Investigating phosphorus loads removed by chemical and biological methods in municipal wastewater treatment plants in Poland

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Abstract

The article presents an analysis of the current methods for phosphorus removal applied in municipal wastewater treatment plants in Poland. Within the study, 131 wastewater treatment plants were investigated, constituting 17 630 500 population equivalent, which is about 1/3 of the overall population equivalent (designed) in Poland. The research was based on a detailed technical questionnaire analysis obtained from wastewater treatment plants operators and calculations of pure metal doses in the applied chemical reagents and their type per a treated wastewater volume, population equivalent and phosphorus load removed. The analysis results show that annually a minimum of 1 470 Mg of phosphorus removed by 35 wastewater treatment plants based entirely on biological treatment methods could be used for phosphorus recovery to produce struvite, calcium phosphate or other highly bioavailable alternative fertilizer products. Moreover, 1 490 Mg of phosphorus removed by other 17 wastewater treatment plants with a minimal coagulant dose (< 1 g of metal per m^3 of wastewater), increase the base for phosphorus recovery to approx. 2 960 Mg per year using the sewage sludge or its dewatering liquors. These results suggest that the implementation of the means mentioned above would significantly increase the possibilities for obtaining phosphorus from secondary sources, especially in wastewater treatment plants without a direct access to sewage sludge incineration plants.

Introduction

Due to the rapid intensification of fertilization with mineral fertilizers and increasing urbanization at the beginning of the second half of the XX-century, many aquatic ecosystems suffered from excessive biogenic substance loads of anthropogenic origin (Hong et al. 2012). One of these substances was phosphorus (P), which is known for its essential biogenic role used for stimulating crops yields growth by applying P-based organic and mineral fertilizers (Rosemarin et al. 2020). However, P biogenic function has a serious drawback, reveled in the aquatic ecosystem in which it simulates algae growth what results in negative consequences as dissolved oxygen deficits, hypoxia, water taste and odor problems, limited photosynthesis and others leading to water eutrophication with harmful algal blooms (HABs) and water bodies degradation in the most severe cases (Ekholm and Krogerus 1998; Dodds et al. 2009; Tang et al. 2018). To mitigate eutrophication process which appeared in a growing number of water ecosystems such as Lake Michigan (Barbiero et al. 2002), Lake Erie (Wilson et al. 2018), Chesapeake Bay (Conley et al. 2009), Constance Lake (AERZEN 2019), Taihu Lake (Wang et al. 2019), the Baltic Sea (Tanzer et al. 2021) and others, in many countries nutrients loads were limited in the final effluent from wastewater treatment plants (WWTPs) (Preisner et al. 2020). In the European Union (EU) Member States based on the Urban Wastewater Treatment Directive (91/271/EEC) (European Commission 1991) limits for P concentration in treated wastewater were established for 2 categories based on the agglomeration size set in population equivalent (PE). Therefore, total phosphorus (TP) concentration was limited to a maximum 2 mg/L in WWTPs which PE is between 10 000 and 100 000 and to a maximum 1 mg/L for > 100 000 WWTPs. These limits were tightened in some cases, e.g. for the direct and indirect discharge to the Baltic Sea based on the Baltic Marine Environment Protection Commission (HELCOM) recommendations to a maximum 0,5 mg/L (HELCOM 2007). More strict regulations are used worldwide in terms of particular sensitive reservoirs such as oligotrophic alpine lakes in Switzerland, Italy and Germany (< 0,3 mgP/L) (Petri 2006; Tu et al. 2019) and in the United States e.g. for Potomac River (0,2 mgP/L) or Lake Occoquan (0,1 mgP/L) (Sedlak 1991). The above limits are usually set for TP (as a sum of organic and inorganic P) or its inorganic form expressed as orthophosphates (PO_4-P) which is the most bioavailable P form to aquatic vegetation (Li and Brett 2013).

P removal is performed in the wastewater treatment process in which mechanical, biological, chemical or physical methods are applied or their combination. Chemical P removal methods are known for low investment costs and high treatment efficiency (Wang et al. 1996). Among the most common coagulants there are: iron chloride (III) ($FeCl_3$), iron chloride (II) ($FeCl_2$), iron sulphate (II) ($FeSO_4$), iron sulphate (III) ($Fe_2(SO_4)_3$), aluminum sulfate ($Al_2(SO_4)_3$), aluminum polychloride ($Al_n(OH)_mCl_{(3n-m))}$), calcium hydroxide ($Ca(OH)_2$) and sodium aluminate ($NaAl_2O$) (Aboulhassan et al. 2006; Kłos and Gumińska 2009; Nowacka et al. 2014).

Phosphates removal by chemical precipitation takes place due to the formation of undissolved iron (Fe) or aluminum (Al) compounds according to the following Formula (1) (Sperczyńska 2016):

$$Me^{3+} + PO_4^{3-} \to MePO_4 \downarrow$$

1

From the chemical reaction it appears that the stoichiometric metal dose (Me) needed to remove 95 g of PO_4-P it is necessary to provide 56 g of Fe^{3+} or 27 g of Al^{3+} (Sperczyńska 2016). However, in practice WWTPs operators apply larger coagulant doses (even 2 or 5 times larger) than the stoichiometric doses due to various and not constant wastewater composition (Henze 1995; Heidrich and Witkowski 2005; Zhou et al. 2008). It is also possible to use lime instead of metal salts, but it leads to a larger amount of produced sludge, additional cost related to lime storage and a high dependence on wastewater pH (Przywara 2006).

