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EFFECTIVENESS OF WASTEWATER POST-TREATMENT IN FILTER COLUMNS WITH THE USE OF MINERAL MATERIALS

Based on the assumptions of the circular economy model and sustainable development, we are currently looking for natural and ecological materials in terms of wastewater treatment of pollutants. This article presents the research of three mineral materials – lava rock (LR), lightweight sintered aggregate (LSA) and lightweight clay aggregates (LCA) used as filling of filtration columns for the treatment of real wastewater. The filtration process was carried out under various hydraulic loads in two columns, one of which was additionally supported by the aeration process. The post-treated sewage was characterized by the following parameters: COD (chemical oxygen demand), TOC (total organic carbon), phosphate phosphorus (P-PO_4), total nitrogen (TN) and total phosphorus (TP). Among the hydraulic loads applied, the most optimal loads were Oh_I = 0.25 $m^3/(m^2 \cdot h)$. Certyd turned out to be the most effective with supporting filtration with the aeration process in removing organic compounds (reduction of 65.1% COD and 38.2% TOC at Oh_I). Lava rock seems to be a promising material reactive in terms of removal of biogenic compounds such as nitrogen (efficiency 23.8% at Oh_I) and phosphorus (64.2% reduction of TP at Oh_I) and organic compounds (21.4% reduction of TOC at Oh_I in conditions without aeration). LCA shows the best efficiency in sorption of phosphorus compounds (41.6% reduction of TP at Oh_I) and organic compounds (21.4% TOC reduction at Oh_I under non-aerated conditions).

Keywords: wastewater treatment, filtration, natural materials, lava rock, lightweight sintered aggregate, lightweight clay aggregates

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1. Introduction

Due to the still progressing development of civilization and the growing awareness of the impact of the necessity to preserve the natural environment as intact as possible, higher and higher requirements are imposed on wastewater treatment plants regarding the quality of treated sewage discharged into receivers [2].

The control of nitrogen and phosphorus content in the wastewater after the treatment process is aimed at preventing the eutrophication process caused by the excessive production of organic matter in water reservoirs that are over-normatively enriched with nutrients. Monitoring of this process is necessary to ensure the continuity of natural aquatic ecosystems, which has a direct impact on the quality of water intended for human consumption [1, 7, 8, 14].

In wastewater treatment technology, verification of parameters such as total organic carbon (TOC) or chemical oxygen demand (COD), in addition to the requirements resulting from legal regulations, is necessary for the proper control of processes (e.g. biological) in order to maximize the efficiency of wastewater treatment plants [6].

Currently, materials are sought that can be used in wastewater treatment processes at various stages of treatment, with particular focus on natural and cheap materials in line with green chemistry and sustainable development. The use of such materials is consistent with the assumption of the circular economy model, which assumes that manufactured products, as well as materials and raw materials, should remain in the economy for as long as possible [5, 9, 10].

This article presents the research of three mineral materials (lava rock, lightweight sintered aggregate, lightweight clay aggregates) in the aspect of wastewater treatment with the use of the filtration process.

2. Materials and methods

2.1. Materials used for research

Certyd® is an commercial name of lightweight sintered aggregate (LSA), which manufactured exclusively by the LSA Aggregate Production Plant in Białystok (Poland). For the production of this material, mainly ash obtained from burning hard coal in fine coal boilers is used, which is subjected to high-temperature sintering at a temperature of 1000–1200°C. LSA is an aggregate that is characterized by high strength and lightness. Certyd® is a material that is neutral to the environment, and the technology of its production is waste-free and safe. The bulk density of this material is approximately 725 kg/m^3, while the water retention capacity ranges from 17% to 19% [3, 11]. Currently, Certyd® in the tests, LSA with a grain diameter from 8 mm to 16 mm was used (fig. 1a).

Lightweight clay aggregates (LCA) or expanded clay so-called of Ceramsite is a lightweight aggregate made by heating clay in a rotary kiln. In study used of expanded clay known under the commercial name Filtralite®, produced by Leca® Norway. LCA has a porosity of 70–80%, while the bulk density is from 650 to 900 kg/m³. This is a chemically inert material with an absorption capacity of about 20%. The raw clay is thermally treated in tube furnaces at a temperature of 1150°C, where it swells and forms spheres with a porous internal structure [4, 12]. In the conducted experiments, LCA with a grain diameter from 2.5 mm to 5 mm was used (fig. 1b). The material is mostly aluminum silicate with few percent of iron and alkali oxides.

