Intensifying Landfill Wastewater and Biodegradable Waste Treatment in Estonia

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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.

/Aare Kuusik/

Prügilareovee ja biolagunevate jäätmete käitlemise tõhustamine Eestis

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CONTENTS

LIST OF ORIGINAL PUBLICATIONS THAT CONSTITUTE THE THESIS AND AUTHORS

The thesis is based on three academic publications, which are referred to in the text as Paper I, Paper II and Paper III. Papers I to III are indexed by the ISI Web of Science:

Paper I: Kuusik, A., Pachel, K., Kuusik, Argo., Loigu, E. (2014). Landfill runoff water and landfill leachate discharge and treatment. In: 9th International Conference Environmental Engineering: Water Engineering: Selected Papers (1-6). Vilnius, Lithuania: VGTU Press "Technika".

Paper II: Kuusik, A., Pachel, K., Kuusik, A., Loigu, E., Tang, W. Z. (2014). Reverse osmosis and nanofiltration of biologically treated leachate. Environmental Technology, 35 (19), 2416-2426.

Paper III: Kuusik, A., Pachel, K., Kuusik, A., Loigu, E. (2016). Assessment of landfill wastewater pollutants and efficiency of different treatment methods. Proceeding of the Estonian Academy of Security Sciences, 65 (4), 1-23. (Accept 12.04.2016, 5.3-010 PEAS)

AUTHOR'S CONTRIBUTION TO THE PUBLICATIONS

Paper	Original idea	Study design and methods	Data collection and handling	Contribution to result interpretation and manuscript preparation	Responsible for result interpretation and manuscript preparation
I	AK	AK	AK	AK, ARK, KP, EL	AK
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III	AK	AK	AK, ARK	AK, ARK, KP, EL	AK

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WT – Walter Z. Tang

The original ideas and study results of the thesis were introduced by the author at the 9th International Conference "Environmental Engineering", 22–23 May 2014, Vilnius, Lithuania.
On the basis of the current research, the landfill wastewater treatment system of the Väätsa landfill was redesigned and reconstructed, and the current research results were taken into consideration in the design and reconstruction of the Torma landfill wastewater treatment system.

OTHER PUBLICATIONS & CONFERENCE PRESENTATIONS

Kuusik, A., Pachel, K., **Kuusik, A.**, Loigu, E. (2014). Anaerobic co-digestion of sewage sludge with fish farming waste. In: 9th International Conference on Environmental Engineering: Water Engineering (1-8). Vilnius, Lithuania: VGTU Press "Technika".

Kuusik, Argo, **Kuusik, A.**, Pachel, K., Loigu, E., Sokk, O. (2013). Generalised Intergation of Solid Waste Treatment Practices to Enhance Methane Productivity, Generate Suspension Fertiliser and Upgrade Biogas. European Scientific Journal, 9 (36), 14-30.

Kuusik, Argo, **Kuusik, A.**, Loigu, E., Sokk, O., Pachel, K. (2013). Selection of Most Promising Substrates for Biogas Production. International Journal of Energy and Environment, 7 (3), 115-124.

Kuusik, Argo, Loigu, E., **Kuusik, A.**, Sokk, O. (2013). Possibility of Enhancing Methane Productivity in Anaerobic Reactors in the Treatment of Excess Sludge from Wastewater Treatment Plants. International Journal of Science and Engineering Investigations, 2 (12), 33-36.

The publications deal with co-digestion of biodegradable waste together with sewage sludge, food industry waste, commercial waste and glycerol. The pilot study was managed by PhD student Argo Kuusik.

ABBREVIATIONS

AD	Anaerobic digestion
AOX	Absorbable organic halides
AMPTS II	Automatic Methane Potential Test System II
AS	Activated sludge
BMP	Biomethane potential
BOD_7	Biochemical oxygen demand
C	Carbon
CH_4	Methane
CO_2	Carbon dioxide
COD	Chemical oxygen demand
EC	European Community
EU	European Union
HCl	Hydrogen chloride
LLE	Landfill leachate
LWW	Landfill wastewater
NF	Nanofiltration
N	Nitrogen
N_2	Nitrogen gas
NH_3	Ammonia (gas) nitrogen
NH_4^+	Ammonium (ion) nitrogen
NO_2^-	Nitrite nitrogen
NO_3^-	Nitrate nitrogen
NO_x	mono-nitrogen oxides NO and NO_2 (nitric oxide and nitrogen dioxide)
oDM	Organic dry matter
ON	Organic nitrogen: TKN - ($NH_3 + NH_4^+$)
P	Phosphorus
Q	Discharge, flow
RO	Reverse osmosis
S	Sulphur
SO_2	Sulphur dioxide
SS	Suspended solids
TK	Total potassium
TKN	Total Kjeldahl nitrogen: organic nitrogen + $NH_3 + NH_4^+$
TN	Total nitrogen: organic nitrogen + $NH_3 + NH_4^+ + NO_2^- + NO_3^-$
TP	Total phosphorus
TUT	Tallinn University of Technology
TOC	Total organic carbon
TS	Total solids
VFA	Volatile fatty acids
VOC	Volatile organic compounds
VS	Volatile solids
WWTP	Wastewater treatment plant

