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The Importance of Media in Wastewater Treatment

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Abstract

The chapter reviews the importance of media in wastewater treatment. The chapter discusses the application of natural fillings (i.e. quartz sand, zeolite and clay) and plastic materials fillings (i.e. PET flakes) for domestic sewage treatment. The effectiveness of removing biogenic compounds (NH_4^+ and PO_4^{3-} ions) and indicator bacteria (*Escherichia coli* and coliform bacteria) by using secondary and tertiary filters have been presented. The effectiveness of one-layer filters and multi-layer filters during the filtration of wastewater pre-treated in a septic tank has been discussed. The possibility of statistical tools (e.g. ANOVA and principal component analysis) to evaluate the filters performance has been described. The phenomena affected the removal of ammonium and phosphates ions from domestic sewage in a vertical flow filter filled with a calcined limestone-silicate rock were also presented.

Keywords: vertical flow filter, ammonium and phosphate ions, indicator bacteria

1. Introduction

Safe water and proper sanitation are crucial for human health and quality of our environment. According to a WHO/UNICEF monitoring program, in 2015 still around 660 million people did not have access to proper quality sources of drinking water. Estimated number of people lacking access to an appropriate sanitary system is 2.4 billion [1, 2].

Leaky septic tanks, introducing pre-treated sewage directly into the ground by drainage or discharging untreated sewage directly into watercourses contaminate groundwater and surface water. Eutrophication of water bodies is a serious problem but their contamination with pathogens is even more dangerous.

There are a number of pathogenic microorganisms transferred through water. They include rotaviruses and polioviruses, pathogenic bacteria or parasitic Protozoa (*Cryptosporidium sp.* oocysts and *Giardia sp.* cysts) [3, 4], as well as an opportunistic pathogen *Pseudomonas aeruginosa* [5–7]. The presence of pathogenic coliform bacteria (*Citrobacter sp.*, *Enterobacter sp.*, *Escherichia coli*, *Klebsiella sp.* and *Proteus sp.*) in aquatic environment indicates fresh contamination with urine and feces.

Escherichia coli is commonly used in the assessment of sanitary condition of water and technological processes. This species is a mesophilous, non-spore forming facultative anaerobe capable of withstanding temperatures between 7 and 45°C and pH 4.7–9.5 [8]. *Escherichia coli* that belongs to the normal flora of the lower intestine in humans and warm-blooded animals may sometimes cause gastroenteritis. Detection of *E. coli* may indicate the presence of other, much more dangerous pathogenic bacteria, such as *Salmonella sp.* (causing typhoid or paratyphoid fever), *Shigella sp.* (causing dysentery) or *Vibrio cholerae* (causing cholera) [9].

It is therefore highly important to use such an on-site wastewater treatment system (OWTS) that is not only effective in removing organic and biogenic compounds but also protects the receiving water against bacterial contamination. Langenbach et al. [10, 11] suggested using a sand filter as a third stage of sewage treatment allowing for removing feces bacteria. A system comprising a settling tank and a sand filter with vertical flow seems to be more cost-effective solution that allows for highly efficient reduction of physical and chemical [12, 13], as well as bacteriological contamination. Using sand as the filter filling may result in elimination level of 1×10^2 – 2×10^4 CFU/100 cm³ for *Escherichia coli* and 5×10^3 – 3×10^5 CFU/100 cm³ for coliform bacteria [14–16]. Other media materials such as clay, zeolite and plastic fillings have also been widely used in wastewater treatment such as moving bed biofilm reactors, trickling filters, rotating biological contactors, etc., which address specific treatment requirements and enhance treatment efficiency.

This chapter discusses the application of natural fillings and plastic materials fillings for domestic sewage treatment. The chapter presents the possibility of using secondary and tertiary filters effective for ammonium and phosphorus ions removal and the pathogen bacteria removal. The effectiveness of one-layer filters and multi-layer filters during the secondary filtration of wastewater pre-treated in a septic tank is also presented.

2. The application of natural materials

2.1. The sand

Effectiveness of the system comprising a septic tank and a sand filter with vertical flow is based on physical and chemical properties of the filter filling. Filtration is a technology commonly used to remove particulate matter and microbial contaminants in the processes of water treatment and sewage purification. It is based on retaining contaminants too big to get through water filled pores of a filter.

Effective operation of a sand filter involves also a formation of a biofilm called *schmutzdecke* on the top layer of the filter filling material [10, 11, 17–21]. This layer is formed at water and sand boundary and is made of biologically active microorganisms and other associated

organic and inorganic substances. The biofilm may grow and reach from a few to a few dozen centimeters inside the bed [10, 11]. Quality of the treated sewage discharged from biosand filters (BSF) depends on sand grain size, filtration rate and intensity of biochemical processes occurring in the filter.

