The Silesian University of Technology No. 4/2012

Agnieszka TUSZYŃSKA a, Katarzyna KOŁECKA b

a Dr.; Faculty of Civil and Environmental Engineering, Gdansk University of Technology, Narutowicza Str. 11/12, 80-233 Gdansk, Poland E-mail address: *atusz@pg.gda.pl*

 b Dr.; Faculty of Civil and Environmental Engineering, Gdansk University of Technology, Narutowicza Str. 11/12, 80-233 Gdansk, Poland E-mail address: *katkolec@pg.gda.pl*

Received: 20.03.2012; Revised: 25.07.2012; Accepted: 26.07.2012

Abstract

In this paper the particle size of suspended solids in the removing of organic matter and phosphorus compounds from lake **water and wastewater treated in the Ecological Treatment Systems (ETS) was analyzed.**

The invesigated materials were taken from two ETS located in Pomerania Region/Poland. Object in Swarzewo is 3rd stage of biological treatment of municipal wastewater, while object in Kartuzy has task to remove a phosphorus from lake water. To deepen the knowledge about migration of pollutants in water and wastewater their particle size analysis based on the laser diffraction method were carried out. Speciation of phosphorus and organic matter expressed in COD was carried out for the purpose of determination of the participation and type of dissolved substances as well as suspended matter.

The granulometric analysis showed that from wastewater and lake water treated in ETS the suspension above size 100 µm is efficiently removed. On the basis of the obtained results it was proved that ETS efficiently remove particulate fractions of phosphorus and organic matter. In wastewater and surface water inflowed to analyzed facilities 30-45% of organic matter and 55-80% of phosphorus was attributed to suspended solids. In the effluent a part of pollutants in suspended fraction decreased and it were 15% for COD and 35% for phosphorus in wastewater and 24% for COD and 50% for phosphorus in **lake water.**

Streszczenie

W pracy podjęto próbę określenia znaczenia granulometrii zawiesiny w usuwaniu substancji organicznej i fosforu z wody jeziornej i ścieków doczyszczanych w ekologicznych systemach oczyszczających (w jęz. ang. Ecological Treatment Systems – ETS). Materiał do badań został pobrany z dwóch obiektów ETS zlokalizowanych w woj. pomorskim. Obiekt w Swarzewie stanowi **III stopień biologicznego oczyszczania ścieków komunalnych, podczas gdy obiekt w Kartuzach ma za zadanie doczyszczać wodę jeziorną ze związków fosforu.**

W celu pogłębienia informacji na temat migracji zanieczyszczeń w środowisku wodnym przeprowadzono ich analizę granulometryczną metodą dyfrakcji laserowej. W celu określenia udziału i rodzaju zanieczyszczeń w zawiesinie i frakcji roz**puszczonej przeprowadzono specjację substancji organicznej wyrażonej jako ChZT oraz związków fosforu.**

Analiza granulometryczna wykazała, że ze ścieków i wody jeziornej poddawanych oczyszczaniu w systemach ETS skutecznie jest usuwana zawiesina o rozmiarze cząstek powyżej 100 μm. Na podstawie uzyskanych wyników stwierdzono, że ekologiczne systemy oczyszczające zapewniają skuteczne usuwanie ze ścieków i wody jeziornej frakcji fosforu i substancji organicznej **w zawiesinie. W ściekach i wodach powierzchniowych dopływających do analizowanych obiektów 30-45% substancji organicznej** i 55-80% fosforu było związanych w zawiesinie. Na odpływie udział zanieczyszczeń we frakcji zawiesinowej uległ obniżeniu i w ściekach wynosił odpowiednio 15% dla ChZT i 35% dla P, zaś w wodzie jeziornej: 24% dla ChZT i 50% dla fosforu.

K e ywo r d s: **Surface water protection; Particle size analysis of suspended solids; Laser diffraction method; Speciation of phosphorus and organic matter.**

1. INTRODUCTION

From Poland about 178 000 tons of organic matter (expressed in COD) flows annually to Baltic Sea. This is about 20% of total load discharged to the sea from all Batlic countries. It is estimated that annual influent of nutrients is 140 000 tons of nitrogen (N) and about 10 000 tons of phosphorus (P) (approximately 30% i 40% of total load, respectively). The highest load of total nitrogen and phosporus is discharged from rivers. Insufficient treated wastewater and surface run-off are the main source of inland water pollution.