1. INTRODUCTION

1.1 Landfilling of biodegradable waste and treatment of landfill wastewater in Estonia

Since 2000, the Väätsa, Torma, Uikala, Jõelähtme and Paikre landfill sites have been built in Estonia in accordance with the environmental requirements applicable in the EU. One of the most important aspects of designing a landfill site is reduction of landfill emissions, including stormwater contaminated by landfilled waste, wastewater and leachate (collectively referred to as landfill wastewater (LWW)), that are harmful to the environment. In essence, this means effective planning and operating a landfill wastewater treatment plant (WWTP) as well as a landfill gas collection and handling system. The processes mentioned above are significantly affected by handling of biodegradable waste (collecting, sorting, composting and landfilling) by waste producers and at landfills. By collecting and sorting domestic waste separately, a significant portion of biodegradable waste which can be composted or digested is separated. Composting takes place on open watertight areas, from which the collected rainwater is highly polluted and the flow of which is extremely variable. The treatment technologies that have been and are currently used (e.g., land filtration, oxidation ponds, activated sludge (AS) treatment, etc.) have not been capable of reducing the pollutant content of the LWW effluent to the level prescribed by law for discharging into public sewerage systems. The ecological state of small rivers receiving the effluent is endangered because their small flow rates during low water periods cannot provide a necessary dilution for the effluent. Domestic wastewater treatment plants have problems with the low temperature of LWW as well as the high concentration of pollutants and their toxicity.

The efficiency of treatment of LWW (as well as leachate) and the volume of biogas emitted by the landfilled waste is significantly affected by the composition of the waste and change of substances in wastewater that accompany the degradation process of biodegradable waste, according to the age of landfilled waste layers. A landfill should be viewed as a reactor, in which aerobic processes take place in turn with anaerobic processes as the waste ages and organic matter decays, creating leachate and landfill gas (biogas). The main components of landfill gas are carbon dioxide (CO_2) and methane (CH_4), while the CH_4 content in the landfill gas is mostly in the range of 50–55 %. The emission of sulphur (S) and nitrogen (N) compounds (SO_2, NO_x, HCl and NH_3) into ambient air causes acidification of soil and water bodies, due to the decay of biodegradable waste. Open composting of biodegradable waste creates a large amount of sulphur compounds and ammonia (NH_3) emissions that cause an unpleasant odour in addition to acidification. The described processes also create greenhouse gas emissions. Study results have shown that digesting biodegradable waste is more efficient than composting.

The key priority when designing landfill sites is reduction of the mentioned emissions and environmental impacts by using new technical and technological solutions. Waste treatment and reduction of emissions that are harmful to the environment must be approached integrally, i.e. waste should be treated in such a way that emissions were minimal in any stage of processing (sorting, recovering as material or for producing new products) and composting replaced by digesting, using landfill gas and digester biogas for producing electricity and thermal energy in combined heat and power plants, as well as using digester digestate for fertilising agricultural land, moistening landfill waste with LWW in order to increase the yield of gas, etc.). If all this is approached integrally, emissions that are harmful to the environment and management costs decrease significantly.

2. STUDY OBJECTIVES

In the last decade, rapid changes have taken place in waste management and treatment throughout the European Union (EU), including Estonia. The share of separate collection, sorting, and incineration of domestic waste has grown, landfilling of biodegradable waste diminished and composting increased. Methane fermentation of biodegradable waste is planned in the near future. In recent years, waste management and treatment was restructured and environmentally hazardous emissions from new landfills have diminished. The flow rates, pollutant concentration and toxicity of LWW have been measured and recommendations for LWW treatment in new conditions and for integral treatment of biodegradable waste at landfills elaborated.

