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Factors Affecting Performance of Municipal Wastewater Treatment Plants

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Declaration:

I hereby declare that this doctoral thesis, my original investigation and achievement, which is being submitted for doctoral degree at Tallinn University of Technology, has not been submitted for any academic degree.

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VALLO KÕRGMAA



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List of publictions

The list of author's publications, on the basis of which the thesis has been prepared. The academic publications are referred in the text as Paper I, Paper II and Paper III:

- Vallo Kõrgmaa; Taavo Tenno; Aimar Kiviruut; Mait Kriipsalu; Mihkel Gross; Priit Tamm; Kristjan Karabelnik; Harri Terase; Vahur Vark; Natalja Lepik; Karin Pachel; Arvo lital (2019). A novel method for rapid assessment of the performance and complexity of small wastewater treatment plants. *Proceedings of the Estonian Academy of Sciences*, 68 (1), 32–42. DOI: 10.3176/proc.2019.1.03.ETIS classification 1.1
- II Vallo Kõrgmaa.; Mait Kriipsalu; Taavo Tenno; Erki Lember; Argo Kuusik; Vallo Lemmiksoo; Karin Pachel; Arvo Lital (2019). Factors affecting SVI in small scale WWTPs. Water Science and Technology, 79 (9), 1766–1776. DOI: 10.2166/wst.2019.177. ETIS classification 1.1
- III Vallo Kõrgmaa, Mailis Laht, Riin Rebane, Erki Lember, Karin Pachel, Mait Kriipsalu, Taavo Tenno, Arvo lital (2020). Removal of hazardous substances in municipal wastewater treatment plants. Water Science and Technology, 81 (9), 2011-2022. DOI: 10.2166/wst.2020.264. ETIS classification 1.1

Author's contribution to the publications

<u>Paper I:</u> Author developed the original idea, coordinated the development of methodology, collected data, analysed results and wrote the paper.

<u>Paper II:</u> Author developed the original idea, analysed the data and wrote the paper. The results were presented by the author at 15th International Specialised Conferences on Small Water and Wastewater Systems in Haifa, Israel, in 2018.

<u>Paper III:</u> Author designed the study, collected data, analysed data and wrote the paper. The results were presented by the author at SETAC Europe 29th Annual Meeting in Helsinki, Finland, in 2019.

Introduction

In 2015 United Nations called for global action in order to achieve a better and more sustainable future for the whole world. One of the Sustainable Development Goals addressed worldwide (SDG 6) was to ensure safe water sources and sanitation for all (United Nations, 2020). There are several objectives set in the European Union level to reduce the water pollution and to ensure public health. Anthropogenic pollution poses a threat to the aquatic environment and to the human health. Wastewater treatment plants are the last defensive line between aquatic environment and emissions of pollutants.

While the wastewater treatment aims to reduce the concentration of pollutants in the effluent, the process itself is rarely studied as a whole. There are numerous studies available about removal efficiencies of certain substances during the wastewater treatment process or its issues (e.g. activated sludge bulking), but all these studies are lacking relevant background information and often are difficult to compare due to the differences in the scale or technologies that have been used.

Regardless of variations between wastewater treatment plants (WWTPs), the performance of WWTP depends on several technical and non-technical factors that can all fail in one time or another. A wastewater treatment plant can produce good quality effluent even if there are several shortcomings. According to the survey in US municipal WWTPs the average facility had 15 performance limiting factors and at no facility was one single factor observed to be limiting the performance (Hegg *et al.*, 1979). Some of the factors triggering issues in WWTP are not easily traced (e.g., low dissolved oxygen concentrations could be the result of sudden load of readily biodegradable organic matter as well as lack of aeration capacity or poor maintenance of aeration system or combination of said factors), but good design and construction quality as well as professional operation and management (O&M) practice can minimize its impact to the effluent quality. The competence of operator has been outlined as one of the key factors for successful plant control (Hegg *et al.* 1979; Muga & Michelic, 2008; Olsson, 2012).

There are many research articles available that focus on certain aspects of wastewater treatment process; however, several scientists have emphasized that implementation of universal performance assessment method in different WWTPs is not possible due to the variations between wastewater treatment plants (different technology and operational conditions). Chen *et al.* (2015) Hao *et al.* (2013) have proposed some universal models like treatment performance index that focus primarily on pollutant removal efficiencies in different treatment steps. Even if the operational conditions are included in these kinds of assessments, the variance of factors is still not taken into account.

The main objective of this study was to examine the wastewater treatment process as a whole system in order to understand the relationships between the influent characteristics, technological complexity, operation and maintenance practices and overall performance of the WWTP in order to achieve satisfactory effluent quality. During this study as many as 246 WWTPs were studied during 2014–2018 and evaluated according to the novel method for rapid assessment of performance and the complexity of WWTPs. The method creates a comparable system of ratings that can be used for different technologies and wide variety of loadings.

Abbreviations

AMPA	α-amino-3-hydroxy-5-methyl-4-isoxazolepropionic acid
ANOVA	Analysis of variance
AX	Anoxic reactor
BOD ₇	Biological oxygen demand (7-days)
ССР	Critical control points
COD	Chemical oxygen demand
CSTR	Continuous stirred-tank reactors
D&C	Design and construction
DEHP	Di-2-ethylhexylphthalate
DO	Dissolved oxygen
f _{AN}	Anaerobic fraction
f _{AX}	Anoxic fraction
F _{crit}	F critical value
fox	Aerobic fraction
HACCP	Hazard analysis and critical control points
HRT	Hydraulic retention time
HRT _{AN}	Hydraulic retention time in anaerobic reactor
HRT _{AX}	Hydraulic retention time in anoxic reactor
HRT _{ox}	Hydraulic retention time in aeration basin
ICA	Instrumentation, Control and Analysis
IVE	Index of effluent violations
KN	Kjeldhal nitrogen
LOQ	Limit of quantification
MLSS	Mixed liquor suspended solids
MWWTP	Municipal wastewater treatment plant
n	The number of events
O&M	Operation and maintenance
ОХ	Aeration basin
PE	Population equivalent
Pi	The harmonized assessment of performance in the treatment step
	on the scale of 10 p
RAS	Returned activated sludge
SBR	Sequencing batch reactor
SRT	Solids retention time
SVI	Sludge volume index
TEC	Total evaluation of complexity
TEP	Total evaluation of performance
ТР	Total phosphorous
TSS	Total suspended solids

V ₃₀	30-minute settling test
WAS	Waste activated sludge
WWTP	Wastewater treatment plant
Xi	The score on the CCP (between 0 and 1)
Xi	A result of effluent analyses for the component i
Уi	An effluent quality standard for the component i

Terms used in the thesis

General complexity	Index describing the situation, where complexity of each individual treatment step is not taken into the consideration, but the number of treatment steps in the specific WWTP is divided by all possible treatment steps which were described in model. General complexity defines all treatment steps that are to be evaluated for performance and complexity.
Index of effluent violations (IVE)	Index describing of compliance with effluent quality standards that are regulated by plant's discharge consent
Sludge volume index (SVI)	Index describing settling characteristics of activated sludge in the aeration basin in activated sludge process
Total evaluation of complexity (TEC)	Index describing how many different treatment steps are involved in the wastewater treatment process and how sophisticated the technology was.
Total evaluation of performance (TEP)	Index describing all the factors that can influence the wastewater treatment process (e.g. quantity and composition of the influent, difference between design values and actual conditions, functionality of equipment, operational problems).

1 Background

The wastewater treatment aims to reduce the concentration of pollutants in the effluent in order to eliminate the threat to the environment. While source reduction of pollutants should be encouraged (EC, 2000), wastewater treatment by physical, chemical and biological means remains to be necessary in order to reduce the negative impact of contaminants to the aquatic environment (Henze *et al.*, 2011).

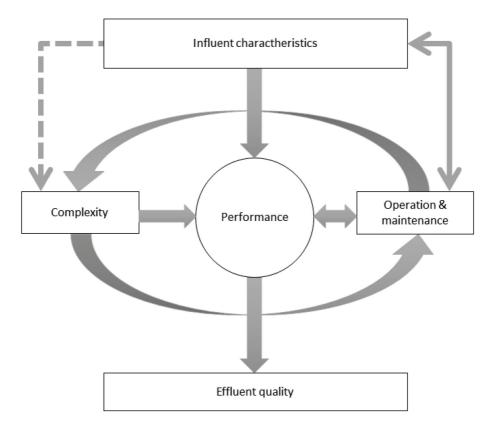


Figure 1. Factors affecting effluent quality in wastewater treatment plant.

Wastewater treatment is a complex process (Fig. 1), where the product (effluent) quality is strongly dependent on the variations in feed (influent) characteristics, complexity of the technology, maintenance procedures and operational decisions. Designers can model the process parameters in wastewater treatment plant (WWTP) in detail based on characteristics of influent or even on bases of metabolic reactions of specific group of microorganisms (Henze *et al.*, 2011).

Regardless of a wide variety of wastewater treatment technologies (e.g. activated sludge, biofilm or constructed wetlands), there are several factors that can reduce the performance of WWTP. The real-life situation on the MWWTP can differ significantly what was expected due to incorrect design input, inevitably changing influent characteristics and their uncertainty (e.g., an exact composition of wastewater is usually unknown), changes in the ecosystem in the biological treatment units, equipment failure or maintenance undoing (Glymph, 2005; Hao *et al.*, 2013; Hegg *et al.*, 1979; Olsson, 2012). Irregularities or even failures in one phase can easily affect the outcome of the

whole system on the negative way. On the other hand, cognizant process control can be able to produce the desired outcome even despite of the equipment failure or irregularities in the influent characteristics. One could argument that process control in a wastewater treatment plant is a "synthetic problem" as defined by Baybrooke and Lindblom (1963, via Turnbull & Hoppe, 2019) – "most of the time a problematic situation is a cluster of interlocked problems with interdependent solutions, turning 'problem solving' into a continuous, complex adjustment of interests process with no definitive answers".

1.1 Issues with performance evaluation

It is difficult to compare fundamentally different treatment systems like small scale manually operated wastewater treatment plant based on biofilm process and a large-scale fully automated sequencing batch reactor (SBR). Biological processes in different MWWTPs could be designed on fundamentally different bases. For example, substrate availability in biofilm processes can be considered to be diffusion-dependent, whereas floc diffusion limitation in activated sludge process is not considered to be relevant (Henze et al., 2011). Still, both of these solutions contain similar design elements like pre-treatment (e.g. sieves), biological treatment, sedimentation and sludge handling. The purpose of using sieves is to remove particles and objects from a flow of wastewater (ISO, 2014) and it does not depend as much on the following process as on quantity and quality of the influent, yet the configuration of used equipment may vary from hand-operated to multi-step and fully automated. The sedimentation is a process of settling and deposition of suspended matter under the influence of gravity (ISO, 2014), but regardless if it is performed in the same reactor (SBR) or in a separate unit (e.g. final clarifier) - the principle of the process remains same. Therefore, the evaluation of the overall efficiency of WWTP constitutes a multi-objective decision chain (Hao et al., 2013).

Hao et al., 2013 and Chen et al., 2015 claimed that implementation of method for comparative evaluation of overall treatment performance of different WWTPs is not practicable due to varying nature of treatment processes used, and operational conditions applied. Several evaluation models and indexes have been proposed, but these models have a dissimilar objective. Sometimes models are focused on assessment of environmental and/or economic benefits, e.g. these models are adaptations of life cycle assessments (Fang et al., 2013; De Faria et al., 2015); sometimes they rely on selection of treatment processes on bases of fuzzy analytical hierarchy process method (Karimi et al., 2011); or multi-criteria analysis (Jozwiakowski et al., 2015). According to Pomies et al. (2013) there are at least 18 models describing micropollutant removal in activated sludge processes available. Chen et al. (2015) has proposed a more general assessment method which is based on treatment performance index; and Hao et al. (2013) has proposed similar method which focuses primarily on achieving pollutant removal efficiency along the treatment steps. A considerable advantage in using these models is that a set of certain operational conditions have been incorporated in the assessment, but an aggregate of investigated factors is still limited. So far, these both methods have been applied only in few cases and only in large scale WWTPs. No studies related to small or medium scale treatment plants were found.

1.2 Factors limiting performance

According to the survey in US municipal WWTPs the average facility had 15 performance limiting factors (e.g. infiltration, process controllability, equipment accessibility for maintenance) and at no facility was one single factor observed to be limiting the performance (Hegg *et al.*, 1979). In general terms, these factors could be divided into groups either by their origin (influent characteristics) or by the impact to the process (complexity of WWTP, the role of an operator). There is a sophisticated relationship between influent characteristics, plant complexity (e.g., reactors shape and working mode) operational parameters in the WWTP and its microbiological population (Henze *et al.*, 2011; Jenkins *et al.*, 2004). Shortcomings in process design and construction phase can have a major impact to the overall performance of biological treatment.

1.2.1 Influent characteristics

The quality and quantity of wastewater is determined by population and industrial activities in the wastewater collection area, the design and condition of the sewer system, but also by sources of wastewater generated internally in the treatment plant (e.g. reject water from sludge dewatering) (Henze *et al.*, 2011). Prasse *et al.* (2015) outlined that besides demographics on the sewer collection area and the number of facilities (e.g. hospitals, laundry services) that use specific substances, the proportion and composition of co-treated industrial wastewater has to be considered having a great impact to certain substances in the influent.

The main characteristics in the influent that dominate the process outcome are (but are not limited to):

- Fluctuations in the flowrate. These could be happening within the day (e.g., people are using more water while taking shower or cooking, industrial discharges are dependent on the production processes) or to be seasonal (e.g. infiltration to the sewer due to the stormwater or snow melting). Fluctuations in the flowrate can cause serious hydraulic overloading issues to the WWTP (Kõrgmaa et al., 2016);
- Load of nutrients to the WWTP. Nutrients are needed in order to keep biological wastewater treatment processes running, but both lack and an abundance of certain nutrients can cause serious changes in the ecosystem in the biological treatment process;
- Hazardous and inhibitory substances in the influent that can cause serious issues in the biological treatment process or failures in meeting effluent quality standards;
- Other factors influencing the successful treatment outcome (e.g., soluble gases, temperature).

1.2.2 The complexity of WWTP

The complexity of WWTP describes the quantity of treatment steps involved in the wastewater treatment process and the level of sophistication of the technology. An addition of more complex treatment steps (e.g., filtration) to the existing WWTP can help to reduce aquatic emissions of hazardous substances with the effluent (Clara et al., 2012) and to increase removal efficiencies of other pollutants. Since nutrient removal has become more common over the years, the complexity of wastewater treatment plants has increased (Olsson & Newell, 2001). This in turn has increased the demands on a skilled workforce and caused new types of issues. For example, activated sludge bulking is one of the most common problems in Estonian WWTPs. In many cases the event of

bulking could be prevented by choosing appropriate solution for system or reactor design (Eikelboom, 2000; Jenkins *et al.*, 2004), but this in turn could stimulate the dominance of other unwanted microorganisms. *Microthrix parvicella* is the most common filamentous bacteria causing bulking in activated sludge processes in Estonia. Although these bacteria cannot simply be controlled by addition of a selector (e.g., anaerobic or anoxic reactors), the negative effect caused by *Microthrix parvicella* can be reduced by avoiding foam traps and by addition of proper pre-treatment (Jenkins *et al.*, 2004). On the other hand, according to Fan *et al.*, (2017) *Microthrix parvicella* favoured alteration between anaerobic and aerobic conditions, i.e. increased system complexity needed for biological phosphorous removal could in turn evoke settling problems in the final clarifier.

1.2.3 The role of an operator

The importance of human factor in wastewater treatment process has been described very briefly in literature (Hegg *et al.*, 1979; Olsson, 2012) and in numerous cases it is considered to be one of the main reasons for poor process performance (Hegg *et al.* 1979, Kõrgmaa *et al.*, 2016). Hegg *et al.* (1979) listed improper operator application of concepts and testing to process control as well as inadequate understanding of the wastewater treatment process as two highest ranking factors contributing to poor plant performance. The competence of operator has been outlined as one of the key factors for successful plant control by several authors (Hegg *et al.* 1979; Muga & Michelic, 2008; Olsson, 2012).

Besides competence there are other constraints that could affect the decision-making process needed for successful process control. For example, financial limitations in small WWTPs often reduce the capability to perform daily analysis of the wastewater quality or measure Mixed Liquor Suspended Solids (MLSS) for process control. The determination of 30-minute settling test (V₃₀) is the most commonly used operational parameter for activated sludge (AS) process control in Estonia (Kõrgmaa et al., 2019). Unfortunately, bulking has a negative impact to the activated sludge settling properties and if MLSS is not analysed concurrently, decisions based on the settling test could lead to inadequate sludge wasting and cause deviation from optimal solids retention time (SRT). Deviations from optimal SRT can lose desired results from nitrification process or lead to expend electricity unnecessarily. The easiest way to detect bulking, is to determine Sludge Volume Index (SVI). SVI is calculated as a quotient between V₃₀ and TSS. According to Bitton (2005), high SVI values (SVI > 150 ml/g) can be associated with bulking sludge. The performance of an activated sludge process is often deteriorated due to sludge separation problems caused by sludge bulking (Jenkins et al., 2004; Guo et al. 2014) which in turn will result in the poor effluent quality.

1.3 Municipal wastewater treatment plants in Estonia

1.3.1 Investments to the MWWTPs

Investments to the wastewater treatment plants have been extensive during last 15 years in Estonia. The main sources for wastewater-related investments have been the Cohesion Fund of the European Union (115.5 M€ was invested between 2004 and 2014) and Estonian Environmental Investment Centre (EIC) (additional 16 M€) (EIC, 2018). About 49 M€ was invested into small scale WWTPs (less than 2 000 PEs) and 115 M€ was invested into 41 larger plants (Kõrgmaa *et al.*, 2016). Between 2004 and 2014 in total 288 WWTPs were constructed or re-constructed by using subsidies.

1.3.2 Overview of MWWTPs

Most of Estonia is sparsely populated. There are a lot of municipal WWTPs (n = 664) for 1.35 million people (Fig. 2). Most of MWWTPs are small (< 300 PE). 51.2 % of pollution load is treated in municipal WWTPs with capacity for more than 100 000 PEs.

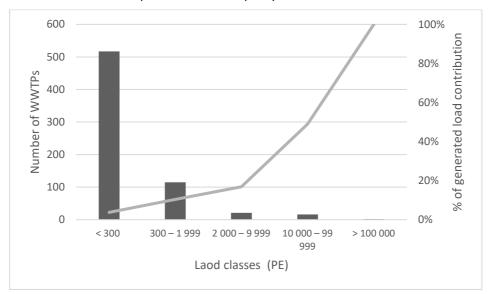


Figure 2. Number of WWTPs per load size class (blocks) and cumulative percentage of generated load contribution (in PE, marked as a line).

The variety of treatment technologies is also dependent of the plant's capacity. An activated sludge (AS) process has been used most (335 plug-flow and 62 sequencing batch reactors), followed by natural-based solutions (incl. 118 oxidation ponds, 22 constructed wetlands) and biofilm reactors (127 plants). The variety of technological solutions is wide in the small-scale (WWTPs < 2 000 PEs), but only activated sludge (plug-flow and SBR) solutions are used in the medium and large-scale municipal wastewater treatment plants (VEKA, 217).

1.3.3 Requirements for the effluent quality in Estonian MWWTPs

Effluent quality is the only parameter that is widely used for an evaluation of successful wastewater treatment. Treated effluents have to meet quality standards. Estonian regulation on effluent quality standards (Minister of Environment, 2019) sets limit values for nutrients for disposed effluent as seen in Table 1. Effluent quality standards for nitrogen and phosphorous in the small-scale (< 300 PE) has in turn caused a situation that complexity of Estonian MWWTPs is rather high compared to similar size MWWTPs in countries where there is no nitrogen or phosphorous removal obligations. In 2014, 45% of Estonian WWTPs were not capable of meeting environmental requirements according to the national monitoring program (Allas, 2014). Reasons of poor performance remained unclear.

Table 1.Effluent quality standards for nutrients according to the WTTPs' load (Minister of Environment, 2019).

PARAMETER	UNIT	< 300 PE	300 –	2 000 –	10 000 -	> 100 000
			1 999 PE	9 999 PE	99 999 PE	PE
BOD ₇	$gO_2 m^{-3}$	40	25	15	15	15
COD	$gO_2 m^{-3}$	150	125	125	125	125
TSS	g m ⁻³	35	35	25	15	15
TN	gN m ⁻³	-	60	45	15	10
TP	gP m ⁻³	-	2	1	0.5	0.5

Although quality standards for hazardous substances in the effluent have been set by the Minister of Environment (2019; see also Appendix I in Paper III), these are rather rarely regulated by water permits and monitored. Serious data gaps exist in the emission estimations. Paper III aimed to increase the knowledge on the occurrence and fate of hazardous substances relevant from the aquatic environment protection perspective (directive 2000/60/EC) in municipal wastewater treatment plants. The studied substances belonged to wide range of chemical groups, e.g. phthalates, pesticides, halogenated flame retardants and volatile compounds.

2 Aims and objectives of the study

The aim of this doctoral study was to examine the wastewater treatment process as a whole system to map the relationships between the influent characteristics, technological complexity, operation and maintenance practices and overall performance of the WWTP in order to achieve satisfactory effluent quality. Previous studies have focused only on certain aspects of wastewater treatment process (e.g., removal efficiencies of certain contaminants, biological issues in activated sludge process), but neglecting certain relevant factors (e.g., operational practices, situation in the plant) simultaneously. Several authors have highlighted that comparative evaluation of overall treatment performance of WWTPs is overly complicated due to varying nature of used treatment processes and applied operational conditions.

The objectives of this doctoral thesis were:

- To develop a universal performance assessment method for all individual wastewater treatment solutions to analyse multiple factors (e.g., influent characteristics, system complexity, operational practices and an accordance between design and real-life process parameters) simultaneously. The method also takes into consideration their impact to the overall system performance and effluent quality. (Paper I).
- To analyse connections between certain issues in the plant (Paper II) and their impact to the activated sludge process while demonstrating the use of novel methodology.
- To analyse the links between removal efficiencies of hazardous substances in the municipal wastewater treatment plants, operators' competency and technological complexity (Paper III).

3 Methodology

3.1 Method for evaluating performance and complexity of WWTPs (Paper I-III)

Biological wastewater treatment is like the certain production processes in food industry – substrate is transformed to the product by biological processes. In that context hazard analysis and critical control points (HACCP) principles used in the food industry could be transmitted to the wastewater treatment processes. The assessment of treatment performance of WWTPs was based on the performance of individual treatment phases. All individual treatment steps were evaluated separately due to the large variability in technologies used as well as different environmental objectives set for the WWTPs. A list of prerequisites regarding e.g. treatment processes and equipment were set to ensure comparability of individual WWTPs (Paper I).

For data collection a questionnaire (Paper I) that included 5 main treatment steps and 21 subcategories was developed to assess WWTPs *in-situ*. A minimum of four critical control points (CCPs) was set for each treatment step. Depending on the complexity of the wastewater treatment process the number of evaluated CCPs ranged from 37 (two treatment steps – septic tank and oxidation ponds) to 170 (9 treatment steps where conventional activated sludge process combined with SBR was used for wastewater treatment and on-site stabilization of sewage sludge was applied). To overcome the problem of assessment subjectivity, all the CCPs were formulated as multiple-choice questions with 2 to 5 alternative answers, but mainly as "yes" or "no".

During the assessment of WWTPs the following parameters were evaluated on a scale of 10 points (Paper I):

- General complexity
- Total evaluation of complexity (TEC)
- Total evaluation of performance (TEP)

Performance and complexity in each treatment step was evaluated based on the CCPs in the same step. CCPs were defined as factors that a) influence performance of the treatment step, b) describe operational conditions or c) the complexity of the treatment step. A minimum of two CCPs were defined for each wastewater treatment step. The list of CCPs were chosen by expert opinion and were based on suggestions in the literature (Baumann *et al.*, 2012; Noorvee *et al.*, 2007; Kuusik *et al.*, 2001; Jenkins *et al.*, 2004; Maastik *et al.*, 2011).

The performance or complexity of a specific treatment step (Pi) was calculated as:

$$P_{i} = \frac{\sum_{i=1}^{n} X_{i} \times Y_{i}}{\sum_{i=1}^{n} Y_{i}} \times 10,$$
 [1]

where P_i is the harmonized assessment of the treatment step on the scale of 10 p, X_i is the score on the CCP (between 0 and 1) and Y_i is the importance of the CCP in the treatment step based on expert opinion.