Heterotrophic bacteria that develop in an aerated sand bed are responsible for removing biodegradable organic substances as determined by BOD. As per literature reports, sand bed filtration of pre-treated wastewater allows for a removal of 92% of organic carbon [22] and a reduction of BOD by over 98% [14, 23–25]. An experiment by Wąsik et al. [25] showed that in a system septic tank/vertical filter with no additional aeration, filled with washed sand with equivalent grain diameter $d_{10} = 0.62$ mm, the removal of organic substances from domestic sewage was the most intense when the filter layers were 15 and 30 cm thick. Apart from heterotrophic bacteria, the sand filters in the presence of ammonium nitrogen are also colonized by nitrifying bacteria responsible for oxidation of ammonium nitrogen to nitrate nitrogen. White [22] reported 91% nitrification of sewage treated with sand filtering of secondary clarifier effluent. Chmielowski [14] demonstrated nitrification effectiveness for treatment of septic tank effluent to reach 92%. Chmielowski and Ślizowski claimed [12, 13] that equivalent diameter of sand bed grain exceeding 1.65 mm lowered sewage treatment effectiveness. Additionally, denitrification may occur in non-aerated zones of the filter. According to literature, the effectiveness of nitrate nitrogen removal in sand filters may be as high as 98% [23].

Properly designed vertical flow sand filters in the system with septic tank also provide a significant reduction of pathogenic bacteria count. In a field study on a technical scale, Chmielowski [14] received treated sewage containing 1×10^2 – 1×10^4 CFU of *Escherichia coli* and on average 10 CFU of *Salmonella sp.* and *Shigella sp.* **Table 1** lists the effectiveness of pathogenic bacteria removal from wastewater on sand filters in various studies [26–30].

Langenbach et al. [10, 11] confirmed usability of a vertical flow sand filter in the removal of feces bacteria from secondary clarifier effluents over 59–148 days of the filter operation. They managed to reduced *Escherichia coli* count by ca. 2 log₁₀ units, while medium count of the bacteria in the filtrate was not higher than 1×10^2 CFU/100 cm³. Such a good performance of

| Type of material, grain size | Scale of work, medium | Removal efficiency, log ₁₀ | References |
|---|---|---|----------------------------|
| Desert sand, river sand, beach sand | Effluent from an anoxic denitrifying reactor treating domestic wastewater | 2.4 log units fecal coliforms | Yettefti et al. 2013 [26] |
| Sand, 1.05 mm | WWTP effluent | 95.32–98.02% | Aloo et al. 2014 [27] |
| Sand, 0–8 mm | Raw municipal post-screen wastewater | 2.2–3.5 log units <i>Escherichia coli</i> | Kauppinen et al. 2014 [28] |
| Sand, $d_{10} = 0.25$ mm; $d_{10} = 0.40$ mm; $d_{10} = 0.63$ mm | Lab model SSF, secondary effluent of WWTP | 1.6–2.2 log units <i>Escherichia coli</i> | Pfannes et al. 2015 [29] |
| Sand, $d_{10} = 0.21$ mm | Lab model SSF, secondary effluent of WWTP | 1.1–4.7 log units <i>Escherichia coli</i> | Seeger et al. 2016 [30] |

Table 1. Summary of pathogenic bacteria removal on sand filters used in various studies.

the filter depends on sand surface, determined by grain size distribution and filter height and on the schmutzdecke layer. In the sand filter, bacteria are slowly removed by their adhesion to the biofilm surface that coats the grains of the filler [31]. In an 8 week study with silica sand Accusand, Elliot et al. [17] found that the growth of schmutzdecke layer was the most important factor enhancing *Escherichia coli* removal by up to 5 log₁₀ units from the drinking water mixed with wastewater. Jenkins et al. [32] reported an average removal of 1.8 log₁₀ units, that is, 98.5% of fecal *coli* bacteria from a river water augmented with wastewater over 10 weeks in a filter filled with fine sand. They identified grain size as a major factor affecting the performance of sand filters. Similar conclusions were drawn by Wąsik and Chmielowski [15] who conducted semi-technical studies in biofilter models the operation of which in variable hydraulic conditions continued for 10–11 months. Observing variable hydraulic load of the filter surface that ranged from 16 to 64 mm·d⁻¹, the authors concluded that the degree of indicator bacteria removal was determined mostly by the range of filling grain size and not its percentage share or type. The filter filled with quartz sand with equivalent diameter $d_{10} = 0.32$ mm was found the most suitable for a reduction of bacterial contamination. The treated domestic sewage in a secondary level for OWTS had a very low count of *Escherichia coli* (10²–10³ CFU). Filling the vertical filters with fine sand allowed for exceptional effective removal of the indicator bacteria by 41–4.8 log₁₀ units, that is, 99.993–99.997% [16]. Similar values were given by Seeger et al. [30] who used a sand with equivalent diameter $d_{10} = 0.21$ mm.