The last Framework Directive (RDW 2000/60/WE) concerning water protection enforces the obligation of ensuring of "good" quality of surface water to year 2015. Taken regulations introduce low concentrations of suspended solids (SS) and nutrients in treated wastewater. In order to protect surface water against the inflowing of organic matter in suspended form and phosphorus compounds permissible concentrations of pollutants in treated wastewater can not exceed 50.0 mg/l and 1.0 mgP/l, respectively. Untill now it was indicated that ensuring in treated wastewater required concentration of phosphorus is very difficult and even impossible. Moreover, it is assumed to prevent algea blooming, phosphorus concentration in surface water should not exceed 0.01 mg/l [10, 11, 12]. Due to the strong association of suspended solids with wastewater Chemical Oxygen Demand (COD) or phosphorus, suspended solids have to be removed very efficiently to achieve the required effluent quality in highly sensitive coastal areas [5]. SS effluent standards in these areas should normally be as low as 10.0 or 5.0 mg/l [14]. According to Authors suspended solids play an important (and often underestimated) role in characterizing the treatability and hence the degree of contaminant removal of a wastewater. The size of the SS has considerable impact on sedimentation and sorption capacity. There are reports that they can fulfill function of organic micropollutants "carriers" as well as phosphorus, nitrogen and heavy metals [16]. Furthemore, the biological degradation rate in terms of COD reduction is influence by particle size distribution [5, 15].

In the last few years it can be observed more often that ecological treatment systems are used to improve the quality of inland waters and renaturalisation of treated wastewater. ETS are usually built along the shore-line in order to protect the rivers and lakes from the inflowing of high pollutants load. ETS are also designed as a third degree of biological treatment in municipal WWTP. These objects are designed to treat the wastewater effluenting from secondary settling tanks from organic matter and nutrients. Previously analyzed ecological treatment systems characterized by very good efficiency of the SS removal greater than 90%. Similarly, the removal efficiency of organic matter is considerable and ranges for BOD5 from 71.5 to 90.5% and for COD from 59.7 to 89.0% [9]. Absorption of mineral forms of N, P and organic carbon by primary producers (phytoplankton, macrophytes) and the deposit of nutrients in bottom sediments is important to reduce the contaminants concentration in wastewater [2].

References mainly relate to determine the efficiency of individual pollutant removal while do not include the information about the dispersed substance – "potential carriers" of organic matter and phosphorus as well as their migration in the environment. It is not known much about their fractions and ability to transform.

The aim of this study was the granulometric analysis of suspended solids in removed organic matter and phosphorus from waste- and surface- water treated in ETS. The obtained results will form the basis to evaluate the removal possibility of contaminants from the environment.

2. MATERIALS AND METHODS

2.1. Sampling locations

The invesigated materials were taken from two ETS located in Pomerania Region. These systems were built for the renaturalisation of Wastewater (in Swarzewo) and lake water (in Kartuzy). Facility in Swarzewo is a third degree of biological treatment in municipal WWTP. The outflow of wastewater is directed directly into the Baltic Sea (average flow $Q = 6000 \text{m}^3/\text{d}$. In Swarzewo there are three ponds which are inhabited by daphnia. The total area of ponds, which are built in series, is 23 189 m2. The average depth of $2nd$ and $3rd$ pond is approximately 1.5 m, while the first pond has a variable depth depending on the amount of the applied sludge. Including the sediment depth it does not exceed 1 m.

In Kartuzy in the section connecting Karczemne Lake and Male Klasztorne Lake the ETS was built in the form of two serpentine pools about 10.0 m wide, 100.0 m long and 0.5 m deep planted with the water plants. The bottom of pools is 0.3 m layer of calcareous sand. ETS has the task to take over the contaminants load (mainly phosphorus) from Karczemne

Lake preventing their entring into the Male Klasztorne Lake. The characteristic of analysed objects is presented in Table 1.

2.2. Methods

Monitoring of most physico-chemical parameters was carried out from June 2009 to May 2011. The wastewater and lake water were sampled once a month at the inlet and outlet from analyzed ETS.