Lava rock (LR) is an aggregate that is produced from the liquid product of a volcanic eruption. It consists mainly of fused oxides of silicon, iron, sodium, potassium and calcium. Lava, due to its porous structure, has a high ability to store and filter water, and at the same time retains high lightness. The red volcanic lava rock from Italy was used in this research. Water absorption after 24 hours is at the level of 15–20%, while its porosity is about 40–60%, which makes it very efficient – the bulk density of this material is 1000 kg/m³ [13]. Figure 1c shows a lava rock with a diameter of 2.5 mm to 5 mm that was used during the tests.

Fig. 1. Materials used for research a) LSA b) LCA c) LR

The research was carried out with the use of real wastewater after the second stage of (biological) treatment, from the Rzeszów WWTP. The quality of this wastewater is presented in tab. 1.

Table 1. Characteristics of wastewater used in research

Indicator	**COD**	**TOC**	**TN**	**$P\text{-}PO_4$**	**TP**
Unit	mg O_2/dm^3	mg C/dm^3	mg N/dm^3	mg $P\text{-}PO_4/dm^3$	mg P/dm^3
Value	44.0	18.1	10.3	0.50	0.73

2.2. Research methodology

Parameters of treated wastewater were determined by determination of organic compounds defined by COD (chemical oxygen demand) and TOC (total organic carbon) indicators. The concentration of phosphate phosphorus ($P\text{-}PO_4$), total nitrogen (TN) and total phosphorus (TP) was also tested.

The chemical oxygen demand was determined by the bichromate method based on the PN-74 C-04578 standard, while the total organic carbon was determined using IR spectrometry, after oxidation to CO_2 at a temperature > 600°C, in accordance with PN-EN ISO 1484:1999.

The total nitrogen content was determined using the peroxodisulphate mineralization method for 1 hour at a temperature of 120°C based on the PN-EN ISO 11905-1:2001 standard.

Total phosphorus and phosphate phosphorus were determined using the method developed on the basis of the PN-EN ISO 6878:2006 standard.

The tests were carried out in two filter columns with an internal diameter of 5 cm, an active height of 50 cm and a total height of 70 cm. The bed filling was 50 cm. In order to rinse possible impurities from the filter bed, the columns were rinsed with deionized water. The filtration process was carried out with various hydraulic loads (tab. 2) through the filtration columns – one of the columns additionally used the aeration process. Samples of post-treated wastewater for laboratory analyzes were collected taking into account the contact time for individual values of hydraulic load.

In each of the collected samples of post-treated sewage, the parameters indicated above were determined as for treated wastewater, i.e. COD, TOC, TN, TP, $P\text{-}PO_4$.

Table 2. The values of hydraulic loads used during the experiments

Indication	Hydraulic load value [$m^3/(m^2 \cdot h)$]
Oh_I	0.25
Oh_{II}	0.50
Oh_{III}	1.00
Oh_{IV}	2.50

3. Results and discussion

3.1. Wastewater filtration with aeration process

The values of individual parameters in the wastewater post-treated in a filter column filled with LSA, LCA and LR additionally supported by the aeration process are presented in tables 3–5.

The research shows that the lowest values of individual parameters in the post-treated wastewater were most often obtained for each of the materials for the hydraulic load of Oh_I. The exception is Lava rock, for which the final concentration of TOC and TN showed the lowest value for the load of Oh_{III}.

Taking into account the organic compounds determined by the COD index, the best efficiency was demonstrated by LSA - with a load of Oh_I, a reduction of 65.1% was achieved. Figure 2 shows a comparison of the effectiveness of individual materials in this aspect. Another effective material was Lava rock with an efficiency of 18.2%. With the increase of the hydraulic load, the ability of each of the materials to remove organic compounds determined by the COD index decreased significantly. LCA turned out to be the least effective material in this respect.