The quality of biochemically treated sewage depends on microbial metabolism that slows down together with falling temperature. This is related, for example, to the climatic conditions occurring during the sewage treatment process. Kauppinen et al. [28] conducted a pilot study to evaluate annual efficacy of three different sand filters (SF) for the clarified raw municipal post-screen wastewater treatment operating in cold moderate climate. The SF with grain size of 0.8 mm removed on average 95.6% of BOD₇. An additional biotite layer with grain size 0.2 mm increased this value up to 98.4%. Both filters provided a removal of ca. 30% nitrogen and ca. 78% phosphorus. Kauppinen et al. [28] confirmed that climatic conditions considerably affected the effectiveness of indicator bacteria and viruses removal. Sand filled filters retained on average 99.994% of *Escherichia coli*, and an additional biotite layer boosted this result to 99.999%. In winter, the values were reduced to 99.987 and 99.985%, respectively. Moreover, virus removal was also less effective in this season.

Aronino et al. [33] investigated the removal of viruses and noticed that a filtration of secondary effluents through a sand filter was associated with higher colloidal and organic loads. This caused a formation of a cake layer on the filter surface but did not change the kinetics of virus filtration process. Upper 40 cm of the filter served as a buffer layer, and actual filtration of the sewage occurred in the lower 60 cm layer of the filter. Microscopic studies confirmed that the size of the viruses was the only factor that determined their removal.

2.2. The clay

To improve biofilter performance, the sand filling may be partially or entirely replaced with porous materials. IUPAC (*International Union of Pure and Applied Chemistry*) defines porous materials as solid bodies with pores, cavities, channels or interstices that are deeper than they are wide [34]. Their adsorption capacity is determined by the internal structure of micropore systems, transition pores and macropores. Inexpensive solids with sorption properties strong enough to be

used for sewage and water treatment include chalcedonite [35, 36] or expanded clay. Expanded clay is produced by heating clay loam. The expanded granules are oval and have a characteristic ceramic coating (bisque) on their surface. Inside the granules, there are evenly distributed small closed pores. Porosity of the material, however, depends on the number of open pores created in the external ceramic coating or on the boundary of the bisque and granule body [37].

Adsorption is a main mechanism of retaining bacteria by porous solids with pore diameter exceeding that of the bacterial cells. Adsorption of the bacterial cells to porous solids depends on three types of parameters: physical (carrier porosity, concentration of organic compounds, temperature and medium flow rate), chemical (ionic strength and pH) and microbiological (hydrophobicity and static charge on the surface of the bacterial cells) [38]. As the microorganisms are retained not only on the surface but also inside the pores, the resulting biofilm may further increase the sorption of contaminants.

Masłoń and Tomaszek [39], who investigated non-granulated expanded clay as a biofilm carrier in *Moving Bed Sequencing Batch Biofilm Reactor*, observed unevenly developed biofilm in both open and closed pores of the filling. Expanded clay with a grain size of 4–8 mm facilitated a stable course of nitrification and a removal of up to 99% of ammonium nitrogen. Lekang and Kleppe [40] investigated a trickling filter filled with lightweight expanded clay aggregate (LECA) of three granule diameters: 2–4, 2–7 and 4–10 mm. After 7–8 weeks of LECA bed operation, 100% of ammonium nitrogen was removed irrespective of either granule size or filtration time.

Wąsik and Chmielowski [15] compared filters working with no additional aeration at variable hydraulic conditions filled with sand or non-granulated expanded clay and achieved ammonium nitrogen removal at the level of 52.4 and 68.4%, respectively. Domestic wastewater inflow changed over a few months from 250 to 2000 m³·d⁻¹ and hydraulic retention time (HRT) varied throughout the study from 1.8 to 56 days.

Jucherski and Nastawny [41] demonstrated that the use of expanded clay as a biofilm substrate for nitrification required optimization of the treatment process by reducing the organic matter load. High BOD₅ intensifies growth of heterotrophic bacteria that compete with nitrifying bacteria and affect the removal of ammonium nitrogen.

Wąsik [71] achieved ca. 40% removal of PO₄³⁻ ions on expanded clay and did not show high affinity of phosphorus compounds to Ca, Fe or Al ions that were LECA components.

