

## Environmental engineering Aplinkos inžinerija

### RESEARCH OF WASTEWATER TERTIARY TREATMENT

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**Abstract.** Small individual household wastewater treatment plants not always operate well. Consequently, the concentrations of ammonium nitrogen and phosphate phosphorus are exceeded. The aim of the work was to find the most effective material for wastewater tertiary treatment and propose environmentally friendly ways to solve fresh water problem. Three filter fillings (foam-glass, zeolite and biochar) were tested. As a result, we have got a following conclusion, foam-glass removed phosphate phosphorus most effectively (efficacy – 14–91%) and against ammonium nitrogen, comparatively effective was zeolite (efficacy – 29–100%).

**Keywords:** tertiary treatment, phosphorus and ammonium removal, zeolite, foam-glass, biochar.

#### Introduction

One of the many consequences of rapid industrialization and urbanization has been the generation and discharge of large volumes of wastewater from municipal, industrial and agricultural sources (Achilli, Cath, Marchand, & Childress, 2009). While the presence of aromatics and heavy metals in wastewater have adverse effects on the environment and living beings, the presence of excessive amounts of innocuous nutrients, such as nitrogen (N) and phosphorus (P), can also upset the balance of aquatic ecosystem through eutrophication. Eutrophication is a process of rapid plant/algae growth in natural water bodies due to nutrient overloading; this leads to oxygen depletion resulting in deterioration of water quality and endangering of aquatic life.

Conventional techniques for N and P removal from wastewater are based on physical and chemical methods. These techniques are not economical and do not facilitate nutrients recycle and reuse (Wu, Yang, & Lin, 2005).

Disposal of improperly treated wastewater often pose risk to the environment and ecology. Using advanced technology to mitigate risk by refine wastewater treatment is a key issue in meeting legislative guidelines, e.g. EU Water Framework Directive. Municipal wastewater treatment typically comprises preliminary treatment, primary treatment and secondary treatment. Preliminary treatment includes a series of screens and grit removal to prepare wastewater for subsequent treatment. Primary treatment

involves the separation of readily-removable suspended solids through gravity sedimentation. Following these two basic processes,

wastewater is then subjected to secondary treatment in which biological and/or chemical processes are involved to remove dissolved constituents. The secondary treatment was previously considered as a complete process, with its effluent being discharged into the receiving environment after disinfection with chlorine gas. However, as environmental regulations are getting stringent and introduction of EU Water Framework Directive in 2000, secondary sewage effluent was no longer a guarantee for discharge. Advanced tertiary treatment is therefore, required for further decreasing the residual constituents in secondary sewage effluent (Choi, 2015; Praveen & Loh, 2015; Sirmerova, Prochazkova, Siristova, Kolska, & Branzyk, 2013).

There also exist important reserches, where activated carbon is used. However, activated carbon is less economically viable as an adsorbent due to the costly activation and regeneration of the spent carbon and disposal of regenerant wastes. As a result, over recent years there has been growing interest in using low-cost natural minerals for treating wastewater (Xu, Bernards, & Hu, 2014). In this article are analyzed three types of filter materials, which can be used for wastewater tertiary treatment. Its aim was to determine which of them is the most effective this way.

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## 1. Methods and materials

### 1.1. Wastewater samples and domestic wastewater treatment plant (WWTP)

In the work was applied one of Lithuania's most popular low-capacity domestic wastewater treatment plant (WWTP) – AT-6, which operating principle is based on the active sludge basis – operational research. The AT-6 cleaning unit is completed in one container. After flowing out from the plant, water is secondary treated.

### 1.2. Samples

Wastewater samples were taken from the AT-6 wastewater treatment plant (WWTP), which is located in Vilnius city (gardening community) next to the dwelling house. The device is operated by a family (four members) for four years. The design capacity of this unit is 0.54 m<sup>3</sup>/d, the design load per day with organic pollutants is 0.24 kg/d. Purified wastewater is discharged into an infiltration well.