The total evaluations of performance (TEP) and complexity (TEC) were calculated on from the weighted scores of the different stages as follows:

$$TEP = \left[\sum_{i=1}^{n} a_i + \frac{\sum_{i=1}^{n} \left(\frac{P_i}{10} \times b_i \right)}{\sum_{i=1}^{n} b_i} \right] \times 10,$$
 [2]

where TEP is harmonized performance assessment of the total treatment process on the scale of 10 points, a_i addresses some general aspects for overall process control, P_i is harmonized performance assessment of the treatment step on the scale of 10 p and b_i describes the expert opinion on the importance of the named treatment step in the whole wastewater treatment process.

Similar calculations were made for TEC (Paper I).

As an assessment of the impact of TEP and TEC to the effluent quality can be problematic due to the varying quality requirements depending on the size of WWTP, the index of effluent violations (IVE) on the 10p scale was developed. The following prerequisites were set to ensure comparability of WWTPs: a) IVE = 10 for the effluents performing < 25% better than required by quality standards, b) the IVE = 0 p for the effluents that exceeded quality standards by more than 300%,

IVE was calculated as:

$$IVE = \frac{\sum_{i=1}^{n} \left(\frac{x_i}{y_i} \times 8\right)}{n},$$
 [3]

where x_i is a result of effluent analyses for the component i, y_i is an effluent quality standard for the component i, n is the number of analysed components.

3.2 Data collection, sampling and laboratory analyses

Sampling campaigns among with data collection were carried out during two different periods (see below).

3.2.1 Data collection for performance evaluation (Papers I and II)

Selected wastewater treatment plants (Fig. 3) were assessed according to a uniform method from October 2014 to March 2015. All together 541 grab samples were collected, including 241 samples from the effluent of the secondary treatment, 137 from the effluent of tertiary treatment units and 163 samples for determination of mixed liquor suspended solids (MLSS). For two WWTPs analysis results from a national monitoring program were used. Average wastewater temperature in studied WWTPs was $8.9 \pm 3.3~^{\circ}\text{C}$.

The study included collection of the design parameters (e.g. flow rate, SRT, sludge volume index), a documentation of the actual situation on the plant (e.g. flow rate, SRT, sludge volume index, equipment failures) and sampling relevant parameters in the effluent and in the process reactors. Operator's competence was evaluated in the form of hidden test. The content of biological oxygen demand (BOD₇), chemical oxygen demand (COD), total suspended solids (TSS), Kjeldahl nitrogen (KN) and total phosphorous (TP) were analysed in laboratory and the additional parameters pH, conductivity (K), dissolved oxygen (DO) and water temperature (T°) were determined in situ.

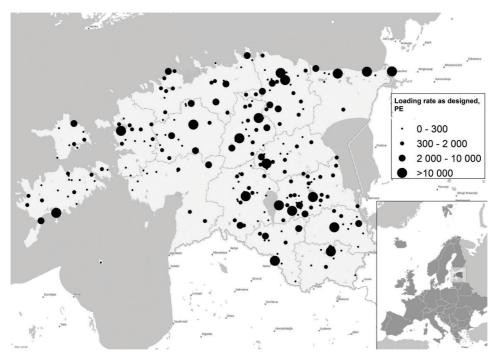


Figure 3. Location and size of evaluated wastewater treatment plants in Estonia.

3.2.2 Data collection for assessment of removal rates of hazardous substances in MWWTPs (Paper III)

This study involved nine MWWTPs (see Table 2) that were selected according to the following criteria: a) loading – this study covered 59.7 % of pollution load from MWWTPs in Estonia, b) treatment technology selection covers most widely used solutions in Estonia including small scale solutions (e.g. biofilm reactors, constructed wetlands) as well as an activated sludge process (continuous flow and sequencing batch reactors, SBR) and c) industrial load had to be less than 50% in order to minimize an impact of industrial wastewaters. Table 2 summarizes main properties of selected wastewater treatment plants.

Seasonal spot samples of influent and effluent water were collected between June 2017 and April 2018 from all MWWTPs according to ISO 5667-10. Samples of sewage sludge were collected according to ISO 5667-13. The flow-proportional composite sampling strategy for both influent and effluent of each MWWTP was not realistic, but from six MWWTPs time proportional composite samples were collected in parallel for certain analyses (PAH, pesticides, alkylphenols, heavy metals and phthalates) in accordance with ISO 5667-3 in order to increase sampling reliability. In total, 72 spot samples, 48 composite samples and 32 sewage sludge samples were collected and analysed for 282 substances. During the sampling event an evaluation of operators' competence and WWTP complexity was carried out similarly as described in chapter 3.2.1.

During the study 282 hazardous substances were analysed, including EU (n = 45) and Estonian (n = 31) priority substances. All analytical methods used in this study were done using accredited methods according to the standard ISO 17025.

Table 2. Simplified description of selected wastewater treatment plants.

WWTP	Capacity (PE)	Primary treatment	Biological treatment	N- remov	P- val removal	Effluent polishing	Comple- xity	Operators' competence
Α	< 300	Septic tank	Constructed wetland	No	No	No	3,3	0,6
В	300 – 1 999	Screen + septic tank	Oxidation pond	No	No	No	4,4	6,6
С	300 – 1 999	Screen + septic tank + grit separator	Biofilm	No	Chemical	Oxidation pond	4,9	0,8
D	2 000 – 9 999	Screen + grit separator	SBR	Yes	Biological + chemical	No	6,4	7,5
E	10 000 – 99 999	Screen + grit separator	SBR	Yes	Chemical	No	6,9	2,0
F	10 000 – 99 999	Screen + grit separator	Activated sludge/SBR	Yes	Biological + chemical	No	7,0	7,8
G	10 000 – 99 999	Screen + grit separator	Activated sludge	Yes	Biological + chemical	No	6,9	8,9
н	> 100 000	Screen + grit separator + primary clarifier	Activated sludge	Yes	Biological + chemical	Drum filter	6,2	8,2
I	> 100 000	Screen + grit separator + primary clarifier	Activated sludge	Yes	Chemical	Post- denitrifi- cation	5,5	8,4

Liquid chromatography (Agilent Technologies Infinity 1290) with tandem mass spectrometric and electrospray ionization (LC-ESI-MS/MS) (Agilent Technologies 6490 with JetStream ESI) was used for analysis of pesticides (including glyphosate), perfluorooctanesulfonate and its derivatives. Water samples were analysed without any sample preparation, solid samples were analysed after extraction procedures. Wastewater and sewage sludge pesticide samples were analysed with addition of OnlineSPE (Agilent Flexible Cube LC module).

Determination of selected organotin compounds was carried out with gas chromatograph and with tandem mass spectrometric analysis (Agilent Technologies 7890B/7000 system). Organotin compounds were alkylated with sodiumtetraethylborate, extracted with hexane and cleaned with silica column. The same was used for different organic substances such as pesticides, chlorobenzenes, PCB-s, PAH-s etc. that have been extracted from the samples either with liquid-liquid extraction or with solid phase extraction. Phthalates were analysed after solid phase extraction with GC/MS/MS according to EVS-EN ISO 18856 in case of water-phase and with CEN/TS 16183 in case of solid phase. Chlorophenols were analysed also with GC/MS/MS according to EVS-EN 12673 in case of water and according to ISO 14154 in case of soil. Extraction was carried out with n-hexane and in case of solid samples with mixture of acetone and n-hexane.

Determination of individual isomers of nonylphenol extracted from the samples was carried out Gas Chromatography/Mass Spectrometry (GC/MS) by Agilent Technologies

(7890B/5977A MSD). Water samples were analysed according to ISO 24293 and solid samples according to CEN/TS 16182. GC/MS was used for determination of benzene and some derivatives with headspace gas chromatographic method. Water samples were analysed according to ISO 11423-1 and solid samples according to ISO 22155.

Determination of hydrocarbon oil index was carried out with gas chromatography with flame ionization detector (Agilent Technologies 7890B). Water samples were prepared according to EVS-EN ISO 9377-2 and solid samples according to EVS-EN ISO 16703. Extraction was carried out with n-hexane and in case of solid samples with mixture of acetone and n-hexane.

Mercury was analysed according to EVS-EN ISO 12846 with Hg Analyzer (RA-915, Lumex). Other metals were analysed according to EVS-EN ISO 11885 with inductively coupled plasma optical emission spectrometry (Vista - MPX Varian).

3.3 Statistical analysis (Papers I-III)

Chemical analyses with measurements below the limit of quantification (LOQ) were substituted with LOQ/V2 prior to statistical analysis. There is a possibility that this censoring will create some bias (Hesel, 2005; Zeghnoun *et al.*, 2007), but according to Vriens *et al.* (2019) in statistical approaches for multipollutant studies the use of more advanced techniques of handling undetectable levels is not supported. United States Environmental Protection Agency (EPA, 2000) guideline states that if the rate of censoring is very high (greater than 50%) then focus should be put on upper quantile of the contaminant distribution, or on the proportion of measurements above a certain critical level that is at or above the censoring limit.

For studying the relationships between the performance, complexity, operators' competence and removal of substances, tools of correlation and regression analysis were applied. For categorical variables, the analysis of variances (ANOVA) was used for comparing the population means in different groups. p-values of 0.05 or lower were considered to indicate statistical significance, but in some occasions results with higher p-values were highlighted.

In order to find the frequency of co-existing problems in studied WWTPs matrix A was formed in a way that findings of performance limiting factors in each WWTP were marked as "1" (the problem was recorded) and "0" (the problem was not recorded). Matrix B was formed as a multiplication of matrices A and its transposed matrix A^T as follows:

$$B=A^{T}A$$
 [4]

Matrix B describes the co-existence of two problems. To find the frequency of co-existence of selected issues (matrix C) all values (b) in each column in the matrix B were divided by the total number of problems (x) that have been described in said column (e.g., SVI > 150 ml/g was observed in 28 % of WWTPs, where the design loading was not based on actual measurements, but measurements were absent in 40 % of WWTPs, where SVI > 150 ml/g was observed) as follows:

$$C = \begin{pmatrix} \frac{b_{11}}{x_1} & \dots & \frac{b_{1n}}{x_n} \\ \vdots & \ddots & \vdots \\ \frac{b_{n1}}{x_1} & \dots & \frac{b_{nn}}{x_n} \end{pmatrix}$$
 [5]

The analysis was carried out by the software programs R and OriginPro.

4 Results and discussion

4.1 Influent characteristics

Most of the problems in the WWTP start from with the public sewer system. According to Kõrgmaa *et al.* (2016) 40 % of WWTPs have issues with serious fluctuations in the wastewater flow or load and 50 WWTPs had issues caused by an industrial wastewater. Estonian municipal wastewater is also often unbalanced in the sense of nutrients – most wastewaters have high content of nitrogen and phosphorous but are lacking carbon sources.

There are several parameters in the influent (e.g., readily degradable COD, lack of nutrients, low temperature) that cause foaming and bulking events in activated sludge process (Eikelboom, 2000; Jenkins *et al.*, 2004). Although event of bulking can be easily determined either by either visual inspection or determination of SVI value, the microscopic examination of activated sludge is often needed to identify the micro-organism responsible for deteriorated settling properties. An identification of dominant filamentous micro-organism is needed in order to make targeted actions (e.g., modification of SRT, addition of nutrients) for reducing the impact of bulking and foaming.

4.1.1 Impact on the biological process (Paper II)

Infiltration to the sewer system had significant impact (p-value 0.03) to the SVI value. Average SVI value in WWTPs, that had infiltration, was 177.9 ml/g while in the plants that did not have any infiltration it was 133.1 ml/g. Industrial sources caused problems in 35 activated sludge plants, but the impact to the SVI value was less significant (p-value 0.07). WWTPs without industrial sources had average SVI value 139.9 ml/g, but industrial sources raised an average SVI value to 180.4 ml/g. An impact of low water temperature (an average wastewater temperature during the sampling session was 8.9 \pm 3.3 °C) could not be evaluated as there was no adjacent group available.

Table 3. One-way ANOVA showed significant (p-value < 0.05) difference in process control parameters and influent characteristics in WWTPs where Microthrix parvicella was found.

Parameter	Unit	With M. parvicella	Variance	Without Variance <i>M.</i> Variance <i>parvicella</i>		Mean square	F- value	P- value
SRT	d	39.10	352.96	14.56	155.91	1 852.96	6.59	0.03
F/M	kgBOD ₇ / kgMLSS	0.033	2.721E-04	0.107	0.005	0.017	8.10	0.02
Filament index	-	4.13	1.55	2.00	0.00	13.89	14.05	0.003
BOD ₇	mgO_2/I	350.00	5 314.29	576.00	13 480.00	157 156.92	18.97	0.001
COD	mgO_2/I	632.50	42 221.43	1 012.00	79 320.00	443 139.23	7.95	0.02
TSS	mg/l	263.00	3 609.14	433.20	23 171.20	89 132.43	8.31	0.01
BOD ₇ /N	-	4.15	1.41	6.07	2.37	11.33	6.44	0.03
BOD ₇ /P	-	28.71	104.46	47.57	147.07	1 094.74	9.13	0.01

It was not possible to analyse influent quality in all WWTPs due to the financial limitations, but 24h composite samples were collected from 15 AS plants. Microscopic analyses of activated sludge were performed in 13 WWTPs. Analyses showed that *Microthrix parvicella* was dominant in 8 WWTPs and three plants had foaming problems caused by *Nocardioforms*. In these 15 plants no correlation between SVI and influent

parameters was observed, but significant correlations were found with the presence of *Microthrix parvicella* (Table 3). One-way ANOVA showed that *M. parvicella* favoured long SRT and low F/M values as described earlier by Fan *et al.* (2018) and Jenkins *et al.* (2004). Table 3 shows that *M.parvicella* favoured wastewaters with lower BOD₇, COD and TSS content and lack of carbon content compared against nitrogen and phosphorous contents (n = 13 and $F_{critical}$ was 4.84 for all parameters).

4.1.2 Hazardous substances in the influent (Paper III)

From 282 substances analysed only 45 (1,2,3,4,6,7,8-H7CDF, 1,2,3,6,7,8-H6CDF, 1,2,3,7,8,9-H6CDD, 1,2,3,7,8,9-H6CDF, 1,2,3,4,7,8-H6CDF, 1,2,3,7,8-P5CDF, 1,2,3trichlorobenzene, 1,3,5-trichlorobenzene, 2,3,4,6,7,8-H6CDF, 2,3,4,6,7,8-H6CDF, 2,3,4,7,8-P5CDF, 2,3,4,7,8-P5CDF, 2,3,7,8-T4CDD, 2,3,7,8-T4CDD, 2,3,7,8-T4CDF, aclonifen, alachlor, aldine, alpha-endosulfan, atrazine, bifenox, deltahexachlorocyclohexane, dieldrin, dichlorophos, dimethoate, endrin, epsilonhexachlorocyclohexane, hexachlorobutadiene, isodrine, chlorofenvinphos, chlorpyrifos, metazachlor, PCB-114, PCB-123, PCB-126, PCB-156, PCB-157, PCB-169, PCB-189, PCB-77, PCB-81, pentachlorobenzene, simazine, trifluralin, cybutrine) were not detected above the limit of quantification (LOQ) from any of analysed samples.

Di-2-ethylhexylphthalate (DEHP), toluene and heavy metals (As, Ba, Ni, Pb, Zn) were detected above LOQ from all samples. 120 substances were found below LOQ in the influent. Ten most frequently found substances in the influent samples are in Table 4. For some of substances seasonal patterns (e.g. diuron, glyphosate, trichloromethane were found in the summer and autumn samples) or connections to population density (e.g. tetrachloroethene) was detected only from MWWTPs with capacity more than 10 000 PEs or median concentrations of trichloromethane increased linearly according to the plant size) could be detected.

Table 4. Substances found most frequently in the influent.

Substance	Unit	MIN	MAX	AVERAGE	MEDIAN	STDEV	N	Frequency in samples
Di-2-ethylhexyl- phthtalate (DEHP)	μg/l	0.3	16	3.93	2.90	3.32	36	100%
Fluoride (F-)	mg/l	0.15	1.5	0.45	0.38	0.26	36	100%
Toluene	μg/l	0.3	75	9.85	4.25	15.92	36	100%
Diisobuthyl- phthalate (DIBP)	μg/l	0.36	7.2	1.39	1.00	1.19	35	97%
Diclofenac	μg/l	0.14	23	4.20	2.90	4.64	35	97%
p/m-cresole	μg/l	9.9	980	201.03	100.00	221.57	35	97%
Phenol	μg/l	2.8	270	60.69	30.00	74.82	34	94%
Diethylphthalate (DET)	μg/l	0.33	2.9	1.27	0.99	0.60	32	89%
PBDE 47	μg/l	5.2E-05	0.0043	7.85E-04	5.90E-04	8.44E-04	31	86%
Resorcinol	μg/l	5	130	53.37	47.00	34.91	31	86%

Substances that are subject to international restrictions (e.g. di-2-ethylhexylphthalate) are still present in raw sewage and treated effluent (Paper III).

4.2 Relationships between performance and complexity (Paper I)

4.2.1 Relationship between complexity, performance and plant size

As bigger WWTPs have more strict effluent quality standards, the general complexity of the WWTPs increased along with loading capacity. In larger MWWTPs specific treatment steps were added (e.g. biological nitrogen removal, effluent filtration). There was a rather good positive relationship between general complexity and designed loading rate for all WWTPs (Paper I). For WWTPs with loading < 100 000 PEs, the relationship was of more significant and monotonic. This suggests that the layout of the wastewater treatment process tends to become more sophisticated in accordance with the growth of design loading rate.

Similarly, TEC was logarithmically growing in accordance with designed loading rate, but the variance was greater. Total evaluation of complexity (TEC) showed a wide variation for each wastewater treatment facility, but it is not always technologically or financially feasible or even rational to choose the most complex solution (the technology on the higher complexity level usually needs higher investment, but frugal alternative could achieve the similar pollutant removal efficiency). For WWTPs with loading rates below 100 000 PE-s there was no great difference between Pearson's r (r = 0.339 and p-value < 0.05) and Spearman's ρ (ρ = 0.386, p-value < 0.05), it can be concluded that regardless of the increase in the number of treatment steps, particularly along with the increasing size of the WTTPs, (more refined technology is needed for nutrient removal), the technical solution for waste water treatment might not be very sophisticated. For example, in 11 % of small (< 300 PEs) WWTPs there were no screens. The type of screening equipment in other WWTPs was different: screw screens (27 %), step screens (26 %), bar screens (11 %) and also various other devices or solutions (25 %). The complexity of the screens varied between 0 and 8 points and this indicates clearly that choice of treatment step's complexity was made in compliance with financial capability.

There was a statistically significant (p-value < 0.05), but weak relationship (Pearson's r = 0.227) detected between total evaluation of performance (TEP) and designed loading rates. The correlation between TEP and designed loading rate for WWTPs with loading rate less than 100 000 PEs (Pearson's r = 0.148, p-value < 0.05) was weak and could be affected by the capability to react against process disturbances in these plants. In three out of four WWTPs exceeding 100 000 PEs there was O&M personnel available 24/7, but in smaller WWTPs (less than 100 000 PEs) there was only one WWTP, where O&M personnel was available likewise. For most of the small plants (less than 300 PEs) the O&M personnel was available for only 2-4 hours per week (during the scheduled supervision) and the ability to react against process disturbances was highly dependent on the discovery of a malfunction.

There was a positive and moderate correlation between TEC and TEP (Pearson's r = 0.413, p-value < 0.05). It could be concluded that higher level of automation (higher complexity) in each treatment step could prevent many process disturbances and therefore improve overall plant performance.

4.2.2 Relationship between performance and complexity of treatment steps

In many cases the complexity of certain treatment step had a great impact on the performance of treatment step. The performance of screens, grit and oil removal, automation, final clarifier and nitrogen removal was most dependent on complexity of treatment step. This shows that for certain treatment steps the use of more sophisticated technology is crucial in order to achieve desired (undisturbed) outcome of said step

(Paper I). For example, the risk for clogging and flooding of the screens can be reduced by using an automatic removal of screenings.

4.3 Factors affecting performance in activated sludge plants (Paper II)

4.3.1 Most common problems in Estonian activated sludge plants

A wastewater treatment plant can produce good quality effluent even if there are several shortcomings. All AS plants (n=195) studied had issues, but the severity of these problems varied in a great magnitude. Most common issues (Fig. 4) were associated with effluent quality (56.4 % of WWTPs had high TSS in the effluent), aeration problems (57.9 %), foaming (38.1 %), bulking (32.3 %) and ensuring of anaerobic (8.2 %) and anoxic (15.4 %) conditions in reactors.

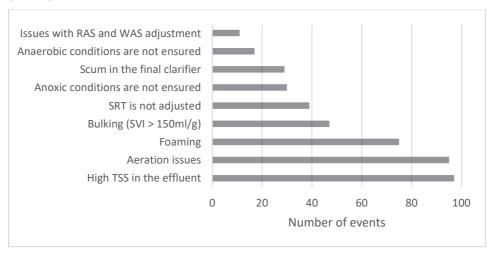


Figure 4. Most common problems in Estonian activated sludge WWTPs.

During the assessment it was observed that five WWTPs were at the stage of start-up of the process due to the loss of activated sludge by serious hydraulic overloading. These plants were excluded from further analyses. The main approach for finding reasons for bulking is to identify the specific filamentous bacterium in the bulking sludge (Martins et al., 2004). Microscopic examination of activated sludge was performed only in 15 WWTPs (Microthrix parvicella was dominant filamentous organism in 61.5 % samples), but as the reasons for foaming and bulking in rest of WWTPs were not known, statistical analyses in Paper II focused mainly on CCPs.

Some of the factors triggering bulking are not easily traced (e.g., low DO concentrations could be the result of sudden load of readily biodegradable organic matter as well as lack of aeration capacity or poor maintenance of aeration system or combination of these factors), good design and construction quality as well as professional O&M practice can minimize the bulking probability or its impact to the effluent quality. In order to understand the complexity of actual situation in the WWTP that could trigger the disturbances in the normal wastewater treatment process and favour bulking, most common problems and their coexistence in each WWTP was assessed.

Table 5. Frequency of co-existence of selected problems in Estonian WWTPs (n=190).