The biggest disadvantage of chemical P precipitation is revealed during P recovery. Application of metal salts based on Fe and Al for coagulation process results in the occurrence of Fe-P or Al-P complexes in the sludge, which lowers the bioavailability of P and limit the recovery possibilities for agriculture needs (Szabó et al. 2008). This issue matters when P is recovered from the sewage sludge, sludge dewatering liquors or digestate. By using crystallization methods for P recovery it is possible to obtain phosphates as calcium phosphate (Ca-P), magnesium phosphate (MP), ammonium phosphate (MAP or struvite) or potassium magnesium phosphate (KMP) (Piekema and Giesen 2001). However, crystallization of struvite or Ca-P has limited recovery efficiency approx. up to 25% regarding influent P content (Melia et al. 2017).

An alternative to the above methods is P recovery from sewage sludge ashes (SSAs) which efficiency reaches over 85% (Amann et al. 2018). However, incineration plants for sewage sludges are mainly located near the largest WWTPs with the highest P load and content in the ash (Smol et al. 2020a).

Moreover, in many countries there are no operating SSA processing plants in which extraction of P and production of phosphate fertilizers is carried (Herzel et al. 2016).

On the other hand, struvite precipitation is currently the most often applied method for P recovery based on the crystalisation process, accounting for more than 75% of the recovery products. Calcium phosphate (Ca-P) is recovered in less than 20% recovery products while phosphoric acid (H_3PO_4) in approx. 5% (Shaddel et al. 2019). However, in Poland there is currently only one ongoing construction in an advanced stage of a P recovery unit (in Jarocin-Cielcza WWTP) which probably from 2022 will produce struvite using the Ostara Pearl technology (PWiK Jarocin 2021).

Due to the general dependence on mineral fertilizers produced from imported phosphate rock many countries such as the EU Member States, including Poland which imports all of the phosphates to produce P fertilizers need to introduce P recovery from alternative sources and among them, wastewater is one of the most promising ones (Smol et al. 2020b).

In the above context, this paper aims to analyze the possibilities to apply P recovery units in WTTPs in Poland based on struvite or similar products with a high content of bioavailable P that can be used in agriculture and replace mineral fertilizers. Within the scope of the present study, loads of P removed from wastewater and doses of metal-based coagulants were considered as the main factors that influence the development of the above-mentioned solutions.

Materials And Methods

The research was based on the developed questionnaire for WWTPs operators. The construction of the questionnaire was divided into 4 steps:

Step 1: Content preparation:

1.1) Primary selection of survey questions.

1.2) Adding suggestions regarding the most appropriate units for collected data (e.g. kg of coagulant per year rather than its volume in cubic meters per day).

1.3) Reduction of the least important or potentially difficult questions for interpretation that might be wrongly understood and led to receiving incorrect data.

The questionnaire included the following survey questions about the WWTP:

(1) the name and location of a WWTP,

(2) the year that the data came from,

(3) daily average of treated wastewater flow in m^3/day,

(4) population equivalent (PE) of the WWTP,

(5) used wastewater treatment technology – selection out of 9 given options based on the list of the most widespread methods: a) anoxic-oxic (AO), b) anaerobic-oxic (A/O), c) 3-stage Bardenpho, d) 5-stage Bardenpho, e) Johannesburg (JHB) or its modification (MJHB), f) University of Cape Town (UCT) or its modification (MUCT), g) Sequencing batch reactor (SBR), h) Membrane bioreactor (MBR), i) other - with additional description of the used method or modification scheme.

Moreover, the questionnaire included the survey questions about the used coagulants:

(6) if the WWTP uses chemical P precipitation in order to achieve the limits set by the valid legal regulations (yes or no),

(7) the amount of used coagulant in m^3/year or L/day,

(8) type of the coagulant - trade name of the product and its chemical formula,

(9) phase of the treatment process in which the coagulant was added (which part of the technological line),

(10) TP and PO_4-P concentration in raw wastewater,

(11) TP and PO_4-P concentration in treated wastewater.

Step 2: Sending the questionnaire to the official electronic addressees of the WWTPs treating municipal wastewater in Poland:

2.1) Development of a database with current email addresses of operating WWTPs in Poland treating municipal wastewater (including also industrial WWTPs treating municipal wastewater).

For the database development mainly the latest available report from the implementation of the National Municipal Wastewater Treatment Programme (NMWTP) from 2017 was used to collect the valid email addresses of WWTPs (Polish Waters 2017; Preisner et al. 2021b). If an email given for contact in the NMWTP report turned to be invalid additional desk research was conducted to identify the correct address.

2.2) Selection of addressees by sorting from the largest WWTPs in terms of PE and secondly in terms of the wastewater flow rate.

The survey was sent to 645 out of 1 930 WWTPs registered by the main wastewater-competent governmental body in Poland – the Polish Waters. The selection was aimed to cover the largest WWTPs and the ones with diverse technological schemes in terms of P removal in 3 PE categories: a) large (> 100

000 PE, max. 1,0 mgP/L), b) medium (100 000–15 000 PE) and c) small (< 15 000 PE). From the above-mentioned categories, the survey was sent to all (105) large WWTPs, to 278 medium WWTPs and 262 small WWTPs.

2.3) Personalization of the query content for each respondent.

Step 3: Collection of received responses:

3.1) Checking the correctness of received questionnaires based on a comparison with the average of other received questionnaire responses in order to avoid typical numerical typos.

3.2) Collecting missing or solving the unclear information obtained from respondents.

Step 4: Data analysis:

4.1) Unification of units of the obtained values (flow rate as m^3/day, mass of coagulant as Mg/year, etc.)

4.2) Calculations of pure metal content in the used coagulants were based on analyzing the Material Safety Data Sheet provided by the coagulant manufacturers as presented in Tables 1 and 2.