Investigations conducted on February 13, 2018– May 17, 2018. The treated wastewater samples (4 to 30 liters) were taken from the outflow pipe of the AT-6 type, once a week at the same time of day (about 7:30 pm) temperature was measured on place. Samples were transported to the VGTU laboratory of the Department of Environmental and Water Engineering. Wastewater samples were allowed to warm up to room temperature at the laboratory, then their pH, chemical oxygen demand (ChDS<sub>Cr</sub>), biochemical oxygen demand (BOD), concentration of suspended matter (SM), nitrate nitrogen (NO<sub>3</sub>-N), nitrite nitrogen (NO<sub>2</sub>-N), ammonium nitrogen (NH<sub>4</sub>-N) and orthophosphate phosphorus (PO<sub>4</sub>-P) concentrations. Measurements are made by metrologically verified meters. Each sample was tested two or three times (after 40 minutes, 1.5 hours of filtering and before the end).



Figure 1. Filtrating columns stand. Fillers: 1 – foam-glass; 2 – biochar; 3 – zeolite (source: compiled by author)

Examination of sewage contamination was reduced by filtering through granular excipients. At VGTU laboratory of Environmental Protection and Water Engineering Department was installed filtrating columns stand (Figure 1).

A composite sample was prepared from the treated wastewater, which was evenly distributed to the filtration columns. Throughout all fillers, the effluent was filtered at 0.87 m/h (1.2 l/h flow), the filtration rate was determined by the volumetric method. During the experiments, samples of the filtrate were taken from all the columns at the same time. The filtrated samples were measured following some same parameters as in the initial wastewater composite sample. The efficiency of the removal of pollutants is calculated according to the formula:

$$E(X)_i = \frac{X_{1,i} - X_{2,i}}{X_{1,i}} \cdot 100 \%,$$

where  $E(X)_i$  – the efficiency of the respective pollutant, %;  $X_{1,i}$  – concentration of the respective pollutant before treatment, mg/l;  $X_{2,i}$  – the concentration of the respective pollutant after treatment, mg/l.

In the work were used following fillers (aggregates) to filter wastewater, which was taken from domestic wastewater treatment plant – AT-6.

### 1.3. Zeolite

The zeolite was selected as is natural, ecological sorbent, passable treated water characteristics (Miladinovic & Weatherley, 2008; Mažeikienė, Valentukevičienė, Rimeika, Matuzevičius, & Daukny, 2008).

Zeolite is a crystalline hydrated aluminosilicate with a framework structure containing pores occupied by water, alkali and alkaline earth cations. Due to their high cation-exchange ability as well as to the molecular sieve properties, natural zeolites (being cheap materials, easily available in large quantities in many places of the world) show special importance in water and gas purification, adsorption and catalysis (Miladinovic & Weatherley, 2008).

The particle size of 0.6–2.0 mm (separated by calibrated sieves) of natural zeolite were used into this experimental research on removal of ammonium ions from wastewater (Figure 2). Clinoptilolite rock from the Sokyrnytskaya deposit (the Transcarpathian region, Ukraine) containing 70–75% clinoptilolite was used in this research. Natural zeolite granules were washed with distilled water for



Figure 2. Zeolite granules (source: compiled by author)

undesirable turbidity removal and dried into laboratory oven, 105°C of temperature. The weight of the material was 442 g (0.5 l volume).

#### 1.4. Foam-glass

Granulated foam-glass – it is an inorganic thermo-insulated material, produced from glass breakage, in small pore granule form. Compound was heated in a furnace in especially high temperature as well as made into various diameter greyish granules. Granulated foam-glass is a unique ecological product produced from waste. Granulated foam-glass can be used in such areas like farming, supply systems, oil extraction, bio filtration and other.