With regard to total organic carbon, the highest removal efficiency – at the level of 30.4% was also achieved for the LSA material. The other materials lava rock and lightweight clay aggregates showed a reduction of 22.2% and 21.4% respectively for the most optimal load Oh_I (fig. 3).

The lowest total nitrogen concentration in the post-treated wastewater was 7.67 mg N/dm^3 – in the case of the treating process in the filtration column with the use of Lava rock as filling and with the hydraulic load of Oh_{III}. The efficiency of TN removal was 25.4%. Additionally, high efficiency in the range from 23.4% to 25.4% was maintained for all load sizes. LCA for the load of Oh_I showed a reduction of total nitrogen at the level of 9.1%, while LSA was 6.6%. The efficiency of total nitrogen removal for individual materials and loads is shown in figure 4.

The efficiency of removing phosphorus compounds (TP and P-PO_4) from wastewater by individual materials is presented in figure 5. In the case of total phosphorus, Lava rock showed the best ability to reduce this pollutant, reaching the efficiency of 50.4%. The next material for which high efficiency was obtained – 41.6%, is LCA. Both materials retained their efficiency not less than 25% for the Oh_{II} and Oh_{III} loads, while for Oh_{IV} a sharp drop in efficiency was observed for Lava rock. With regard to phosphate phosphorus, LR showed even greater efficiency for the Oh_I-Oh_{II} loads ranging from 57.5% to 34.3%, while LCA ranged from 19.4% to 38.8%. In the case of LSA, the greatest removal efficiency, both for total phosphorus and phosphate phosphorus, was achieved for the Oh_I load and was at the level of 8.1% and 12.2% respectively.

Table 3. The quality of wastewater post-treated with the use of the LSA material

Indicator	Unit	Treated wastewater	$O_{h\,I}$	$O_{h\,II}$	$O_{h\,III}$	$O_{h\,IV}$
COD	mg O_2/dm^3	44.0	14.8	23.6	32.8	36.0
TOC	mg C/dm^3	18.1	14.0	15.9	15.6	17.0
TN	mg N/dm^3	10.3	10.66	10.88	10.80	10.97
P-PO_4	mg P-PO_4/dm^3	0.50	0.43	0.44	0.46	0.47
TP	mg P/dm^3	0.73	0.68	0.70	0.71	0.72

Table 4. The quality of wastewater post-treated with the use of the LCA material

Indicator	Unit	Treated wastewater	$O_{h\ I}$	$O_{h\ II}$	$O_{h\ III}$	$O_{h\ IV}$
COD	mg O_2/dm^3	44.0	40.0	40.4	41.6	41.6
TOC	mg C/dm^3	18.1	15.2	16.5	16.8	16.9
TN	mg N/dm^3	10.3	9.57	9.80	9.70	9.89
P-PO$_4$	mg P-PO_4/dm^3	0.50	0.52	0.67	0.69	0.76
TP	mg P/dm^3	0.73	0.67	0.82	0.86	0.93

Table 5. The quality of wastewater post-treated with the use of the LR material

Indicator	Unit	Treated wastewater	$O_{h\ I}$	$O_{h\ II}$	$O_{h\ III}$	$O_{h\ IV}$
COD	mg O_2/dm^3	44.0	36.0	37.6	40.4	41.6
TOC	mg C/dm^3	18.1	14.1	14.0	13.8	14.3
TN	mg N/dm^3	10.3	7.84	7.84	7.67	7.87
P-PO$_4$	mg P-PO_4/dm^3	0.50	0.21	0.31	0.33	0.48
TP	mg P/dm^3	0.73	0.36	0.50	0.54	0.72

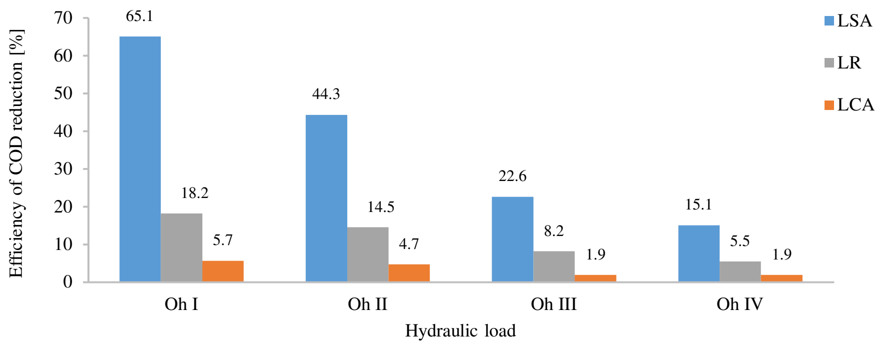

Fig. 2. The efficiency of removal of organic compounds, determined by the COD, depending on the filling material and hydraulic load – filtration process with aeration