Treatment of domestic sewage in the expanded clay filled filter [16] allowed for 98.33% reduction of *Escherichia coli* (to the mean level of 2×10^4 – 1×10^6 CFU/100 cm³), and 99.71% reduction of coliforms (to the mean level of 1×10^3 – 1×10^6 CFU/100 cm³). Fine expanded clay with granule size 1.0–2.5 mm facilitated reduction of indicator bacteria by ca. 4 log₁₀ units to an average level of 3.08×10^3 CFU/100 cm³ for *Escherichia coli* and 5.05×10^4 CFU/100 cm³ for coliform bacteria [16].

2.3. The zeolite

Natural zeolites are aluminosilicates with a skeletal structure comprising free spaces filled with ions and water molecules with high freedom of movement. They have net negative structural charge due to an isomorphic substitution of cations in crystal lattice [42, 43]. This negative charge is balanced by such cations as Na⁺, which results in their high cation-exchange

capacity, for example, toward ammonium ions NH_4^+ . Apart from their ion-exchange properties, zeolites have also excellent sorption capacity. Efficiency of contamination removal with zeolites is determined by zeolite chemical composition, granule size, hydraulic load, concentration of the removed ions and pH of the reaction environment [44, 45].

The most common natural zeolite is clinoptilolite with molecular formula $(\text{Na,K,Ca})_{2-3}\text{Al}_3(\text{Al,Si})_2\text{Si}_{13}\text{O}_{36}\cdot 12\text{H}_2\text{O}$ [46]. Due to their considerable ion-exchange and adsorption capacity, clinoptilolites are mainly used to remove NH_4^+ from water [47–51] and sewage [52–58]. Clinoptilolites may replace quartz sand and their selective properties may be successfully used to filter water or sewage. Clinoptilolite filled filters help in cleaning water not only from ammonium nitrogen but also from suspended solids, colloidal particles or bacteria. **Table 2** presents ammonium exchange capacities of clinoptilolites used for wastewater treatment in various studies [58–65].

Effectiveness of ammonium ion removal depends on the type and dose of the zeolite, time of its exposure to sewage, temperature, pH and the presence of other anions and cations in the solution [66]. Kalló [67] showed a removal of ammonium nitrogen from biologically treated wastewater via ion exchange in a column filled with Hungarian clinoptilolite with granule size 0.5–2.0 mm. The author reported that in the column filled with 0.5–1.6 mm granules, the ion exchange was controlled by diffusion as the ion-exchange rate increased for smaller granule sizes.

Wiśniowska et al. [63] evaluated zeolite suitability as a supportive measure of nitrogen removal in the systems based on activated sludge. They concluded that zeolite was an effective sorbent in emergency situations as it prevented disturbances in the removal of biogenic

| Type of zeolite | Medium | Sorption capacity | References |
|---|----------------------|---|--|
| P1 zeolite | Wastewater | 0.1–0.18 meq NH_4^+ /g | Juan et al. 2009 [61] |
| K-F zeolite | | 0.09–0.15 meq NH_4^+ /g | |
| K-Chabazite/K-Phillipsite zeolite (mixture) | | 0.1–0.16 meq NH_4^+ /g | |
| Hungarian clinoptilolite | Wastewater | 3.79 mg NH_4^+ /g | Zabochnicka-Swiatek and Malinska 2010 [59] |
| Zeolite type A-carbon | Greywater | 115.213 mg NH_4^+ /g | Widiastuti et al. 2011 [58] |
| Nanozeolite - palygorskite nanocomposite | Synthetic wastewater | 237.6 mg NH_4^+ /g | Wang et al. 2014 [61] |
| Clinoptilolite ECOLIN | Wastewater | 0.3 mg NH_4^+ /g | Ferronato et al. 2015 [62] |
| Clinoptilolite | Wastewater | 7.80 mg N- NH_4^+ /g | Wisniowska et al. 2015 [63] |
| Synthetic zeolite | Synthetic wastewater | 12.5–44.3 mg NH_4 /dm ³ | Turan 2016 [64] |
| Commercial zeolite 13X | Synthetic wastewater | 131.04–184.8 mg NH_4 /dm ³ | Das et al. 2017 [75] |
| Fly ash zeolite | | 115.36–155.68 mg NH_4 /dm ³ | |

Table 2. Summary of ammonium ions sorption capacities of zeolites used in various studies.

compounds. A comparison of ammonium nitrogen removal with zeolite and bentonite identified zeolite, with absorption level of $7.80 \text{ mg N-NH}_4^+ \cdot \text{g}^{-1}$, as 11% more effective than bentonite. This effectiveness of N-NH_4^+ removal was within the range for natural zeolite reported by other authors, that is, $0.4\text{--}25.5 \text{ mg} \cdot \text{g}^{-1}$ of the sorbent [40, 66, 68]. A study by Wąsik et al. [69] showed higher efficiency of zeolite than sand filters in removing biogenic compounds from domestic sewage. The use of zeolite allowed for effective average elimination of ammonium nitrogen (73.31%) and orthophosphates (62.93%).