To deepen the knowledge about migration of pollutants in water and wastewater their particle size analysis by laser diffraction method were carried out. The laser granulometer Malvern Instruments Ltd 2000 with measuring range from 0.02 to $2000 \mu m$ was used to measure the content and size of dispersed particles. This equipment thanks to optical unit analyses of light dispersion on a particle and next based on Mie theory and Fraunhofer model, calculates the particle size [3]. The measurement results are presented as a histogram that allows to assess the distribution of "grain" pollutions.

Speciation of phosphorus and organic matter expressed as COD was carried out for the purpose of determination of the participation and type of dissolved and suspended fraction. Fractionation of COD and P was performed by filtration through polycarbonate membrane filters with pore diameters $0.45 \mu m$.

Determinations of organic matter concentration (expressed as BOD5 and COD), phosphorus and organic and total suspended solids were made according to obligatory European standards.

Efficiency of analysed pollutant removal was calculated as the quotient of the difference of concentration on the influent (C_i) and effluent (C_e) to the concentration on the influent (C_i) , $\eta = (C_i - C_e) / C_i$. To develop the obtained results the Excel program was used. The results were statistically analyzed in the linear correlation. For each essential correlation the critical value of the correlation coefficient at $P_{0.05}$ was given. For calculations modules Statistica Pl program was used.

3. RESULTS AND DISCUSSION

3.1. Size distribution of particle volume

Figures 1 and 2 show the particle size distribution of contaminants in wastewater and lake water before and after treatment in two analysed facilities. Figures show that wastewater and surface water of the influent to ETS during the study was characterised by the highest volume part of pollutants in suspended fraction. Whereas in effluent part of colloidal and dissolved fractions increased.

Particle size distribution in the influent of wastewater at ETS – Swarzewo and in lake water at ETS – Kartuzy (average values at least N=20 samples

e

Figure 2.

Particle size distribution in the effluents of wastewater and lake water from ETS – Swarzewo and ETS – Kartuzy (average values at least N=20 samples)

The pollutants inflowing to the facility in Swarzewo included fraction with a equivalent diameter about 0.02 µm and larger diameter about 2.4 mm. Influent of wastewater had the highest volume part of pollutants in range from 80 to 500 µm and it was 80% of total particles volume (Fig. 1). The lake water from Karczemne Lake had the highest volume part of pollutants with a equivalent diameter in range from 30 to 400 µm (Fig. 1). The granulometric analysis showed that the particles of the size up to 400 µm comprised the overwhelming part (about 90%) of the samples volume.

Treated wastewater and lake water had lower volume part of suspended fraction than influents to the analyzed objects. This was due to processes occurring in ETS such as oxidation and reduction, as well as sorption and sedimentation. Particles of equivalent diameter highest than 100 microns underwent these processes. The fractions of pollutants in range from 10 to 70 μ m had the highest volume part in a sample of treated wastewater (about 60%, Fig. 2). Particle size distribution of contaminants in effluent of lake water was similar to the composition of treated

wastewater discharged into the Baltic Sea. Similar like in Swarzewo the highest volume part – 70% had the fraction of equivalent diameter in range from 10 to $80 \mu m$.

3.2. Association of Chemical Oxygen Demand (COD) and phosphorus (P) with suspended solids (SS)

The analysis of waste and lake water samples revealed that considerable amounts of COD and phosphorus were particulates (Table 2). 45% of the influents wastewater COD and 30% of the influents lake water COD, were attributed to suspended solids.

On the effluent the particle associated COD fraction decreased and it was from 15% do 24%. Average ratios of particulate COD/particle mass decreased from 1.1-1.3 mgCOD/mgSS in the influent to 0.6-1.1 mgCOD/mgSS in the final effluents due to biodegradation processes. In case of phosphorus, the particle associated fraction was 55-80% in influent and decreased in effluent to 35% in wastewater and 50% in lake water due to sorption and sedimentation processes. Particulate P/particle mass ratios were between 0.04 and 0.08 mgP/mgSS in influents. The ratios of particulate P/particle in final effluent increased and were 0.06 mgP/mgSS for lake water and 0.11 mgP/mgSS for wastewater (Tabele 2).

3.3. The efficiency of suspended solids, organic matter and phosphorus removal in analyzed ETS

The study allowed to determine the efficiency of contaminants removal in ecological treatment systems. The ability of ETS to remove suspended solids, organic matter and phosphorus compounds is presented in Figure 3.