Table 1
Fe-based coagulants used in the analyzed WWTPs

Product name	**PIX 100**	**PIX 109**	**PIX 110**	**PIX 113**	**PIX 123**	**PIX C40**
Total iron [%]	10,30	10,50	12,50	11,80	12,60	13,60
Density (ρ)	1,265	1,32	1,46	1,52	1,556	1,42
Chemical formula	$FeCl_2$	$FeCl_3$	$FeClSO_4$	Fe_2SO_4	$Fe_2(SO_4)_3$	$FeCl_3$

Table 2
Al-based coagulants used in the analyzed WWTPs

Product name	**PAX 16**	**PAX 18**	**PAC Donau**	**FLOKOR 1,2A**
Al_3^+ [%]	8,20	9,00	15,50	12,00
Al_2O_3 [%]	15,50	17,00	-	22,60
Density (ρ)	1,33	1,37	1,37	1,29
Chemical formula	$Al(OH)_xCl_y + H_2O$	$Al(OH)_xCl_y + H_2O$	$Al(OH)_xCl_y + H_2O$	$Al(OH)_xCl_y + H_2O$

The calculations allowed to obtain the pure metal content in the applied Fe-based and Al-based coagulants for all WWTPs were chemical P removal was used following Formula 2 and 3 respectively (Bratby 2006):

$$\mathrm{M_{Fe}} = \mathrm{V_{PIX}} \bullet \rho \bullet \mathrm{C_{Fe}}$$

2

Where:

M_{Fe} – pure Fe mass [Mg],

V_{PIX} – applied PIX volume [m^3/year],

ρ – coagulant density [g/m^3],

C_{Fe} – Fe content in PIX [%].

$$\mathrm{M_{Al}} = \mathrm{V_{PAX}} \bullet \rho \bullet \mathrm{C_{Al}}$$

3

Where:

M_{Al} – pure Al mass [Mg],

V_{PAX} – applied PAX volume [m^3/year],

ρ – coagulant density [g/m^3],

C_{Al} – Al content in PAX [%].

4.3) Basic statistical analysis (averages, standard deviations, regression and Pearson's correlation coefficient analysis for chosen parameters in order to prepare the output data album used for further substantive analysis.

In response to the 645 sent surveys, a total of 141 questionnaires were sent back with all survey questions answered referring to the newest possible data from 2020. The only exception was the information about the PO_4-P concentration which was included in only 13 replies regarding both, raw and treated wastewater, which results from valid legal regulations that obligate WWTPs to monitor only TP concentration and removal ratio, so PO_4-P content is analyzed only on a voluntary basis. Moreover, 10 received questionaries’ were excluded from the further analysis due to uncertain data provided by the WWTP operators. Therefore, complete datasets collected from 131 WWTPs account for approx. 17 630 500 PE in Poland which states for about 1/3 of the overall designed value of municipal WWTPs together with industrial WWTPs treating municipal wastewater (Polish Waters 2017). To clarify the above, according to the inventory by the national water authority – Polish Waters (Wody Polskie), the total designed PE in Poland is 53 148 678 but in fact the actual PE as the total generated and collected wastewater is approx. 38 500 000 PE (European Environmental Agency 2021).

The complete questionnaires were obtained from 39 large (> 100 000 PE), 74 medium (100 000–15 000 PE) and 18 small (< 15 000 PE) WWTPs by PE category.

Results

Nutrient removal processes are applied in various technological treatment systems which can have a decisive effect on the final effluent and sewage sludge parameters. In Poland, enhanced biological nutrient removal (EBNR) technology is the leading method in terms of municipal WWTPs. However, to meet the current legal regulations for P removal set by the latest Regulation of the Ministry of Marine Economy and Inland Navigation (Ministry of Marine Economy and Inland Navigation 2019), chemical P precipitation is applied often as an integrated treatment method. Thus, the first step of the research stage was to check which treatment technology are used in the analyzed WWTPs and what is their average PE (Table 3).

Table 3
Basic characteristics of the analyzed WWTPs

No.	Wastewater treatment technology	PE (average)	Number of plants
1.	3-stage Bardenpho	111199	87
2.	SBR	16206	12
3.	A/O	94214	8
4.	5-stage Bardenpho	281076	8
5.	AO	71732	7
6.	UCT/MUCT	411000	3
7.	JHB	171878	1
8.	MBR	14950	1
9.	BIOLAK-VOX (A/O system with 5-zones for BioP removal)	57450	1
10.	AAOOE (anaerobic-anoxic-oxic-oxic-endogenic)	580000	1
11.	AOA (anaerobic-oxic-anoxic)	100000	1
12.	Biodenipho (BioP + BioDenitro)	2100000	1

Based on the survey responses it can be stated that the most often used (~ 66,5%, 87 out of 131 plants) wastewater treatment technology in Poland is the 3-stage Bardenpho system (also called the “AAO” system due to the sequence of anaerobic-anoxic-oxic chambers) which is mainly used in medium and large WWTPs (111 199 PE on average). The similar but more complex, 5-stage Bardenpho system consisting of additional anoxic and oxic chambers with internal sludge recirculation is used by 8 analyzed WWTPs with a higher average in terms of PE (281 076). SBR technology is applied in smaller plants (16 206 PE average) and is the second in terms of occurrence and is used by 12 of the analyzed WWTPs. The A/O and AO systems were used in 8 (94 214 PE average) and 7 (71 731 PE average) WWTPs respectively. The 3 of the analyzed plants were based on the advanced UCT technology (including the 4th largest plant in Łódź with 1 026 000 PE). The other 6 WWTPs covered by the survey were based on various modifications of activated sludge technology including JHB and MBR systems, BIOLAK-VOX system based on the A/O system with 5-zones for biological P removal, AAOOE system with anaerobic-anoxic-oxic-oxic-endogenic chamber sequence, AOA system with anaerobic-oxic-anoxic chambers. The largest WWTP in Poland “Czajka” in Warsaw (2 100 000 PE) uses the dedicated BioDenipho process by Veolia which consists of biological P removal and BioDenitro – used for alternating oxic and anaerobic conditions for improved nitrogen removal (Veolia 2019).

Another important feature was to investigate the dosing point locations in the technological line of the analyzed WWTPs applying chemical P precipitation. Basically, 3 schemes of coagulation can be distinguished (Henze 1995).