The main focus of this thesis is on the treatment of biodegradable waste and LWW at landfills. In Estonia to date, three landfills (Torma, Väätsa and Uikala) are equipped with modern LWW treatment equipment. LWW from the Jõelähtme and Paikre landfills is discharged into the public sewerage system of Tallinn and Pärnu where it is treated together with domestic wastewater. The main focus in this thesis was placed on the Torma, Väätsa and Uikala landfills because the purification capacity of the WWTP-s of the Torma and Väätsa landfills had been exhausted, and the Uikala landfill had the only working LWW treatment plant in Estonia at the time. The in-depth study on the pollutant content and volume of LWW, on different modern methods of LWW treatment and biodegradable waste digestion, and on the RO concentrate was conducted at the Väätsa landfill site and in the labs of the Tallinn University of Technology (TUT).

Taking into account the current legislation in place in the EU and Estonia, the rapid changes in the last decade in the field of waste management and treatment throughout the EU countries, including Estonia, and the experience of Estonia and some of the world's leading countries in solid waste and LWW treatment (Sweden, UK, Denmark, Finland, etc.) that are located in the same climatic zone as Estonia, the main directions of this thesis were:
- formation and composition of LWW (Paper I);
- decreasing and equalising of the pollutant content, flow rate and toxicity of LWW (Paper III);
- treatment of LWW and determining the most efficient LWW treatment method (Papers II and III);
- reverse osmosis (RO) and nanofiltration (NF) of biologically treated leachate (Paper II);
- toxicity of landfill leachate and RO concentrate (Papers I, II and III);
- treatment of biodegradable waste and reducing the volume of biodegradable waste composted and deposited in landfills (Paper III);
- biodegradable waste methane fermentation in landfills (Paper III);
- use of landfill and digester gas as well as digester digestate (Paper III).

3. METHODOLOGY AND EXPERIMENTS

Most of the LWW and biodegradable waste treatment efficiency studies and studies of the environmental impact caused by treatment were conducted as a series of long-term studies (Studies, 2010; Elaboration, 2013; Kuusik, Aare et al., 2007; Kuusik, Aare et al., 2013; Studies, 2007; Studies, 2008) (Papers I–III). The results from studies conducted during the evaluation of the environmental impact of biogas plants (Environmental, 2004; Environmental, 2008; Strategic, 2009) were also used.

Studies on LWW and leachate generated in Estonian landfills were performed in 2007–2014. The flow rate and pollutant content of LWW (together with leachate, water collected from composting areas and landfill territories, as well as of domestic wastewater from the Väätsa, Uikala, Jõelähtme and Paikre domestic waste landfills) were measured in the years 2007–2010 and climatic conditions registered. The water samples were also used for analysing the following ingredients (Papers I–III) (Studies, 2007; Studies, 2008; Studies, 2010):

Water from the paved landfill territory, composting areas and landfill leachate:
- pH;
- electrical conductivity;
- alkalinity;
- chemical oxygen demand (COD);
- biochemical oxygen demand (BOD_7);
- total organic carbon (TOC);
- suspended solids (SS);
- total nitrogen (TN);
- ammonium nitrogen (NH_4-N);
- total phosphorus (TP);
- sulphate (SO_4^{2-});
- chloride (Cl^-);
- dioxins.

Metals:
- Fe^{2+} ja Fe^{3+};
- Mn, Na, Mg;
- Cu, Pb, Ni, Cd, Sn, Hg, Ag, Cr.

Hydrocarbons and phenols:
- hydrocarbons;
- phenols.

Domestic wastewater:
- pH;
- COD;
- BOD_7;
- SS;
- TN;
- TP;
- hydrocarbons.

During sampling and a week before, the daily precipitation (mm) and air temperature were registered. In winter, the melting intensity of snow was described. At each landfill, the toxicity of LWW for AS organisms was determined. LWW samples were analysed in the water chemistry laboratories of the Estonian Environmental Research Centre and the Department of Environmental Engineering of TUT. Dioxins were determined in a laboratory in Czech Republic that holds a corresponding accreditation. The toxicity of LWW was measured (by the Estonian Environmental Research Centre) with the help of ecotoxicological tests on the basis of the impact on *Protozoa*; subsequently, the impact on the bacteria in the AS was concluded (Studies, 2010).