Porous granulated foam-glass granule research was carried out in the work (Figure 3). The research used foam-glass granules produced by UAB “Stikloporas” (Druskininkai, Lithuania) production line. The main characteristics of foam-glass granules during the research were explored. It was determined whether grey foam-glass granules fit according to their characteristics related to water effect to material using contaminated water cleaning technologies. The weight of the material was 184 g (0.5 l volume).



Figure 3. Granulated foam-glass (source: compiled by author)

#### 1.5. Biochar

Biochar filling was made from Pine (*Pinus sylvestris* L.) – wood biomass. In the experiment was used dried biochar material, which was formed on 450±5 °C of temperature for 2 hours. Initial biochar granules size was 4.5 cm<sup>3</sup>, according to needs, we granulated to the smaller cubes – 0.6–0.8 cm<sup>3</sup> (Figure 4). The weight of the material was 300 g (0.5 l volume).



Figure 4. Biochar cubes (source: compiled by author)

#### 1.6. Phosphate determination

Laboratory samples were taken after filtration through zeolite, foam-glass and biochar fillers. Samples were placed into the 50 ml marked glass flasks and were added the following reagents by us (Figure 5).

Every flask was filled with filtered wastewater (1–20 ml). After 1 ml of ascorbic acid and 2 ml of ammonium molybdate solution (I) were added. Last step of the mixing was to fill the flask with distilled water up to 50 ml and put for 10–30 minutes at the room temperature (20–25 °C). After passing this time we used to put these mixed samples into the spectrophotometer ( $\lambda = 880$ ) and measure concentration. The obtained result is applied in the formula:

$$\rho_p = \frac{(A - A_0) \cdot V_{max}}{f \cdot V_s},$$

where  $\rho_p$  – phosphate phosphorus concentration; A – the absorbance value of the test portion;  $A_0$  – the absorbance value of the blank sample;  $V_{max}$  – volume, to which disseminated test portion, ml;  $f$  – calibration curve inclination, mg/l;  $V_s$  – the volume of the test portion taken for analysis, ml.



Figure 5. Tested samples (source: compiled by author)

#### 1.7. Ammonium determination

During the process of ammonium determination, wastewater samples are placed in the 50 ml volume flask. Basically, an amount of the wastewater sample was 1 ml. After putting of sample in the marked flasks, we used to add 39 ml of distilled water (to be filled 40 ml).

Then, there were added 4 ml of colour reagent and mixed well. Second reagent is sodium dichloroisocyanurate solution and it was necessary to add 4 ml and mix well again. After all these procedures, we used to put the mixed samples at the room temperature (20–25 °C) for 1 hour and then we used to make the last step of the practical work – measuring the concentration of the sample with spectrophotometer ( $\lambda = 660$ ) and the obtained result is applied in the formula:

$$\rho_a = \frac{(A - A_0)}{f \cdot V_s},$$

where  $\rho_a$  – ammonium concentration;  $A$  – the absorbance value of the test portion;  $A_0$  – the absorbance value of the blank sample;  $f$  – the calibration curve inclination, mg/l;  $V_S$  – the volume of the test portion, taken for analysis, ml.

## 2. Results and discussion

Initial data of investigated wastewater is presented in Table 1.

Table 1. Initial data of wastewater

No.	Litres	NH <sub>4</sub> -N, mg/l	PO <sub>4</sub> -P, mg/l
1	4	50.8	25.8
2	9	78.8	11.1
3	9	76.4	8.8
4	12	61.5	10.8
5	15	100.5	9.7
6	18	76.8	2.69

Results according to amount of filtered wastewater (liters) are shown on graphics. Variation of ammonium nitrogen (Figure 6) and phosphate phosphorus (Figure 7) concentrations in wastewater samples after filtration through foam-glass.

Variation of ammonium nitrogen (Figure 8) and phosphate phosphorus (Figure 9) concentrations in wastewater samples after filtration through zeolite.