The first scheme is dosing coagulant directly to the grit removals or before the primary settling tanks (Pre-coagulation). This method removes P, suspended solids and limits the effluent biochemical oxygen demand (BOD) and chemical oxygen demand (COD) up to 90% which may adversely affect the further course of biological wastewater treatment due to needed organics content (Yang et al. 2019). Effectiveness of P removal with pre-coagulation ranges from 70 to 90% (Dong et al. 2019)

Simultaneous P precipitation is performed by adding a coagulant into the bioreactor. A special example of simultaneous P precipitation is when the coagulants are dosed directly before or after the bioreactor chambers. The precipitated P is removed from wastewater together with the secondary (excess) sludge. The effectiveness of this method is high and ranges from 85% up to 95% (Kallqvist et al. 2002).

Post-coagulation is performed in mixing chambers located after the secondary settling tanks. It is completely independent of biological P removal and its efficiency reaches 90–95% but it requires additional settling tank (Klaczyński 2013).

Within the analyzed WWTPs, pre-coagulation (point A) and simultaneous coagulation (points B, C, D) were applied for P removal from wastewater with minor examples of coagulant addition into the recirculated sludge (point E) as presented in Fig. 1.

The results presented in Table 4 show that 35 analyzed WWTPs (~ 26,7%) were operating without chemical P removal using only mechanical-biological processes. Moreover, 4 plants used pre-coagulation for P removal while in 2 WWTPs the coagulant was dosed into the recirculated sludge. The rest of 90 WWTPs (~ 68,7%) applied a simultaneous coagulation process. Detailed data regarding each coagulant dosing point, annual wastewater flow, PE and removed P loads were presented in Table 4.

Table 4
Coagulant dosing points in the analyzed WWTPs

Main coagulant dosage point	No. of WWTPs	Wastewater flow [m^3/day]	Share (wastewater flow)	PE	Share (PE)	P load removed annually
A – Before primary settling tank	4	127 786	5,40%	1 182 074	6,70%	569,47
B – After primary settling tank	3	24 321	1,03%	315 446	1,79%	158,84
C – Into the bioreactor	19	344 100	14,55%	2 485 721	14,10%	1 112,02
D – Before secondary settling tank	68	1 036 620	43,84%	7 877 655	44,68%	4 298,01
E – Into recirculated sludge	2	382 686	16,18%	2 130 000	12,08%	911,46
No coagulation	35	449 015	18,99%	3 639 601	20,64%	1 469,69
Total	131	2 364 528	100,00%	17 630 496	100,00%	8 519,48

Referring to the data about wastewater flow and PE, 18,99% of analyzed wastewater reflected by 20,64% PE was treated without chemical addition for P removal, using mechanical-biological methods exclusively. In the case of 5,40% (6,70% PE) pre-coagulation was applied and 16,18% of the analyzed influent flow (12,08% PE) coagulant was added to the recirculated sludge, while 59,42% of the wastewater flow (60,57% PE) was treated using simultaneous coagulation process.

One of the main research goals was to calculate a load of removed P without chemical precipitation, which can potentially reduce P recovery efficiency based on struvite or Ca-P crystallization (Eggers et al. 1991). Therefore, 5 categories in terms of applied pure metal dose per a cubic meter of treated waterer were used for WWTPs classification: a) WWTPs without chemical P removal (0 g/m^3), b) from 0 to 1 g/m^3, c) from 1 to 2 g/m^3, d) from 2 to 5 g/m^3, e) > 5 g/m^3. The classification results were presented in Fig. 2 with additional information about average PE of WWTPs in each pure metal dose category and the percentage of the complete P load removed.

Moreover, pure metal doses within the applied coagulants for P removal in terms of total WWTPs PE and wastewater flow were presented in Table 5.

Table 5
Overview of pure metal doses applied in the WWTPs analyzed in the study

Pure metal dose [g/m^3]	Number of WWTPs	Total PE	Total flow [m^3/d]
0	35	3 639 601	449 015
< 1	17	1 878 094	360 015
1–2	15	5 734 303	813 167
2–5	31	2 524 665	282 258
> 5	33	3 853 834	460 073

Theoretically, 1 469,69 Mg/year of P is removed without any chemical addition (in WWTPs serving 3 639 601 PE), which is a great base for struvite or Ca-P crystallization application for P recovery. Moreover, 1 491,01 Mg/year of P is removed from wastewater (in WWTPs serving 1 878 094 PE) with a pure metal dose under 1 g/m^3 (EBNR plants supported with chemical P removal), which also can be used for a plant bioavailable fertilizer (Lahav et al. 2013), so those 2 categories state for nearly 3 000 Mg of P annually (Fig. 3). The material for P recovery from the other categories in terms of pure metal dose (> 1 g/m^3) will most probably limit the recovery methods to sewage sludge incineration and producing fertilizers from their ashes, while too high Fe or Al content in the sludge or its dewatering liquors limits the P bioavailability (Wilfert et al. 2018).

Moreover, the questionnaire results show differences between the 3 categories of WWTPs in terms of their PE which have been presented in Table 6. The first feature is that the large WWTPs received less concentrated influent than the medium and small size plants with TP concentrations of 11,48 mgP/L, 12,82 mgP/L and 17,39 mgP/L respectively. This might be an effect of the combined sewage system in large cities in Poland served by WWTPs with over 100 000 PE, that collects sanitary wastewater and relatively clean rainwater together in one sewer. Due to the current legal regulations for P removal depending on PE values, large and medium plants discharge lower P load than small plants (0,46; 0,62; 1,14 mgP/L on average respectively).

An interesting point of view is given by the results of the pure metal dose used in each category. The unit dose of pure metal applied per a cubic meter of treated wastewater decreases in inverse proportion to the PE category of analyzed WWTPs with the lowest dose of pure metal in large plants (2,88 g/m^3), then in medium plants (3,21 g/m^3) and the highest in small plants (4,17 g/m^3). These results may be explained by the higher treatment efficiency observed in most of the large WWTPs that in Poland are usually more advanced and based on EBNR technology (Castellet-Viciano et al. 2018; Preisner et al. 2021a).