Table 2.

Suspended solids, total and particulate COD and P in wastewater and lake water treatment in ETS (average values at least n=20 sam**ples with standard deviation**

Sample source	COD			Phosphorus			Suspended Solids	
							organic	mineral
	total [mg/l]	$>0.45 \mu m$ $[\%]$	COD/SS [mg/mg]	total $[mg/l]$	$>0.45 \mu m$ $[\%]$	P/SS [mg/mg]	$>0.45 \mu m$ [mg/l]	
ETS – Swarzewo								
Influent	65.6 ± 16.8	45	1.3	2.27 ± 0.5	80	0.08	22.1 ± 7.8	1.5 ± 0.4
Effluent	40.2 ± 4.1	15	1.0	1.64 ± 0.5	35	0.11	5.1 ± 1.1	0.2 ± 0.1
ETS - Kartuzy								
Influent	31.8 ± 4.8	30	1.1	1.41 ± 0.3	55	0.04	20.4 ± 4.2	1.7 ± 0.4
Effluent	27.6 ± 6.7	24	0.6	1.03 ± 0.3	50	0.06	12.4 ± 3.7	1.2 ± 0.7

3.3.1. Suspended solids removal

The average value of suspended solids concentration in object in Swarzewo was 22.1 ± 7.8 mg/l in influent and 5.1 ± 1.1 mg/l in effluent (Table 2). The part of organic suspended solids in total suspension in influent of wastewater was about 95%. The ETS ponds with daphnia had more than 80% removal efficiency of pollutants from wastewater (Fig. 3). The efficiency of suspended solids removal in ETS-Swarzewo was similar to the efficiency of the filtration process. Tchobanoglous et al. [13] on the basis of studies on WWTP in Las Vegas proved that filters which were the 3rd stage of biological treatment (filled with anthracite, sand and grave) reduced the suspended solids from 30.0 to 3.0 mg/l in wastewater. According to Authors the best quality of effluent, which can be achieved using simple filtration is from 5.0 to 10.0 mg of SS per liter.

The part of organic suspened solids in lake water was similar to their part in wastewater from Swarzewo and it was approximately 93%. However, the efficiency of suspended solids removal in ETS – Kartuzy was two times lower compared with ETS – Swarzewo. This was probably caused by the secondary formation of suspended solids. The ETS objects planted with water plants created some amount of "residual" organic suspended solids, which may occur in the outflow from the object. Residual suspensded solids have nothing to do with an anthropogenic suspension suplied with wastewater and they result from biochemical changes of natural organic matter. Based on the results of the analysis of previously operated ETS it has been shown that the concentration of the residual suspensed solids varied from 7.0 to 25.0 mg /l [7, 8]. Therefore, the concentration of suspended solids in effluent from object in Kartuzy was more than two times higher than in effluent from Swarzewo (Fig. 3).

3.3.2. Organic matter removal

The wastewater inflow to the ETS-Swarzewo was of low biodegradability. This is demonstrated by the value of $\text{COD}_{\text{tot}}/\text{BOD}_5$ ratio which is 2.8. This is caused by a high concentration of dissolved not biodegradable organic matter in wastewater. The part of this substance in COD_{tot} was approximately 60%. Therefore, the efficiency of organic matter removal was low, only about 38% (Fig.3).

Also, the inflow of lake water to ecological treatment system in Kartuzy was not susceptible to biodegradation. The value of the $\text{COD}_{\text{tot}}/\text{BZT}_5$ ratio of lake water was 4.4. Therefore, the efficiency of removal of organic matter expressed as COD was very low, only 13% (Fig. 3).

