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BIOLOGICAL STABILITY OF WATER IN WATER DISTRIBUTION SYSTEMS. THE EFFECT OF WATER TREATMENT TRIALS

Research into the removal efficiency of biogenic substances, necessary for the growth of heterotrophic microorganisms, was carried out for the following water treatment trials: surface water, infiltrative water and mixed surface water after microsieving with aerated underground water. The results have shown that the efficiency of removal of organic substances, independently of the type of treated water, increased along with the organic carbon concentrations in raw water. The average effectiveness of phosphate removal was: 92.1%, 88.8% and 83.7% for surface, infiltrative and mixed water, respectively. In all analyzed systems, presence of phosphates was an limiting factor for the regrowth of microorganisms in the distribution system. In none of the water treatment trials, effective removal of inorganic nitrogen has been recorded.

1. INTRODUCTION

It is widely acknowledged that the main cause of regrowth of microorganisms in distribution systems (DS) is the lack of water biostability in the water network [1]. This issue is common in many water distribution systems which are utilised globally, and often leads to human health hazards due to the presence of pathogenic microorganisms in tap water [2]. Therefore, the greatest human health risk stems from water recontamination by heterotrophic, including potentially pathogenic microorganisms. Concentrations of organic substances and removal efficiency in water treatment trials [3] are considered the main parameters determining potential for regrowth of microorganisms. Much of the conducted scientific research provides an underpinning for the necessity of effective removal of all organic substance fractions in treatment systems [4], thus limiting the regrowth of microorganisms. Despite the biodegradation of low molecular weight compounds – defined as biodegradable dis-

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solved organic carbon (BDOC) [5] – by microorganisms during metabolic processes high molecular weight organic substances can become precursors of nutrients. This is due to the capacity for the formation of BDOC from larger organic molecules during the oxidation process, including disinfection [6]. Therefore, processes applied during water treatment should guarantee a maximum removal efficiency of organic carbon and all its fractions. Conventional water treatment (coagulation + sedimentation + sand filtration + disinfection) guarantee a high effectiveness of removal of high molecular weight organic compounds, mostly of hydrophobic compounds [7]. Coagulation and adsorption processes are usually referred to as the most effective at removing total organic carbon (TOC) and dissolved organic carbon (DOC), whereas the chemical oxidation process (especially ozonation) has been indicated as mainly guaranteeing the transformation of high molecular weight into lower mass particles [8], while marginally mineralising organic compounds. However, removal of BDOC requires the use of biological processes, mainly biofiltration through granular activated carbon filters (GAC) [9]. Increased efficiency of this process can be provided by preceding it with an ozonation process [10]. However, the effectiveness of eliminating organic substances from water is not always sufficient to limit regrowth of microorganisms.

This is why factors such as low concentration of inorganic nutrients, i.e. nitrogen or phosphorus compounds, can also limit regrowth of microorganisms. Phosphate ions are effectively removed in conventional water treatment trials through the elimination of insoluble phosphates, e.g. with the use of coagulation. Therefore, it has been widely recognized that the concentration of phosphates is the factor limiting the growth of microorganisms [11].

Unfortunately, conventional water treatment trials do not guarantee effective removal of inorganic nitrogen present in water. Wastewater treatment technologies where biogenic substances are present in high concentrations indicate that effective methods of nitrogen removal are denitrification, ion exchange, membrane separation and electrochemical processes [12]. However, these processes are not widely used in water treatment plants. There is also insufficient available information regarding the effectiveness of removal of inorganic nitrogen compounds during treatment processes.

Disinfection is used in the last step of water treatment as a final method of protecting water from regrowth of microorganisms. However, as research has proven, this is not a sufficient condition [13, 14] for the prevention of regrowth of microorganisms.

The main goals of this study were to determine the level of biostability of treated water and the influence of unit treatment processes on the efficiency of nutrient removal.