Ferronato et al. [62] investigated the capability of granulated clinoptilolite manufactured by ECOLIN in removing pathogenic microorganisms and NH_4^+ from wastewater. In a short-term (24 h) experiment, they evaluated the adsorption rate of clinoptilolite in a laminar flow bed. The initial count of *Escherichia coli* and total coliform was 1.2×10^5 and 1.77×10^5 CFU/100 cm³, respectively, while the concentration of ammonium ions was $13.9 \text{ mg} \cdot \text{dm}^{-3}$. The experiment demonstrated a decrease in the adsorption of NH_4^+ from 0.3 to 0.06 mg/g/l due to the availability of clinoptilolite binding sites for these ions. High degree of ammonium ion adsorption in clinoptilolite bed was in line with the data reported by other authors [70]. A reduced count of pathogenic microorganisms was also observed, by 90.4–95.2% for *Escherichia coli* and 89.9–94.8% for total coliforms.

According to the literature, the processes of filtration and adsorption control immobilization of pathogenic bacteria contained in the sewage flowing through a porous substrate [11, 38]. The first mechanism is highly controlled by the size of the filter filling. Stevik [38] reported that the effectiveness of bacteria retention due to filtration was inversely proportional to the grain size of a filtration material. Adsorption is the main mechanism of bacteria retention in porous media with pore diameter larger than the bacteria. As the microorganisms are retained not only on the surface but also inside the pores, the resulting biofilm may serve as an additional sorbent and increase adhesion of the bacterial contaminants. Natural zeolites are capable of entrapping microorganisms thanks to micropores [71], Van der Waals forces, hydrogen bonding or ion bridging [71–73]. Additionally, selective, positively charged materials may attract Gram-negative bacteria such as *Escherichia coli* [74, 75]. However, it should be taken into account that soluble organic compounds contained in the sewage may block the substrate surface and consequently the charges that attract *Escherichia coli* [76].

Wąsik and Chmielowski [16] reported an increased count of pathogenic bacteria in treated wastewater together with increasing size of zeolite granules. Enlarging the zeolite equivalent diameter d_{10} from 1.0 to 3.6 mm resulted in rising the count of *Escherichia coli* and coliform bacteria, respectively, from 5.75×10^2 to 8.67×10^3 CFU/100 cm³ and from 1.85×10^4 to 3.47×10^4 CFU/100 cm³. The highest removal rate of pathogenic bacteria at the level of 99.995% was observed for zeolite with granule size 1.0–2.5 mm.

2.4. Multi-layer filters

New solutions based on biological beds filled with porous or modified materials often increase the efficiency of wastewater treatment but they may be inadequate in terms of their microbiological quality. Therefore, supplementation of the porous materials with a layer of quartz sand seems a simple solution to this problem. Quartz sand is inexpensive and provides an effective barrier for the pathogenic bacteria. A study by Kanawade [77] focused on using a multi-layer filter to

remove ammonium and suspended solids from effluents of a domestic wastewater plant. The filter was filled with sand of grain size 0.5–1.0 mm that filtered out suspended solids and the top layer was made of clinoptilolite that removed ammonium nitrogen. Turkish clinoptilolite with adsorption capability of $10.4 \text{ mg} \cdot \text{g}^{-1}$ was used. As a result, 100% of ammonium nitrogen and 75% of suspended solids were removed by the multi-layer filter over 38 hours of its operation.

Kalenik [78] investigated treatment of model wastewater in a sandy soil bed with a layer of clinoptilolite. He showed that phosphorus removal efficiency was 53.1% in a 0.10 m thick layer and as high as 89.2% when the bed was 0.20 m thick. The use of medium sand alone (without additional layer of clinoptilolite) allowed for a removal of 23% of total phosphorus.

Syafalni et al. [79] filtrated dyed wastewater on granular activated carbon (GAC) and zeolite with particle size range of 1.18–2.00 mm. A filter comprising GAC as a top layer and zeolite as a bottom layer removed 59.46% COD, 60.82% of ammonia and 58.4% of the dye.