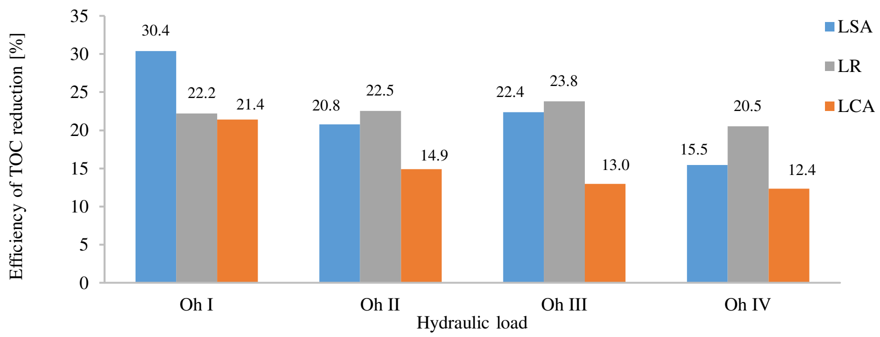

Fig. 3. Effectiveness of TOC reduction depending on the filling material and hydraulic load – filtration process with aeration

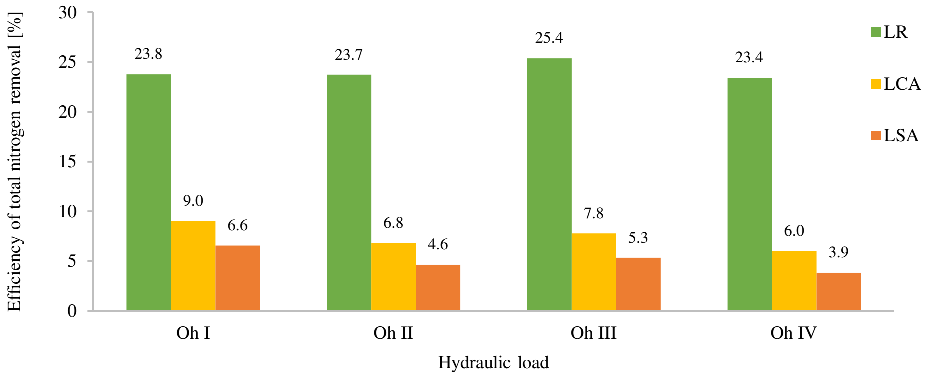

Fig. 4. Effectiveness of total nitrogen removal depending on the filling material and hydraulic load – filtration process with aeration

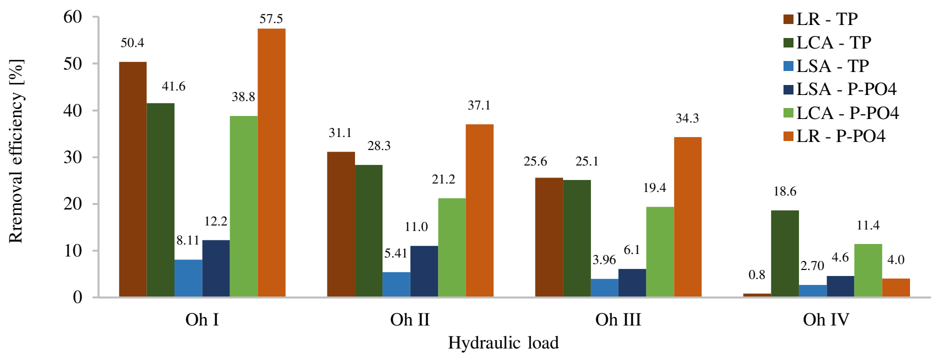

Fig. 5. Effectiveness of phosphorus compounds removal depending on the filling material and hydraulic load – filtration process with aeration

3.2. Wastewater filtration without the aeration process

The quality of wastewater post-treated in the filtration columns filled with LSA, LCA and LR, without the support of the aeration process, is presented in tab. 6–8.