2. EXPERIMENTAL

Research into the variations of biological stability levels were conducted in technical scale on three water treatment trials, i.e.: surface (WTT1), infiltrative (the surface water after filtration through the ground) (WTT2) and mixed surface water after microsieving with underground aerated water (WTT3). Surface water was treated through: volume coagulation, sedimentation, sand filtration, ozonation, filtration through biologically active GAC deposits, and disinfection with a mix of chlorine and chlorine dioxide. A pre-hydrolized poly-aluminum chloride was used as a coagulant, while sodium hydroxide was introduced into the water prior to disinfection in order to correct the pH level. Infiltrative water underwent the following processes: aeration, sand filtration, ozonation, adsorption through the use of biologically active GAC deposits, and disinfection with a mix of chlorine and chlorine dioxide. This trials also involved correction of pH with the use of sodium hydroxide. Meanwhile, mixed water was treated through the processes of contact coagulation, indirect chlorination, two--step filtration, successively through sand and dolomites, and disinfection with chlorine dioxide. Corrections of pH was conducted during the second filtration step. Iron sulfate was used as the active coagulant. All three trials were exploited with variable output levels, which, during conduction of research, fell within the ranges of: 1750–3000 m^3/h (for WTT1), 2487–3725 m^3/h (for WTT2) and 782–1215 m^3/h (for WTT3). This caused variable technical parameters during the implementation of unit processes. All discussed water treatment trials took into account variables, derived from the water quality of processed water and the doses of used reagents. Research in the selected trials was conducted for a period of 2 years. The subject of research were raw water samples, samples taken from each unit water treatment process, and of treated water, collected regularly once a month.

The water biostability level was determined based on the content of organic and inorganic nutrient substrate necessary for the growth of heterotrophic microorganisms, i.e.: biodegradable dissolved organic carbon, phosphates and inorganic nitrogen (N_{inorg} – NH_4^+, NO_2^-, NO_3^-). Based on available literature [15–17] and results of experiments, the threshold values limiting of regrowth of microorganisms were evaluated: BDOC – 0.25 g C/m^3, PO_4^{3-} – 0.1 g P/m^3, N_{inorg} – 0.2 g N/m^3. The assumption was made that biologically stable water has concentration levels of each of the nutrients equal or below the threshold value. Additionally, when assessing the stability level of water introduced into distribution systems, the disinfectant concentration levels was taken into account.

Apart from nutrient content, the concentration of total and dissolved organic carbon and pH values in water samples were determined, while the concentration of remaining disinfectants was also recorded in water samples after disinfection.

Concentrations of inorganic nutrients were determined by the colorimetric methods with the use of a Shimadzu spectrophotometer. TOC and DOC were determined with the use of a TOC Hach Lange analyzer. BDOC was determined by the standard methods.

The concentration of non-biodegradable organic carbon (NBDOC) was calculated as the difference between concentrations of DOC and BDOC.

3. RESULTS AND DISCUSSION

Raw water, regardless of the type, was characterized by high concentration of organic substances, including nutrients (Table 1). During the whole research period, surface and mixed water contained much more organic substances than infiltrative water.

Table 1

Ranges of parameters of water quality in raw and treated waters

Parameter	Raw water			Treated water		
	WTT1	WTT2	WTT3	WTT1	WTT2	WTT3
pH	7.1–8.0	6.6–7.2	7.0–7.9	7.2–8.1	7.2–7.9	7.4–7.9
TOC, g C/m^3	2.61–11.11	1.44–5.16	4.50–9.71	0.70–3.94	0.75–3.30	1.84–3.95
DOC, g C/m^3	2.12–9.55	1.21–4.83	3.27–9.54	0.65–3.45	0.38–3.30	1.43–3.90
BDOC, g C/m^3	0.19–0.88	0.11–0.39	0.18–1.23	0.10–0.34	0.05–0.24	0.11–0.88
NBDOC, g C/m^3	1.80–8.96	1.02–4.47	2.92–9.01	0.44–3.34	0.21–3.06	1.25–3.03
NH_4, g N/m^3	0.02–0.40	0.11–0.31	0.07–0.47	0.01–0.16	0.01–0.11	0.01–0.05
NO_3^-, g N/m^3	0.95–4.70	0.26–0.95	0.26–3.79	0.45–3.68	0.16–0.68	0.15–2.91
NO_2^-, g N/m^3	0.00–0.02	0.00–0.01	0.00–0.01	0.00	0.00	0.00
N_{inorg}, g N/m^3	1.10–4.78	0.45–1.24	0.33–4.06	0.51–3.83	0.21–0.70	0.15–2.96
PO_4^{3-}, g PO_4^{3-}/m^3	0.04–0.23	0.05–0.37	0.05–0.31	0.00–0.03	0.01–0.06	0.00–0.03
Cl_{2F}[a], g Cl_2/m^3	–	–	–	0.21–0.97	0.06–0.61	0.02–0.70
ClO_2, g ClO_2/m^3	–	–	–	0.00–0.23	0.00–0.18	0.17–0.44

[a]Free chlorine.