3.3.3. Phosphorus compounds removal

The average value of total phosphorus concentration in wastewater from Swarzewo was 2.27 ± 0.5 in influent and 1.64 ± 0.5 mgP/l in effluent (Table 2). The part of phosphorus attributed to SS was approximately 80% in influent, while the part of this fraction in effluent significantly decreased and it was approximately 35%. The content of suspended organic matter significantly affected the content of total phosphorus in wastewater. The significant positive linear correlation (correlation coefficient: $r = 0.80$, $N = 20$, $r_{\text{crit}} = 0.396$) between the concentrations of suspended organic matter and total phosphorus was concluded. The results show that the phosphorus attributed to SS was removed from wastewater. While the opposite situation was observed for phosphates. The concentration of dissolved phosphorus fraction was 0.86 ± 0.3 mgP/l in the influent and 1.16 ± 0.3 mgP/l in the effluent. The part of phosphates in the total phosphorus was approximately 20% in influent, while the part of this fraction increased nearly two times in effluent (Fig. 3). Probably the reason of this effect was the secondary release of dissolved phosphorus fraction from sediments into wastewater. Obtained results may indicate that the organic suspensions which adsorbed phosphorus contained in the sediments caused the phosphates re-pollution in wastewater. ETS do not have the proper equipment to remove suspensions from the bottom, which result in secondary pollution of dissolved phosphorus. Some of phosphorus in suspended fraction remained at the bottom, while some have been re-released into the liquid phase, which caused increase of the value of total phosphorus in effluent of wastewater. Therefore, the efficiency of total phosphorus removal

e

in ETS was low (about 30%) (Fig. 3). Similar observations have been described among others by Arias et al. [1] and Gomez et al. [4]. According to Authors ecological treatment systems show significant differences in the efficiency of phosphorus compounds removal – ranging from 20 to 70%. Reddy and D'Angelo [8] proved that at the beginning of operation ETS fulfill their function in the removal of phosphorus compounds, but their efficiency was reduced over the period of time. These observations were confirmed among others Maurer et al. [6]. Authors proved that the impact of accumulation of phosphorus in the living biomass (macrophytes, algae and peryfiton) is variable and may lead to both short- and long-term decline of phosphorus in the ETS. The release of nutrients from macrophytes may occur during decomposition of dead plants or by feeding of animals. Athroping water plants and algae (as a solid bottom) can become an internal source of nutrients, and therefore caused the increase of the phosphorus concentration in water.

The average value of the total phosphorus concentration in lake water flowing into the ETS – Kartuzy was 1.41 ± 0.3 mgP/l. In effluent Ptot concentration decreased and was 1.03 ± 0.3 mgP/l (Table 2). Similar like in Swarzewo, the content of suspended organic matter significantly affected the concentration of total phosphorus in lake water. The significant positive linear correlation (correlation coefficient: $r = 0.72$, $N = 18$, $r_{crit} = 0.396$) between the concentrations of suspended organic matter and total phosphorus was concluded. In influent almost half of the total phosphorus in lake water consituted dissolved phosphorus fraction, while in effluent that part of phosphate has decreased down to 20.0% of P_{tot} . The application of calcareous sand as an absorbent caused that the efficiency of dissolved phosphorus fraction removal was very high (about 70.0%) (Fig. 3). Obtained results confirm the observations described by Arias et al. [1]. According to Authors the calcareous sand as filling of filter bed can absorb about 2-3 kg of phosphorus per m3 of sand.

4. CONCLUSIONS

This study demonstrates that a remarkable part of COD and phosphorus from wastewater and lake water is related to the suspended solids. Therefore, efficient removal of suspended solids is necessary to protect highly sensitive coastal areas. Granulometric analysis of wastewater and lake water showed that the particle size of contaminants respectively in range from 80.0 to

 $400 \mu m$ in inflow and from 10.0 to $80.0 \mu m$ in outflow dominated. The increase of organic matter and phosphorus content related to suspension of its lower size was not observed either. The basis for this conclusion was the similar values of the P/SS and COD/SS ratio in wastewater and lake water before and after treatment. Due to the more or less random distribution of particulate contaminants, it is important to obtain efficient overall particle mass removal.

Analysis of the work of two ecological treatment systems found that these objects remove up to 70% of suspended solids of particle size above 100 μ m from wastewater and lake water. On the basis of obtained results it was found that ETS efficiently remove a phosphorus and an organic matter attributed to suspended solids. In wastewater and surface water that inflowed to analyzed facilities 30-45% of organic matter and 55-80% of phosphorus were attributed to suspended solids. In the effluent a part of pollutants in suspended fraction decreased and it was 15% for COD and 35% for phosphorus in wastewater and 24% for COD and 50% for phosphorus in lake water.