With regard to the content of organic compounds, defined as COD, the highest removal efficiency was achieved on the filling made of lava rock (30.9%) and LSA (23.6%). In both cases, these values were recorded for the hydraulic load Oh_I (fig. 6). This shows the opposite tendency than in the treatment with aeration, where LSA turned out to be the most effective, and the remaining materials showed a low COD reduction efficiency. In the case of TOC (fig. 7), all materials retained a similar efficiency as in the process assisted by aeration, ranging from 20.0% to 38.2%.

The lowest final content of total nitrogen was noted in the wastewater post-treated with a filter made of lava rock material, it was 7.78 mg N/dm^3 (value for Oh_I), which allowed to achieve a similar level of efficiency for all loads – in the range of 23.1–24.4%, in accordance with the results presented in figure 8. The other two materials also retained similar efficiency as in the process with aeration.

Phosphorus compounds were most effectively removed in the treating process in a filter filled with Lava rock material, achieving a reduction of 64.2% for TP and 65.1% for P-PO_4, thus showing an increase in efficiency compared to the process with aeration by approximately 10%. For the remaining materials, comparable reductions in both TP and P-PO_4 were achieved as in the case of the aeration assisted filtration process. Lightweight sintered aggregate turned out to be the least effective, showing for the most optimal load (Oh_I) a reduction of 8.1% for TP and 12.2% for P-PO_4. The effectiveness of individual materials in the removal of phosphorus compounds is shown in figure 9.

Table 6. The quality of wastewater post-treated with the use of the LSA material

Indicator	Unit	Treated wastewater	$O_{h\,I}$	$O_{h\,II}$	$O_{h\,III}$	$O_{h\,IV}$
COD	mg O_2/dm^3	44.0	41.6	40.4	35.2	32.4
TOC	mg C/dm^3	18.1	12.4	13.7	14.9	15.9
TN	mg N/dm^3	10.3	10.70	10.89	10.86	10.96
P-PO_4	mg P-PO_4/dm^3	0.5	0.42	0.43	0.45	0.47
TP	mg P/dm^3	0.73	0.65	0.67	0.68	0.71

Table 7. The quality of wastewater post-treated with the use of the LCA material

Indicator	Unit	Treated wastewater	$O_{h\,I}$	$O_{h\,II}$	$O_{h\,III}$	$O_{h\,IV}$
COD	mg O_2/dm^3	44.0	32.4	35.2	37.6	39.2
TOC	mg C/dm^3	18.1	13.4	15.5	14.5	15.5
TN	mg N/dm^3	10.3	9.57	9.77	9.66	9.89
P-PO_4	mg P-PO_4/dm^3	0.5	0.53	0.67	0.70	0.84
TP	mg P/dm^3	0.73	0.70	0.81	0.96	0.99

Table 8. The quality of wastewater post-treated with the use of the LR material

Indicator	Unit	Treated wastewater	$O_{h\,I}$	$O_{h\,II}$	$O_{h\,III}$	$O_{h\,IV}$
COD	mg O_2/dm^3	44.0	30.4	35.6	38.8	40.4
TOC	mg C/dm^3	18.1	12.8	13.2	13.1	13.5
TN	mg N/dm^3	10.3	7.78	7.85	7.85	7.91
P-PO_4	mg P-PO_4/dm^3	0.5	0.17	0.23	0.24	0.36
TP	mg P/dm^3	0.73	0.26	0.36	0.41	0.57