The dissolved fraction was predominant amongst organic substances in all three types of water, constituting 54.2–96.0%, 62.8–99.2% and 72.7–98.6% for WTT1, WTT2 and WTT3, respectively. Meanwhile, nutrients (BDOC) amounted to 4.4–19.3%, 3.4–15.5% and 3.0–24.0% of DOC for WTT1, WTT2 and WTT3, respectively, and in general their contents were similar to those found in natural waters [4, 18]. BDOC concentration in one sample of raw water from WTT3 was lower than the threshold value for the biological stability. The percentage of such samples in infiltrative waters was

37.5% and in surface waters it was 20.8%. This results from the lower organic contamination levels in infiltration water (Table 1). High content of dissolved organic carbon, especially NBDOC in TOC prove the predominance of hydrophobic substances in all types of raw water [19]. Therefore, processes used in the discussed trial systems guaranteed an effective elimination of TOC. In all three treatment trials this concentration was reduced below the minimum allowed in drinking water in Poland (5 g C/m^3). The removal efficiency of all TOC fractions increased with higher concentration of organic substances in raw water. This dependence confirms a linear correlation with the confidence level α of 0.05 (Table 2).

Table 2

Correlations between removal efficiencies of organic fractions and their concentrations in raw water

Fraction	WTT1	WTT2	WTT3
ΔTOC	0.648TOC – 0.342	0.372TOC + 0.174	0.738TOC – 1.185
ΔDOC	0.657DOC – 0.386	0.235DOC + 0.420	0.670DOC – 0.791
ΔNBDOC	0.676NBDOC – 0.450	0.242NBDOC + 0.374	0.709NBDOC – 0.842

Further unit processes, in all of the three trials, guaranteed reduction of TOC, DOC and NBDOC. The coagulation proved to be the most effective in removal of TOC and its high molecular weight fractions, guaranteeing an average reduction of organic substances of 58.1% and 55.3% for surface and mixed water, respectively. The oxidation process used in each of the trials caused an increase in the BDOC concentration, while simultaneously causing a very small decrease in the TOC concentration. Low molecular weight organic substances, which are formed during oxidation (Fig. 1), were effectively removed in the biofiltration process, in which reduction of DOC and NBDOC concentrations were also recorded.

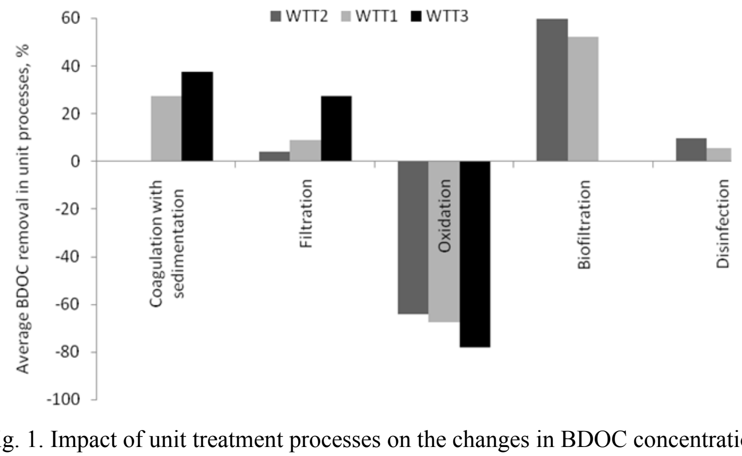

Fig. 1. Impact of unit treatment processes on the changes in BDOC concentration

The BDOC removal efficiency in analyzed trials was however insufficient to guarantee biological stability in all water samples. This could be due to the low content of phosphate ions in waters flowing into biofiltration. This hypothesis is corroborated with research conducted by Yu et al. [20] which identified a dependence between the biological activity of microorganisms on deposit surfaces and the phosphates content in flowing water.

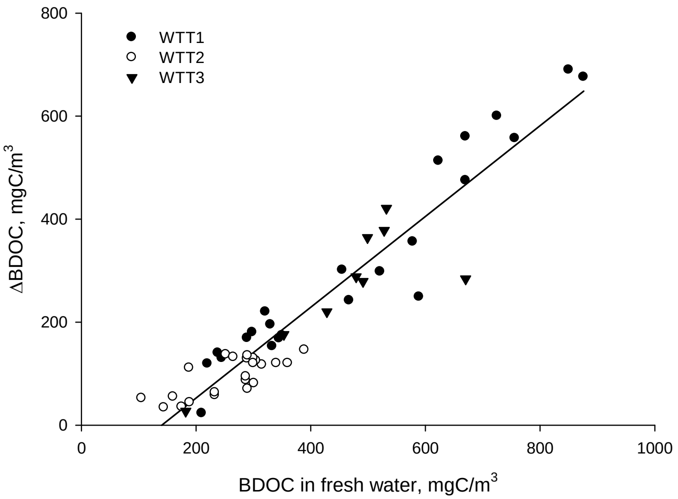

Fig. 2. Dependence of BDOC reduction on BDOC concentration in raw water

The effectiveness of BDOC elimination in the discussed water treatment trials was also directly proportional to their concentration in raw water (Fig. 2) independently of the water type. A consequence of the above relation was the lowest efficiency of BDOC removal from infiltrative water. The final average effectiveness of BDOC removal from water was measured to be 58.0% (WTT1), 33.8% (WTT2) and 44.4% (WTT3). The increase in BDOC concentrations during oxidation, based on one sample from each WTT type, was significant enough for organic nutrient content in water flowing into the mains system to be higher than in raw water.