By analyzing the dose of each pure Fe and Al dose per a mass of removed P it can be stated that in all analyzed WWTPs Fe-based coagulants were much more often used than Al-based ones. However, these proportions vary between categories and in large and medium WWTPs Fe:Al ratio in the used coagulants was 0,27:0,02 and 0,28:0,03 respectively, while in small WWTPs it was only 0,22:0,06.

Thus, a sum of Fe and Al content in the applied coagulants shows that per a Mg of removed P in large, medium and small WWTPs 0,29 Mg; 0,31 Mg; 0,28 Mg respectively of pure metal was used.

Table 6
Summary of the survey results for 3 PE-based categories

Average PE	TPin [mg/L]	TPout [mg/L]	P removal	P load removed [Mg]	Fe/P load removed	Al/P load removed	Pure Fe & Al used [Mg]	Pure metal dose [g/m^3]	n
> 100 000	11,48	0,46	95,5%	179,21	0,27	0,02	45,60	2,88	39
100 000 - 15 000	12,82	0,62	94,0%	19,45	0,28	0,03	4,85	3,21	74
< 15 000	17,39	1,14	91,5%	6,00	0,22	0,06	1,24	4,17	18

Following the above results, a statistical analysis based on Pearson's correlation coefficient was conducted including the pure metal consumption per P load removed and the pure metal consumption per treated wastewater volume as shown in Fig. 4.

A statistically significant correlation at a significance level (α) = 0,05 was observed between the analyzed parameters in all 3 WWTPs categories with Pearson's correlation coefficient (r) = 0,915 for large WWTPs, 0,826 for medium WWTPs and 0,739 for small WWTPs. The above results can be interpreted as follows: in large WWTPs coagulants are used more rationally and most probably their dose is lower due to the higher biological P removal processes efficiency than in medium and small WWTPs.

Discussion

It is a well-known fact that coagulant dosing is depending on various factors including the capacity of the biological P removal process, effluent quality standards, wastewater pH other technical factors e.g. adding coagulant to ensure that the sewage sludge is free of filamentous bacteria. Moreover, economic aspects are critical for WWTPs operators (especially for private companies responsible for ensuring the effluent parameters are in line with the binding legal regulations). According to Jiang et al. (2004) the unit costs of P removal are connected with the desired final effluent P concentration and the capacity (size) of a WWTPs. It should be noted that these values are out of date, but the proportions between the individual TP levels remain at a similar level. P removal costs based on the above-mentioned study followed by an estimation including the inflation rates between 2004 and 2020 in the United States were presented in Fig. 5.

While, values of final P concentration between 2,0 and 1,0 mg/L are achievable in most conventional activated sludge bioreactors, limiting P below 0,5 mg/L requires advanced EBNR technology use and concentration below 0,13 and 0,05 mg/L are available only in complex biochemical systems with tertiary treatment, final adsorption or post-filtration (Jiang et al. 2004). Therefore, with the rising P elimination level the treatment cost increase similarly to an exponential function (Neverova-Dziopak 2018).

The costs of using a certain coagulant are also different regarding Fe and Al based products. According to (Lema and Suarez Martinez 2017) the average cost of PIX is 250–350 (EUR/Mg) while for PAX it is about 175–200 (EUR/Mg) what results in a unit cost of 0,007 – 0,021 and 0,003 – 0,010 EUR/m^3 of treated wastewater respectively. The selection of coagulant is also dependent on the wastewater pH where PIX is mainly used if the pH is between 3,5–7,0 and over 8,5 and PAX is applied when pH is from 4,0–7,0 (Wang et al. 1996).

It should be noticed that the location of coagulant dose is also an important factor that affects the final P bioavailability. Chemically enhanced primary treatment (CEPT), also known as pre-coagulation is increasing in popularity due to the carbon redirection opportunities and limited metal transfer to excess sewage sludge (Shewa et al. 2020). In 4 of the analyzed WWTPs CEPT process was applied, and therefore in Lublin-Hajdów (600 000 PE), Elbląg (141 500 PE), Brwinów (190 000 PE) and Zamość (250 000 PE) WWTPs, where 569,47 Mg of P per year is removed P recovery with crystallization-based methods using the sludge from their secondary settling tanks, might be used, while it should have a lower content of metal-P complexes (Dong et al. 2019). However, the economic reasonability of this solution needs to be investigated based on the actual P load removed in the excess sludge.

As for biological P removal without chemical support, a COD/P ratio > 50 is needed to provide efficient dephosphatation and final P concentration < 2 mg/L (Miksch and Sikora 2012). However, in Polish conditions regarding municipal wastewater the COD/P ratio is rather high with an average COD/P = 83,38 from all of the analyzed WWTPs and specifically, COD/P in large, medium and small WWTPs was 88,94; 81,08; 73,48 on average respectively. In only 9 cases of the analyzed WWTPs this ratio was < 50 and it mainly occurred in small plants where 2 mg/L or above was the limit for TP. The only exception was Sosnowiec-Radocha II WWTP (441 500 PE) where COD/P = 47,37 and approx. 72, 96 Mg of PIX113 was applied (pure metal dose of 0,5 g/m^3).

To obtain high-value fertilizers which contain plant bioavailable phosphorus compounds such as struvite or Ca-P the wastewater treatment process should be treated without our with minimum coagulant addition e.g. by the EBNR technology (Nättorp et al. 2018). The ENBR methods are not widely applied due to some barriers such as stringent effluent regulations, investment costs and more complex operation processes compering to conventional activated sludge systems with chemical P precipitation (Wilfert et al. 2015). The recovery rate is relatively low while only 10% to a maximum 50% of all influent P load can be recovered (Egle et al. 2016) which is often limited by high ammonia demand (Xue and Huang 2007).