Wąsik and Chmielowski [80, 81] investigated a multi-layer filter filled with sand and granulated activated carbon exposed to variable hydraulic load (from 43 to $88 \text{ mm} \cdot \text{d}^{-1}$). They noticed huge variations in the efficiency of reduction of BOD_5 (6–99%), COD_{Cr} (31–90%) and total suspended solids (55–95%) due to variable conditions prevailing in individual layers of the filter. They concluded that a monolayer filter filled with granulated activated carbon was the most suitable for treatment of domestic sewage over a 3-month study cycle. Average efficiency of BOD_5 , COD_{Cr} and suspended solids elimination was very high irrespective of rising hydraulic load and reached, respectively, 98, 97 and 87%. This was consistent with the reports of other authors on biologically active carbon filters [82, 83]. Mean efficiency of bacterial elimination in a two-layer filter comprising 75% of fine sand ($d_{10} = 0.32 \text{ mm}$) in its bottom layer and 25% of fine zeolite ($d_{10} = 1.8 \text{ mm}$) in the top layer was 97% for BOD_5 , 92% for COD_{Cr} , 99.993% for *Escherichia coli* and 99.953% for other coliform bacteria.

3. Plastic materials

Natural materials commonly used as a filling for biological systems may be replaced with a plastic filling. Compared with conventional media (quartz sand, gravel, clay and rock) plastic fillings have high specific surface area and lower tendency to clogging. Modern biological filters have a large specific area of up to $150\text{--}200 \text{ m}^2/\text{m}^3$ (filter media in trickling filters), which provides more space for growth of heterotrophic and nitrifying bacteria [84]. Reportedly, the plastic media in a Moving Bed Biofilm Reactor present up to $1200 \text{ m}^2/\text{m}^3$ specific area [85].

Plastic filter media are light and can be constructed to greater depths, thus increasing the hydraulic load capacity and improving mass transfer. Plastic fillings are characterized by the highest abrasion resistance and better gas transfer due to the greater draft [86, 87]. Filters with natural filling, such as rock or sand are often poorly aerated as they contain less empty/hollow fractions [66]. Currently used plastic fillings of biological systems are produced as random or modular packing media.

Galbraith et al. [88] discussed high costs of obtaining molten mineral material, that is, sand of grain size suitable for a construction of filters meeting legal requirements (VDH 2011). The

authors claimed materials such as organic fiber, synthetic foam or textile to be more economically advantageous. Systems based on non-mineral materials may be smaller than filters filled with mineral materials. Several systems based on the use of artificial materials sold by commercial vendors have been approved for use in Virginia. Non-mineral media systems can be prefabricated, transported and assembled locally from modules, while mineral-filled filters are typically built on-site.

Harwanto et al. [89] evaluated the use of a polystyrene microbead filter (PF) and Kaldnes filter (KF) in trickle filters. They determined mean efficiency of ammonium ion removal to be $35.0\text{--}310.5\text{ g}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ for PF and $32.1\text{--}288.1\text{ g}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ for KF. Nijhof [90], who investigated the efficiency of a leaching system filled with Filterpack CR50 Mass Transfer filling with specific area of $200\text{ m}^2\cdot\text{m}^{-3}$, established the nitrification index as ranging from 0.1 to $0.8\text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$. Moulick et al. [91] used nylon pot scrubber media as a filling in trickling filters. They reported 28–68% efficiency of ammonia removal and nitrification indices within the range $0.11\text{--}1.29\text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$.

Kishimoto et al. [92] researched nitrification efficiency in restaurant wastewater treated in trickling filters filled with plastic media of the same material, the same shape but different roughness. One media type had a smooth surface (KT-15, Dainippon Plastics, Japan) and the other a rough surface (LT-15, Dainippon Plastics, Japan). They found that the removal of organic compounds (defined as COD) and nitrification were more effective in rough surface media filling (LT-15) than in smooth surface media filling (KT-15). Better performance of LT-15 filling was concluded to be due to twice larger biomass of microorganisms attached to this media.

Stephenson et al. [93] examined eight different plastic media (acrylonitrile butadiene styrene, nylon, polycarbonate, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), polyvinyl chloride and tufnol) in a reactor receiving settled domestic wastewater. They found that nitrification rates did not correlate with biomass concentration or surface roughness of the media. The use of PTFE, that is, a material with the lowest surface adhesion force, allowed for development of a biofilm with the highest nitrification rate of $1.5\text{ g}\cdot\text{m}^{-1}\cdot\text{d}^{-1}$.