	A01	A02	A03	A04	Δ 05	Δ06	A07	A 0.8	A 00	Δ 10	Δ11	A12	Δ12	Δ 1.4	Δ15	A16	Δ17	Δ19	Δ10	Δ 20	
A01	1.00	0.28	0.32	0.31			0.28					0.28				0.30					A 01
A02	0.40	1.00	0.51	0.47			0.44					0.46				0.43				0.44	
A03	0.21	0.24	1.00	0.22	0.21		0.28					0.20			0.08			0.25		0.21	A03
A04	0.73	0.80	0.80	1.00			0.76					0.74	-			0.72		-			A04
A05	0.49	0.43	0.37	0.37	1.00	0.47	0.28	0.31	0.38	0.36	0.35	0.38				0.44					A05
A06	0.62	0.57	0.54	0.57	0.71	1.00	0.53	0.57	0.60	0.61	0.57	0.56	0.52	0.57	0.42	0.56	0.55	0.54	0.57	0.51	A06
A07	0.35	0.39	0.54	0.40	0.30	0.37	1.00	0.72	0.44	0.40	0.46	0.55	0.38	0.48	0.42	0.45	0.50	0.45	0.43	0.49	A07
A08	0.57	0.63	0.80	0.55	0.47	0.56	1.00	1.00	0.55	0.60	0.59	0.66	0.58	0.72	0.42	0.62	0.55	0.63	0.57	0.61	A08
A09	0.59	0.57	0.51	0.55	0.59	0.61	0.64	0.57	1.00	0.65	0.73	0.70	0.49	0.57	0.58	0.55	0.55	0.56	0.52	0.63	A09
A10	0.40	0.36	0.46	0.36	0.36	0.40	0.37	0.39	0.42	1.00	0.46	0.46	0.45	0.41	0.33	0.33	0.36	0.33	0.43	0.35	A10
A11	0.60	0.56	0.44	0.57	0.53	0.57	0.65	0.60	0.71	0.71	1.00	0.74	0.51	0.74	0.42	0.60	0.55	0.58	0.52	0.58	A11
A12	0.41	0.49	0.46	0.47	0.48	0.47	0.65	0.56	0.58	0.60	0.63	1.00	0.49	0.68	0.50	0.53	0.68	0.55	0.48	0.53	A12
A13	0.41	0.34	0.49	0.40	0.27	0.34	0.36	0.39	0.32	0.46	0.34	0.39	1.00	0.46	0.42	0.36	0.41	0.38	0.48	0.40	A13
A14	0.44	0.51	0.44	0.41	0.47	0.41	0.50	0.53	0.41	0.46	0.55	0.59	0.51	1.00	0.33	0.48	0.50	0.44	0.43	0.42	A14
A15	0.13	0.06	0.02	0.07	0.04	0.05	0.06	0.05	0.06	0.06	0.05	0.06	0.07	0.05	1.00	0.07	0.09	0.06	0.00	0.14	A15
A16	0.44	0.45	0.54	0.46	0.56	0.48	0.54	0.53	0.46	0.43	0.51	0.54	0.47	0.56	0.58	1.00	0.95	0.84	0.57	0.70	A16
A17	0.08	0.07	0.12	0.12	0.10	0.11	0.14	0.11	0.11	0.11	0.11	0.16	0.12	0.14	0.17	0.22	1.00	0.20	0.10	0.19	A17
A18	0.51	0.52	0.66	0.55	0.60	0.53	0.63	0.63	0.55	0.50	0.58	0.65	0.58	0.59	0.58	0.98	1.00	1.00	0.71	0.75	A18
A19	0.11	0.10	0.15	0.11	0.11	0.11			0.10		0.10	0.11	0.14	0.11	0.00	0.13	0.09	0.14	1.00	0.16	A19
A20	0.30	0.28	0.29	0.28	0.33		0.36				0.30	0.32	0.32		0.67			0.39		1.00	A20
	A01	A02	A03	A04	A05	A06	A07	A08	A09	A10	A11	A12	A13	A14	A15	A16	A17	A18	A19	A20	
	SVI > 150	Design loading was not based on actual measurements	AS process tanks are over- or undersized	Industrial wastewater causes problems to the WWTP	Infiltration into the sewer system	Extreme peak flows	F/M > 0,15 g BHT7/g MLSS	Real SRT is less than needed	Problems with aeration system	Floating sludge in aerobic tank	Floating sludge in final clarifier	Mass surface loading rate is greater than 500 L/m2*h	Operator does not regulate RAS rate	Operator does not know WAS rate	WAS is not removed	Effluent BOD over the limit	Effluent COD over the limit	Effluent TSS over the limit	Effluent TN over the limit	Effluent TP over the limit	

Matrix of co-existing problems (Table 5) gives an overview of frequency of co-existence of selected problems (n=20) in investigated AS plants. In total, 40 problems were initially analysed and similarly to situation reported by Hegg $et\ al.$ (1979), an average Estonian AS WWTP had 14.6 ± 3.9 issues. It can be seen from Table 5 that if certain problems exist in the WWTP, the probability for the connected issue could be high. E.g., floating sludge in final clarifier (A11) occurs often in a situation where there are problems with aeration system (A09), high F/M (A07) and operator is lacking the knowledge about controlling the WAS rate (A14).

4.3.2 Factors correlating with SVI

SVI is calculated as a quotient between V_{30} and MLSS, but statistical analyses revealed that there were statistically significant, but weak relationships between SVI and V_{30} (Pearson's r=0.215, p-value = 0.004) and between SVI and MLSS (Pearson's r=-0.301, p-value = 4.11e-5), if looked separately. This suggests that operational decisions based only on the measurement of sludge settleability can mislead the operator's judgement in process control strategies.

For statistical analyses, the SVI was divided into two groups with "good" (SVI < 150 ml/g) and "bad" settleability (SVI > 150 ml/g) and was evaluated with "1" or "0" respectively.

All questions regarding CCPs were formed to be answered as "yes" or "no" and were evaluated with "1" or "0" respectively. Further analyses showed correlations as described in Table 6. Settleability was influenced by biological phosphorous removal, performance of process parts and influent sources.

The factors correlating with SVI could be divided into two groups (see also Table 6): a) factors that affect SVI (e.g., WAS removal, usage of remote control) and b) factors that are affected by SVI (e.g., performance of the final clarifier, effluent quality). A negative correlation shows that an occurrence of the factor induces the bulking (e.g., if there are industrial sources in the influent, the bulking is more likely to occur).

Table 6. Factors that had a statistically significant (p-value < 0.05) or important (p-value < 0.10) correlation with "good" SVI.

Davamatav		1	Spearman	
Parameter	R p-value		Р	p-value
Performance of biological reactor (on the 10p scale)	0.247	0.001	0.234	0.003
30-minute settling test (V ₃₀), ml	-0.244	0.002	-0.226	0.004
Is the real mass surface loading rate in final clarifier less than 500 kg/(m ² ·d)?	-0.215	0.012	-0.215	0.012
Industrial sources in the influent cause problems to the WWTP	-0.380	0.022	-0.380	0.022
MLSS, g/l	-0.307	0.021	0.168	0.031
Effluent TSS, mg/l	0.096	0.226	0.163	0.038
Does an operator measure TP for process control?	-0.161	0.039	-0.161	0.039
Effluent TN, mgN/l	0.185	0.018	0.162	0.039
Is effluent quality within limits?	-0.146	0.061	-0.146	0.061
Are there any hydraulic problems in final clarifier?	0.160	0.062	0.160	0.062
Is biological P-removal possible?	-0.148	0.064	-0.148	0.064
SRT(real)/SRT(designed), d/d	-0.088	0.358	0.172	0.071
Real SRT, d	-0.117	0.204	0.172	0.071
Performance of final clarifier (on the 10p scale)	-0.103	0.212	-0.148	0.083
Are WAS pumps working?	0.087	0.293	0.145	0.091
Is there any infiltration to the sewer system?	-0.132	0.093	-0.132	0.093
Volumetric fraction of anaerobic reactor (AN/OX)*	0.304	0.080	0.282	0.106
Anaerobic fraction (f _{AN})*	0.313	0.071	0.264	0.132
Effluent TP, mgP/I	0.139	0.077	0.051	0.496

^{*}correlations calculated without certain package plants (n = 10). The reason for excluding these WWTPs was that these plants had very big anaerobic reactor ($f_{AN} = 0.5$) that was not suitable for enhanced biological phosphorous removal (Gašparikova *et al.*, 2005).

The reasons for bulking could be divided into three groups: a) influent characteristics, b) design and construction (D&C) and c) operation and maintenance (O&M). While influent characteristics involve factors like nutrient deficiency, low temperature and pH that have been reported to be the reasons for bulking (Eikelboom, 2000; Gerardi, 2008; Jenkins *et al.*, 2004), the factors resulting from misgivings in the fields of D&C or O&M are sparsely reported.

4.3.3 Design and construction

Design and construction (D&C) shortcomings are not always easily found and need an inspection to be assessed. Even if an expertise has been conducted, the correlation between D&C issues and bulking is not often reported. It can be also debated if the simplicity of the process as a consequence of minimal investment possibilities could be the main reason for bulking (e.g., absence of bypasses or grease separators). Absence of grease separators is a common problem in small WWTPs. Nielsen *et al.* (2002) reported that *Microthrix parvicella* took up oleic acid under both anaerobic and aerobic conditions, while only a few floc formers were able to take it up under anaerobic conditions.

One-way ANOVA showed that the statistically relevant (p-value < 0.05) possible causes for bulking were a) the type of biological reactor used, b) infiltration to the sewer system and c) use of phosphorous removal (bulking was observed 45.9 % of AS plants with bio-P). The choice of reactor type was important (p-value 0.07) if compared against "good" SVI with one-way ANOVA. 69.8 % of plug-flow and SBR systems had SVI < 150 ml/g. Meanwhile, only 56.3 % of continuous stirred-tank reactors (CSTR) had similar SVI values. The balancing tank is usually needed to reduce variations in wastewater flow and concentrations. In many cases it helps to reduce the effect of peak flows and the risk for hydraulic overloading. In small WWTPs, where balancing tank was absent, the average SVI value was 134.7 ml/g (n = 90), whereas in WWTPs with balancing tank the average SVI value was 173.0 ml/g (n = 57). The effect was significant (p-value 0.05) but could be misleading as an average HRT in balancing tanks was 1.6 d-1. This period could be long enough for wastewater septicity to develop.

Selectors are usually considered to be an effective way for bulking control, but they do not work for all filamentous micro-organisms (Martins et al., 2004). Due to the high requirements of effluent quality 65.7 % of Estonian AS WWTPs are designed with the possibility for biological nitrogen removal and 20.0 % of plants have enhanced biological phosphorous removal. In total, there were 33 WWTPs included in this study with AAO configuration. Although one-way ANOVA showed that the volume fraction of an anaerobic reactor compared to volumes of an aerobic (AN/OX) and/or an anoxic reactors (AN/AX or AN/(AX+OX)) could play crucial part in the probability for sludge bulking (Table 7), the data was influenced by certain package plants that do have big anaerobic tanks which are not suitable for enhanced biological phosphorus removal (Gašparikova et al., 2005). When these plants were excluded from the ANOVA analyses, the p-value of an anaerobic volume fraction was 0.14 for AN/OX and 0.07 for AN/(AX+OX), but for both cases the F-value was smaller than F_{crt}. This means that although excluded package plants were not suitable for biological P-removal, an increased anaerobic fraction favoured bulking reduction. An anaerobic fraction and volumetric fraction of anaerobic reactor compared to the aerobic reactor gave both positive correlation with SVI < 150 ml/g values, even without mentioned package plants (for f_{AN} Pearson's r 0.313, p-value 0.07 and for AN/OX Pearson's r 0.304, p-value 0.08). Analyses of hydraulic retention time in anaerobic and anoxic reactors revealed that in small WWTPs the contact time was much higher than recommended by Henze et al. (2011) with average values for anaerobic reactor 10.5 \pm 6.8 h and for anoxic reactor 27.9 \pm 24.0 h respectively.

Table 7. The design parameters of anaerobic reactors showed significant impact to the bulking according to the one-way ANOVA.

		SVI <		SVI >		Number	r Mean	F-	P-	
Parameter	Unit	150	Variance	150	Variance	of	square	value	value	\mathbf{F}_{crit}
		ml/g		ml/l		WWTPs				
f_{OX}	-	0.772	0.038	0.739	0.032	188	0.045	1.250	0.265	3.90
f_{AN}	-	0.340	0.052	0.312	0.063	38	0.007	0.127	0.724	4.11
f_{AX}	-	0.356	0.019	0.363	0.019	121	0.002	0.090	0.764	3.92
AN / OX	m^3/m^3	0,400	0.135	0.187	0.011	38	0.428	5.352	0.027	4.12
AN / OX*	m^3/m^3	0.259	0.030	0.187	0.011	35	0.048	2.243	0.143	4.12
AX / OX	m^3/m^3	0.954	0.871	0.521	0.086	121	1.571	2.979	0.094	3.94
AN / AX	m^3/m^3	1.054	0.955	0.521	0.086	32	2.199	4.107	0.052	4.18
AN / (AX + OX)	m³/m³	0.291	0.066	0.135	0.005	37	0.223	5.820	0.021	4.12
AN / (AX + OX)*	m³/m³	0.195	0.013	0.135	0.005	29	0.031	3.392	0.074	4.13
HRT _{AN}	d ⁻¹	0.399	0.051	0.369	0.058	32	0.007	0.131	0.719	4.17
HRT_{AX}	d ⁻¹	1.240	1.277	1.091	0.624	113	0.596	0.582	0.447	3.92
HRT _{OX}	d ⁻¹	3.717	14.186	2.478	3.174	165	58.671	5.752	0.018	3.90

While the impacts of reactor type and infiltration to the SVI could be explained by kinetic selection theory (e.g., infiltration causes the dilution of nutrients and therefore gives growth advantage to filamentous organisms with lower K_s values over floc forming bacteria with higher K_s values), the role of phosphorous removal process as the cause for bulking remains uncertain. Some of the possible explanations for connections between bulking and phosphorous removal have been presented by Nielsen *et al.* (2002, 2010) and Wang *et al.* (2014). Nielsen *et al.* (2010) suggested that soluble components, either from the wastewater or produced by hydrolysis in the anaerobic tank, are taken up for storage as polyhydroxyalkanoates (PHA)/lipids by the filamentous *Microthrix parvicella*, by PAOs (*Accumulibacter*), and by GAOs (*Competibacter*, *Defluviicoccus*). Wang *et al.* (2014) suggested that *M. parvicella* could take part in enhanced biological phosphorous removal. *M. parvicella* is the most dominant organism in Estonian WWTPs that is causing bulking.

Fan *et al.* (2017) reported that *Microthrix parvicella* favoured lower temperature, alteration between anaerobic and aerobic conditions and long chain fatty acids (LCFA) in batch tests. In the anaerobic/aerobic alternation experiment reported by Fan *et al.* (2017) the AN/OX ratio was 0.5 which seems to be favouring SVI < 150 ml/g as shown in Table 7, their study still showed that this was enough to initiate *M. parvicella* bulking with the presence of LCFAs as the sole carbon source. Their experiments with different water temperatures (13°C and 20°C) combined with anaerobic-aerobic conditions and LCFA feed showed also great differences in *M. parvicella* abundance favouring colder temperature. In real conditions the number of factors occurring simultaneously could be unlimited (Tables 5 and 6) and the event of bulking could be initiated or even suppressed. Still, the role of volume fraction of an anaerobic reactor could be significant and needs further investigation.

4.4 Operation and maintenance

The competence of the operators was evaluated during the data collection in the form of hidden test on the scale of 10 points. The average result was 7.31 and the minimum result 1.5. However, the difference between small water companies (less than 2,000 people served) and large ones (more than 2,000 people served) was remarkable – an average competence of small water company operators was 6.23 p (median - 6.66 p), with results ranging from 1.14 p to 9.65 p. The competence of the operators of large water companies was somewhat better (average - 7.56 p, median - 7.70 p, minimum - 1.45 p and maximum 10.0 p), which in turn may be due to the fact that a) large water companies company employees have the opportunity to specialize more – the wastewater treatment plant operator does not have to deal with e.g. accounting and customer service, b) 87% of large water company operators have received wastewater treatment plant operator training, while only 66% of small water company operators have done so (Kõrgmaa *et al.*, 2016).

The performance of the wastewater treatment plant did not depend on the operator's level of education, experience, or the number of wastewater treatment plants he operates (Kõrgmaa *et al.*, 2016). Operators who were able to adequately assess loads and were familiar with the meaning of basic professional terms (e.g. load, concentration or flow rate) also received a higher performance rating for their treatment plant (Kõrgmaa *et al.*, 2016). From this it can be concluded that those operators who were able to apply the knowledge gained during the trainings on a daily basis were also more capable in achieving better performance in WWTP.

Statistical analysis showed that performance of AS WWTPs (Kõrgmaa *et al.*, 2016) was dependent on O&M practices and operators' familiarity with WWTP. Most critical O&M practices were controlling the SRT and F/M, but also activities made during the maintenance and its frequency. According to Kõrgmaa *et al.* (2016) it was concluded that operators' competency had significant (p-value < 0.05) linear correlation with TEP in activated sludge process.

4.4.1 Effect on process control (Paper II)

Although statistical analyses revealed that operators' competence did not influence the bulking directly, it could have severe consequences to the WWTPs performance. E.g., in one SBR that was treating wastewater from dairy factory, an operator observed that the activated sludge had poor settling properties and added external activated sludge with good settling properties into his WWTP. The settling did not improve as an operator had not removed any WAS and as a result MLSS was 12 mg/l during the visit.

Procedures for O&M with operators' competency constitute the key factors for successful pollution removal. The chemical precipitation of phosphorous thickens and compacts the activated sludge (Lind, 1998), but it might also initiate toxic effects and the control over chemical addition is crucial (Suresh et~al., 2018). During the study a negative weak correlation (Pearson's r = - 0.161, p-value 0.039) was found between bulking and the operators' claim about adjusting chemicals for phosphorous removal. Further analyses showed that contradiction between desired effect of chemical precipitation (less bulking) and operators' claim about controlled chemical adjustment was driven by operators' tendency to add too much chemicals. In WWTPs where operator claimed to be making adjustments according to the real measurements the ratio of chemicals added to the amount actually needed was 1.6 ± 1.5 , but in the other group the same ratio was 1.3 ± 1.2 . Although the difference between groups was not statistically relevant (p-value

0.29), it shows that while operators make decisions for chemical addition based on effluent results (according to ANOVA average TP was 2.4 mgP/l in the group that made adjustments against 4.3 mgP/l in the group that did not, p-value 0.02), in order to minimise TP concentration in the effluent operators tended to add too much iron salts and as a result it affects activated sludge properties negatively.

Instrumentation, control and automation (ICA) plays an important role to reach operational goals (Olsson & Jeppsson, 2006). 66.7 % studied WWTPs used ICA for the process control. Statistical analysis showed that in WWTPs, where ICA was used bulking was less often observed (Paper II).

4.4.2 Effect on hazardous substances removal (Paper III)

Hegg *et al.* (1979) listed improper operator application of concepts and testing to process control as well as inadequate understanding of the sewage treatment as two highest ranking factors contributing to poor plant performance.

Statistical analyses showed that operators' competency had significant (p-value < 0.05) correlation with removal efficiencies of several substances (e.g., COD, BOD₇, heavy metals, PAHs).

Operators' competence had a strong influence on the stability of the wastewater treatment efficiency. A competent operator was more successful in ensuring stable COD removal and in avoiding decrease in process performance. While taking into consideration that there was significant (p-value < 0.05) moderate correlation (Pearson's r=0.663) between COD and removal of organic hazardous substances, it can be concluded that operators' competency in conjunction with O&M practices applied plays crucial role in successful removal of hazardous substances.

4.5 Relationship between performance and effluent quality

4.5.1 Removal of hazardous substances in the municipal WWTP (Paper III)

Estonian regulation on effluent quality standards (Ministry of Environment, 2019) sets limit values for disposed effluent. Municipal wastewater treatment plants are designed to reduce the pollution load to the environment. While nutrients and many other substances could be efficiently and consistently eliminated, the removal of hazardous substances is often insufficient (Luo *et al.*, 2014).

In order to understand whether the control of emissions of hazardous substances might be reduced by better process control, limiting the industrial discharges to the public sewer system or upgrading the existing technology it is crucial to understand the fate and removal efficiency of hazardous substances during the wastewater treatment process (Paper III). Still, it has to be underlined that while discussing the removal of hazardous substances during the wastewater treatment process the discussion generally refers to the removal of parent compounds from aqueous phase (Luo *et al.*, 2014). Sewage sludge analyses strongly indicate that for most substances an accumulation to the biomass has taken place. E.g., during this study, di-2-ethylhexylphthalate (DEHP) was detected from all influent samples above LOQ and only 25 per cent of effluent results exceeded LOQ, but all sewage sludge samples contained high levels of DEHP. Average removal efficiency was 69.1 % for DEHP. This means that while DEHP is quite successfully removed from aqueous phase, it is merely transferred to the sewage sludge.

Removal efficiencies calculated based on grab and composite samples respectively are presented in Paper III Although Prasse *et al.* (2015) stated that grab sampling of influent and effluent wastewater is inappropriate to determine elimination efficiencies of

MWWTPs as concentrations of hazardous substances might vary significantly over time, it was not possible avoid grab sampling in order to satisfy the sampling conditions set in ISO 5667-3 for certain substances. Removal efficiencies expressed in Table 10 should be therefore regarded with certain reservations as grab sampling can be considered random. As a result, for some of the substances (e.g., boscalid, diclofenac) the removal efficiency was negative. Negative removal efficiency (-85.5 %) of aminomethylphosphonic acid (AMPA) indicates that this substance is formed during the biological wastewater treatment process. AMPA is a metabolite of microbial degradation of widely used herbicide glyphosate (Struger et al., 2015). Some of negative removal efficiencies in Paper III present the situation when MWWTP A was hydraulically overloaded during the snow melting period resulting in wash-out of filter solids. During this event it was noticed that MWWTP A was still able to remove 91.8 % of BOD7, but COD removal had reduced to 53.3 %. TSS and PAH removal efficiencies were negative. During the event two-ring compound naphthalene was still degraded (49.5 % was removed from wastewater), but removal efficiencies for four-ring compounds fluoranthene and pyrene that need longer degradation time (Bouches et al., 1996; Moscoso et al., 2015) were negative and washout of these compounds was observed.

4.5.2 Factors affecting effluent quality (Paper I)

It was found that by interpreting the results of statistical analyses of CCPs and by studying their impact on the effluent quality, there were several CCPs which were extremely critical for the quality of effluent water. As much as 50% of WWTPs with poor effluent quality had a thick layer of scum on the surface of the final clarifier. In AS plants IVE was dependent on the performance of the final clarifier:

$$IVE = 5,00 + 0,29 P_{FC} + 0,21 \cdot P_{EFF} + 0,60 \cdot P_{OAE},$$
 [6]

where PFC is a performance of final clarifier, PEFF is a CCP that describes whether the results of effluent analyses are in consent with quality standards and POAE is a CCP that shows whether there were any sensory indicators (colour, turbidity, odour) of poor effluent quality. It has to be underlined that low ratings for PEFF and POAE could be the direct result of poorly performing final clarifier. This empirical model (r = 0.48, p-value < 0.05) clearly showed, that in AS plants it is the final clarifier which is likely to be the weakest point in WWTP. Obviously, the effluent quality is directly depending on several operating and maintaining aspects. It is important whether the surface of the clarifier is kept clean or the floating scum is also discharged together with the effluent; whether there are hydraulic problems with flow or a malfunction of any pumps that influences the sludge volume surface loading rate. As shown in Paper I, the Pearson's product-moment correlation between TEC and TEP was found significant (p-value < 0.05) particularly for the final clarifier. As concluded, several problems could be easily prevented using more sophisticated technology and/or by relying on more automated facilities. For example, installing automatic scrapers for surface scum removal to the final clarifier (increasing complexity) would help to prevent TSS escaping with the effluent (increased performance).

5 Conclusions

This doctoral thesis aimed to examine the wastewater treatment process as a whole system in order to map the relationships between the influent characteristics, technological complexity, operation and maintenance practices and overall performance of the WWTP for achieving satisfactory effluent quality. The main focus was on the performance of activated sludge process.

The main results of this study are summarized as follows:

- A novel method was developed to rapidly assess the performance (TEP) and complexity (TEC) of WWTPs. TEC and TEP are new standardized tools that make the comparison of a wide range of WWTPs using different technologies and treating different loadings possible.
- An average Estonian activated sludge WWTP had 14.6 ± 3.9 performance limiting factors and it is common that several problems occur simultaneously.
 In real conditions the number of factors occurring simultaneously could be unlimited and the reduction in performance could be initiated or even suppressed by combination of several issues.
- There was a significant positive correlation between TEC and TEP. Thus, increasing complexity by combining automation with a more advanced equipment (higher complexity) could improve the performance of the WWTPs. Moreover, higher complexity could prevent several serious process disturbances. Particularly the complexity of equipment of some treatment steps has a highly significant impact to the performance of the whole facility. It was found that the final clarifier appears be the weakest point among all the treatment steps in WWTP. Performance of final clarifier in AS process is in turn dependent on the influent characteristics and SVI values.
- Most of the problems in the WWTP start from with the public sewer system –
 40 % of WWTPs had issues with serious fluctuations in the wastewater flow or
 load and 20 % WWTPs had issues caused by an industrial wastewater. Most
 wastewaters had a high content of nitrogen and phosphorous but were lacking
 carbon sources. 84 % of hazardous substances analysed were found from the
 influent above LOQ. Influent characteristics often cause reduction of
 performance.
- Microthrix parvicella dominated in 65 % of WWTPs where microscopic examination was performed. Statistical analysis showed that factors triggering Microthrix parvicella growth were connected to the operational conditions (long SRT, low F/M) and influent characteristics (lack of carbon sources compared against nitrogen and phosphorous content).
- Effluent quality was dependent on several factors (incl. operational and maintenance aspects), but the performance of final clarifier was most critical.
 Wastewater treatment systems that had greater level of complexity were more successful in removing hazardous substances.
- This study showed that operators' competency had a strong influence on the stability of the wastewater treatment process. Procedures for O&M with operators' competency constitute the key factors for successful pollution removal and in avoiding decrease in process performance.
- Performance of wastewater treatment plant is dependent of influent characteristics, operator's competency and complexity of the plant. The best

result in wastewater treatment process is achieved by combining stable influent with good design solutions, excellent operation and maintenance practices.