As it has been shown in this paper, in Poland a relatively large share of municipal WWTPs is based on EBNR technology (20,6% PE of the examined plants) resulting in a high potential for P recovery using crystallization methods (3 639 601 PE out of 17 630 500 PE included in the analysis). To compare with other countries, the following share in PE is accounted for chemical P removal or biochemical integrated methods: The Netherlands – 83%, France – 83%, Germany – 92%, United Kingdom – 95,3%, Sweden 99,8% (Korving et al. 2019).

In Poland, mainly Fe salts are used (86%) for P precipitation similarly to other countries such as Germany – 77%, France and the United Kingdom – 88% (Paul et al. 2001) due to lower costs and higher environmental concerns related to Al toxicity, however in Japan Al salts are more common (Ohtake and Tsuneda 2018).

Finally, it should be noted that chemical P removal is the main reason for Fe or Al content in the sludge but there are other reasons for their presence in wastewater such as: adding Fe for controlling sulfide concentration in the sewage system (Ganigue et al. 2011) or Al to eliminate bacteria in the sludge (Zhang et al. 2019), groundwater infiltration with high Fe content into the sewage system (Zhang et al. 2020), and due to typical Fe content in human excrete (Korving et al. 2019).

A separate issue that was not covered by this study is related to the limitations of P recovery in WWTPs caused by the possible presence of toxic heavy metals such as cadmium (Cd), lead (Pb), mercury (Hg), nickel (Ni), chromium (Cr), copper (Cu), zinc (Zn) and arsenic (As) in wastewater and sewage sludge. This problem is currently considered so urgent that limiting the sources of heavy metals was indicated as one of the main goals of the European Green Deal strategy aiming to achieve a toxic-free environment.

Conclusions

The results presented in the study based on the 131 analyzed WWTPs serving a total of 17 630 500 PE indicate great possibilities for P extraction from wastewater in Poland using struvite or Ca-P precipitation. The relatively large share of entirely biological-based WWTPs (3 639 601 PE) in Poland combined with WWTPs using minimal chemical support in form of $> 1 g/m^3$ of pure metal (1 878 094 PE) provides a sum of approx. 5,5 million PE with an annual P load of 2 960 Mg as a base for P recovery methods based on struvite, Ca-P precipitation or similar products. Additionally, 4 WWTPs (1 181 500 PE in total) using CEPT could be considered for applying partly P recovery from the secondary sewage sludge. However, no clear policy targets to promote P recovery and the current very low development level of similar systems in Poland set a difficult barrier to pass. On the other hand, rising environmental awareness of various stakeholders and increasing interest in P recovery methods among WWTPs operators gives hope that in the coming years the removed pollutants will become an asset for the economy and Polish WWTPs will be transformed into resource and energy recovery plants.

Declarations

Funding declaration

No funding was received for this study

Conflicts of interest/Competing interests declaration

No conflicts were identified

Author contributions

M.P. collected the data and developed research methods. M.P. and M.S. wrote the main manuscript text. M.P. prepared all figures and tables. M.P. and M.S. reviewed the manuscript.