In the years 2007–2014, experiments on percolation and RO of LWW and on biological filter technology with submerged support media of light gravel treatment were conducted at the Väätsa landfill and the Department of Environmental Engineering of TUT. The operation WWTP of existing landfills at Väätsa, Torma and Uikala was supervised. Some in vitro experiments were

conducted at the Department of Chemistry of TUT (Studies, 2010). The remainder of in vitro experiments processes included the aerobic biological oxidation process, ozonation reactor process, coagulation process, post-ozonation of the coagulated wastewater, post-ozonation of the effluent from the treatment plant, post-ozonation of the wastewater that had been treated biologically in the AS plant, lime coagulation and post-ozonation of LWW, coagulation with oil-shale ash and the Fenton process (Paper III).

At the Väätsa landfill, the concentrate from RO is pumped to the waste deposit. A series of experiments (in TUT) with methane fermentation was carried out with the aim of determining the toxicity of the concentrate produced in RO and its influence on different phases of degradation in the waste deposit and degradation of organic substances during fermentation (Studies, 2008; Studies, 2010). The concentrate discharged from RO was co-digested anaerobically in a mixture with Tallinn WWTP sewage sludge to evaluate the degradability and CH_4 productivity in various mixing ratios. The RO discharging concentrate additions have a negative effect on the anaerobic digestion (AD) of sewage sludge (Papers III).

An experimental study under laboratory conditions and using pilot reactors was performed at TUT to find more efficient solutions for the AD process and to choose suitable substrates for co-digestion (Studies, 2010; Kuusik, Aare et al., 2014). These experiments were conducted in the TUT Department of Environmental Engineering. The biomethane potential tests were carried out in anaerobic mesophilic conditions, measuring the maximum volume of biogas or biomethane produced per gram of volatile solids (VS) contained in the organic matter used as substrate for the AD process. These tests were conducted using either pure substrates or a mixture of two substrates in order to investigate the effect of the combination of different organic wastes on the digestion process (co-digestion). Biomethane potential (BMP) tests were conducted with Automatic Methane Potential Test System II (AMPTS II). The AMPTS II follows the same measuring principles as conventional methane potential tests, which make the analysis results fully comparable with standard methods (Papers III).

Experimental studies in laboratory conditions and pilot reactors have been performed in many research centres to find better solutions for the AD process and for the choice of substrates and their co-digestion. Such experiments in the course of the Sustainable Utilisation of Waste and Industrial Non-core Materials (SUSBIO) project were carried out in TUT and in the Turku University of Applied Sciences (Kuusik, Aare et al., 2013; Kuusik, Argo et al., 2013; Kuusik, Argo et al., 2013b; Kuusik, Argo et al., 2013c; Kuusik, Argo et al., 2013d; Kuusik, Aare et al., 2012). The biogas production was analysed using specially made and purchased (AMPTS II) laboratory equipment. Screening of the experimental methods was undertaken. Of available experimental methods, the biomethane potential (BMP) tests proved to be the most successful, mainly thanks to their ease in setting up and conduction of tests as well as obtainable useful information. BMP tests were conducted in batch conditions and in bench scale, measuring the maximum volume of biogas or biomethane produced per gram of VS contained in the organics used as substrates in the AD process. These tests were conducted using either pure substrates or a mixture of two substrates in order to investigate also the effect that the combination of different organic waste has on the digestion process (co-digestion). In fact, according to recent studies, the concurrent presence in the same anaerobic reactor of different organic wastes can improve the performance of the digestion process. The results of co-digestion of different studied organic substrates have demonstrated a synergic effect of combined treatment as the biodegradability of the resulting mixture was higher than that of single substrates when investigated separately. In particular, the combination of different substrates with proper percentages of each fraction can result in the production of a mixture having a carbon to nitrogen (C:N) ratio in the optimal range

20:1–30:1. Analogous results were obtained with regard to the carbon to phosphorous (C:P) ratio. Therefore, the above-cited improvement of the biodegradability characteristics of a solid mixture is substantially influenced by the adjustment of the C:N:P ratio. Further benefits of co-digestion include a higher biogas and energy production as well as a decrease in the volume of solid waste to be disposed due to the gasification of a higher percentage of the substrate (Papers III).

