equal to $0.08 \pm 0.09 \text{ mgN-NH}_4 \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$. If the sludge loading with ammonium nitrogen is much higher than the nitrification rate, then N-NH₄ removal is less efficient, and the ammonium form cumulates in the bioreactor. The rate of nitrification was positively correlated with the sewage temperature in the bioreactor.
- (4) The highest rate of denitrification was observed during variant II, and was equal to $7.56 \pm 5.71 \text{ gN-NO}_x \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$ in comparison to variant I ($3.47 \pm 5.13 \text{ gN-NO}_x \cdot \text{gsmo}^{-1} \cdot \text{d}^{-1}$).

(5) The dephosphatation denitrification process was observed especially during variant II. The result of dephosphatation denitrification, apart from the high N-NO₃ reduction, was the phosphorus removal from the sewage. On average, the reduction level for total phosphorus was equal to 18.2%.

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