Despite the lowest efficiency of BDOC removal only WTT2 guaranteed sufficient BDOC elimination to ensure biological stability in all water samples introduced into the distribution system. The presence of biofiltration processes in the surface and infiltrative water treatment system resulted in a higher percentage of samples with BDOC concentration below 0.25 g/m^3 (Table 3) than in mixed water. The most effective method of removing BDOC in mixed water treatment trial (WTT3) proved to be the coagulation process, while the lack of biofiltration in this technological system resulted in over half of the samples containing BDOC concentration above the threshold value (Table 3).

Table 3

Percentage of biologically stable samples with regards to specific nutrient concentration [%]

Sample	Item	Raw	After coagulation	After filtration	After oxidation	After biofiltration	Treated
WTT1	PO_4^{3-}	0.0	25.0	62.5	62.5	100.0	100.0
	N_{inorg}	0.0	0.0	0.0	0.0	0.0	0.0
	BDOC	20.8	50.0	50.0	0.0	91.6	95.8
WTT2	PO_4^{3-}	0.0	–	57.5	57.5	100.0	100.0
	N_{inorg}	0.0	0.0	0.0	0.0	0.0	0.0
	BDOC	37.5	–	62.5	0.0	83.3	100.0
WTT3	PO_4^{3-}	0.0	46.2	100.0	46.2[a]	–	100.0
	N_{inorg}	0.0	7.7	7.7	7.7	7.7	7.7
	BDOC	7.7	38.5	0.0	0.0	–	46.2

[a]For WTT3, the oxidation process occurred before filtration.

Regardless of the type of water treatment trial, only phosphate ions were effectively removed, the removal effectiveness of which was directly proportional to the PO_4^{3-} concentration in raw water. The high efficiency in phosphate ion removal from water is connected with their precipitation into weakly soluble phosphates which are removed during sedimentation and filtration (Fig. 3). Such high efficiency of the above processes guaranteed a significant percentage increase in biologically stable samples with regards to phosphate concentration even after the first treatment steps (Table 3).

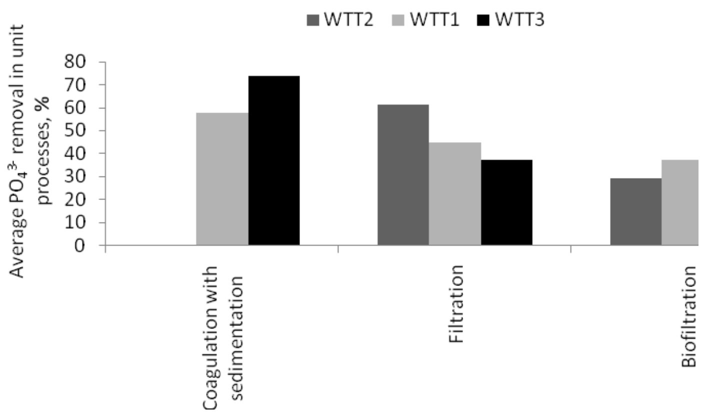

Fig. 3. Efficiency of unit processes in phosphate removal

Additionally, phosphate ions in WTT1 and WTT2 were assimilated by microorganisms inhabiting the surface of GAC deposits [8, 19]. The effectiveness of removal of phosphate ions averaged at 83.7% (WTT1), 88.8% (WTT2) and 92.1% (WTT3).

The highest efficiency of phosphate removal in WTT3 should be explained by the effective sedimentation of iron phosphate precipitated during coagulation with iron sulphate as a coagulant. As a result, in all of the discussed water treatment trials, the phosphate ion concentration was a limiting for regrowth of microorganisms in DS.