Further studies are required to apply these indexes in different geographical regions and climatic conditions, as well as in larger WWTPs. It can also be suggested to introduce the usage of the proposed assessment method on WWTPs with different equipment and installation to improve the daily O&M practices (providing that CCPs are regularly checked and critical parameters for system control are simultaneously calculated). By developing a wide-range database the researchers and designers would get comprehensive tool for further design improvements.

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Abstract

Factors affecting performance of municipal wastewater treatment plants

The performance of wastewater treatment plants (WWTP) depends on various technical, non-technical, and human factors. As many as 246 small and medium size WWTPs were studied during 2014–2018 and evaluated according to the novel method for rapid assessment of performance and the complexity of small WWTPs. The method creates a comparable system of ratings for all treatment solutions by analyzing simultaneously influent characteristics, system complexity, O&M practices and process parameters in comparison with designed and/or standardized values and their impact to the overall system performance. Total evaluation of complexity (TEC) and total evaluation of performance (TEP) are new unified tools which were applied for comparing WWTPs with different technologies and wide variety of loadings.

The study analysed the interrelationships between performance of WWTP, influent characteristics, operation and maintenance practices and complexity of plant and their impact to theeffluent quality. To this end, both the problems in biological treatment process (bulking and foaming of activated sludge) and the efficiency of removing hazardous substances were examined in depth. In both cases, statistical analysis methods were used to identify the factors that affect the performance of the wastewater treatment plant.

The study revealed that the performance of wastewater treatment plants and the quality of effluent are primarily affected by the characteristics of wastewater, the complexity of the treatment plant and operating practices. A positive correlation was found between performance and complexity – WWTPs with higher complexity were more efficient. This shows that by introducing automation and more sophisticated equipment for process control, it is possible to prevent many process failures and to improve the performance of the entire plant.

Characteristics of influent caused problems, especially in the biological treatment unit, which in turn could lead to the sedimentation problems in the final clarifier. *Microthrix parvicella* dominated in 65% of activated sludge treatment plants where microscopic examination was performed. Statistical analysis showed that the proliferation of *M. parvicella* was caused by several operating conditions (long SRT, low F/M) and influent characteristics (carbon deficiency compared to nitrogen and phosphorus contents).

According to the developed empirical model, the quality of effluent depends on the performance of the wastewater treatment plant. WWTPs with higher complexity were more efficient in removing hazardous substances from the water phase. The competence of the operator had a strong effect on the stability of the treatment process and thus on the removal of contaminants.

Lühikokkuvõte

Reoveepuhastite tõhusust mõjutavad tegurid

Reoveepuhastite (WWTP) tõhusus sõltub mitmetest tehnilistest, mittetehnilistest ning inimlikest faktoritest. Aastatel 2014–2018 uuriti kokku 246 asulareoveepuhastit, mida hinnati uudse tõhususe ning komplekssuse hindamise metoodikaga. Meetod võimaldab samadel alustel hinnata erinevaid puhastustehnoloogiaid ning analüüsib reovee omadusi, süsteemi komplekssust, käitamispraktikaid ning võrdleb protsessi reaalseid parameetreid projekteeritud väärtuste suhtes. Komplekssus (TEC) ja tõhusus (TEP) on uued universaalsed tööriistad, mida rakendati erinevate koormuste ning tehnoloogiliste lahenduste hindamiseks.

Uuringu käigus analüüsiti reoveepuhasti tõhususe, reovee parameetrite, käitamistingimuste, operaatori teadmiste ja puhasti komplekssuse omavahelisi seoseid ning nende mõju heitvee kvaliteedile. Selleks võeti süvendatud vaatluse alla nii probleemid bioloogilises puhastusprotsessis (aktiivmuda pundumine ja vahutamine) kui ka ohtlike ainete ärastamise efektiivsus. Mõlemal juhul kasutati statistilise analüüsi meetodeid, et välja selgitada tegurid, mis mõjutavad reoveepuhasti tõhusta tööd.

Uuringu tulemusena selgus, et reoveepuhastite tõhusust ning heitvee kvaliteeti mõjutavad eelkõige reovee omadused, puhasti komplekssus ning käitamispraktikad. Tõhususe ning komplekssuse vahel tuvastati positiivne korrelatsioon – suurema komplekssusega puhastid olid tõhusamad. See näitab, et kui võtta protsessi juhtimisel kasutusele automaatika ning suurema keerukusastmega seadmed, on võimalik ennetada mitmeid protsessi seisakuid ning parandada kogu puhasti tööd.

Reovee omadused põhjustavad probleeme eelkõige bioloogilise puhastuse etapis, mis omakorda võivad kaasa tuua settimise probleeme järelsetitis. *Microthrix parvicella* domineeris 65 % aktiivmudapuhastites, kus aktiivmuda mikroskopeerimine läbi viidi. Statistiline analüüs näitas, et *M. parvicella* vohamist kutsusid esile mõningad käitamistingimused (pikk SRT, madal F/M) ning reovee omadused (süsiniku vaegus võrreldes lämmastiku ja fosfori sisaldustega).

Loodud empiirilise mudeli kohaselt sõltub heitvee kvaliteet reoveepuhasti tõhususest. Suurema komplekssusega puhastid said ohtlike ainete eemaldamisega vee faasist paremini hakkama. Operaatori kompetentsus avaldas tugevat mõju puhastusprotsessi stabiilsusele ning seeläbi saasteainete eemaldamisele. Samuti aitas hea käitamispraktika ning operaatori kõrge teadlikkus puhastusprotsessist kaasa probleemide ennetamisele bioloogilise puhastuse etapis.

Appendix: Publications

Paper I

Vallo Kõrgmaa; Taavo Tenno; Aimar Kiviruut; Mait Kriipsalu; Mihkel Gross; Priit Tamm; Kristjan Karabelnik; Harri Terase; Vahur Vark; Natalja Lepik; Karin Pachel; Arvo Iital (2019). A novel method for rapid assessment of the performance and complexity of small wastewater treatment plants. *Proceedings of the Estonian Academy of Sciences*, 68 (1), 32–42. DOI: 10.3176/proc.2019.1.03.



Proceedings of the Estonian Academy of Sciences, 2019, **68**, 1, 32–42

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A novel method for rapid assessment of the performance and complexity of small wastewater treatment plants

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Abstract. The performance of wastewater treatment plants (WWTPs) depends on various technical, non-technical, and human factors. A total of 245 small and medium-size WWTPs were studied during 2014–2015 and evaluated according to a novel method for rapid assessment of their performance and complexity. The suggested method creates a comparable system of ratings for all treatment solutions by analysing simultaneously influential characteristics, system complexity, operational practices, and process parameters in comparison with designed and/or standardized values and their impact on the overall system performance. Total evaluation of complexity and total evaluation of performance are new unified tools, which were applied for comparing WWTPs that applied different technologies and had a wide variety of loadings.

The study revealed that the greater the designed loading, the more treatment steps were usually needed and employed. The complexity of these treatment steps can vary a lot depending on the plant capacity. There was a positive relationship between complexity and performance: a higher complexity provided a better performance of WWTPs. This suggests that combining automation as a tool for the process control with a more advanced equipment (higher complexity) could prevent many process disturbances and therefore improve the overall plant performance.

Key words: wastewater treatment, performance assessment, critical control points, total evaluation of complexity, total evaluation of performance.

1. INTRODUCTION

The main goal of wastewater treatment is to reduce the amount of pollutants below the admissible level in order health. Regardless of a large variety of wastewater treatment technologies (e.g. activated sludge, sequencing batch reactors, biofilm, or constructed wetlands), the performance of wastewater treatment plants (WWTPs) depends on various technical and non-technical factors

not to pose any threat to the environment or to public

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such as characteristics of influent wastewater and how well the designed treatment process is in agreement with them, operational and management practices, reliability of equipment, and flexibility of the process (Hegg et al., 1979; Olsson, 2012; Hao et al., 2013). It is extremely difficult to compare fundamentally different treatment systems such as the biofilm process for 150 personal equivalents (PE) and a sequencing batch reactor for 10 000 PE. The former process relies on attached microorganisms, but the latter operates with free-swimming bacteria known as activated sludge. One is small and moderately automated, but the other is much larger and well automated. As a consequence. the two processes are designed on different bases. For example, substrate availability in biofilm processes can be considered to be diffusion-dependent, whereas floc diffusion limitation in activated sludge is not relevant. Still, both of these solutions contain similar design elements like pretreatment (e.g. sieves), biological treatment, sedimentation, and sludge handling. The function of sieves is to remove particles and objects from the flow of wastewater (ISO, 2014). The performance of the sieve depends on the quantity and quality of the influent, yet the configuration of sieving equipment may vary from hand-operated to multi-step and fully automated. Sedimentation is a process of settling and deposition, under the influence of gravity, of suspended matter carried by water or wastewater physical process that relies on gravity to remove suspended solids from water (ISO, 2014). Regardless of whether it is performed in the same tank of sequencing batch reactors or in a separate tank, the principle of the process is the same. Therefore, the evaluation of the overall efficiency of a WWTP constitutes a multi-objective decision chain (Hao et al., 2013).

It has been argued that implementation of the same method for comparative evaluation of overall treatment performance of different WWTPs is not possible due to the varying nature of treatment processes used and operational conditions applied (Hao et al., 2013; Chen et al., 2015). In many cases the process parameters in a WWTP can be modelled in detail based on designed values, characteristics of the influent, or even on the basis of metabolic reactions of the specific group of microorganisms (Copp, 2002; Henze et al., 2011). However, the actual situation on the plant can differ dramatically from the modelled result due to incorrect design parameters, inevitably changing input parameters, equipment failure, or neglect of maintenance. As there are many factors that can influence the effectiveness of a WWTP (e.g. infiltration, bulking sludge, broken sensors, or air diffusers), comparative evaluation is difficult to perform.

Some evaluation models and indexes have been proposed, but the aim of these models is quite different. Many authors focus on environmental or economic benefit evaluation and the models then are adaptations of life cycle assessments (De Faria et al., 2015; Fang et al., 2016), and the selection of treatment process is based on fuzzy analytical hierarchy process method (Karimi et al., 2011) or multi-criteria analysis (Jozwiakowski et al., 2015). The more universal models like treatment performance index proposed by Chen et al. (2015) and a similar model described by Hao and co-authors (2013) focus mainly on pollutant removal efficiencies in different treatment phases. Advantages of these models are that operational conditions are included in the assessment, but the variance of factors is still limited. In the above-mentioned studies the suggested models have been applied only on a handful of very large treatment plants. No studies treating small-scale WWTPs could be found.

The aim of this study was to create a comparable system of ratings for all wastewater treatment solutions by analysing simultaneously influential characteristics, system complexity, operational practices, and process parameters in comparison with designed and/or standardized values and their impact on the overall system performance.

In total, there are 1.35 million people and 664 municipal WWTPs in Estonia (VEKA, 2016). Between 2004 and 2014 about 49 million euros was invested into small-scale (less than 2000 PE) WWTPs and 115 million euros into 41 bigger plants. The main sources of investments were the Cohesion Fund of the European Union (2004 to 2007, 53.8 million euros, and 2007 to 2013, 61.7 million euros) and national investment programmes (approximately 16 million euros through the Estonian Environmental Investment Centre) (EIC, 2016). In total 288 WWTPs were constructed or modernized by using subsidies. According to national monitoring programmes, 45% of these WWTPs were not capable of meeting environmental requirements (Allas, 2014), and according to water enterprise self-monitoring programmes about 10% of the WWTPs were not capable of meeting these requirements most of the time even after the investments (VEKA, 2016). Reasons for their poor performance remained unclear. The difference between national programmes and self-monitoring programmes could be a result of differences in the sampling time (operators themselves could choose the sampling time for selfmonitoring analyses) and the performance of the WWTP during that specific period, or caused by some other factors, e.g. sampling strategy and analytical methods, as highlighted by Prasse et al. (2015).

Prasse et al. (2015) showed that chemical and biological assessment of wastewater treatment technologies

are influenced by the sampling strategy and analytical methods used, and therefore they suggested flow-proportional composite sampling. However, for us the flow-proportional composite sampling strategy for both influent and effluent of each WWTP was not realistic due to the funding and time limitations. Moreover, Estonian regulation on effluent quality (Vabariigi Valitsus, 2013) states that all wastewater effluents have to meet quality standards all the time. Therefore, a completely different approach to analyse the performance and effectiveness of WWTPs was chosen.

2. MATERIAL AND METHODS

2.1. Overview of the studied facilities

In total 245 WWTPs that had been built or modernized by using subsidies were evaluated (Fig. 1). Activated sludge process was the process most commonly used (163 conventional activated sludge treatment plants and 34 using sequencing batch reactors), followed by ecological wastewater treatment systems (incl. 13 oxidation

ponds, 28 constructed wetlands for secondary treatment, and 104 maturation ponds for tertiary treatment) and biofilm reactors (21 plants). Most evaluated WWTPs are small: 109 WWTPs are designed for loadings less than 300 PE, 91 for 300–2000 PE, 26 for 2000–10 000 PE, 15 for 10 000–100 000 PE, and 4 WTTPs for more than 100 000 PE. Of all studied WWTPs 88 (24%) were prefabricated package plants and 157 WWTPs were of special design.

There were three aspects that significantly complicated assessment of the performance: (i) most of the WWTPs were very small (see Fig. 2); (ii) technical solutions for wastewater treatment were different, and (iii) no unified evaluation methodology was available.

The suggested integrated method creates a comparable system of ratings for all individual technological steps making it possible to simultaneously analyse several factors such as influent characteristics, system complexity, operational practices, and process parameters. It also simultaneously considers their impact on the overall system performance. In addition, sampling results were linked to this rating-based model.

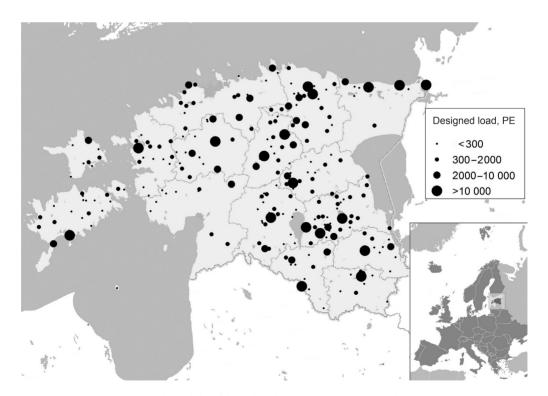


Fig. 1. Location and size of the evaluated wastewater treatment plants in Estonia.

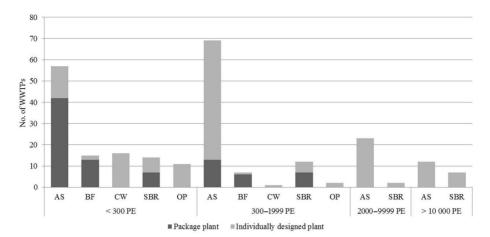


Fig. 2. Overview of treatment technologies in the studied facilities (<10 000 PE). Technologies applied: AS – activated sludge, BF – biofilm, CW – constructed wetlands, SBR – sequencing batch reactors, OP – oxidation ponds.

2.2. A novel method for evaluating performance

The overall assessment of the treatment performance of WWTPs was based on the performance of individual treatment phases, but unlike Chen et al. (2015), the individual treatment steps had to be evaluated separately due to the great variance in technologies used and environmental objectives set for the evaluated WWTPs. To ensure comparability of individual WWTPs, the following prerequisites were set:

- The steps of treatment processes (primary, secondary, and tertiary treatment) are characteristic of all WWTPs.
- Different equipment and processes have the same function in the same treatment step (e.g. the bar screen and screw screen are both devices for removing particles from wastewater).
- All the processes and equipment having the same purpose at the same treatment step have to be comparable by setting specific critical control points (CCPs) for each treatment step.

A questionnaire was developed to assess WWTPs in situ. The questionnaire was divided into five main categories and 21 subcategories (Table 1). For each subcategory (treatment step), a minimum of four CCPs were set. Depending on the complexity of the wastewater treatment process the number of CCPs to be evaluated varied between 37 (two treatment steps – septic tank and oxidation ponds) and 170 (9 treatment steps – primarily treated wastewater was divided into two parallel treatment lines using activated sludge process or sequencing batch reactors, and the sewage sludge was stabilized on site).

To overcome the problem of assessment subjectivity, all the CCPs were formulated as multiple-choice questions with two to five alternative answers, but mainly in the form 'yes' or 'no'.

During the assessment of WWTPs the following parameters were evaluated on a scale of 10 points (Fig. 3):

- General complexity describes the situation where
 the complexity of each individual treatment step
 is not taken into consideration, but the number of
 treatment steps in the specific WWTP is divided
 by all possible treatment steps described in the
 model. General complexity defines all treatment
 steps that are to be evaluated for performance and
 complexity.
- Total evaluation of complexity (TEC) describes how many different treatment steps are involved in the wastewater treatment process and how sophisticated the technology is.
- Total evaluation of performance (TEP) describes all the factors that can influence the wastewater treatment process (e.g. quantity and composition of the influent, difference between design values and actual conditions, functionality of equipment, operational problems).

Evaluations of performance and complexity in each treatment step were established on the basis of the evaluation of the CCPs in the same step. In this paper CCPs are defined as factors that (i) influence the performance of the treatment step (e.g. growth of filamentous micro-organisms), (ii) describe operational conditions (e.g. surface of final clarifier is kept clean, pumps are in

Table 1. C	ategorization	of treatment s	stens used in	assessment
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Treatment step	Mandatory CCPs to be evaluated for all WWTPs	Selected steps of individual WWTPs
Wastewater characterization		
Characteristics of the influent	X	
Additional loading due to excess loading		X
Effluent quality	X	
2. Primary treatment		
Screens and sieves		X
Primary clarifier		X
Grit chamber and oil trap		X
Septic tank		X
3. Secondary treatment		
Activated sludge process		X
Sequencing batch reactor		X
Constructed wetlands		X
Biological filter		X
Submerged bed reactor		X
Biological contactor		X
Final clarifier		X
4. Tertiary treatment		
Nitrogen removal		X
Phosphorus removal		X
Effluent polishing		X
5. Other factors		
General aspects of wastewater treatment	X	
Automation		X
Sewage sludge treatment		X
Operator's competence	X	

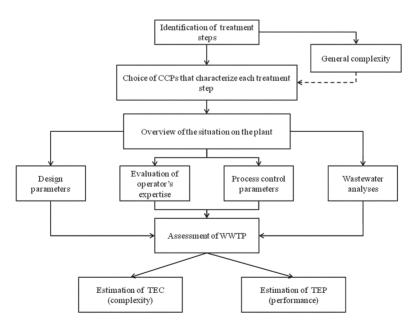


Fig. 3. Flow sheet of activities and data collection in WWTPs.

working order), or (iii) describe the complexity of the treatment step (e.g. screens are pressed and washed). For each wastewater treatment step, a minimum of two CCPs were defined. All CCPs were chosen by a group of experts following suggestions in the literature (Kuusik et al., 2001; Jenkins et al., 2004; Noorvee et al., 2007; Maastik et al., 2011; Baumann et al., 2012).

The relevance and values for each CCP that described the performance or complexity of a specific treatment step (P_i) were defined empirically. The following formula was used to calculate P_i :

$$P_{i} = \frac{\sum_{i=1}^{n} X_{i} \times Y_{i}}{\sum_{i=1}^{n} Y_{i}} \times 10,$$
 (1)

where P_i is the harmonized assessment of the treatment step on the scale of 10 points, X_i is the score on the CCP (between 0 and 1), and Y_i describes the expert opinion on the importance of the given CCP in the treatment step.

Based on each treatment step, TEP and TEC were calculated. A summary of the evaluations was formed from the weighted scores of the different stages as follows:

TEP =
$$\left[\sum_{i=1}^{n} a_i + \frac{\sum_{i=1}^{n} \left(\frac{P_i}{10} \times b_i\right)}{\sum_{i=1}^{n} b_i}\right] \times 10, \quad (2)$$

where TEP is harmonized performance assessment of the total treatment process on the scale of 10 points, a_i addresses some general aspects for the overall process control, P_i is harmonized performance assessment of the treatment step on the scale of 10 points, and b_i describes the expert opinion on the importance of the given treatment step in the whole wastewater treatment process.

For TEC similar calculations were made.

As assessing the impact of performance and complexity on the effluent quality can be problematic due to the varying quality requirements for the WWTPs by size, the index of effluent violations (IVE) was developed. The value of IVE was set on the 10 point scale. With the purpose of ensuring comparability of different WWTPs on the same basis, the following prerequisites were set: (i) for the effluents performing by up to 25% better than required by the discharge consent, IVE = 10; (ii) for the effluents exceeding quality standards by more than 300%, IVE = 0.

IVE is formed as an average compliance with effluent quality standards that are regulated by the plant's discharge consent:

IVE =
$$\frac{\sum_{i=1}^{n} \left(\frac{x_i}{y_i} \times 8 \right)}{n},$$
 (3)

where x_i is the result of effluent analysis for the component i (e.g. BOD_7), y_i is the effluent quality standard for the component i (e.g. BOD_7), and n is the number of components that were analysed and regulated by discharge consent.

2.3. Data collection, sampling, and laboratory analyses

The selected WWTPs were visited and assessed according to a uniform method over a period of six months, between October 2014 and March 2015. A total of 541 grab samples were collected, of which 241 samples were taken from the effluent of secondary treatment units and 94 from the effluent of tertiary treatment units. For the determination of mixed liquor suspended solids (MLSS) 94 samples were collected. No effluent samples were taken from two WWTPs as there was no outflow during the plant visit. For these two WWTPs analysis results from a national monitoring programme were used. The average wastewater temperature during the sampling session was 8.9 ± 3.3 °C.

The assessment was performed in the presence of a local operator. The following actions were performed (Fig. 3): (i) design parameters (e.g. flow rate, solids retention time (SRT) sludge volume index) were collected; (ii) actual situation on the plant (e.g. flow rate, SRT, sludge volume index, equipment failures) was documented (taking photos, filling in Excel sheets of the model); (iii) samples were collected from the effluent and process reactors to determine biological oxygen demand, chemical oxygen demand, total suspended solids, Kjeldahl nitrogen, and total phosphorous. The parameters pH, conductivity, dissolved oxygen, and water temperature were determined in situ. In addition to the data needed for the TEC and TEP, some additional data were collected during the plant inspection. These included the operator's knowledge about the process and evaluation of the operator's competence, the operator's contentment with each treatment step, and additional data that were not used in any assessment but were expected to be relevant in the interpretation of results.

All the wastewater grab samples were collected from the effluents of secondary and tertiary treatment processes, where available, according to ISO 5667-10 (ISO, 1992). Wastewater samples were stored and transported to the accredited laboratory according to ISO 5667-3 (ISO, 2018) and immediately analysed according to the standard methods in the laboratory.

2.4. Statistical analyses

For studying the relationship between the plant performance and WWTP complexity, tools of correlation and regression analyses were applied. Together with the Pearson coefficient, the Spearman correlation coefficient was also applied in those cases where the dependence between study variables was of monotonic type instead of linear. For categorical variables, analysis of variances (ANOVA) was used for comparing the population means of performance in different groups. In some cases where assumptions of ANOVA were not satisfied, the Kruskal–Wallis test was applied as an alternative. It decides whether the population distributions of the performance are identical in study groups or not. The analysis was carried out using the software R.

3. RESULTS AND DISCUSSION

3.1. Effluent quality

Final effluents have to meet effluent standards. Estonian regulation on effluent quality standards (Vabariigi Valitsus, 2013) sets limit values for the disposed effluent. In treatment plants with a loading rate of less than 300 PE only constructed wetlands met consent effluent standards. whereas WWTPs with different technologies did not perform that well. As many as 70% of activated sludge, 79% of sequencing batch reactors, and only 60% of fixed film (biofilm) reactors and oxidation ponds met consent effluent standards. In the smallest settlements (WWTP 300-1999 PE), where total N and total P limit values are also set, the effect of these two parameters is easily observed. A large number of WWTPs failed in all categories: 67% of sequencing batch reactors, 57% of activated sludge plants, 50% of oxidation ponds, only 33% of biofilm reactors, and none of the constructed wetlands met the consent of the effluent standards in this study.

3.2. Relationship between the performance and complexity of WWTPs

As expected, the complexity of the WWTPs increased with plant size and specific treatment steps (e.g. screens, primary clarifiers, biological nitrogen removal).