Variation of ammonium nitrogen (Figure 10) and phosphate phosphorus (Figure 11) concentrations in wastewater samples after filtration through biochar.

The results show that the foam-glass filler reduced the phosphate phosphorus concentration in filtered 20 liters of sewage. The natural zeolites filling reduced the concentration of ammonium nitrogen in the filtered sewage.

Using foam-glass filler, the efficiency of phosphate phosphorus removal at the beginning of the study was 91%, in the middle of the study – 14%. This filler slightly decreased the concentration of ammonium nitrogen in filtered wastewater (14%). Using zeolites filler, the efficiency of ammonia nitrogen removal at the beginning of

Table 2. Removal efficiency

Results	E %, removing NH <sub>4</sub> -N			E %, removing PO <sub>4</sub> -P		
	I*	II*	III*	I*	II*	III*
foam-glass	14.9	13.9	2.98	90.6	14.4	-15.5
zeolite	100	29	-7.4	44.9	8.2	4.1
biochar	-72	-27	-86	-13.4	-13.4	13.3

Note: I\* – at the beginning of the study; II\* – in the middle of the study; III\* – at the end of the study.

the study was 100%, in the middle of the study – 29%. The biochar filler was not effective in discharging these pollutants from wastewater (Table 2). The efficiency of foam-glass and of zeolite was reduced eliminating the contamination of biogenic materials during the filtration process. The sorbic capacity of these materials was exhausted: after 31 liters of wastewater filtration – foam-glass; after 63 liters of wastewater filtration – zeolite.

By analysing the results obtained, it is possible to compare them with the results obtained for tertiary wastewater treatment using osmotic membrane photobioreactor (OMPBR) and mineral diatomite (Xu et al., 2014). OMPBR operating at HRTs varying from 2 to 4 days exhibited high removal efficiencies of more than 90% NH<sub>4</sub><sup>+</sup>-N, 50% NO<sub>3</sub><sup>-</sup>-N and 85% PO<sub>4</sub><sup>3-</sup>-P through microalgal assimilation and membrane rejection. Results after treatment with OMPBR were 1,1 mg/l NH<sub>4</sub><sup>+</sup>-N, 0,4 mg/l NO<sub>3</sub><sup>-</sup>-N and 0,66 mg/l PO<sub>4</sub><sup>3-</sup>-P (Xu et al., 2014).

The results indicate that OMPBR may be promising application in nutrients removal from wastewater. Raw diatomite has potential for removing COD, BOD<sub>5</sub>, and suspended solid, but poor performance for nutrient constituents (e.g. ammoniacal nitrogen, total nitrogen and total phosphorus). The quality of water from the treatment with modified diatomite can meet the discharge standards and results were – 8 mg/l NH<sub>4</sub><sup>+</sup>-N, 14 mg/l TN, 0,55 mg/l TP. After evaluation of the average efficiency of foam-glass, zeolite, biochar, in all cases of efficiency and considering percentages – treatment with membrane (OMPBR) was more effective than treatment with investigated fillers.

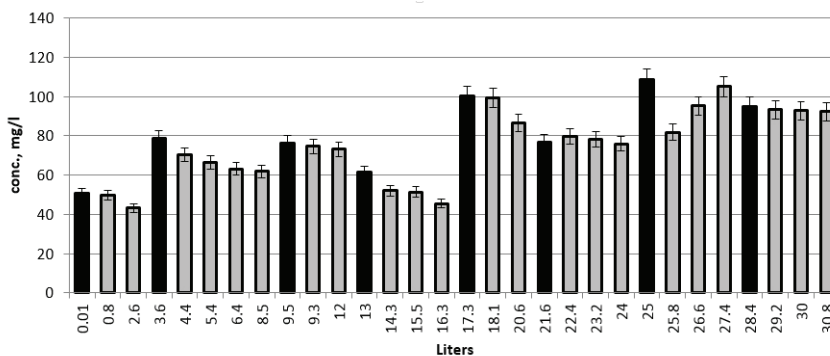


Figure 6. Concentration: ■ – initial; □ – after filtration of NH<sub>4</sub>-N, when foam-glass filler was used