Unfortunately, the effectiveness of removal of inorganic nitrogen compounds was much lower than that of organic substances or phosphate ions, averaging 44.2% (WTT1), 46.2% (WTT2) and 42.6% (WTT3). Nitrate ions were predominant amongst inorganic nitrogen compounds found in all raw water samples, measured at 73.9–99.2%, 51.7–89.6%, and 54.6–94.1% N_{inorg} for WTT1, WTT2 and WTT3, respectively. The concentration of inorganic nitrogen was above the threshold value for biological stability in all raw water samples (Table 1). The elimination effectiveness of NH_4^+ in all water treatment trials was similar, and the maximum values recorded were 95.5% (WTT1), 95.7% (WTT2) and 96.3% (WTT3). High effectiveness in removal of ammonium ions is most likely caused by their adsorption on the surface of solid particles [21], hence removed during sedimentation and filtration. These ions could also be assimilated by microorganisms existing on the surface of GAC deposits, which was consequently observed for WTT1 and WTT2. The effective removal of nitrate ions from water, in accordance with available scientific literature [21, 22], is possible through processes of ion exchange and/or denitrification of gas nitrogen. None of the analyzed water treatment trials met the necessary conditions for the denitrification process. As a result, the NO_3^- concentration was above the threshold value for biological stability in all trials.

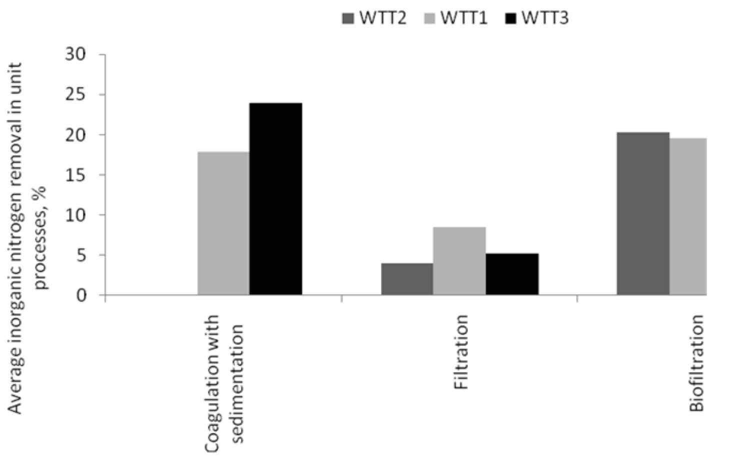

Fig. 4. Effectiveness of unit treatment processes in inorganic nitrogen removal

Due to the predominant role of nitrate ions in inorganic nitrogen and the low effectiveness in their removal through water treatment processes, the treated water samples were still characterised by concentration levels of inorganic nitrogen, too excessive for the limitation of regrowth of microorganisms (Table 3). Observations showed that the most effective processes for the removal of inorganic nitrogen were coagulation and biofiltration (Fig. 4). Coagulation principally guaranteed the removal of ammonium ions, while the biofiltration process effectively eliminated all forms of inorganic nitrogen.

Due to the insufficient elimination of nutrient in water treatment trials, water disinfection with increased doses of chlorine and chlorine dioxide were used in order to lower the potential for regrowth of microorganisms. Unfortunately, this is not sufficient to guarantee biological stability of water, especially within vast distribution systems [23].

4. CONCLUSIONS

The analyzed water treatment trials ensured the removal of organic and inorganic nutrients, thus decreasing the potential for regrowth of microorganisms in the water DS. The use of coagulation and/or biofiltration in water treatment trials guaranteed significant decreases in concentration of organic nutrients. Consequently, most of samples from all three WTT contained less than 0.25 g C/m^3, which can be a factor limiting the regrowth of microorganisms in the distribution system. All of the technologies effectively eliminated phosphate ions from treating waters. Through the precipitation and removal of insoluble phosphates, all water samples had biological stability with regards to this specific inorganic nutrient. Regardless of the type of treatment processes, the phosphate concentrations were the inhibiting factors for regrowth of microorganisms.

Ammonium ions were also effectively removed in the discussed water treatment trials; however, this did not guarantee stability to the water introduced into the water supply flow. The reduction of nitrogen ion content resulted from the use of coagulation and biofiltration processes. The effectiveness of N_{inorg} removal in all technologies was insufficient to ensure water stability of water introduced into the distribution system.

None of the water treatment technologies guaranteed a simultaneous elimination of organic and inorganic nutrients effective enough to limit the regrowth of microorganisms. However in most samples BDOC and phosphate removal efficiency were sufficient.

Additionally, the water treatment trials discussed in this paper should start using processes which above all ensure the effective elimination of nitrogen compounds, because inorganic nitrogen concentrations in all samples were too high.

In conclusion, regardless of water type, the coagulation with sedimentation and biofiltration are two most effective unit processes in removal of BDOC and phosphates.

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