Statistical analysis revealed that all four WWTPs with designed loading exceeding 100 000 PE caused statistically significant distortion in the overall tendencies of data interpretation. Therefore, some of the following results were submitted with and some without these four WWTPs.

There was a rather good positive relationship between general complexity and design loading rate for all WWTPs (Fig. 4a) $(R^2 = 0.434, p\text{-value} = 1.863e^{-0.5}, Pearson's <math>r = 0.279$). For WWTPs with loading rates less than 100 000 PE, the relationship was more significant $(p\text{-value} = 1.35e^{-11}, Pearson's <math>r = 0.432)$ and monotonic (Spearman's $\rho = 0.631, p\text{-value} = 2.2e^{-16}$). This suggests that the structure of the wastewater treatment process tends to become more sophisticated with the growth of the design loading rate. It can easily be explained by the increasingly stricter requirements for effluent quality as plants (design) loading rates become higher.

As it is not always financially rational or feasible to choose the most complex solution for each wastewater treatment facility, the total evaluation of complexity showed a wide variation. Similarly to general complexity, the TEC was logarithmically growing in accordance with the design loading rate (Fig. 4b), but the variance was greater ($R^2 = 0.225$, p-value = 1.023e⁻⁰⁶, Pearson's r = 0.317). There was no great difference in Pearson's r $(r = 0.339 \text{ and } p\text{-value} = 2.025e^{-07})$ and Spearman's ρ $(\rho = 0.386, p\text{-value} = 2.249e^{-0.9})$ for WWTPs with loading rates less than 100 000 PE. This suggests that although the number of treatment steps increases with the plant size, the technical solution chosen for a certain treatment step might not be very sophisticated. For example, there were no screens in 11 out of 99 WWTPs with a design loading rate of less than 300 PE. In other WWTPs, the screening equipment was screw screens (27), step screens (26), bar screens (11), and also various other devices or solutions (24). The complexity of the screens varied between 0 and 8 points, which indicates clearly that the choice of treatment steps complexity was made in compliance with the availability of financial resources.

A statistically significant but weak relationship $(R^2 = 0.062, p\text{-value} = 5.566e^{-4}, Pearson's r = 0.227)$ was detected between TEP and the design loading rates (Fig. 4c). There was only a weak correlation between TEP and the design loading rate for WWTPs with the loading rate less than 100 000 PE (Pearson's r = 0.148, p-value = 0.027, and Spearman's $\rho = 0.144$, p-value = 0.031), which could be due to the capability to react to process disturbances in these plants. Three out of four WWTPs exceeding 100 000 PE had operations and maintenance personnel available 24/7, but among smaller WWTPs there was only one where this personnel were available all the time. At most of the small plants (less than 300 PE) such personnel were available for only 2-4 hours per week (during the scheduled supervision), and their ability to react to process disturbances was highly dependent on the discovery of a malfunction.

Figure 4d shows that there was a positive and moderate correlation between TEC and TEP ($R^2 = 0.228$,

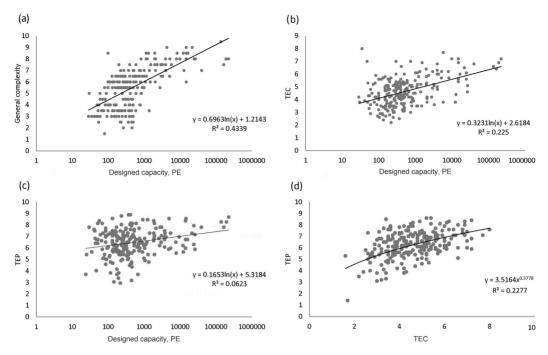


Fig. 4. Relationships between complexity, performance, and design loading rates.

Pearson's r = 0.413, p-value = $1.658e^{-11}$). This suggests that automation as a tool for the process control and equipment complexity in each treatment step could prevent many process disturbances and therefore improve the overall plant performance. For example, if the screens are not removed, either automatically or manually by an operator, clogging can happen, and as a result, the next biological process could be affected.

3.3. Relationship between the performance and complexity of treatment steps

For each treatment step, Pearson's correlation r, Student's t-test, and p-value were found for weighted complexity and performance in the given step (Table 2). In Table 2, r represents Pearson's product-moment correlation, t represents the values of Student's t-distribution, df represents the number of WWTPs where statistical analyses could be made (e.g. not all WWTPs had screens), and p-value shows the significance of this relationship.

Our study did not reveal any statistically significant (p-value < 0.05) correlation between the values of complexity and performance for some treatment steps

(Table 2). This can be explained by the characteristics of the influent. In this step the performance assessment is based on the condition of the sewer system, rate of infiltration, variations in the influent flow rate due to seasonal or weather fluctuations, and the influence of industrial wastewaters. The complexity assessment in this step is based on the available loading control methods in the WWTP (e.g. use of balancing tanks or overflows. difference between actual and design loadings). Pearson's product-moment correlation between complexity and performance in this treatment step is statistically not significant (p-value = 0.56) and very weak (r = 0.037)as the quality and quantity of the influent have a great impact on the following treatment steps; however, the possibilities for balancing and loading control of wastewater are not directly connected to the origin of the wastewater.

The complexity of a certain treatment step has a great impact on the performance of several treatment steps. The most significant relationships between complexity and performance were detected for automation $(p\text{-value} < 2.2e^{-16})$, screens $(p\text{-value} = 6.08e^{-12})$, final clarifier $(p\text{-value} = 1.39e^{7})$, and nitrogen removal $(p\text{-value} = 1.33e^{-6})$. The performance of a treatment step

Treatment step	t	df	<i>p</i> -value	r
Characteristics of the influent	0.582	243	0.561	0.037
Additional loading due to exhaustion	0.481	69	0.632	0.058
Screens and sieves	7.274	220	$6.083e^{-12}$	0.440
Primary clarifier	2.270	55	0.027	0.293
Grit chamber and oil trap	3.616	12	0.004	0.722
Septic tank	1.121	49	0.268	0.158
Activated sludge process	3.330	160	$1.263e^{-4}$	0.297
Sequencing batch reactor	0.775	32	0.444	0.136
Constructed wetlands	1.898	26	0.069	0.349
Biological filter	-0.856	15	0.406	-0.216
Submerged bed reactor	NA	2	NA	NA
Biological contactor	NA	1	NA	NA
Nitrogen removal	5.124	107	$1.333e^{-06}$	0.444
Phosphorus removal	1.051	199	0.295	0.074
Final clarifier	5.496	171	$1.394e^{-7}$	0.387
Automation	12.783	205	$< 2.2e^{-16}$	0.666
Effluent polishing	3.424	142	$8.059e^{-4}$	0.276
Sewage sludge treatment	2.862	30	0.008	0.463

Table 2. Pearson's product-moment correlation between TEC and TEP for each treatment step

NA – not applicable.

depended most strongly on the complexity in the given step for grit and oil removal (r = 0.72) and automation (r = 0.67). This suggests that for some treatment steps the complexity of equipment has a significant impact on their performance. For example, the risk for clogging and flooding of the screens can be reduced by using automatic removal of screens.

3.4. Relationship between performance and effluent quality

Performance assessment indexes proposed by Hao et al. (2013) and Chen et al. (2015) mainly focus on the effectiveness of pollutant removal, but they do not give any information about the overall situation on the plant. While Hao et al. (2013) took the operational situation into consideration (evaluation index K9), they did not specify what the 39 items that were inspected were and what their impact on the overall plant performance was. Although TEP does not give any specific information about problematic CCPs in the plant, conducting an assessment gives an operator a good indication how to start identifying problematic treatment steps within the WWTP.

Interpreting the results of statistical analyses of CCPs and studying their impact on the effluent quality revealed that several CCPs were extremely critical for effluent quality. For example, 50% of the WWTPs where effluents did not meet quality standards had a

scum layer on the surface of the final clarifier. Analysis of the factors that influence the IVE in activated sludge plants showed that IVE was dependent on three factors: the performance of the final clarifier ($P_{\rm final\ clarifier}$), the CCP that describes whether the results of effluent analyses meet the quality standards ($P_{\rm effluent\ analyses}$), and the CCP that describes whether there are any sensory indicators (colour, turbidity, smell) of poor effluent quality ($P_{\rm visual\-organoleptic\ assessment\ of\ effluent}$). The following model is used to estimate a WWTP's IVE:

$$IVE = 5.00 + 0.29 P_{\text{final clarifier}} + 0.21 \cdot P_{\text{effluent analyses}} + 0.60 \cdot P_{\text{visual-organoleptic assessment of effluent}}$$
(4)

It has to be underlined that low ratings for $P_{\text{effluent analyses}}$ and $P_{\text{visual-organoleptic assessment of effluentt}}$ could be a direct result of a poor performance of the final clarifier. This empirical model (r = 0.48, p-value 2.72e⁻¹⁰) showed clearly that for activated sludge plants the final clarifier could be the weakest point as its effluent quality was directly dependent on several operational and maintenance aspects (e.g. whether the surface of the clarifier was clean or the floating scum was discharged together with the effluent, whether there were any hydraulic problems or a malfunction of activated sludge pumps that influenced the sludge volume surface loading rate). In addition, as it was shown in Table 2, Pearson's product-moment correlation between TEC and TEP was significant $(p\text{-value} = 1.39e^{-7})$ for the final clarifier. It can be concluded that several problems could be prevented in the initial design phase or by using more automated facilities. For example, using balancing tanks for the reduction of fluctuations in the wastewater flow could prevent hydraulic problems in the final clarifier.

4. CONCLUSIONS

A novel method was developed to rapidly assess the performance and effectiveness of a large number of small WWTPs. The objective of the integrated assessment method was to create a comparable system of ratings for all types of wastewater treatment technologies and steps. Evaluations of performance and complexity were formed on the basis of treatment steps used in each WWTP and assessment of specific CCPs in the treatment steps.

Based on the evaluations of performance and complexity of all 245 inspected WWTPs, the following conclusions were drawn:

- The greater the design loading rate, the more treatment steps are usually needed, but the complexity of
 these treatment steps can vary significantly depending
 on plant size;
- No direct relationship was detected between the plant capacity and its performance. This suggests that operation and maintenance practices play an important role in the performance of a WWTP regardless of its size;
- There was a positive relationship between TEC and TEP: increasing complexity provided better performance of the WWTPs. This suggests that combining automation as a tool for the process control with a more advanced equipment (higher complexity) could prevent many process disturbances and therefore improve the overall plant performance (e.g. infiltration to the sewer system during the rainy period can cause bulking by lowering food to microorganisms ratios if no automatic bypasses had been built);
- In particular, the complexity of equipment of some treatment steps (e.g. screens, final clarifier) has a highly significant impact to the performance of the whole facility.

The proposed performance assessment method makes it possible to simultaneously analyse several factors such as influent characteristics, system complexity, operational practices, and process parameters, and so their impact on the overall system performance can be taken into consideration. The new standardized tools TEC and TEP make it possible to compare a wide range of WWTPs using different technologies and treating different loadings. In further analyses the impact of several factors (e.g. operator's competence, level of

automation, different wastewater treatment strategies) on the performance and effluent quality of the WWTP can be modelled.

Since the development process of TEC and TEP involved data from small-scale WWTPs only in Estonia and because of the varying maintenance and operational practices, further work would be suggested to apply these indexes in other regions and climatic conditions as well as to larger-scale WWTPs. It can also be suggested that the usage of the new assessment method be introduced on a large variety of WWTPs to improve the daily maintenance and operational practices (CCPs are always checked and critical parameters for system control are calculated simultaneously) and to develop a wide-ranging database that gives scientists and designers inputs for further research.

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Uudne meetod väikeste reoveepuhastite tõhususe ja komplekssuse kiireks hindamiseks

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Reoveepuhastite (WWTP) tõhusus sõltub mitmest tehnilisest, mittetehnilisest ja inimlikust faktorist. Aastatel 2014–2015 uuriti 245 väikest ja keskmise suurusega reoveepuhastit, mida hinnati uudse tõhususe ning komplekssuse hindamise metoodikaga. Meetod võimaldab samadel alustel hinnata erinevaid puhastustehnoloogiaid ja analüüsib reovee omadusi, süsteemi komplekssust ning käitamispraktikaid ja võrdleb protsessi reaalseid parameetreid projekteeritud väärtuste suhtes. Üldine komplekssus (TEC) ja tõhusus (TEP) on uued universaalsed tööriistad, mida rakendati erinevate koormuste ning tehnoloogiliste lahenduste hindamiseks.

Uuringu käigus selgus, et mida suurem on puhasti koormus, seda rohkem puhastusastmeid rakendatakse. Nende puhastusastmete komplekssus võib varieeruda suures ulatuses sõltuvalt puhasti koormusest. Tõhususe ja komplekssuse vahel tuvastati positiivne korrelatsioon: suurema komplekssusega puhastid olid tõhusamad. See näitab, et kui protsessi juhtimisel võtta kasutusele automaatika ja suurema keerukusastmega seadmed, on võimalik ennetada mitmeid protsessi seisakuid ning parandada kogu puhasti tööd.

Paper II

Vallo Kõrgmaa.; Mait Kriipsalu; Taavo Tenno; Erki Lember; Argo Kuusik; Vallo Lemmiksoo; Karin Pachel; Arvo Lital (2019). Factors affecting SVI in small scale WWTPs. *Water Science and Technology*, 79 (9), 1766–1776. DOI: 10.2166/wst.2019.177.

Factors affecting SVI in small scale WWTPs

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ABSTRACT

This paper analyses factors associated with bulking in 195 small scale wastewater treatment plants (WWTPs) in Estonia. Operational data from each plant were collected and analysed statistically. The key factors associated with bulking were infiltration into sewage pipes, the type and purpose of process reactor, operational practices and influent characteristics. Both anaerobic fraction and volumetric fraction of the anaerobic reactor compared to the aerobic reactor resulted in a positive correlation with sludge volume index (SVI) <150 ml/g values. Good operation and maintenance practice as well as an operator's competence play a crucial role in bulking prevention. Using the 30 minute settling test (V₃₀) as the single process control parameter can mislead an operator's judgement in process control strategies and cause effluent violations. Misjudgements in process control decisions can lead to unwanted conditions in small WWTPs (e.g. excessive chemical addition favoured bulking). Use of instrumentation, control and automation helped to keep the process conditions more stable and reduce the probability of bulking. Analyses of variance showed that the factors associated with Microthrix parvicella growth were long solids retention time (SRT), low foodto-microorganism ratio (F/M) and lack of carbon content compared against nitrogen and phosphorus contents

Key words | activated sludge process, analyses of variance, bulking, sludge volume index

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INTRODUCTION

The main goal of wastewater treatment is to reduce the concentration of pollutants in the effluent below the admissible level in order to eliminate the threat to the environment or to public health. Regardless of a large variety of wastewater treatment technologies (e.g. activated sludge (AS), biofilm or constructed wetlands), the performance of wastewater treatment plants (WWTPs) depends on various technical and non-technical factors such as characteristics of influent wastewater and how well these factors are in accordance with the designed treatment process, operational and management practices, reliability of equipment and flexibility of the process. In many cases, the process parameters in a WWTP can be modelled in detail based on designed values, characteristics of influent or even on the basis of metabolic reactions of specific groups of microorganisms (Henze et al. 2008). However, the actual situation at the plant can differ dramatically from the modelled result due to incorrect design parameters, inevitably fluctuating input parameters, equipment failure, maintenance requirements or sludge bulking and foaming.

Operators in small WWTPs often do not have resources for analysing mixed liquor suspended solids (MLSS). The determination of the 30 minute settling test (V₃₀) is the most commonly used operational parameter for AS process control in small scale (less than 50,000 PE) WWTPs in Estonia (Kõrgmaa et al. 2016). Unfortunately, if MLSS is not analysed, decisions based on the settling test could lead to inadequate sludge wasting and cause deviation from the targeted solids retention time (SRT). Sludge volume index (SVI), calculated as a quotient between V₃₀ and TSS, has an impact on the good performance of the final clarifier (Jenkins et al. 2004) and might cause deterioration from effluent quality limits. According to Bitton (2005), high SVI values (SVI >150 ml/g) can be associated with bulking sludge. The performance of an AS system for biological wastewater treatment is often

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deteriorated due to sludge separation problems caused by sludge bulking (Guo et al. 2014).

The common practice in describing reasons for bulking has been towards isolating a single cause, evaluating its impact and finding a solution for its removal. Unfortunately, bulking is often a result of several favourable factors happening at the same time (Guo et al. 2012). Bulking consists of filamentous bulking due to excess proliferation of filamentous bacteria (Eikelboom 2000) and non-filamentous bulking resulting from certain microbes that produce large amounts of extracellular material (Jenkins et al. 2004). There are several reasons for filamentous bulking, such as factors like low dissolved oxygen (DO) concentrations (Jenkins et al. 2004; Martins et al. 2004; Gerardi 2008), nutrient deficiency (Vaiopoulou et al. 2007; Gerardi 2008), low temperatures (Rosetti et al. 2005), low pH (Glymph 2005; Gerardi 2008) and septic wastewater (Glymph 2005; Gerardi 2008). Operational parameters like long SRT and low food-to-microorganism (F/M) values favour bulking (Guo et al. 2012; Li et al. 2016) even in AS systems where anaerobic selectors are present. In order to obtain well-settling sludge, the ratio of biochemical oxygen demand (BOD) to nitrogen (N) to phosphorus (P) in influent should generally satisfy 100:5:1 (Eikelboom 2000; Guo et al. 2014).

Research of AS bulking and foaming problems has a long history, even though the operational conditions under which bulking sludge occurs are usually only marginally documented (Martins et al. 2004). Although poor quality of influent creates prerequisites for bulking, it is important not to underestimate the operating conditions. The performance of WWTPs depends on various technical and non-technical factors such as characteristics of influent wastewater and how well these are in accordance with the designed treatment process, operational and management practices, reliability of equipment and flexibility of the process (Hegg et al. 1979; Olsson 2012; Hao et al. 2013). According to a survey in US municipal WWTPs, the average facility had 15 performance-limiting factors (e.g. infiltration, process controllability, equipment accessibility for maintenance) and at no facility was one single factor observed to be limiting the performance (Hegg et al. 1979).

This paper focuses on performance-limiting factors associated with bulking sludge. This study is based on the national survey of 245 small and medium size WWTPs that were studied during 2014-2015 and evaluated according to a novel method for rapid assessment of the

performance and complexity of small WWTPs. This paper focuses only on the findings from AS systems (n = 195).

MATERIALS AND METHODS

Method for evaluating performance

The overall assessment of the treatment performance of WWTPs was based on the performance of individual treatment phases, but unlike that described by Chen et al. (2015), the individual treatment steps had to be evaluated separately due to the great variance in technologies used and environmental objectives set for the evaluated WWTPs. For the purpose of ensuring comparability of individual WWTPs, the following prerequisites were set:

- The steps of treatment processes (primary, secondary and tertiary treatment) are characteristic for all WWTPs.
- Different equipment and processes have the same function in the same treatment step (e.g. the bar screen and screw screen are both devices for removing particles from wastewater).
- · All the processes and equipment having the same purpose at the same treatment step have to be comparable by setting specific critical control points (CCPs) for each treatment step.

A questionnaire was developed to assess WWTPs in situ. The questionnaire was divided into five main categories and 21 subcategories. For each subcategory (treatment step), a minimum of four CCPs were set. Depending on the complexity of the wastewater treatment process, the number of CCPs to be evaluated varied between 37 (two treatment steps – septic tank and oxidation ponds) and 170 (nine treatment steps – primarily treated wastewater was divided into two parallel treatment lines using an AS process or sequencing batch reactor (SBR), and the sewage sludge was stabilized on site). To overcome the problem of assessment subjectivity, all the CCPs were formulated as questions containing the choice of answers, which was set between two to five variables, mainly in the form of 'yes' or 'no'.

Evaluations of performance and complexity in each treatment step were established on the basis of evaluation of the CCPs in the same step (Kõrgmaa et al. 2019). In this paper, CCPs are defined as factors that (a) influence performance of the treatment step (e.g. growth of filamentous microorganisms), (b) describe operational conditions (e.g. the surface of the final clarifier is kept clean, pumps are in working order) or (c) describe the complexity of the

treatment step (e.g. screenings are pressed and washed). For each wastewater treatment step, a minimum of two CCPs were defined. All CCPs were chosen by a group of experts according to the literature (Kuusik *et al.* 2001; Jenkins *et al.* 2004; Maastik *et al.* 2011; Baumann *et al.* 2012).

Data collection, sampling and laboratory analyses

Most of Estonia is sparsely populated and, as a result, there are 664 municipal WWTPs for 1.35 million people (VEKA 2017). In total, 195 small scale (less than 50,000 PE) AS WWTPs were assessed in Estonia according to a uniform method over a period of 6 months between October 2014 and March 2015 (Figure 1). In total, 479 grab samples were collected, of which 193 samples were taken from the effluent of the secondary treatment and 94 from the effluent of tertiary treatment units. One hundred and ninety-two samples were collected for determination of MLSS. Effluent samples were not collected from two WWTPs, as there was

no outflow during the plant visit. Analysis results from a national monitoring program were used for these two WWTPs. In 15 WWTPs, composite samples from influent and effluent were collected and the microscopic examination of AS was carried out.

During the investigation, the following actions were performed: (a) design parameters (e.g. flow rate, SRT) were collected; (b) the actual situation of the plant (e.g. flow rate, SRT, SVI, equipment failures) was documented (taking photos, filling Excel sheets of the model); (c) samples from effluent and process reactors were collected to determine biological oxygen demand (BOD₇), chemical oxygen demand (COD), total suspended solids (TSS), total nitrogen (TN) and total phosphorus (TP). The following parameters: pH, conductivity (K), DO and water temperature (T°) were determined *in situ*. In addition to the data needed for evaluation of the WWTPs' performance, some additional data was collected during the plant inspection: (a) the operators' knowledge about the process and

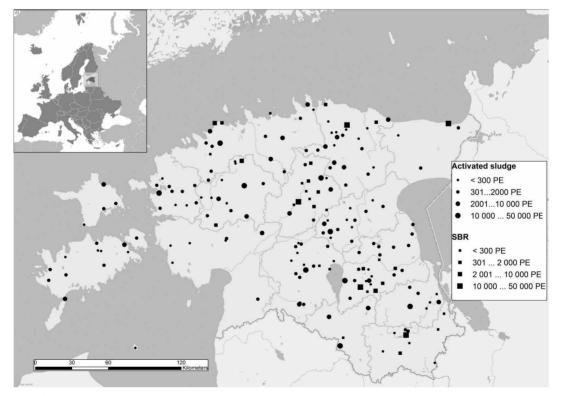


Figure 1 | Location and size of evaluated wastewater treatment plants in Estonia

evaluation of the operators' competence and (b) additional data that was not used in any assessment, but was expected to be relevant in the interpretation of results.

All the wastewater samples were collected according to ISO 5667-10. Wastewater samples were stored and transported to the accredited laboratory according to ISO 5667-3 and immediately analyzed according to the standard methods in the laboratory. Due to the financial limits, influent samples were not collected from all WWTPs and the data on influent quality was gathered from water companies using their analytical results. Statistical analyses do not include these results, as influent analyses did not often reflect an actual situation in the plant during the visit.

Statistical analyses

For studying the relationship between the plant performance, SVI and WWTP complexity, tools of correlation and regression analysis were applied. Together with the Pearson coefficient, the Spearman correlation coefficient was also applied in those cases, where the dependence between study variables was of the monotonic type instead of linear. For categorical variables, analysis of variance (ANOVA) was used for comparing the population means in different groups.