According to the Estonian Waste Law (RT I, 23.03.2015, 204), sewage sludge is a biodegradable waste. The goal of the "Elaboration of the strategy for processing sediments from wastewater treatment, including safeguarding harmless reusage by applying efficient supervision, chemical and biological indicators and quality assurance systems" study (Elaboration, 2013) conducted by the Estonian Environmental Research Centre and TUT was to find the most suitable sewage sludge treatment technology for Estonia and to be in accordance with regulation No 78 from 30.12.2002 of the Minister of the Environment "Requirements for using sewage sludge in agriculture, landscaping and recultivation" (RTL 2003, 5, 48). The treatment of sewage sludge was viewed as a separate part of waste treatment. In order to evaluate the potential for recycling, the environmental safety (people, animals and plants) of current sewage sludge composting technologies and the properties of sewage sludge were examined. The volume and quality of sewage sludge generated in Estonia was analysed and evaluated in terms of nutrient content, heavy metals and hygienic characteristics. Other aspects that were studied included the following: the potential value of sewage sludge and other biodegradable waste that result in decreasing the volume of biodegradable waste dumped in the landfills; the potential value of biodegradable waste generated in Estonia in the form of energy (biogas) and soil improvers (digestate) obtained by AD on the basis of the end of waste criteria (Paper III).

A study "Elaboration of methodology for experiments on deforestation and peatland renovation with the sludge from Tallinn domestic wastewater treatment by the Tallinn Water Utility" was conducted in 2002–2007, with the aim to elucidate the influence of the treatment of alvar soil and peat with different doses of sewage sludge on different seedlings under experimental conditions (Kuusik, Aare et al. 2007). The sludge from Tallinn WWTP had passed methane fermentation and centrifugal drying. Soil and peat with different doses of sewage sludge (8.9, 26.6 and 44.3 kg/m²) and control variants were used. The tests showed that methane fermented and dewatered sewage sludge from the Tallinn WWTP can be used in afforestation (Paper III).

During the evaluation of the environmental impact of constructing the Valjala, Oisu and Vinni biogas plants (Environmental, 2004; Environmental, 2008; Strategic, 2009), pollutants from composting and the methane fermentation of biodegradable waste (dung, liquid manure, green waste and biodegradable waste from processing of agricultural products) were studied.

4. LANDFILL WASTEWATER COMPOSITION AND ENVIRONMENTAL STANDARDS

4.1 Formation and composition of landfill wastewater

Studies on the LWW and leachate generated at Estonian solid waste landfills (Väätsa, Uikala, Jõelähtme and Paikre) were performed in 2007–2010 (Studies, 2007; Studies, 2008; Studies, 2010). LWW is polluted water collected from the landfill territory that consists of leachate, i.e., a liquid that moves through or drains from a landfill, precipitation that passes over the landfill site,

vehicle washing water and water drained from the sanitation devices. Landfill leachate (LLE) is the water that has percolated through contaminated material, e.g., tipped refuse. (Kriipsalu et al., 2016).

LWW and leachate generally have a very high concentration of pollutants and present a big threat to the water quality of receiving waters. As legislation becomes more and more strict, proper treatment of LWW is very important. The Väätsa, Uikala, Jõelähtme and Paikre landfills meet the EU's new environmental requirements. The landfill is separated from groundwater and untreated rainwater from paved areas or leachate is not discharged into the environment.

The pollutant content and dynamics of flow of LWW have been scarcely researched throughout the world. The focus is mainly put on leachate (its formation, flow rate and pollutant content) as generally the most polluted part of LWW. If we collect all the leachate and other wastewater (including stormwater from composting areas) of the landfill territory, we get LWW with a very uneven flow rate and pollutant content that requires different technical and technological approaches in order to treat it effectively all year round. The reason that there are so few studies on the amount and pollutant content of LWW is that they depend on many parameters that change over time and vary between landfills.

Amount of landfill wastewater
Studies show that the amount of LWW depends on the characteristics of the site – age of the waste layer(s), size of landfill territory and watertightness of the pavement of the composting areas; on the physical characteristics of the waste – the percentage of biodegradable waste (change of amount over time); on the climatic and meteorological conditions of the site – amount of precipitation, on the intensity of rainfall and snowmelt, as well as the intensity of evaporation, but also on the activities at the landfill – waste sorting and treatment techniques (composting, dumping, AD), cleansing possibilities of vehicles and containers, number of workers, amount of disinfection water, etc. For all these reasons, it is not possible to employ uniform calculation formulas for calculating the amount of LWW. In order to obtain data on flow rates and the pollutant content of LWW, one must conduct long-term and rather costly measurements that need to be repeated according to the changes in the conditions that take place over time, as mentioned above (Paper III).