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Figures

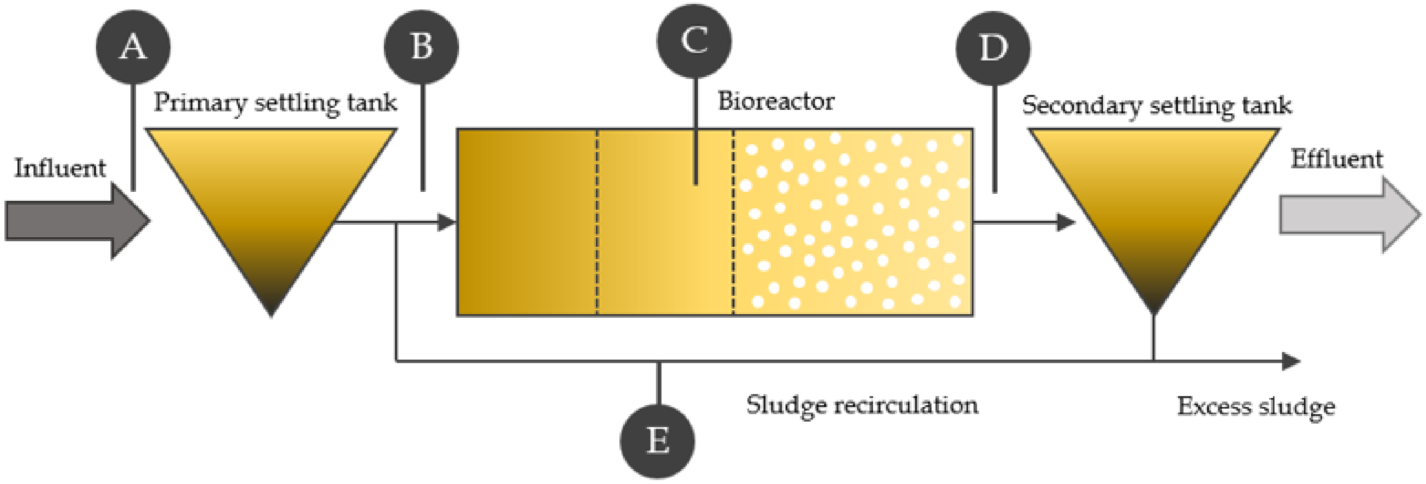

Figure 1

Location of the coagulant dosing point in the WWTPs technological line

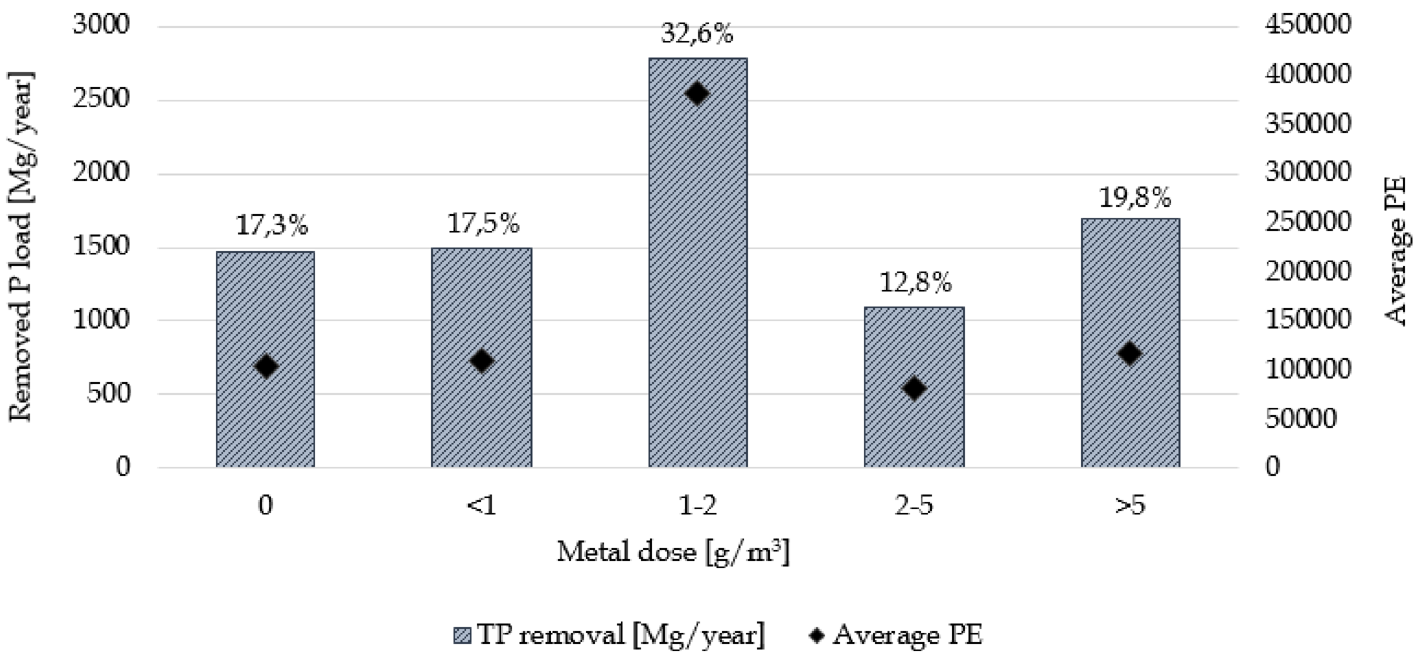

Figure 2

Relation between pure metal dose and removed P load

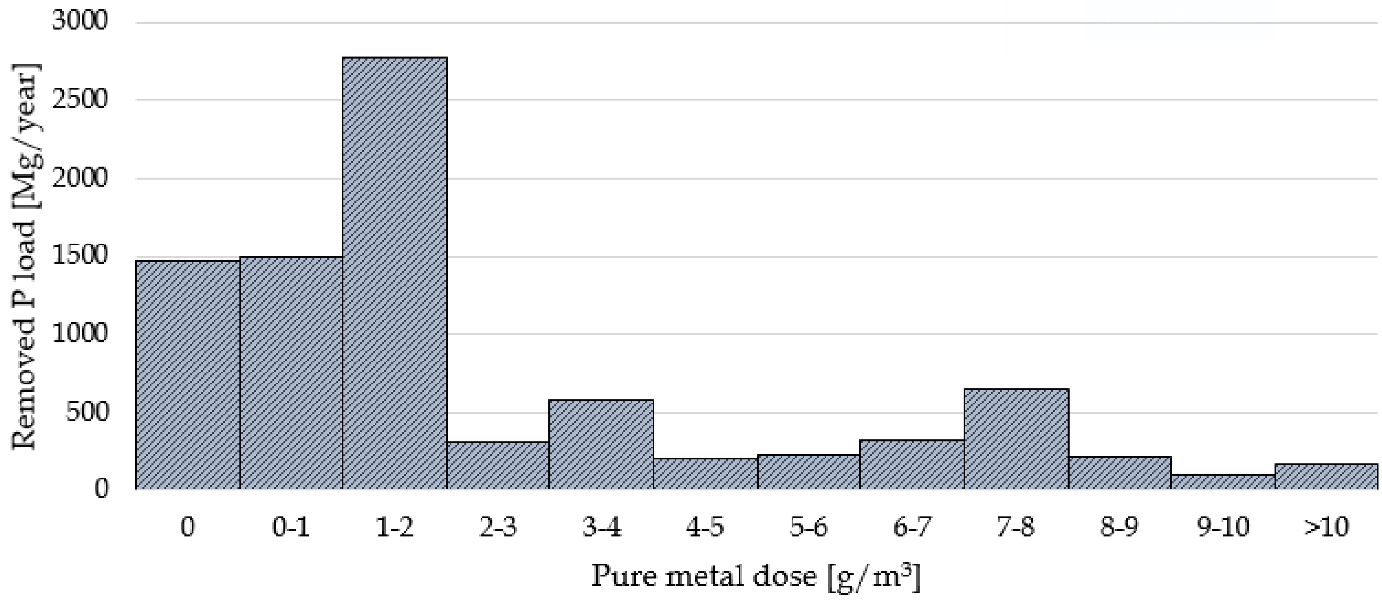

Figure 3

Pure metal dose and removed P load in the analyzed WWTPs