In order to find the frequency of coexisting problems in studied WWTPs (see Figure 2), matrix A was formed in such a way that findings of performance limiting factors in each

	A01	A02	A03	A04	A05	A06	A07	A08	A09	A10	A11	A12	A13	A14	A15	A16	A17	A18	A19	A20	
A01	1.00	0.28	0.32	0.31		0.35						0.28						0.29			A01
A02	0.40	1.00	0.51	0.47	0.52	0.45	0.44	0.50	0.44	0.44	0.45	0.46	0.41	0.56	0.42	0.43	0.27	0.42	0.43	0.44	A02
A03	0.21	0.24	1.00	0.22	0.21	0.20	0.28	0.30	0.19	0.26	0.16	0.20	0.27	0.22	0.08	0.23	0.23	0.25	0.29	0.21	A03
A04	0.73	0.80	0.80	1.00	0.75	0.76	0.76	0.74	0.72	0.75	0.76	0.74	0.81	0.74	0.83	0.72	0.82	0.74	0.76	0.74	A04
A05	0.49	0.43	0.37	0.37	1.00	0.47	0.28	0.31	0.38	0.36	0.35	0.38	0.27	0.42	0.25	0.44	0.32	0.40	0.38	0.42	A05
A06	0.62	0.57	0.54	0.57	0.71	1.00	0.53	0.57	0.60	0.61	0.57	0.56	0.52	0.57	0.42	0.56	0.55	0.54	0.57	0.51	A06
A07	0.35	0.39	0.54	0.40	0.30	0.37	1.00	0.72	0.44	0.40	0.46	0.55	0.38	0.48	0.42	0.45	0.50	0.45	0.43	0.49	A07
A08	0.57	0.63	0.80	0.55	0.47	0.56	1.00	1.00	0.55	0.60	0.59	0.66	0.58	0.72	0.42	0.62	0.55	0.63	0.57	0.61	A08
A09	0.59	0.57	0.51	0.55	0.59	0.61	0.64	0.57	1.00	0.65	0.73	0.70	0.49	0.57	0.58	0.55	0.55	0.56	0.52	0.63	A09
A10	0.40	0.36	0.46	0.36	0.36	0.40	0.37	0.39	0.42	1.00	0.46	0.46	0.45	0.41	0.33	0.33	0.36	0.33	0.43	0.35	A10
A11	0.60	0.56	0.44	0.57	0.53	0.57	0.65	0.60	0.71	0.71	1.00	0.74	0.51	0.74	0.42	0.60	0.55	0.58	0.52	0.58	A11
A12	0.41	0.49	0.46	0.47	0.48	0.47	0.65	0.56	0.58	0.60	0.63	1.00	0.49	0.68	0.50	0.53	0.68	0.55	0.48	0.53	A12
A13	0.41	0.34	0.49	0.40	0.27	0.34	0.36	0.39	0.32	0.46	0.34	0.39	1.00	0.46	0.42	0.36	0.41	0.38	0.48	0.40	A13
A14	0.44	0.51	0.44	0.41	0.47	0.41	0.50	0.53	0.41	0.46	0.55	0.59	0.51	1.00	0.33	0.48	0.50	0.44	0.43	0.42	A14
A15	0.13	0.06	0.02	0.07	0.04	0.05	0.06	0.05	0.06	0.06	0.05	0.06	0.07	0.05	1.00	0.07	0.09	0.06	0.00	0.14	A15
A16	0.44	0.45	0.54	0.46	0.56	0.48	0.54	0.53	0.46	0.43	0.51	0.54	0.47	0.56	0.58	1.00	0.95	0.84	0.57	0.70	A16
A17	0.08	0.07	0.12	0.12	0.10	0.11	0.14	0.11	0.11	0.11	0.11	0.16	0.12	0.14	0.17	0.22	1.00	0.20	0.10	0.19	A17
A18	0.51	0.52	0.66	0.55	0.60	0.53	0.63	0.63	0.55	0.50	0.58	0.65	0.58	0.59	0.58	0.98	1.00	1.00	0.71	0.75	A18
A19	0.11	0.10	0.15	0.11		0.11	0.12			0.13	0.10	0.11	0.14	0.11				0.14		0.16	
A20		0.28	0.29	0.28	0.33	0.26	0.36				0.30	0.32	0.32		0.67			0.39		1.00	A20
	A01	A02	A03	A04	A05	A06	A07	A08	A09	A10	A11	A12	A13	A14	A15	A16	A17	A18	A19	A20	
	SVI > 150	Design loading was not based on actual measurements	AS process tanks are over- or undersized	Industrial wastewater causes problems to the WWTP	Infiltration into the sewer system	Extreme peak flows	F/M > 0.15 g BHT7/g MLSS	Real SRT is less than needed	Problems with aeration system	Floating sludge in aerobic tank	Floating sludge in final clarifier	Mass surface loading rate is greater than 500 L/m2*h	Operator does not regulate RAS rate	Operator does not know WAS rate	WAS is not removed	Effluent BOD over the limit	Effluent COD over the limit	Effluent TSS over the limit	Effluent TN over the limit	Effluent TP over the limit	

Figure 2 | Frequency of co-existence of selected problems in Estonian WWTPs (n = 190).

WWTP were marked as '1' (the problem was recorded) and '0' (the problem was not recorded). Matrix B was formed as a multiplication of matrices A and its transposed matrix A^T as follows:

$$B = A^T A$$

Matrix B describes the co-existence of two problems (e.g. in total there were 63 WWTPs where SVI >150 ml/g and design loading was not based on actual measurements in 88 WWTPs; there were 25 cases where both problems co-existed). To find the frequency of co-existence of selected issues (matrix C), as described in Figure 2, all values (b) in each column in matrix B were divided by the total number of problems (x) that have been described in said column (e.g. SVI >150 ml/g was observed in 28% of WWTPs, where the design loading was not based on actual measurements, but measurements were absent in 40% of WWTPs, where SVI >150 ml/g was observed) as follows:

$$C = \begin{pmatrix} \frac{b_{11}}{x_1} & \cdots & \frac{b_{1n}}{x_n} \\ \vdots & \ddots & \vdots \\ \frac{b_{n1}}{x_1} & \cdots & \frac{b_{nn}}{x_n} \end{pmatrix}$$

RESULTS AND DISCUSSION

Most common problems in Estonian activated sludge plants

A wastewater treatment plant can produce good quality effluent even if there are several shortcomings. All the plants (n=195) visited during this study had some kind of problems, but the severity of these problems varied to a great magnitude. Most common issues were associated with effluent quality (56.4% of WWTPs had high TSS in the effluent), aeration problems (57.9%), foaming (38.1%), bulking (32.3%) and ensuring anaerobic (8.2%) and anoxic (15.4%) conditions in said reactors.

During the assessment it was observed that five WWTPs were at the stage of start-up of the process due to the loss of AS by serious hydraulic overloading. These plants were excluded from further analyses. The main approach for finding reasons for bulking is to identify the specific filamentous bacterium in the bulking sludge (Martins *et al.* 2004). Microscopic examination of AS was performed only in 15 WWTPs

(*Microthrix parvicella* was the dominant filamentous organism in 61.5% of samples), but as the reasons for foaming and bulking in the rest of the WWTPs were not known, statistical analyses in this paper focused mainly on CCPs.

Some of the factors triggering bulking are not easily traced (e.g. low DO concentrations could be the result of sudden load of readily biodegradable organic matter as well as lack of aeration capacity or poor maintenance of the aeration system or a combination of said factors), good design and construction quality as well as professional O&M practice can minimize the probability of bulking or its impact on effluent quality. In order to understand the complexity of the actual situation in the WWTP that could trigger the disturbances in the normal wastewater treatment process and favour bulking, most common problems and their coexistence in each WWTP were assessed.

Figure 2 gives an overview of the frequency of coexistence of the selected problems (n=20) in investigated AS plants. In total, 40 problems were initially analysed and similarly to the situation reported by Hegg *et al.* (1979), an average Estonian AS WWTP had 14.6 ± 3.9 issues. It can be seen from Figure 2 that if certain problems exist in the WWTP, the probability for the connected issue could be high. For example, column A15 shows that on 67% of occasions when the WAS was not removed, SVI >150 ml/g was observed, meanwhile the event of bulking could have several other initiators (only in 13% of WWTPs, where SVI >150 ml/g, was WAS not removed).

Although only 10 factors give significant (*p*-value <0.05) correlation with 'bad' SVI values (Table 1), the coexistence of multiple factors, as shown in Figure 2, does not necessarily mean the occurrence of bulking. For example, in 98 WWTPs the balancing tank was absent and 112 WWTPs had extreme peak flows, 78 WWTPs had F/M greater than 0.15 g BOD₇/g MLSS. There were 28 plants with combinations of all said problems. In all these 28 plants, SRT was less than needed, but only eight of them had SVI >150 ml/g. Figure 2 allows assessment of the possibility of bulking. For example, 42.4% of plants that had infiltration to the sewer system had SVI >150 ml/g and 66.7% of WWTPs, where waste AS was not removed, had the same issue.

Factors correlating with SVI

SVI is calculated as a quotient between V_{30} and MLSS, but statistical analyses revealed that there were statistically significant, but weak relationships between SVI and V_{30} (Pearson's r=0.215, p-value =0.004) and between SVI

Table 1 | Factors that had a statistically significant (p-value <0.05) or important (p-value < 0.10) correlation with 'good' SVI

	Pearson		Spearmar	1
Parameter	R	p- value	P	p- value
Performance of biological reactor (on the 10p scale)	0.247	0.001	0.234	0.003
30 minute settling test (V ₃₀), ml	-0.244	0.002	-0.226	0.004
Is the real mass surface loading rate in the final clarifier less than $500 \text{kg/(m}^2 \cdot \text{d})$?	-0.215	0.012	-0.215	0.012
Industrial sources in the influent cause problems to the WWTP	-0.380	0.022	-0.380	0.022
MLSS, g/l	-0.307	0.021	0.168	0.031
Effluent TSS, mg/l	0.096	0.226	0.163	0.038
Does an operator measure TP for process control?	-0.161	0.039	-0.161	0.039
Effluent TN, mgN/l	0.185	0.018	0.162	0.039
Is effluent quality within limits?	-0.146	0.061	-0.146	0.061
Are there any hydraulic problems in the final clarifier?	0.160	0.062	0.160	0.062
Is biological P-removal possible?	-0.148	0.064	-0.148	0.064
SRT(real)/SRT(designed), d/d	-0.088	0.358	0.172	0.071
Real SRT, d	-0.117	0.204	0.172	0.071
Performance of final clarifier (on the 10p scale)	-0.103	0.212	-0.148	0.083
Are WAS pumps working?	0.087	0.293	0.145	0.091
Is there any infiltration to the sewer system?	-0.132	0.093	-0.132	0.093
Volumetric fraction of anaerobic reactor (AN/OX) ^a	0.304	0.080	0.282	0.106
Anaerobic fraction (f _{AN}) ^a	0.313	0.071	0.264	0.132
Effluent TP, mgP/l	0.139	0.077	0.051	0.496

a Correlations calculated without certain package plants (n = 10). The reason for excluding these WWTPs was that these plants had a very big anaerobic reactor (f_{AM} = 0.5) that was not suitable for enhanced biological phosphorus removal (Gašparikova et al. 2005).

and MLSS (Pearson's r = -0.301, *p*-value = $4.11e^{-5}$), if looked at separately. This suggests that operational decisions based only on the measurement of sludge settleability can mislead the operator's judgement in process control strategies.

For statistical analyses, the SVI was divided into two groups with 'good' (SVI <150 ml/g) and 'bad' settleability (SVI >150 ml/g) and was evaluated with '1' or '0' respectively. All questions regarding CCPs were formed to be answered as 'ves' or 'no' and were evaluated with '1' or '0' respectively. Further analyses showed correlations as described in Table 1. Settleability was influenced by biological phosphorus removal, performance of process parts and influent sources.

Table 1 shows that the factors correlating with SVI could be divided into two groups: (a) factors that affect SVI (e.g. WAS removal, usage of remote control) and (b) factors that are affected by SVI (e.g. performance of the final clarifier, effluent quality). A negative correlation shows that an occurrence of the factor induces the bulking (e.g. if there are industrial sources in the influent, the bulking is more likely to occur).

The reasons for bulking could be divided into three groups: (a) influent characteristics, (b) design and construction (D&C) and (c) operation and maintenance (O&M). While influent characteristics involve factors like nutrient deficiency, low temperature and pH that have been reported to be the reasons for bulking (Eikelboom 2000; Jenkins et al. 2004; Gerardi 2008), the factors resulting from misgivings in the fields of D&C or O&M are sparsely reported.

Design and construction

Design and construction (D&C) shortcomings are not always easily found and need an inspection to be assessed. Even if expertise has been used, the correlation between D&C issues and bulking is not often reported. It can be also debated whether the simplicity of the process as a consequence of minimal investment possibilities could be the main reason for bulking (e.g. the absence of bypasses or grease separators). Absence of grease separators is a common problem in small WWTPs. Nielsen et al. (2002) reported that Microthrix parvicella took up oleic acid under both anaerobic and aerobic conditions, while only a few floc formers were able to take it up under anaerobic conditions.

One-way ANOVA showed that the statistically relevant (p-value <0.05) possible causes for bulking were (a) the type of biological reactor used, (b) infiltration to the sewer system and (c) use of phosphorus removal (bulking was observed at 45.9% of AS plants with bio-P). The choice of reactor type was important (p-value 0.07) if compared against 'good' SVI with one-way ANOVA. 69.8% of plugflow and SBR systems had SVI <150 ml/g. Meanwhile, only 56.3% of continuous stirred-tank reactors (CSTR) had similar SVI values. The balancing tank is usually needed to reduce variations in wastewater flow and concentrations. In many cases it helps to reduce the effect of peak flows and

Table 2 The design parameters of anaerobic reactors showed significant impact on the bulking according to the one-way ANOVA

Parameter	Unit	SVI <150 ml/g	Variance	SVI >150 ml/l	Variance	Number of WWTPs	Mean square	F-value	P-value
f_{OX}	-	0.772	0.038	0.739	0.032	188	0.045	1.250	0.265
f_{AN}	-	0.320	0.052	0.312	0.063	37	0.001	0.011	0.916
f_{AX}	-	0.355	0.019	0.363	0.019	122	0.002	0.101	0.751
AN/OX	$\mathrm{m}^3/\mathrm{m}^3$	0,400	0.135	0.187	0.011	37	0.428	5.352	0.027
AN/OX ^a	m^3/m^3	0.259	0.030	0.187	0.011	35	0.048	2.243	0.143
AX/OX	$\mathrm{m}^3/\mathrm{m}^3$	0.954	0.871	0.521	0.086	122	1.571	2.979	0.094
AN/AX	m^3/m^3	1.054	0.955	0.521	0.086	32	2.199	4.107	0.052
AN/(AX + OX)	m^3/m^3	0.291	0.066	0.135	0.005	32	0.223	5.820	0.021
$AN/(AX+OX)^a$	m^3/m^3	0.195	0.013	0.135	0.005	29	0.031	3.392	0.074
HRT_{AN}	d^{-1}	0.399	0.051	0.369	0.058	32	0.007	0.131	0.719
HRT_{AX}	d^{-1}	1.240	1.277	1.091	0.624	113	0.596	0.582	0.447
HRT _{OX}	d^{-1}	3.717	14.186	2.478	3.174	165	58.671	5.752	0.018

^aValues calculated without certain package plants. The reason for excluding these WWTPs was that an anaerobic tank used in these plants was not suitable for enhanced biological phosphorus removal.

the risk of hydraulic overloading. In small WWTPs, where the balancing tank was absent, the average SVI value was 134.7 ml/g (n = 90), whereas in WWTPs with a balancing tank the average SVI value was 173.0 ml/g (n = 57). The effect was significant (p-value 0.05), but could be misleading as an average HRT in balancing tanks was 1.6 d⁻¹. This period could be long enough for wastewater septicity to develop.

Selectors are usually considered to be an effective way of bulking control, but they do not work for all filamentous microorganisms (Martins et al. 2004). Due to the high requirements of effluent quality, 65.7% of Estonian AS WWTPs are designed with the possibility for biological nitrogen removal and 20.0% of plants have enhanced biological phosphorus removal. In total, there were 33 WWTPs included in this study with AAO configuration. Although one-way ANOVA showed that the volume fraction of an anaerobic reactor compared to volumes of aerobic (AN/OX) and/or anoxic reactors (AN/AX or AN/(AX+ OX)) could play a crucial part in the probability of sludge bulking (Table 2), the data was influenced by certain package plants that have big anaerobic tanks which are not suitable for enhanced biological phosphorus removal (Gašparikova et al. 2005). When these plants were excluded from the ANOVA analyses, the p-value of an anaerobic volume fraction was 0.14 for AN/OX and 0.07 for AN/ (AX + OX). An anaerobic fraction and volumetric fraction of anaerobic reactor compared to the aerobic reactor both give positive correlation with SVI <150 ml/g values, even without the mentioned package plants (for f_{AN} Pearson's r 0.313, p-value 0.07 and for AN/OX Pearson's r 0.304, p-value 0.08). Analyses of hydraulic retention time in anaerobic and anoxic reactors revealed that in small WWTPs the contact time was much higher than recommended by Henze et al. (2008) with average values for anaerobic reactors of $10.5 \pm 6.8 \, h$ and for anoxic reactors of 27.9 ± 24.0 h respectively.

While the impacts of reactor type and infiltration to the SVI could be explained by kinetic selection theory (e.g. infiltration causes the dilution of nutrients and therefore gives growth advantage to filamentous organisms with lower Ks values over floc-forming bacteria with higher K_s values), the role of the phosphorus removal process as the cause of bulking remains uncertain. Some of the possible explanations for connections between bulking and phosphorus removal have been presented by Nielsen et al. (2002, 2010) and Wang et al. (2014). Nielsen et al. (2010) suggested that soluble components, either from the wastewater or produced by hydrolysis in the anaerobic tank, are taken up for storage as polyhydroxyalkanoates (PHA)/lipids by the filamentous Microthrix parvicella, by PAOs (Accumulibacter), and by GAOs (Competibacter, Defluviicoccus). Wang et al. (2014) suggested that M. parvicella could take part in enhanced biological phosphorus removal. M. parvicella is the most dominant organism in Estonian WWTPs that is causing bulking.

Fan et al. (2017) reported that M. parvicella favoured lower temperature, alteration between anaerobic and aerobic conditions and long chain fatty acids (LCFA) in batch tests. In the anaerobic/aerobic alternation experiment

reported by Fan et al. (2017), the AN/OX ratio was 0.5 which seems to be favouring SVI <150 ml/g. As shown in Table 2, their study still showed that this was enough to initiate M. parvicella bulking with the presence of LCFAs as the sole carbon source. Their experiments with different water temperatures (13 °C and 20 °C) combined with anaerobic-aerobic conditions and LCFA feed also showed great differences in M. parvicella abundance, favouring colder temperatures. In real conditions, the number of factors occurring simultaneously could be unlimited (Figure 2 and Table 1) and the event of bulking could be initiated or even suppressed. Still, the role of the volume fraction of an anaerobic reactor could be significant and needs further investigation.

Operation and maintenance

O&M is dependent on the human factor. The importance of the human factor in wastewater treatment process has been described very briefly in literature (Hegg et al. 1979; Olsson 2012) and in many cases it is considered to be the main reason for poor process performance (Hegg et al. 1979). Hegg et al. (1979) listed improper operator application of concepts and testing to process control as well as inadequate understanding of sewage treatment as the two highest ranking factors contributing to poor plant performance. The competence of the operator has been outlined as one of the key factors for successful plant control (Hegg et al. 1979; Muga & Michelic 2008; Olsson 2012). The competence of the operators was evaluated during the data collection in the form of a hidden test on a scale of 10 points. The average result was 7.31 and the minimum result 1.5. Although statistical analyses revealed that operators' competence did not influence the bulking directly, it could have severe consequences for the WWTPs' performance. For example, in one SBR that was treating wastewater from a dairy factory, an operator observed that the AS had poor settling properties and added external activated sludge with good settling properties into his WWTP. The settling did not improve as the operator had not removed any WAS and as a result MLSS was 12 mg/l during the visit.

Procedures for operation and maintenance constitute the key factors for successful pollution removal. Table 1 shows that O&M factors also have low correlation with the event of bulking; these factors are statistically important (p-value >0.10), but might also be misleading. Some of the statistically important factors (e.g. control of chemical precipitation, use of back-up generators for instrumentation, control and automation (ICA)) show negative correlations where the desired effect should be positive. In some cases, correlations shown in Table 1 can be also misleading without further analysis.

The chemical precipitation of phosphorus thickens and compacts the AS (Lind 1998), but it might also initiate toxic effects and control over chemical addition is crucial (Suresh et al. 2018). Table 1 shows negative weak correlation (Pearson's r = -0.161, p-value 0.039) between bulking and the operators' claim regarding adjusting chemicals for phosphorus removal. Further analyses showed contradiction between the desired effect of chemical precipitation (less bulking) and the operators' claim was driven by the operators' tendency to add too much of chemicals. In WWTPs where operator claimed to be making adjustments, according to the real measurements the ratio of chemicals added to the amount actually needed was 1.6 ± 1.5 , but in the other group the same ratio was 1.3 ± 1.2 . Although the difference between groups was not statistically relevant (p-value 0.29), it shows that while operators make decisions on chemical addition based on effluent results (according to ANOVA, average TP was 2.4 mgP/l in the group that made adjustments against 4.3 mgP/l in the group that did not, p-value 0.02), they tend to add too much of iron salts and as a result it affects AS properties negatively.

ICA plays an important role in reaching operational goals (Olsson & Jeppsson 2006). 66.7% of studied WWTPs used ICA for process control. ANOVA showed that in WWTPs where ICA was used the average SVI was 136.0 ml/g, while in the other group it was 173.8 ml/g (p-value 0.02).

According to the ANOVA, there was no difference in SRT values if compared against plants with or without bulking, even though Spearman's P was 0.172 with a p-value of 0.071. Some of the filamentous organisms grow on a wide range of SRT (Jenkins et al. 2004; Martins et al. 2004). As microscopic examination was performed only in 13 WWTPs, it remains open how much bulking was influenced by SRT values.

Influent characteristics

Infiltration to the sewer system had a significant impact (p-value 0.03) on the SVI value. Average SVI value in WWTPs that had infiltration was 177.9 ml/g, while in the plants that did not have any infiltration it was 133.1 ml/g. Industrial sources caused problems in 35 WWTPs and the impact on the SVI value was important (p-value 0.07), causing bulking (an average SVI value of 180.4 ml/g). WWTPs without industrial sources had an average SVI value of 139.9 ml/g. Average wastewater temperature during the sampling session was 8.9 ± 3.3 °C and the impact of cold

 Table 3 | One-way ANOVA showed significant (p-value <0.05) difference in process control parameters and influent characteristics in WWTPs where Microthrix parvicella was found</th>

Parameter	Unit	With M. parvicella	Variance	Without M. parvicella	Variance	Number of WWTPs	Mean square	F-value	<i>P</i> -value
SRT	d	39.10	352.96	14.56	155.91	13	1,852.96	6.59	0.03
F/M	kgBOD ₇ / kgMLSS	0.033	2.721×10^{-4}	0.107	0.005	13	0.017	8.10	0.02
Filament index	_	4.13	1.55	2.00	0.00	13	13.89	14.05	3.22×10^{-3}
BOD_7	mgO_2/l	350.00	5,314.29	576.00	13,480.00	13	157,156.92	18.97	$1.14\!\times\! 10^{-3}$
COD	mgO_2/l	632.50	42,221.43	1,012.00	79,320.00	13	443,139.23	7.95	0.02
TSS	mg/l	263.00	3,609.14	433.20	23,171.20	13	89,132.43	8.31	0.01
BOD ₇ /N	_	4.15	1.41	6.07	2.37	13	11.33	6.44	0.03
BOD ₇ /P	-	28.71	104.46	47.57	147.07	13	1,094.74	9.13	0.01

water temperature could not be evaluated as there was no adjacent group available.

It was not possible to analyse influent quality in all WWTPs due to the financial limitations, but 24 h composite samples were collected from 15 AS plants. Microscopic analyses of AS were performed in 13 WWTPs. Analyses showed that Microthrix parvicella was dominant in eight WWTPs, and three plants had foaming problems caused by Nocardioforms. In these 15 plants, no correlation between SVI and influent parameters was observed, but significant correlations were found with the presence of M. parvicella (Table 3). One-way ANOVA showed that M. parvicella favoured long SRT and low F/M values, as described earlier by Fan et al. (2018) and Jenkins et al. (2004). Table 3 shows that M. parvicella favoured wastewaters with lower BOD7, COD and TSS content and a lack of carbon content compared against nitrogen and phosphorus contents.

CONCLUSIONS

Usually it is not possible to point out only a single reason behind high SVI values. An average Estonian AS WWTP had 14.6 ± 3.9 issues, as described above, and it is common that several problems occur simultaneously. In real conditions, the number of factors occurring simultaneously could be unlimited and the event of bulking could be initiated or even suppressed by a combination of the said issues. The best result is achieved by combining stable influent characteristics with good design solutions, and excellent operation and maintenance practices.