ACKNOWLEDGEMENT

Research was carried out thanks to research project No N N523 452036 of Ministry of Science and Higher Education and 2 projects of Fund for Environmental Protection and Water Management in Gdansk: WFOŚ/D/201/105/2009 and WFOŚ/D/201/199/2010

REFERENCES

- [1] *Arias C.A., Del Bubba M., Brix H*.; Phosphorus removal by sands for use as media in subsurface flow constructed reed beds. Wat. Res., Vol.35, 2001; p.1159-1168
- [2] *Craft C. B., Richardson C. J.*; Peat accretion and N, P and organic C accumulation in nutrient-enriched and unenriched everglades peatlands. Ecological Applications, Vol.3, No.3, 1993; p.446-458
- [3] *De Boer G., De Weerd C., Thoenes D., Goossens H.*; Laser diffraction spectrometry: Fraunhofer versus Mie scattering. Particle Syst. Charact, Vol.4, 1987; p.14-19
- [4] *Gomez E., Durillon C., Rofes G., Picot B*.; Phosphate adsorption and relase from sediments of brackish lagoons: pH, O₂ and loading influence. Wat. Res., Vol.33, No.10, 1999; p.2437-2447
- [5] *Levine A.D., Tchobanoglous G. and Asano T.*; Characterization of the size distribution of contaminants in wastewater: treatment and reuse implications. J. Wat. Pollut. Control Fed., Vol.57, 1985; p.805-816
- [6] *Maurer M., Abramomovich D., Siegrist H., Gujer W.*; Kinetics of biologically induced phosphorus precipitation in wastewater treatment. Wat. Res., Vol.33, No.2, 1999; p.484-493
- [7] *Moore B. C., Lafer J., Funk W.*; Influence of aquatic macrophytes on phosphorus and sediment pore water chemistry in a freshwater wetland. Aquatic Botany, Vol.49, 1995; p.137-148
- [8] *Reddy K.R., D'Angelo E.M*.; Bichemical indicator to evaluate pollutant removal efficiency in constructed wetlands. In: 5th International Conference on Wetland System for Water Pollution Control, Uni. Fuer Bodenkultur Wien and International Association on Water Quality, Vienna, 1996; p.1-21
- [9] *Richardson C. J., Craft C. B*.; Effective phosphorus retention in Wetland: Fact or fiction? Constructed wetlands for water quality improvement. Edited by G. A. Moshiri. Lewis Publishers, 1993; p.271-282
- [10] Rozporządzenie Ministra Środowiska z dnia 27 listopada 2002 r. w sprawie wymagań, jakim powinny odpowiadać wody powierzchniowe wykorzystywane do zaopatrzenia ludności w wodę przeznaczoną do spożycia. (Regulation of the Minister of Environment of 27.11. 2002 on the requirements regarding surface water used for public supply of water intended for human consumption). Dz.U. nr 204 poz. 1728 z 2002 r. (in Polish)
- [11] Rozporządzenie Ministra Środowiska z dnia 11 lutego 2004 r. w sprawie klasyfikacji dla prezentowania stanu wód powierzchniowych i podziemnych, sposobu prowadzenia monitoringu oraz sposobu interpretacji wyników i prezentacji stanu tych wód. (Regulation of the Minister of Environment of 11.02. 2004 on the classification of the present status of surface water and groundwater, the way of monitoring and interpretation of the results and presentation of status of these waters). Dz. U. Nr 32, poz. 284 z 2004 r. (in Polish)
- [12] *Schroeder et al*.; Using Phytoremediation Technologies to Upgrade Wastewater Treatment in Europe. Env. Sci. Pollut. Res, 2006; p.1-8
- [13] *Smith S.V*.; Phosphorus versus nitrogen limitation in the marine environment. Limnol. Oceanogra. Vol.29, No.6, 1984; p.1149-1161
- [14] *Tchobanoglous G., Burton F., Stensel D*.; Wastewater Engineering. Proc.Metcalf&Eddy, 4th Edition, 2006; p.1818
- [15] *Tuszyńska A., Obarska-Pempkowiak H*.; Dependence between quality and removal effectiveness of organic matter in hybrid constructed wetlands. Bioresource Technology, Vol.99, 2008; p.6010-6016
- [16] *Yeoman S., Lester J.N. and Perry R.*; Phosphorus removal and its influence on particle size and heavy metal distribution during wastewater treatment. Environ. Technol. Vol.13, 1992; p.901-924

e