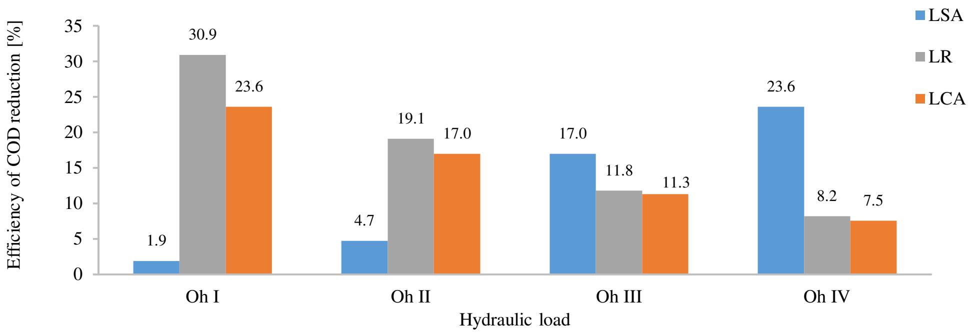

Fig. 6. The efficiency of removing organic compounds, determined by the COD, depending on the filling material and the hydraulic load – filtration process without aeration

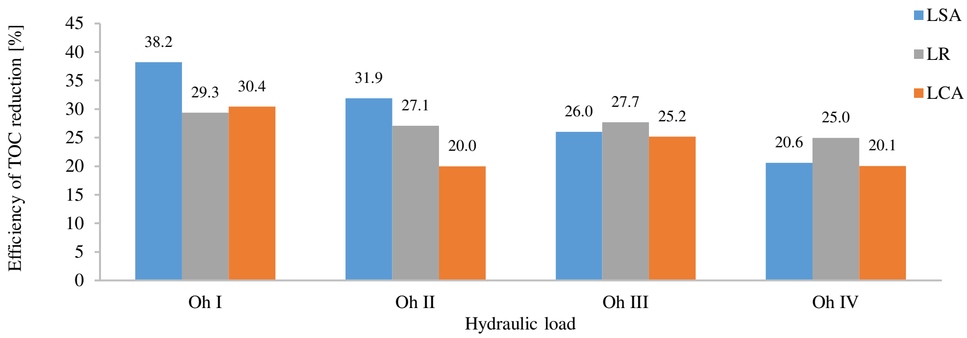

Fig. 7. The efficiency of TOC reduction depending on the filling material and hydraulic load – filtration process without aeration

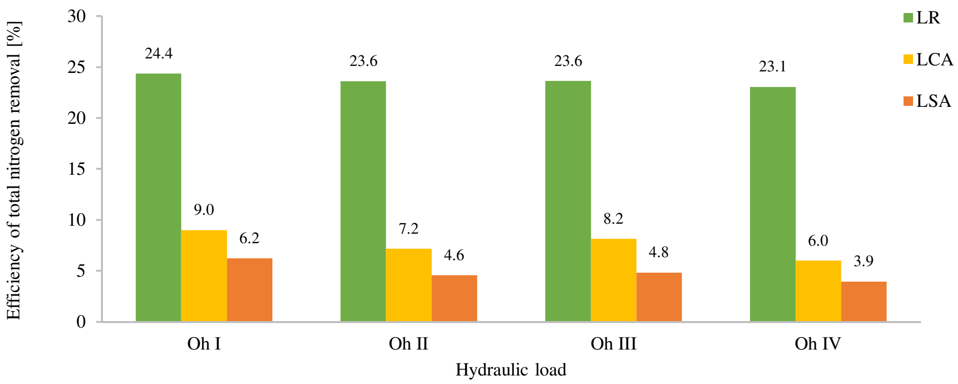

Fig. 8. Effectiveness of total nitrogen removal depending on the filling material and hydraulic load – filtration process without aeration

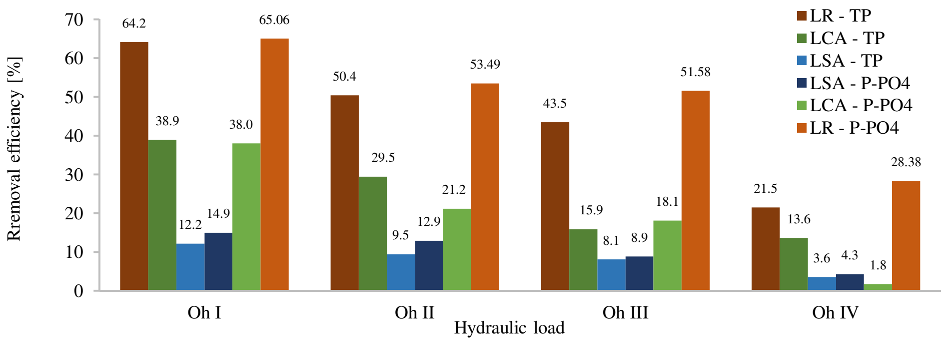

Fig. 9. Effectiveness of removing phosphorus compounds depending on the filling material and hydraulic load – filtration process without aeration