Influent characteristics have a significant influence on filamentous growth. *Microthrix parvicella* dominated in 65% of WWTPs where microscopic examination was performed. ANOVA showed that factors triggering *M. parvicella* growth were long SRT, low F/M and lack of carbon sources compared against nitrogen and phosphorus content. Infiltration had significant correlation with bulking in all WWTPs.

The increasing need for nitrogen and phosphorus removal has evoked an even wider use of selectors. In order to avoid bulking, designers should consider the purpose and type of the reactor. The reactor type was important (p-value 0.07) if compared against 'good' SVI with one-way ANOVA. 69.8% of plug-flow and SBR systems had SVI <150 ml/g; meanwhile, only 56.3% of continuous stirred-tank reactors (CSTR) had similar SVI values. An anaerobic fraction and volumetric fraction of anaerobic reactor compared to the aerobic reactor resulted both in a positive correlation with SVI <150 ml/g values (for f_{AN} Pearson's r 0.313, p-value 0.07 and for AN/OX Pearson's r 0.304, p-value 0.08). Analyses of hydraulic retention time in anaerobic and anoxic reactors revealed that in small WWTPs the contact time was much higher than recommended by Henze et al. (2008), with average values for an anaerobic reactor of 10.5 ± 6.8 h and for an anoxic reactor of $27.9 \pm 24.0 \, h$ respectively. The role of the volume fraction of the anaerobic reactor could be significant and needs further investigation.

Good operation and maintenance practice as well as operators' competence plays a crucial role in bulking prevention. Using V_{30} as the only process control parameter can mislead operators' judgement in process control strategies and cause effluent violations. Misjudgements in

process control decisions can lead to unwanted conditions in small WWTPs (e.g. in order to reduce effluent phosphorus too much, chemicals were added and this favoured bulking). Use of ICA helped to keep process conditions more stable and reduce the probability of bulking.

In this study, we demonstrated, that this approach could be used more widely. Statistical analyses of operational conditions (including influent characteristics and identification of filamentous organisms) on the broad range of WWTPs could simplify ascertainment and impact the assessment of the factors that affect bulking.

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Paper III

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Removal of hazardous substances in municipal wastewater treatment plants

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ABSTRACT

Chemical pollution poses a threat to the aquatic environment and to human health. Wastewater treatment plants are the last defensive line between the aquatic environment and emissions of pollutants. This study focuses on identification of most relevant hazardous substances in Estonian municipal wastewater and their fate in the treatment process. During this study, seasonal wastewater and sewage sludge samples were collected from nine municipal wastewater treatment plants and analyzed for 282 hazardous substances, including EU (n=45) and Estonian (n=31) priority substances. Results of this study show that several substances that are subject to international restrictions (e.g. Stockholm Convention) are still present in untreated sewage. Wastewater treatment systems that had a greater level of complexity (TEC >5) were more successful in removing hazardous substances. Statistical analyses showed that removal efficiency of organic hazardous substances had significant (p-value <0.05) linear correlation with removal efficiencies of chemical oxygen demand (COD) and total suspended solids (TSS), but a monotonic relationship with operators' competency. This study showed that operators' competency had a strong influence on the stability of the wastewater treatment efficiency and removal of organic hazardous substances. **Key words** | hazardous substances, municipal wastewater treatment, removal efficiency

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HIGHLIGHTS

- Wide range of hazardous substances (n = 282) was analysed from nine wastewater treatment plants. Removal efficiencies for said substances were found.
- Wastewater treatment systems that had a greater level of complexity were more successful in removing hazardous substances. Statistical analyses showed that removal efficiency of organic hazardous substances had significant linear correlation with removal efficiencies of COD and TSS, but a monotonic relationship with operators' competency.
- This study showed that operators' competency had a strong influence on the stability
 of the wastewater treatment efficiency and removal of organic hazardous
 substances.

INTRODUCTION

In Europe, the Water Framework Directive (2000/60/EC) established provision for a list of Priority Substances (EC 2000). Directive 2008/105/EC laid down environmental quality standards (EQS) for the first 33 priority substances

and eight other pollutants that were already regulated at Union level (EC 2008). Directive 2013/39/EU updated this list, concluding environmental quality standards to 45 priority substances (EC 2013). In addition to these

European-wide regulated substances Estonia has set quality standards (Minister of Environment 2019a) for six heavy metals (As, Ba, Cr, Sn, Zn, Cu), 11 pesticides (glyphosate, MCPA, chloromequat chloride, metazachlor, tebuconazole, dimethoate, clopyralid, spiroxamine, mancozeb, prothioconazole and 2,4-D), eight phenolic compounds, three volatile compounds (o-xylene, m,p-xylene, toluene) and for some other substances (C₁₀-C₄₀ hydrocarbons, fluorides). In order to ensure good quality of receiving bodies, Estonia has set quality standards for wastewater discharges (Minister of Environment 2019b) considering effluent from the wastewater treatment plants to be one of the most important point sources of hazardous substances to the aquatic environment (Clara et al., 2012).

There are two factors that basically define the effluent quality - characteristics of an influent and the performance of the municipal wastewater treatment plant (MWWTP). The performance of a WWTP depends on various technical and non-technical factors such as characteristics of influent wastewater and how well these are in accordance with the designed treatment process, operational and management practices, reliability of equipment and flexibility of the process (Hegg et al. 1979; Olsson 2012; Hao et al. 2013). Prasse et al. (2015) emphasized that as treatment technologies can also differ it is not clear to what extent the removal efficiencies of individual substances vary. Prasse et al. (2015) also outlined that chemical and biological assessment of wastewater treatment technologies were influenced by the sampling strategy and analytical methods used, therefore they suggested flow-proportional composite sampling in order to increase reliability. There are several articles available about removal of hazardous substances during the wastewater treatment process on a more general level (Clara et al. 2012; Gardner et al. 2013; Luo et al. 2014) or either focusing on certain substances (Clara et al. 2010; Pan et al. 2016; Ejhed et al. 2018), technologies (Gasperi et al. 2010; Gorito et al. 2017; Gruchlik et al. 2018) or smallscale solutions (Gros et al. 2017). According to Pomies et al. (2013) there are at least 18 models describing micropollutant removal in activated sludge processes available. Treatment technologies and conditions among MWWTPs can be standardized by indexes (Kõrgmaa et al. 2019), but influent characteristics are always community specific. Prasse et al. (2015) outlined that besides demographics on the sewer collection area and the number of facilities (e.g. hospitals, laundry services) that use specific hazardous substances, the proportion and composition of co-treated industrial wastewater has to be considered as having a great impact on certain substances in the influent.

In recent years there has been discussion on upgrading existing MWWTPs by adding tertiary treatment steps (e.g. filtration, ozonation) in order to reduce emissions of hazardous substances (Clara et al. 2012; Luo et al. 2014). Still, the decision about upgrading MWWTP should be made with regard to the situation where the control of influent wastewater is not possible and the 'polluter pays' principle does not have any desired effect. Whereas emissions of hazardous substances in industrial (indirect) discharges could be controlled by environmental permitting (2010/75/EU) (EC 2010) or by the contracting conditions between the industry and MWWTP (Finnish Water Utilities Association 2018), emissions in domestic wastewater are rather difficult to control. The mechanisms that are targeting reduction of the use of hazardous substances (e.g. REACH, Stockholm Convention) are working well on an industrial production level, but they are not capable of repealing emissions from widely used products that already contain prohibited hazardous substances.

In total, there are 668 municipal wastewater treatment plants in Estonia. 51.2% of the pollution load is treated in MWWTPs with capacity for more than 100,000 person equivalents (PEs) and 16.7% in MWWTPs that are less than 10,000 PEs. The variety of treatment technologies is also dependent on the plant's capacity. The activated sludge (AS) process has been the process most commonly used (335 plug-flow and 62 sequencing batch reactors), followed by natural-based solutions (including 118 oxidation ponds and 22 constructed wetlands) and biofilm reactors (127 plants). Most of the MWWTPs are small - 503 MWWTPs have a capacity less than 300 PEs, 128 for 300-2,000 PEs, 20 for 2000-10,000 PEs, 15 for 10,000-100,000 PEs and two MWWTPs are designed for more than 100,000 PEs. Maximum concentrations of nitrogen and phosphorous in effluents are legally determined for MWWTPs exceeding 300 PEs in Estonia. The performance of MWWTPs has been varying over a wide scale based on a survey of 245 Estonian MWWTPs (Kõrgmaa et al. 2019).

This study aims to increase the knowledge on the occurrence and fate of hazardous substances relevant from the aquatic environment protection perspective (Directive 2000/60/EC) in municipal wastewater treatment plants. The studied substances belonged to a wide range of chemical groups; for example, phthalates, pesticides, halogenated flame retardants and volatile compounds (Appendix A). This study was a part of the inventory of sources and flows of priority hazardous substances in Estonia. In order to improve the understanding of removal efficiencies of hazardous substances during the wastewater treatment

2013

Table 1 | Simplified description of selected wastewater treatment plants

Plant	capacity (PE)	Primary treatment	Biological treatment	N-removal	P-removal	Effluent polishing	Sludge treatment	Complexity	Operator
A	<300	Septic tank	Constructed wetland	No	No	No	No	3.3	0.6
В	300-1,999	$Screen + septic \ tank$	Oxidation pond	No	No	No	No	4.4	6.6
С	300-1,999	Screen + septic tank + grit separator	Biofilm	No	Chemical	Oxidation pond	No	4.9	0.8
D	2,000-9,999	Screen + grit separator	SBR	Yes	Biological + chemical	No	Composting	6.4	7.5
E	10,000 -99,999	Screen + grit separator	SBR	Yes	Chemical	No	Composting	6.9	2.0
F	10,000 -99,999	Screen + grit separator	Activated sludge/SBR	Yes	Biological + chemical	No	Anaerobic digestion	7.0	7.8
G	10,000 -99,999	Screen + grit separator	Activated sludge	Yes	Biological + chemical	No	Anaerobic digestion	6.9	8.9
Н	>100,000	Screen + grit separator + primary clarifier	Activated sludge	Yes	Biological + chemical	Drum filter	Anaerobic digestion	6.2	8.2
I	>100,000	Screen + grit separator + primary clarifier	Activated sludge	Yes	Chemical	Post- denitrification	Anaerobic digestion	5.5	8.4

process most widely used technologies in Estonia were included: constructed wetlands, soil beds, biofilm and activated sludge solutions. Additionally, the complexity of treatment plants and operators' competence was evaluated to estimate possible gaps between technological potential and achieved pollutant removal rate.

MATERIALS AND METHODS

Selection of wastewater treatment plants

This study involved nine MWWTPs (see Table 1) that were selected according to the following criteria: (a) loading this study covered 59.7% of the pollution load from MWWTPs in Estonia; (b) treatment technology selection covered most widely used solutions in Estonia, including small-scale solutions (e.g. biofilm reactors, constructed wetlands) as well as an activated sludge process (continuous flow and sequencing batch reactors (SBRs)); and (c) industrial load had to be less than 50% in order to minimize the impact of industrial wastewaters.

Method for assessment of the complexity of WWTPs and operators' competence

During the sampling event, an evaluation of operators' competence and WWTP complexity was carried out according to the methodology described by Kõrgmaa et al. (2019). This evaluation was based on the following prerequisites: (a) the steps of the treatment processes (primary, secondary and tertiary treatment) are characteristic for all WWTPs; (b) different equipment and processes have the same function in the same treatment step (e.g. the bar screen and screw screen are both devices for removing particles from wastewater); and (c) all the processes and equipment having the same purpose at the same treatment step have to be comparable by setting specific critical control points (CCPs) for each treatment step. In this paper, CCPs are defined as factors that describe the complexity of the treatment step (e.g. screenings are pressed and washed). For each wastewater treatment step, a minimum of two CCPs were defined. To overcome the problem of assessment subjectivity, all the CCPs were formulated as questions containing a choice of answers, which was set between two to five variables, mainly in the form of 'ves' or 'no'. In order to form an overall assessment each CCP was given a score (between 0 and 1) and a summary of evaluation (on the scale of 10 points) was formed from the weighted scores of the different treatment stages. The complexity of a WWTP was defined by Kõrgmaa et al. (2019) as the total evaluation of complexity (TEC) and it describes how many different treatment steps are involved in the wastewater treatment process and how sophisticated the technology is.

The assessment was performed in the presence of a local operator. During the plant visit, operator's competence (on a

scale of 10 points) was assessed in the form of a hidden test similarly to the complexity assessment.

Table 1 summarizes the main properties of selected wastewater treatment plants.

Sampling

Seasonal spot samples of influent and effluent water were collected between June 2017 and April 2018 from all MWWTPs according to ISO 5667-10. Samples of sewage sludge were collected according to ISO 5667-13. The flow-proportional composite sampling strategy for both influent and effluent of each MWWTP was not realistic, but from six MWWTPs time-proportional composite samples were collected in parallel for certain analyses (PAH, pesticides, alkylphenols, heavy metals and phthalates) in accordance with ISO 5667-3 in order to increase sampling reliability. In total, 72 spot samples, 48 composite samples and 32 sewage sludge samples were collected and analysed for 282 substances.

Chemical analysis

During the study, 282 hazardous substances were analyzed, including EU (n=45) and Estonian (n=31) priority substances. All analytical methods used in this study were accredited methods according to the standard ISO 17025.

Liquid chromatography (Agilent Technologies Infinity 1290) with tandem mass spectrometric and electrospray ionization (LC-ESI-MS/MS) (Agilent Technologies 6490 with JetStream ESI) was used for analysis of pesticides (including glyphosate), perfluorooctanesulfonate and its derivatives. Water samples were analyzed without any sample preparation, solid samples were analyzed after extraction procedures. Wastewater and sewage sludge pesticide samples were analyzed with the addition of OnlineSPE (Agilent Flexible Cube LC module).

Determination of selected organotin compounds was carried out with gas chromatograph and with tandem mass spectrometric analysis (Agilent Technologies 7890B/7000 system). Organotin compounds were alkylated with sodiumtetraethylborate, extracted with hexane and cleaned with a silica column. The same was used for different organic substances such as pesticides, chlorobenzenes, PCB-s, PAH-s etc, which had been extracted from the samples either with liquid-liquid extraction or with solid phase extraction. Phthalates were analysed after solid phase extraction with GC/MS/MS according to EVS-EN ISO 18856 in the case

of the water phase and with CEN/TS 16183 in the case of the solid phase. Chlorophenols were analyzed derivatization also with GC/MS/MS according to EVS-EN 12673 in the case of water and according to ISO 14154 in the case of soil. Extraction is carried out with n-hexane and in the case of solid samples with a mixture of acetone and n-hexane.

Determination of individual isomers of nonylphenol extracted from the samples was carried out by gas chromatography/mass spectrometry (GC/MS) by Agilent Technologies (7890B/5977A MSD). Water samples were analyzed according to ISO 24293 and solid samples according to CEN/TS 16182. GC/MS was used for determination of benzene and some derivatives with the headspace gas chromatographic method. Water samples were according to ISO 11423-1 and solid samples to ISO 22155.

Determination of the hydrocarbon oil index was carried out using gas chromatography with a flame ionization detector (Agilent Technologies 7890B). Water samples were prepared according to EVS-EN ISO 9377-2 and solid samples according to EVS-EN ISO 16703. Extraction was carried out with n-hexane and in the case of solid samples with a mixture of acetone and n-hexane.

Mercury was analyzed according to EVS-EN ISO 12846 with an Hg Analyzer (RA-915, Lumex). Other metals were analyzed according to EVS-EN ISO 11885 with inductively coupled plasma optical emission spectrometry (Vista – MPX Varian).

Statistical analyses

Chemical analyses with measurements below the limit of quantification (LOQ) were substituted with LOQ/ $\sqrt{2}$ prior to statistical evaluation. There is a possibility that this censoring will create some bias (Hesel 2005; Zeghnoun et al. 2007), but according to Vriens et al. (2019), in statistical approaches for multipollutant studies the use of more advanced techniques for handling undetectable levels is not supported. The Environmental Protection Agency (EPA 2000) guideline states also that if the rate of censoring is very high (greater than 50%) then focus should be put on the upper quantile of the contaminant distribution, or on the proportion of measurements above a certain critical level that is at or above the censoring limit. For studying the relationship between the MWWTP complexity, operators' competence and removal of substances, tools of correlation and regression analysis were applied. Together with the Pearson coefficient, the Spearman correlation coefficient was also applied in those cases, where the dependence between study variables was of the monotonic type instead

Table 2 | Substances found most frequently in the influent

Substance	Unit	Min	Мах	Average	Median	STDEV	N	Frequency in samples
Di-2-ethylhexylphthalate (DEHP)	μg/l	0.3	16	3.93	2.90	3.32	36	100%
Fluoride (F ⁻)	mg/l	0.15	1.5	0.45	0.38	0.26	36	100%
Toluene	μg/l	0.3	75	9.85	4.25	15.92	36	100%
Diisobuthyl-phthalate (DIBP)	μg/l	0.36	7.2	1.39	1.00	1.19	35	97%
Diclofenac	μg/l	0.14	23	4.20	2.90	4.64	35	97%
p/m-cresole	μg/l	9.9	980	201.03	100.00	221.57	35	97%
Phenol	μg/l	2.8	270	60.69	30.00	74.82	34	94%
Diethylphthalate (DET)	μg/l	0.33	2.9	1.27	0.99	0.60	32	89%
PBDE 47	μg/l	5.2E-05	0.0043	7.85E-04	5.90E-04	8.44E-04	31	86%
Resorcinol	μg/l	5	130	53.37	47.00	34.91	31	86%

of linear. For categorical variables, analysis of variance (ANOVA) was used for comparing the population means in different groups. p-values of 0.05 or lower were considered to indicate statistical significance.

RESULTS AND DISCUSSION

Overview of detected hazardous substances

From 282 substances, only 45 (1,2,3,4,6,7,8-H7CDF, 1,2,3,6, 7,8-H6CDF, 1,2,3,7,8,9-H6CDD, 1,2,3,7,8,9-H6CDF, 1,2,3,4, 7,8-H6CDF, 1,2,3,7,8-P5CDF, 1,2,3-trichlorobenzene, 1,3, 5-trichlorobenzene, 2,3,4,6,7,8-H6CDF, 2,3,4,6,7,8-H6CDF, 2,3,4,7,8-P5CDF, 2,3,4,7,8-P5CDF, 2,3,7,8-T4CDD, 2,3,7,8-T4CDD, 2,3,7,8-T4CDF, aclonifen, alachlor, aldine, alphaendosulfan, atrazine, bifenox, delta-hexachlorocyclohexane, dieldrin, dichlorophos, dimethoate, endrin, epsilon-hexachlorocyclohexane, hexachlorobutadiene, isodrine, chlorofenvinphos, chlorpyrifos, metazachlor, PCB-114, PCB-123, PCB-126, PCB-156, PCB-157, PCB-169, PCB-189, PCB-77, PCB-81, pentachlorobenzene, simazine, trifluralin, cybutrine) were not detected above the LOQ from any of the analysed samples.

Di-2-ethylhexylphthalate (DEHP), toluene and heavy metals (As, Ba, Ni, Pb, Zn) were detected above LOQ from all samples. One hundred and twenty substances were found below LOQ in the influent. Table 2 shows the ten most frequently found substances in the influent samples. For some of the substances, seasonal patterns (e.g. diuron, glyphosate, trichloromethane were found in the summer and autumn samples) or connections to population density (e.g. tetrachloroethene was detected only from MWWTPs with capacity more than 10,000 PEs or median concentrations of trichloromethane increased linearly according to the plant size) could be detected.

One hundred and fifty-seven substances were found below the LOQ in effluent. Fluoride and diclofenac were the most frequently detected substances (Table 3).

Sewage sludge contained most of the substances above LOQ. In total, 16 substances (see Table 4) were found from all samples. Only 65 substances were not detected from any of the samples.

Removal efficiency of hazardous substances in municipal wastewater treatment plants

Municipal wastewater treatment plants are designed to reduce the pollution load to the environment. While nutrients and many other substances could be efficiently and consistently eliminated, the removal of hazardous substances is often insufficient (Luo et al. 2014).

In order to understand whether the control of emissions of hazardous substances might be reduced by better process control, limiting the industrial discharges to the public sewer system or upgrading the existing technology, it is crucial to understand the fate and removal efficiency of said substances during the wastewater treatment process. Still, it has to be underlined that while discussing the removal of hazardous substances during the wastewater treatment process, the discussion generally refers to the removal of parent compounds from the aqueous phase (Luo et al. 2014). Sewage sludge analyses strongly indicate that for most substances an accumulation to the biomass has taken place. For example, during this study DEHP was detected from all influent samples above

Table 3 | Substances found most frequently in the effluent

Substance	Unit	Min	Мах	Average	Median	STDEV	N	Frequency in samples
Fluoride (F ⁻)	mg/l	0.15	0.8	0.39	0.34	0.19	36	100%
Diclofenac	μg/l	0.18	22	3.84	3.10	3.94	35	97%
Tebuconazole	μg/l	0.0052	0.054	0.02	0.01	0.01	24	67%
Trichloromethane	μg/l	0.03	1	0.22	0.09	0.30	24	67%
Propiconazole	μg/l	0.005	1.05	0.14	0.01	0.32	22	61%
4-tert-octylphenol	μg/l	0.003	0.039	0.01	0.01	0.01	17	47%
Toluene	μg/l	0.1	5.8	0.87	0.20	1.66	15	42%
Arsenic (As)	μg/l	0.25	0.67	0.46	0.45	0.14	14	39%
Acenaphthylene	μg/l	0.005	0.033	0.01	0.01	0.01	14	39%
Barium (Ba)	μg/l	11	150	57.79	51.50	44.60	14	39%
Nickel (Ni)	μg/l	0.54	5.6	3.19	3.30	1.76	14	39%

Table 4 | Substances found most frequently in the sewage sludge

Substance	Unit	Min	Мах	Avearge	Median	N	Frequency in samples
2.4-Dichlorophenol/2.5-Dichlorophenol	μg/kg dm	5.3	110	28.2	22.5	30	100%
4-chlorophenol	μg/kg dm	1.5	75	18.1	12.0	30	100%
4-Nonylphenol	mg/kg dm	0.095	299	19.7	0.84	30	100%
4-tert-Octylphenol	mg/kg dm	0.002	87	3.7	0.07	30	100%
Barium (Ba)	mg/kg dm	93	1,100	374.4	300.0	30	100%
Di-2-ethylhexylphthalate (DEHP)	mg/kg dm	0.54	54	12.9	11.0	30	100%
Dibuthylphthalate (DBP)	mg/kg dm	0.07	0.65	0.2	0.19	30	100%
Diclofenac	μg/kg dm	3.6	1,920	404.4	245.0	30	100%
Mercury (Hg)	mg/kg dm	0.05	1.3	0.4	0.35	30	100%
Chromium (Cr)	mg/kg dm	1.6	60	27.7	23.0	30	100%
Nickel (Ni)	mg/kg dm	5	29	15.5	15.0	30	100%
Nonylphenols	mg/kg dm	0.095	904	51.2	0.84	30	100%
p.p´-DDE	μg/kg dm	1.9	24	8.3	6.40	30	100%
Lead (Pb)	mg/kg dm	5	23	11.7	11.0	30	100%
Zinc (Zn)	mg/kg dm	120	830	534.5	555.0	30	100%
Copper (Cu)	mg/kg dm	14	320	162.2	157.5	30	100%

LOQ and only 25 per cent of effluent results exceeded LOQ, but all sewage sludge samples contained high levels of DEHP. Average removal efficiency was 69.1% for DEHP. This means that while DEHP is quite successfully removed from the aqueous phase, it is merely transferred to the sewage sludge.