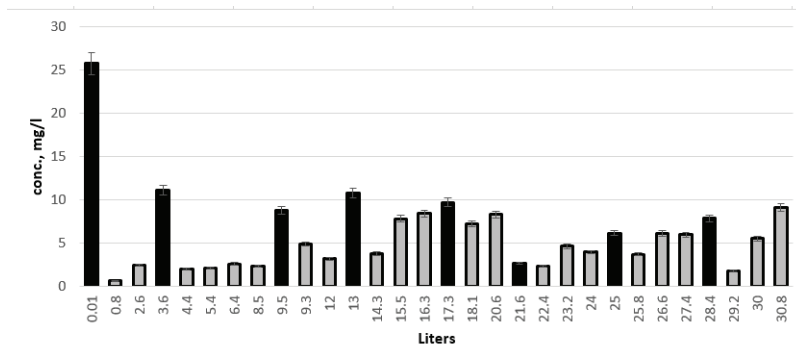


Figure 7. Concentration: ■ – initial; □ – after filtration of PO<sub>4</sub>-P, when foam-glass filler was used

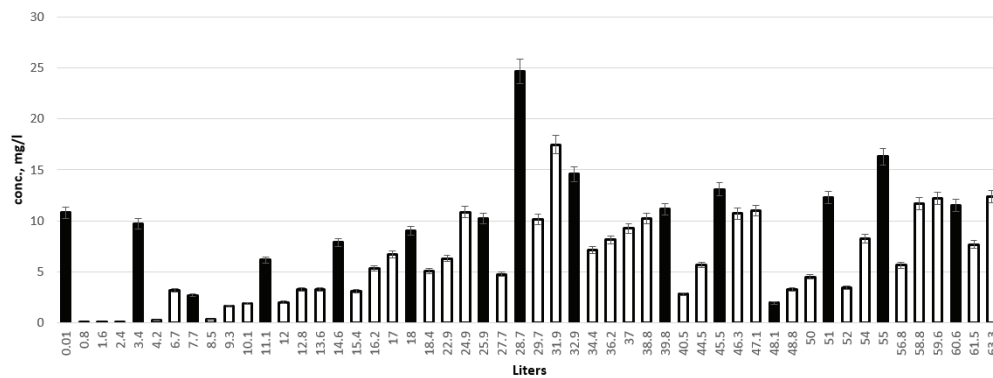


Figure 8. Concentration: ■ – initial; □ – after filtration of NH<sub>4</sub>-N, when zeolite filler was used

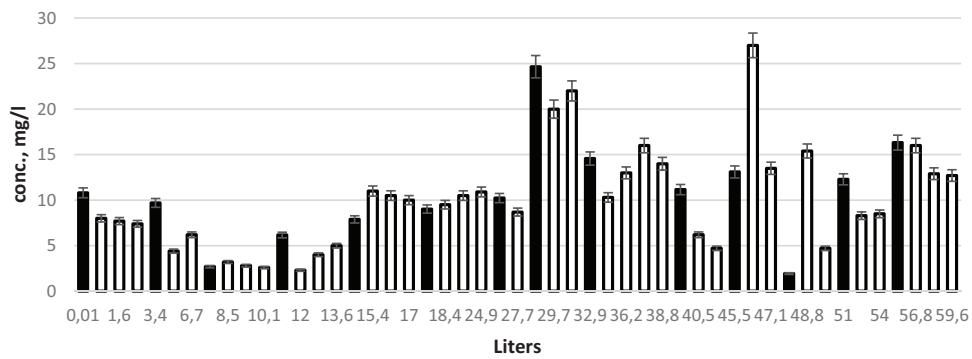


Figure 9. Concentration: ■ – initial; □ – after filtration of PO<sub>4</sub>-P, when zeolite filler was used