Wąsik and Chmielowski [15] determined the effects of ammonia and indicator bacteria removal during the treatment of domestic sewage on a vertical flow filter filled with plastic material (PET flakes). The experiments were performed in previously developed models that continuously operated for a few months at variable hydraulic conditions ($250\text{--}2000\text{ cm}^3\cdot\text{d}^{-1}$) and hydraulic retention time (HRT) from 1.8 to 56 days. PET flakes provided favorable conditions for nitrifying bacteria, as mean ammonium nitrogen removal rate for this material was 66.74%. The filters with plastic filling reduced the count of *Escherichia coli* by 98.08% and of coliform bacteria by 98.41%.

4. The calcined limestone-silicate rock

The calcined limestone-silicate rock is formed in a thermal processing as a result of calcium carbonate decomposition to calcium oxide and carbon dioxide. The process is associated with an increase in sorption capacity of limestone-silicate rock (so called gaize) from 19.6 to $119.6\text{ g P}\cdot\text{kg}^{-1}$ for the material burnt at 1000°C [94]. The presence of calcium ions and high pH make the calcined

rock suitable for the removal of phosphorus compounds. Alkaline environment (pH ca. 8) facilitates binding of orthophosphate ions by calcium ions and formation of hydroxyapatite crystals. Renman [95] confirmed the presence of amorphous tricalcium phosphate in an exhausted filter filling commercially known as Polonite®. She also demonstrated that 82% of the exhausted filling was calcium and phosphorus compounds in the form of hydroxyapatite.

Most studies investigating the use of calcined rock focused on the removal of phosphates [96, 97]. **Table 3** presents the use of limestone-silicate rock or its calcined form in the removal of phosphate ions as reported by various researchers [98–101]. In their study on the use of calcined rock (Polonite®) in the removal of phosphates from wastewater, Renman et al. [98, 102] noticed also about 18% removal of inorganic forms of nitrogen, which they considered to be losses associated with their evaporation.

An experiment of Wąsik et al. [103] investigating the use of calcined rock as a filling of a vertical flow filter operating under variable hydraulic retention time (HRT) identified chemical processes, and not biofiltration, as the basic cause of PO_4^{3-} and NH_4^+ removal. A few months long filtration of pre-treated domestic sewage through calcined limestone-silicate rock revealed a very high (95%) positive correlation between the removal of phosphates and ammonium

| Type of material, grain size | Wastewater treatment system; phosphates concentration in a influent | Sorption capacity, removal efficiency | References |
|--|---|--|--------------------------------|
| Polonite (2–5.6 mm) mixed with 8% peat | Septic tank + biofilter; 1.9–4.9 mg P- PO_4^{3-} /l | 89% PO_4^{3-} | Renmann 2008 [95] |
| Calcined opoka (Polonite), 2–6 mm | Septic tank (domestic wastewater) | >90% PO_4^{3-} | Cucarella et al. 2009 [98] |
| Polonite®, 2–5.6 mm | Septic tank (domestic wastewater) + column with Polonite; 1) 4.68 ± 1.88 mg PO_4^{3-} /l and 2) 5.19 ± 1.86 mg PO_4^{3-} /l | (1) $91 \pm 11\%$ PO_4^{3-} (2) $87 \pm 19\%$ PO_4^{3-} | Renmam A, Renmam G. 2010 [96] |
| Calcined Opoka, 10–50 mm | Wetland + opoka; 4–9.1 mgP/l | 0.727–1.258 g/kg, 18.2–35.7% P | Jóźwiakowski 2012 [99] |
| Polonite® (Bioptech, Hallstavik), 2–5.6 mm | Septic tank (domestic wastewater) + column with Polonite (1) mean BZT ₇ 120 mg/l, total phosphorus 8.0 ± 1.7 mg/l; (2) + SBR; mean BZT ₇ 20 mg/l, total phosphorus 5.3 ± 2.6 mg/l | Mean: (1) 47–97% TP (2) 76–97% TP | Nilsson et al. 2013 [97] |
| Calcined carbonate-silica rock (opoka) (1) 1–2 mm; (2) 2–5 mm; (3) 5–10 mm | Vertical flow filter; domestic wastewater | (1) 0.38 g TP/kg; (2) 0.30 g TP/kg (3) 0.28 g TP/kg | Jóźwiakowski et al. 2017 [100] |
| Polonite® (Ecofiltration NORDIC), 2–6 mm LECA, 4–10 mm | Mechanically treated wastewater, after septic tank in on-site wastewater treatment system | 40.9 mg P- PO_4^{3-} /g 5.1 mg P- PO_4^{3-} /g | Karczmarczyk et al. 2017 [101] |

Table 3. Summary of phosphorus ions sorption capacities for carbonate-silica rocks used in various studies.