Table 5 shows removal efficiencies calculated based on grab and composite samples respectively. Although Prasse *et al.* (2015) stated that grab sampling of influent and effluent wastewater is inappropriate to determine elimination

efficiencies of MWWTPs as concentrations of hazardous substances might vary significantly over time, it was not possible to avoid grab sampling in order to satisfy the sampling conditions set in ISO 5667-3 for certain substances. Removal efficiencies expressed in Table 5 should therefore be regarded with certain reservations, as grab sampling can be considered random. As a result, for some of the substances (e.g. boscalid, diclofenac), the removal efficiency was negative. Negative removal efficiency (-85.5%) of aminomethylphosphonic acid (AMPA) indicates that

2017

 Table 5 | Removal efficiencies of selected hazardous substances in municipal wastewater treatment plants

Substance	Min (%)	25-Percentile (%)	Median (%)	75-Percentile (%)	Max (%)
Heavy metals					
Barium (Ba)	14.0	40.7	54.5	73.0	84.5
Mercury (Hg)	-85.0	65.2	85.3	90.1	97.5
Nickel (Ni)	1.0	63.6	74.5	86.7	92.9
Lead (Pb)	84.4	97.0	98.4	99.5	99.6
Zinc (Zn)	-720.2	57.9	67.8	81.0	94.8
Copper (Cu)	-34.4	60.2	70.7	82.9	95.1
Organotin compounds					
Monobutyltin (MBT)*	30.0	100.0	100.0	100.0	100.0
Monooctyltin (MOT)*	-130.0	80.2	100.0	100.0	100.0
Pesticides					
AMPA*	-288.9	-114.3	-66.9	0.7	40.9
Boscalid*	-800.0	-425.0	-180.4	-45.0	50.0
Glyphosate*	-178.3	-51.3	30.7	100.0	100.0
Propiconazole	-517.6	0.0	13.4	34.8	100.0
Tebuconazole	-182.4	-32.5	0.9	64.9	100.0
Terbutryn	-100.0	-47.9	-27.3	6.9	100.0
Phenols					
o-Cresol*	-72.7	100.0	100.0	100.0	100.0
p,m-Cresol*	-110.0	99.4	100.0	100.0	100.0
Phenol*	-18.5	100.0	100.0	100.0	100.0
Resorcinol*	52.3	100.0	100.0	100.0	100.0
Phthalates					
Di-2-ethylhexylphthalate (DEHP)	-84.8	63.2	84.8	94.7	98.6
Dibutyl phthalate (DBP)	-201.7	45.6	51.8	65.8	83.7
Diethyl phthalate (DET)	44.4	82.3	83.7	87.0	93.4
Diisobutyl phthalate (DIBP)	12.0	67.2	76.4	79.8	92.1
Polycyclic aromatic hydrocarbons					
Acenaphthene	55.8	61.7	79.0	89.3	92.5
Acenaphthylene	-296.0	41.5	48.1	79.4	99.6
Anthracene	49.5	64.3	70.5	85.7	96.0
Fluoranthene	-578.8	70.5	80.4	85.9	95.6
Fluorene	72.8	84.5	90.8	92.9	98.1
Naphthalene	49.5	90.9	96.5	98.7	99.9
Phenanthrene	-324.3	91.2	94.8	96.6	98.8
Pyrene	-861.7	80.4	88.6	93.3	98.1
Volatile organic compounds					
Styrene*	-70.0	68.1	100.0	100.0	100.0
Tetrachloroethene (PER)*	17.1	79.5	92.7	99.0	100.0
Toluene*	-262.5	96.3	100.0	100.0	100.0
Trichloromethane*	-100.0	28.0	55.6	73.0	100.0

(continued)

Substance	Min (%)	25-Percentile (%)	Median (%)	75-Percentile (%)	Max (%)
Other substances					
Hydrocarbon oil index (hydrocarbons C10-C40)*	68.4	100.0	100.0	100.0	100.0
Perfluorobutanoic acid (PFBA)*	-17.4	27.3	46.4	89.2	100.0
Diclofenac*	-1285.0	-26.0	4.3	30.9	100.0
4-tert-octylphenol	22.8	45.3	61.4	79.5	89.2
4-nonylphenol	23.1	54.1	79.0	90.9	95.0
Fluoride (F-)	-233.3	-4.1	10.7	26.0	74.7
COD	53.3	87.3	92.9	96.4	99.5
BOD_7	90.0	97.4	98.6	99.4	99.8
TSS	-160.0	93.8	97.7	98.6	99.7

^{*}Calculated from grab samples.

Table 6 | One-way ANOVA showed that higher complexity increases removal efficiencies

	TEC < 5			TEC > 5	TEC > 5					
Parameter	Average	Variance	Count	Average	Variance	Count	Mean square	F-value	P-value	F-critical
\sum (238 substances)	76.8	1028.5	11	91.2	83.8	24	1558.4	4.2	0.048	4.1
COD	72.6	679.7	12	93.2	19.3	21	3239.1	12.8	0.001	4.2
BOD	90.1	156.7	12	98.3	4.8	21	507.1	8.6	0.006	4.2
TSS	62.0	5215.4	12	96.8	8.4	21	9254.3	5.0	0.033	4.2

this substance is formed during the biological wastewater treatment process. AMPA is a metabolite of microbial degradation of widely used herbicide glyphosate (Struger et al. 2015). Some of the negative removal efficiencies in Table 5 present the situation when MWWTP A was hydraulically overloaded during the snow melting period, resulting in wash-out of filter solids. During this event, it was noticed that MWWTP A was still able to remove 91.8% of BOD₇, but chemical oxygen demand (COD) removal had reduced to 53.3%. Total suspended solids (TSS) and PAH removal efficiencies were negative. During the event, two-ring compound naphthalene was still degraded (49.5% was removed from wastewater), but removal efficiencies for four-ring compounds fluoranthene and pyrene, which need longer degradation time (Bouchez et al. 1996; Moscoso et al. 2015), were negative and wash-out of these compounds was observed.

Statistical analyses showed that removal efficiency of 238 organic hazardous substances had significant (p-value <0.05) linear correlation with removal efficiencies of COD (Pearson's r=0.663) and TSS (Pearson's r=0.869), but a monotonic relationship with operators' competency (Spearman's P=0.339).

One-way ANOVA showed that there was significant difference in COD, BOD₇, TSS and organic hazardous substances removal efficiencies between less complex (TEC <5) and more advanced treatment technologies (Table 6). MWWTPs with greater complexity (TEC >5) had higher removal efficiency for said substances. This indicates that activated sludge systems could be more successful in removing pollutants from the water phase.

Estimation of yearly emissions to the environment

For Estonia, an estimation of loads of hazardous substances that end up in the different environmental compartments (mainly water and soil) was made based on the study results. The annual flow rates for each MWWTP were retrieved from the national water usage database (estimation based on 2017 flow rates). To estimate emissions of hazardous substances for all the country, all Estonian municipal WWTPs were divided into three groups according to their loadings (group I < 10,000 PEs, group II – 10,001 to 99,999 PEs, group III – more than 100,000 PEs). To calculate emissions for individual MWWTPs not having measured results,

Table 7 Load estimation for some relevant hazardous substances in Estonian MWWTPs in 2017 (kg/year)

Substance	Influent kg/y	Effluent kg/y	Influent – effluent kg/y	Sewage sludge kg/y
Zinc (Zn)	4,923	1,533	3,390	14,888
Nickel (Ni)	648	171	477	477
Resorcinol	2,041	116	1,926	125
Chromium (Cr)	1,030	87	943	943
AMPA	27	48	-21	12
Di-2-ethylhexylphthalate (DEHP)	133	42	91	302
Glyphosate	16	26	-10	6
Lead (Pb)	406	18	388	388
Trichloromethane (chloroform)	31	15	16	4
4-Nonylphenol	5	0	5	26
Tetrachloroethylene	6	1.5	4	1.4
Toluene	169	8	161	27
Prothioconazole	4.38	2.71	1.67	0.74
Diuron	1.5	1.9	-0.4	0.1

median concentrations were calculated for each hazardous substance within the group (e.g. to calculate pyrene concentrations for group B, median concentrations from MWWTPs E, F and G were used) and multiplied by the annual flow rate of said MWWTP.

Table 7 shows an estimation of loadings of some hazardous substances. The determination of hazardous substances depends on their properties and their behaviour in analytical matrices. There are a number of compounds which, for example, do not dissolve well in water and therefore may even be adsorbed to particulate matter. This in turn might result in a situation where these compounds are determined, for example, in sewage sludge but not from the influent. Table 7 shows in *italics* the loads of hazardous substances entering the wastewater treatment plant that were below the LOQ in all influent samples, but were present in the effluent and/or sewage sludge. These loads were recalculated based on the total load of said substance in the effluent and in the sewage sludge.

Due to the randomness of spot sampling and the behaviour of substances in the wastewater treatment plant (depending on the properties of the substance it accumulates, volatilizes or decomposes during the treatment process), the loss of substances (see Table 7 column 'Influent – effluent') during the treatment process was also assessed when compiling the mass balance of hazardous substances.

The term 'loss' herein does not describe so much the loss of material in the process as it was used to evaluate the difference between the efficiency of the purification process and the accumulation in the sediment. For some substances, this allows an assessment of their fate in the wastewater treatment plant. For example, a total of 4.38 kg/year of prothioconazole is conveyed to Estonian WWTP through the influent and 2.71 kg/year is discharged into the environment with effluent. 1.67 kg/year of prothioconazole should be accumulated to the sewage sludge based on the difference between influent and effluent loads, but 0.74 kg/year of prothioconazole was estimated based on analytical results. However, further analysis of the results revealed that degradation of prothioconazole had taken place and 0.63 kg/year of the prothioconazole-destio was present in the sewage sludge. Similarly, influent contained 1,433 kg/year of phenol, 114 kg/year was emitted with effluent and 92 kg/ year was found from sewage sludge. The 1,227 kg/year that was missing from the balance was most likely degraded during the treatment process.

Effect of operators' competency

The importance of the human factor in the wastewater treatment process has been described very briefly in literature (Hegg et al. 1979; Olsson 2012) and in many cases it is considered to be the main reason for poor process performance (Hegg et al. 1979). Hegg et al. (1979) listed improper operator application of concepts and testing to process control as well as inadequate understanding of sewage treatment as the two highest ranking factors contributing to poor plant performance. The competence of the operator has been outlined as one of the key factors for successful plant control (Hegg et al. 1979; Muga & Michelic 2008; Olsson 2012). The competence of the operators was evaluated during the data collection in the form of a hidden test on the scale of 10 points according to the methodology described in Kõrgmaa et al. (2019). The operators' competence was in the range between 0.6 and 8.9 with an average result of 5.7 points.

Statistical analyses showed that operators' competency had moderate, but significant (p-value <0.05) correlation with removal efficiencies of COD (Pearson's r=0.474), arsenic (r=0.485), BOD $_7$ (r=0.526), and naphthalene (r=0.530). Strong correlation was observed for removal efficiencies of lead (Pearson's r=0.603), fluoranthene (r=0.617), pyrene (r=0.660) and chrysene (r=0.696). Significant (p-value <0.05) monotonic relationship was observed between operators' competency and removal efficiencies of

nickel (Spearman's $\rho=-0.504$), COD ($\rho=0.472$), 4-tert-octylphenol ($\rho=-0.658$), phentanthrene ($\rho=0.569$), fluorene ($\rho=0.603$), fluoranthene ($\rho=0.641$), and anthracene ($\rho=0.731$).

Operators' competence had a strong influence on the stability of the wastewater treatment efficiency. A competent operator is more successful in ensuring stable COD removal and in avoiding decrease in process performance. While taking into consideration that there was significant (p-value $3.54e^{-5}$) moderate correlation (Pearson's r = 0.663) between COD and removal of organic hazardous substances, it can be concluded that operators' competency plays the crucial role in successful removal of hazardous substances.

CONCLUSIONS

This study showed that many substances that are subject to international restrictions (e.g. di-2-ethylhexylphthalate, hexabromocyclododecane) are still present in raw sewage and treated effluent. For most of the substances, removal from the water phase in MWWTPs was observed but the chemical analyses of sewage sludge indicate that pollutants are often transmitted from water to the biomass. In order to understand whether the control of emissions of hazardous substances might be reduced by better process control, limiting industrial discharges to the public sewer system or upgrading the existing technology, it is crucial to understand the removal efficiency of said substances during the wastewater treatment process.

Wastewater treatment systems that had a greater level of complexity (TEC >5) were more successful in removing hazardous substances. Statistical analyses showed that removal efficiency of organic hazardous substances had significant (p-value <0.05) linear correlation with removal efficiencies of COD and TSS, but a monotonic relationship with operators' competency. Increasing operators' competency will help to reduce emissions of hazardous substances to the environment. This study showed that operators' competency had a strong influence on the stability of the wastewater treatment efficiency and removal of organic hazardous substances.

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SUPPLEMENTARY MATERIAL

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2017–2020 Estonian Waterworks Association, leading expert

2018—... Järvamaa Vocational Education Centre, lecturer (wastewater

treatment)

2013–2016 Tallinn University of Technology, lecturer (wastewater

treatment)

R&D related managerial and administrative work

2019- ... Head of Water Quality Management Working Group (within

LIFE IP CleanEst project)

2019-... Member of River Basin Management Plan Working Group

(within LIFE IP CleanEst project)

2019– ... Member of Ecosystem Services Working Group (within LIFE IP

CleanEst project)

2017–... Member of the professional committee of the water

treatment operator

2015-... Member of the evaluation committee for the water

management programme in Estonian Investment Centre

2015–2019 Member of the Water Sampling Attestation Committee

In-service trainings

2019 Interpreting Surface Water Monitoring Data: Terminology,

Models and Tiered Schemes in Mixture Risk Assessment,

SETAC, Helsinki, Finland

2017 13th Summer School on Toxic Compounds in the Environment

2017, Brno, Czech Republic

2017	Kenttätoimintakoulutus – haitalliset aineet vesinäytteenotossa, SYKE, Helsinki, Finland
	, ,
2013	"Operation and control of activated sludge processes using
	microbiological analysis", 24th International Specialized
	Course, Perugia, Italia
2013	Online Course on Biological Wastewater Treatment, UNESCO-
	IHE, Deflt University
2010	Environmental Technology for Treatment and Management of
	Bio-waste, Syddansk Universiteet, Denmark
2009	Quantum Eesti AS – principles of microscopy
2009	Ministry of Environment – training and certification of water
	samplers
2008	Tartu Folk High School – project management

Supervised dissertations

Greta Nurk, MSC, 2015, Excipients in plant protection products as possible sources of hazardous substances for the aquatic environment, Tallinn University of Technology

Participation in scientific projects

articipation in scien	ittilic projects
2019–2028	LIFE17 IPE/EE/000007 LIFE IP CleanEst
2017-2020	CWPharma – Clean Waters from Pharmaceuticals
2017-2020	BEST – Better Efficiency for Sewage Treatment
2019–2020	"Study of drinking water quality and systems in sparsely populated areas"
2017–2018	"Inventory of sources of substances hazardous to the aquatic environment"
2015–2016	"Improving the quality of monitoring and research on priority and hazardous substances hazardous to the aquatic environment through the introduction of innovative sampling methodologies"
2014–2016	"Evaluation of the efficiency of wastewater treatment plants built and reconstructed with the help of the EU and EIC in 2004–2014"
2013–2015	"Source analysis of priority hazardous substances in the Pärnu River to determine the source of pollution and to stop pollution"
2009–2012	"Development of sewage sludge treatment strategy, including ensuring safe recovery through enhanced monitoring, implementation of chemical and biological indicators and introduction of quality systems, III phase"
2009–2011	COHIBA – Control of hazardous substances in the Baltic Sea region
2008–2009	"Development of sewage sludge treatment strategy, including ensuring safe recovery through enhanced monitoring, implementation of chemical and biological indicators and introduction of quality systems, II phase"

Publications

- Vallo Kõrgmaa; Taavo Tenno; Aimar Kiviruut; Mait Kriipsalu; Mihkel Gross; Priit Tamm; Kristjan Karabelnik; Harri Terase; Vahur Vark; Natalja Lepik; Karin Pachel; Arvo lital (2019). A novel method for rapid assessment of the performance and complexity of small wastewater treatment plants. Proceedings of the Estonian Academy of Sciences, 68 (1), 32–42. DOI: 10.3176/proc.2019.1.03.ETIS classification 1.1
- Vallo Kõrgmaa.; Mait Kriipsalu; Taavo Tenno; Erki Lember; Argo Kuusik; Vallo Lemmiksoo; Karin Pachel; Arvo Lital (2019). Factors affecting SVI in small scale WWTPs. Water Science and Technology, 79 (9), 1766–1776. DOI: 10.2166/wst.2019.177. ETIS classification 1.1
- Vallo Kõrgmaa, Mailis Laht, Riin Rebane, Erki Lember, Karin Pachel, Mait Kriipsalu, Taavo Tenno, Arvo lital (2020). Removal of hazardous substances in municipal wastewater treatment plants. Water Science and Technology, DOI: 10.2166/wst.2020.264 (accepted 22.05.2020]. ETIS classification 1.1
- Vallo Kõrgmaa, Mailis Laht, Erki Lember, Karin Pachel, Mait Kriipsalu, Arvo Iital (2019) Identification and removal of hazardous substances in Estonian municipal wastewater treatment plants. SETAC Europe 29th Annual Meeting, Helsinki, 26 to 30 May 2019. Society of Environmental Toxicology and Chemistry (SETAC), 385. ETIS classification 5.2
- Mailis Laht, Vallo Kõrgmaa (2019) Improving water-dependent habitats in Northeast Estonia by removing the residual pollution from rivers and wetlands. SETAC Europe 29th Annual Meeting, Helsinki, 26 to 30 May 2019. Society of Environmental Toxicology and Chemistry (SETAC), TH227. ETIS classification 5.2
- Pitk, P.; Pürjer, J.; Kõrgmaa, V.; Vilu, R. (2011). Bio-waste resource for sustainable anaerobic waste treatment solution on Island Saaremaa, Estonia. International IWA-Symposium on Anaerobic Digestion of Solid Waste and Energy Crops, Vienna, Austria. ETIS classification 5.2
- Pitk, P.; Kõrgmaa, V.; Vilu, R. (2011). Methane Potential of Sterilized Solid Slaughterhouse Wastes. 8th IWA International Symposium on Waste Management Problems in Agroindustries, Çeşme, Turkey. ETIS classification 5.2

Elulookirjeldus

Isikuandmed

Nimi: Vallo Kõrgmaa Sünniaeg: 18.03.1984 Sünnikoht: Tallinn Kodakondsus: Eesti

Kontaktandmed

E-post: vallo.korgmaa@gmail.com; vallo.korgmaa@klab.ee

Hariduskäik

2013–2020 Tallinna Tehnikaülikool, PhD

2007–2009 MSC, Tallinna Tehnikaülikool, rakenduskeemia ja

biotehnoloogia

2002–2006 BSC, Tallinna Tehnikaülikool, rakenduskeemia ja

biotehnoloogia

1995–2002 Keskharidus, Jakob Westholmi Gümnaasium

Keelteoskus

Eesti keel emakeel lnglise keel kõrgtase Vene keel kesktase Hispaania keel algtase

Teenistuskäik

2008-... Eesti Keskkonnauuringute Keskus OÜ, keskkonnakeemia

osakonna peaspetsialist

2017–2020 Eesti Vee-Ettevõtete Liit, juhtivekspert

2018–... Järvamaa Kutsehariduskeskus, lektor (reoveekäitlus)
2013–2016 Tallinna Tehnikaülikool, lektor (reoveekäitlus)

Teadusorganisatsiooniline ja administratiivne tegevus

2019–... Veekäitluse töörühma juht (projekti LIFE IP CleanEst raames)2019–... Veemajanduskavade töörühma liige (projekti LIFE IP CleanEst

raames)

2019-... Ökosüsteemi teenuste töörühma liige (projekti LIFE IP

CleanEst raames)

2017–... Veekäitlusoperaatori kutsekomisjoni liige

2015–... KIK SA Veemajandusprogrammi hindamiskomisjoni liige

2015–2019 Proovivõtjate atesteerimiskomisjoni liige

Täiendkoolitused

2019 Interpreting Surface Water Monitoring Data: Terminology,

Models and Tiered Schemes in Mixture Risk Assessment",

SETAC, Soome

2017 13th Summer School on Toxic Compounds in the Environment

2017, Brno

2017 Kenttätoimintakoulutus – haitalliset aineet vesinäytteenotossa,

Soome, SYKE

2013 "Operation and control of activated sludge processes using

microbiological analysis", 24th International Specialized

Course, Perugia, Italia

2013	Online Course on Biological Wastewater Treatment, UNESCO-IHE, Deflt University
2010	Environmental Technology for Treatment and Management of Bio-waste, Syddansk Universiteet, Taani
2009	Quantum Eesti AS – mikroskoopia põhimõtted
2009	Keskkonnaministeerium – proovivõtjate koolitus ja atesteerimine
2008	Tartu Rahvaülikool – projekti kirjutamine ja juhtimine

Juhendatud väitekirjad

Greta Nurk, MSC, 2015, Abiained taimekaitsevahendites kui võimalikud ohtlike ainete allikad veekeskkonnale, Tallinna Tehnikaülikool

Osalemine teadusprojektides

2019-2028	LIFE17 IPE/EE/000007 LIFE IP CleanEst
2017-2020	CWPharma – Clean Waters from Pharmaceuticals
2017-2020	BEST – Better Efficiency for Sewage Treatment
2019–2020	"Hajaasustuspiirkondade joogivee kvaliteedi ja -süsteemide uuring."
2017-2018	"Veekeskkonnale ohtlike ainete allikate inventuur"
2015–2016	"Veekeskkonnale ohtlike prioriteetsete ja ohtlike ainete seirete ja uuringute kvaliteedi tõstmine uuenduslike proovivõtumetoodikate kasutuselevõtu kaudu"
2014–2016	"Aastatel 2004–2014 EL ja KIK abirahaga rajatud ja rekonstrueeritud reoveepuhastite tõhususe hindamine" tegevuste koordineerimine ja juhtimine
2013–2015	"Prioriteetsete ohtlike ainete allikaanalüüs Pärnu jões reostusallika kindlaks määramiseks ning reostuse lõpetamiseks"
2009–2012	"Reoveesette töötlemise strateegia väljatöötamine, sh ohutu taaskasutamise tagamine järelevalve tõhustamise, keemilisteja bioloogiliste indikaatornäitajate rakendamise ning kvaliteedisüsteemide juurutamise abil, III etapp"
2009–2011	COHIBA – Control of hazardous substances in the Baltic Sea region
2008–2009	"Reoveesette töötlemise strateegia väljatöötamine, sh ohutu taaskasutamise tagamine järelevalve tõhustamise, keemilisteja bioloogiliste indikaatornäitajate rakendamise ning kvaliteedisüsteemide juurutamise abil, II etapp"

Publikatsioonid

Vallo Kõrgmaa; Taavo Tenno; Aimar Kiviruut; Mait Kriipsalu; Mihkel Gross; Priit Tamm; Kristjan Karabelnik; Harri Terase; Vahur Vark; Natalja Lepik; Karin Pachel; Arvo lital (2019). A novel method for rapid assessment of the performance and complexity of small wastewater treatment plants. Proceedings of the Estonian Academy of Sciences, 68 (1), 32–42. DOI: 10.3176/proc.2019.1.03.

Vallo Kõrgmaa.; Mait Kriipsalu; Taavo Tenno; Erki Lember; Argo Kuusik; Vallo Lemmiksoo; Karin Pachel; Arvo Lital (2019). Factors affecting SVI in small scale WWTPs. Water Science and Technology, 79 (9), 1766–1776. DOI: 10.2166/wst.2019.177.

- Vallo Kõrgmaa, Mailis Laht, Riin Rebane, Erki Lember, Karin Pachel, Mait Kriipsalu, Taavo Tenno, Arvo lital (2020). Removal of hazardous substances in municipal wastewater treatment plants. Water Science and Technology, DOI: 10.2166/wst.2020.264 (vastu võetud 22.05.2020].
- Vallo Kõrgmaa, Mailis Laht, Erki Lember, Karin Pachel, Mait Kriipsalu, Arvo Iital (2019) Identification and removal of hazardous substances in Estonian municipal wastewater treatment plants. SETAC Europe 29th Annual Meeting, Helsinki, 26 to 30 May 2019. Society of Environmental Toxicology and Chemistry (SETAC), 385.
- Mailis Laht, Vallo Kõrgmaa (2019) Improving water-dependent habitats in Northeast Estonia by removing the residual pollution from rivers and wetlands. SETAC Europe 29th Annual Meeting, Helsinki, 26 to 30 May 2019. Society of Environmental Toxicology and Chemistry (SETAC), TH227.
- Pitk, P.; Pürjer, J.; Kõrgmaa, V.; Vilu, R. (2011). Bio-waste resource for sustainable anaerobic waste treatment solution on Island Saaremaa, Estonia. International IWA-Symposium on Anaerobic Digestion of Solid Waste and Energy Crops, Vienna, Austria.
- Pitk, P.; Kõrgmaa, V.; Vilu, R. (2011). Methane Potential of Sterilized Solid Slaughterhouse Wastes. 8th IWA International Symposium on Waste Management Problems in Agroindustries, Çeşme, Turkey.