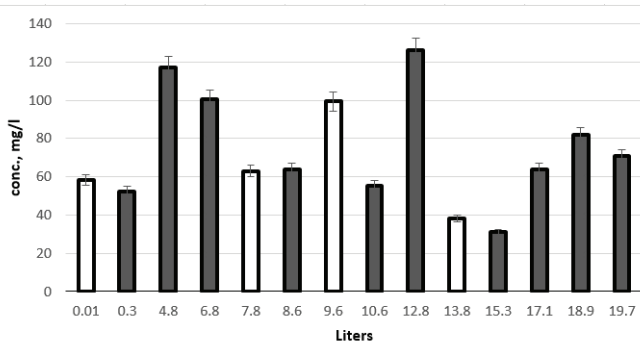


Figure 10. Concentration: □ – initial; ■ – after filtration of NH<sub>4</sub>-N, when biochar filler was used

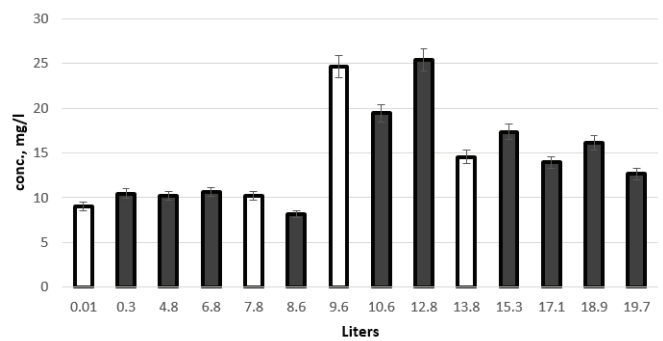


Figure 11. Concentration: □ – initial; ■ – after filtration of PO<sub>4</sub>-P, when biochar filler was used

Over recent years, there has been growing interest in using low-cost natural minerals for treating wastewater. Among these minerals is zeolite. The results indicate that foam-glass and zeolite may be promising application in nutrients removal from wastewater.

## Conclusions

Using foam glass-filler, the efficiency of phosphate phosphorus removal at the beginning of the study was 91%, in the middle of the study – 14%. This filler slightly decreased the concentration of ammonium nitrogen in filtered wastewater (14%).

Using a foam-glass filler (184 g) the concentration of phosphate phosphorus was reduced filtering 31 liters of wastewater.

Using zeolites filler, the efficiency of ammonia nitrogen removal at the beginning of the study was 100%, in the middle of the study – 29%.

Using the zeolites filler (442 g) the concentration of ammonia nitrogen was reduced filtering 63 liters of wastewater.

The results indicate that foam-glass and zeolite may be promising application in nutrients removal from wastewater.

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## TRETINIO NUOTEKŲ VALYMO TYRIMAI

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### Santrauka

Individualaus buitinių nuotekų valymo įrenginiai ne visada dirba efektyviai. Dėl to fiksuojami nitratų azoto, amonio azoto, bendrojo azoto, bendrojo fosforo ir fosfatų fosforo koncentracijų išvalytose nuotekose viršijimai. Darbo tikslas surasti efektyvias medžiagas tretiniam nuotekų valymui ir pasiūlyti šiai problemai spręsti aplinkai draugiškas technologijas, kad išvalytos nuotekos nepakenktų aplinkos kokybei. Buvo tirti trys filtro užpildai (pustiklis, ceolitas, bioanglis). Gauti rezultatai ir išvados, kad pustiklis efektyviau (efektyvumas – 14–91%) šalina iš nuotekų fosfatų fosforą, o ceolitas – amonio azotą (efektyvumas – 29–100 %).

**Reikšminiai žodžiai:** tretinis nuotekų valymas, bendrojo fosforo ir bendrojo azoto valymas, ceolitas, pustiklis, bioanglis.