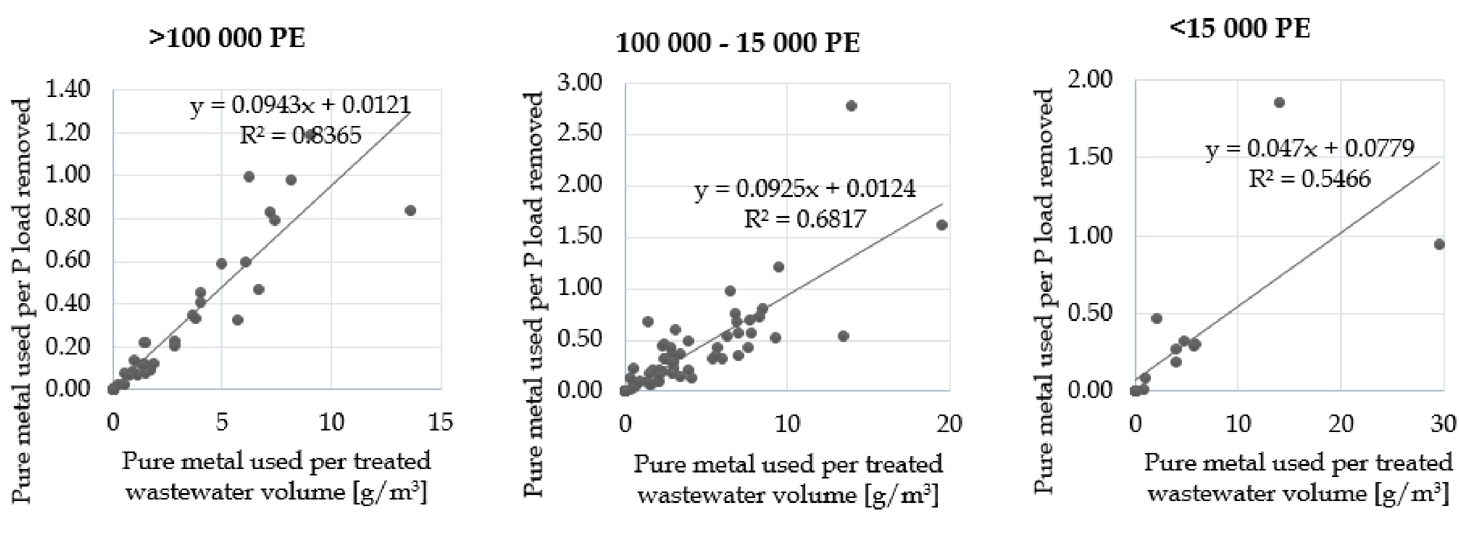

Figure 4

Scatter plots of the relationships between the pure metal used per P load removed and pure metal used per a treated wastewater volume

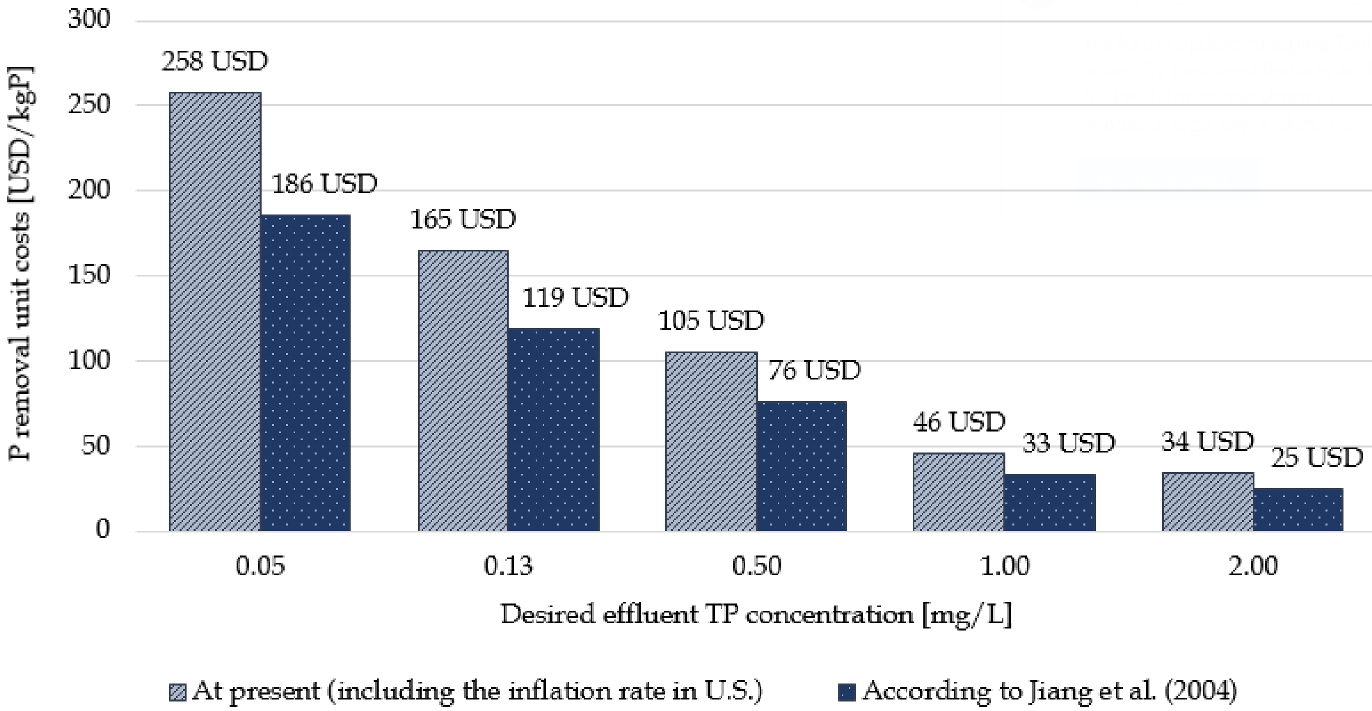

Figure 5

Estimated unit costs of P removal for a 75 000 m^3/day flow rate WWTP (Own figure based on Jiang et al. 2004)