3.3. Discussion of the results

Lightweight sintered aggregate achieved the best results for the removal of organic compounds expressed by the COD index in the case of the aeration assisted process. This indicates that aeration supports the process of decomposition of organic compounds through their additional mineralization on the LSA surface, which, without this process, proved to be much less effective. In the case of TOC, the aeration process negatively influenced the reduction of pollutants expressed by this index, resulting in a reduction in the efficiency of the process for each of the applied loads. In relation to nitrogen and phosphorus compounds, LSA showed a much lower ability to reduce these impurities (less than 10%) in comparison to organic compounds. The influence of the aeration process or the variable load was of little importance in this aspect.

The use of LCA gave the most satisfactory results in the case of removal of phosphorus compounds with the aeration process for all hydraulic loads, which may indicate that the aeration process facilitates the sorption or precipitation of phosphorus by this material. Nevertheless, without the aeration process, this material also achieved satisfactory results for this parameter. In addition, the high efficiency of phosphorus removal means that LCA may contain calcium, aluminum, iron or magnesium compounds, which have a natural ability to sorb and precipitate phosphorus compounds [5]. In the case of this material, additional aeration allowed to obtain a higher degree of reduction of organic compounds (determined by the COD) thanks to their additional mineralization. The efficiency of lowering the TOC parameter slightly decreased in the case of support with an additional aeration process, but still maintaining the desired effect. The removal of nitrogen compounds by lightweight clay aggregates was on the level of 6.0% to 9.1% without any significant influence of the additional aeration process.

Lava rock is a material that has distinguished itself significantly from the others in its high efficiency in removing nitrogen and phosphorus compounds.

In the case of total nitrogen, the degree of reduction was not influenced by either the aeration process or the size of the load. Removal of phosphorus compounds was slightly more efficient with the additional aeration process, which may indicate that the nitrogen and phosphorus binding processes by Lava are not dependent on system aeration. Lava rock, like lightweight clay aggregates, is also a reactive material that has the ability to selectively remove phosphorus by sorption or precipitation [9]. In the removal of organic compounds, both in relation to COD and TOC, the aeration process negatively influenced their removal, especially in the case of the former.

4. Conclusions

In the conducted experiments, each of the three materials used as a filling in the filtration columns, showed a satisfactory efficiency depending on the conditions and type of pollution.

1. The most optimal hydraulic load for wastewater treatment in removing pollutants covered by the research is $Oh_I = 0.25\ m^3/(m^2 \cdot h)$. In almost every case, for this load value, the efficiency of removing individual pollutants was the highest, and the efficiency decreased with increasing hydraulic load. This may be caused by the disturbance of the favorable conditions of the impurities sorption process by the reduced availability of substances to the grains of the bed, as well as by the clogging of the filling as a result of increasing loading.
2. Supporting the process of wastewater treatment with aeration was advantageous mainly in the case of removing organic pollutants, due to the additionally ongoing process of mineralization of these substances.
3. LSA proved to be most effective material in assisting filtration with the aeration process in removing organic compounds.
4. Lava rock seems to be a promising material reactive in the removal of biogenic compounds such as nitrogen and phosphorus, due to its high efficiency in removing these compounds.
5. LCA shows satisfactory efficiency both in sorption of phosphorus compounds and organic compounds.
6. In the case of post-treating wastewater with a small amount of impurities, the obtained effectiveness of each of the tested materials indicates the possibility of their use in real conditions. It is worth emphasizing that the process of post-treating biologically treated wastewater will allow for the removal of residual pollutants, which may ultimately be important when using post-treated wastewater, e.g. for the purpose of irrigating areas.
7. Further research is recommended to determine the conditions and mechanisms of binding individual pollutants by the investigated mineral materials.

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