nitrogen. The study revealed also a considerable (73–74%) dependency between the reduction of coliform count and biogenic compounds. Authors claimed that alkalization of the environment and chemical reactions involving NH_4^+ and PO_4^{3-} in the presence of magnesium and calcium ions as well as carbonates and coliform bacteria created suitable conditions to the formation of struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) and apatite $\text{Ca}_5(\text{PO}_4)_3 \cdot \text{OH}$ and $\text{Ca}_5(\text{PO}_4)_3 \cdot \text{CO}_3$ crystals. The formation of struvite crystals by microorganisms in the presence of ammonia ions and magnesium phosphate was first described by Robinson [104]. Struvite is spontaneously precipitated during domestic sewage treatment [105] in the presence of high concentration of soluble phosphorus, ammonium and magnesium, and low concentration of total suspension solids and alkaline pH. If the formation and accumulation of struvite was controlled, it could have a market potential as a slow release fertilizer [106, 107].

Wąsik et al. [103] showed that microscopic examination of sediment samples taken from the surface and interior of the filter confirmed the formation of magnesium ammonium phosphate (struvite) and apatite crystals. The crystallization process was carried out both on the surface and inside of the bacterial cells and total elimination of the coliform bacteria confirmed their role as nuclei of crystallization. Microscopic research confirmed tricalcium phosphates were more abundant than struvite.

5. The importance of statistical tools

Statistical tools helps us analyze the data generated in lab scale systems and full scale installations, and are able to identify the critical factors that govern the process treatment efficiencies, and provide engineering design guidance with confidence.

Variance analysis ANOVA performed by Wąsik [69] identified selectivity, porosity and grain size of the filling as the factors responsible for effective removal of ammonium nitrogen, orthophosphates and *Escherichia coli* and coliform bacteria from domestic sewage treated in a septic tank and a vertical flow filter. Natural selective and porous materials were found to be the most effective in the removal of biogenic compounds. The filling of grain size from 1.0 to 2.5 mm provided highly efficient removal of ammonium (75.34%) and orthophosphate (>79%) ions. The filter filled with natural porous material of fine grain size was the most suitable for removing pathogenic bacteria, and allowed for elimination of 99.98% of *Escherichia coli* and 99.94% of coliform bacteria.

Wąsik and Chmielowski [15, 16] used the principal component analysis (PCA) to determine the mechanisms of pathogenic bacteria removal. PCA showed that in the case of natural materials the effectiveness of *Escherichia coli* elimination depends mainly on the filling grain size and not the filling type.

6. Summary

Treatment of domestic sewage in the areas with scattered development remains a serious issue. Discharging ineffectively treated sewage into the environment may cause an increase in the count of pathogenic organisms. In developing countries, where water and sanitation

infrastructure is still inadequate, billions of people are exposed to diseases resulting from using unsafe water. The World Health Organization (WHO) estimates that around 1.7 million deaths globally are related to inadequate Water, Sanitation and Hygiene (WaSH) [2, 7]. It is therefore crucial to prevent the spread of gastric and infectious diseases via water. Humans are exposed not only as a result of consumption of contaminated water but also via skin contact during various forms of recreation (e.g. swimming and diving).

While one of popular approaches to on-site wastewater treatment system (OWTS) are activated sludge processes such as sequential batch reactors (SBR) due to their cost effectiveness, an inexpensive and simple solution for the treatment of domestic sewage especially in rural areas is septic tank followed by vertical flow filters with different natural media fillings, that is, quartz sand, zeolite and clay. This process route seems to be an economically attractive alternative to sewage treatment for residents without access to a public sewerage system.

Natural materials like zeolite offer the highest mean rate of ammonium nitrogen removal compared with sand or clay media. Sand filters are the most effective in reduction of the indicator bacteria but they have the highest tendency for clogging. It is recommended to undertake research in the field of developing an idea that would protect the sand filter against clogging. Natural materials commonly used as a filling of vertical flow filters may be partly replaced with a plastic filling like PET flakes. Compared with conventional media plastic fillings have high specific surface area and lower tendency to clogging. It is proposed to conduct further research using different materials used alternatively in the multi-layer vertical filters (i.e. sand and plastic media) [108].

Filter media cost is one of the most important factor that affects the cost of vertical flow filters. In areas where the sand are commonly found, its cost is nominal. Alternative filling of filters like clay are moderately cheap. Zeolite is the most expensive filling. Cost of clay, zeolite, calcium-silicate rock is all subject to availability and cost of transport. PET flakes are obtained in the recycling process of commercially available bottles. Detailed cost and long-term costs of plastic media is not available because the PET flakes media is still under study.

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