During the study, the amount, pollutant content and toxicity of LWW and leachate were measured and analysed. Studies show that the amount of LWW could be zero or negligible in the short term as well as long-term. The impact of the leachate amount and duration of the discharge is directly dependent on the age of the landfill and the landfill density. The flow begins approximately 3–5 hours after the beginning of a rainfall or snowmelt. 90 % of rainwater from paved areas is quickly discharged to sewers (Studies, 2010). In new landfills the stormwater percolates quickly through the waste and ends up in sewers, taking along a large part of the easily washable pollutants. According to research data the amount of leachate drained from old landfills contains up to 20 % of precipitation water, compared to an avg. 60 % from new landfills. The older the landfill and the better the waste is compacted, the greater is the intensity of evaporation and the smaller the amount of leachate (Paper I).

Every porous material, including waste, has the ability to detain and hold a certain volume of water. The biggest volume of water that waste can hold is called maximum water retentive capacity. The amount of water held in a landfill waste layer after gravitational water has drained away is called field capacity (Kriipsalu et al., 2016). The field capacity is presented as a ratio of the mass of water in the waste and the dry mass of the waste, and it is represented as a percentage. The initial water content of domestic waste is considered to be 0.35 kg per one kg of dry mass of waste. According to publications, leachate begins to emerge when the water content of waste

exceeds the field capacity (in the case of domestic waste, it is usually 50–60 %, i.e. 0.5–0.6 kg of water per kg of dry mass of waste. Before it begins to leak, a waste layer can hold 0.15–0.25 kg/kg of additional water. However, if waste is already wet when dumped, it can hold less stormwater than dry waste (Crawford and Smith, 1985; Kriipsalu et al., 2016). The volume of leachate also depends on the water absorbing capacity (hygroscopicity) of the waste. The biggest amount of leachate is generated when the whole mass is saturated with water (water content 0.8–1.15 kg/kg). The amount of leachate from closed landfills in Germany is estimated to be 13 % of the annual precipitation (White et al., 1997; Kriipsalu et al., 2016). After watertight covering and closing of the landfill, the amount of leachate significantly decreases, but increases if the landfill is being moistened in order to stimulate the production of biogas.

The diurnal, weekly and annual flow rates of LWW and leachate in Estonia are variable, as is typical for the northern part of the temperate climate zone (ESTONICA). The long-term avg. annual precipitation for Estonia is 750 mm (Reihan, 2008) and varies considerably temporally and across the territory, e.g., at Pärnu 435 mm in 1947 and 953 mm in 2001 (EMHI).

In this study the impact of different weather conditions on the amount of LWW and leachate was analysed. Depending on the duration of rainfall and on the purpose for which watertight areas were used, 60 to 80 % of rainwater falling on watertight areas and 10 to 30 % of rainwater falling on landfill waste lifts reaches sewers. A large part of the territory of new solid waste landfills in Estonia (including composting areas) have a impermeable cover and the LWW studies conducted in the years 2007–2010 demonstrate that (Studies, 2010):

- the water of small showers (up to 2 mm) usually evaporate completely;
- daily leachate flow rates at Jõelähtme and Väätsa landfills depended in September to December 2007 on the intensity of precipitation as follows: 11–525 m^3/d at Jõelähtme and 21–128 m^3/d at Väätsa. The largest flow rates were measured in September. At Jõelähtme, a leachate flow rate of 525 m^3/d was measured, and at Väätsa only 16 m^3/d. Rainwater flow rates from composting areas were at Jõelähtme 51 m^3/d and at Väätsa 108 m^3/d, and from the landfill territory 130 m^3/d at Jõelähtme and 4 m^3/d at Väätsa, altogether amounting to 706 m^3/d at Jõelähtme and 128 m^3/d at Väätsa. During the measurements, the total precipitation of the three days was 16 and 13 mm respectively;
- daily leachate flow rates at the Uikala and Pakre landfills depended in September to December 2007 on the intensity of precipitation as follows: 221–298 m^3/d and 42–55 m^3/d. The largest flow rates were measured in December and October. At Uikala, the flow rate of wastewater from the paved area in front of the office and the office were also measured to be 4.5 m^3/d. At Paikre, only leachate is collected. During the measurements, the three day total precipitation was 3.2 and 7.4 mm respectively.

LWW flow rate fluctuations were measured at periods with different intensity of rainfall and snowmelt at the Väätsa landfill, where precipitation water and leachate were collected from an area of 4.1 ha. The study results were the following: Q_{min} = 0–2 m^3/d, Q_{avg} = 10–20 m^3/d (1.4 to 2.9 m^3/ha per day) and Q_{max} = 50–95 m^3/d (7.1 to 13.6 m^3/ha). In some cases, Q_{max} increased up to 150 m^3/d (21.4 m^3/ha). The fluctuations of leachate flow were smaller: Q_{min} = 0–2 m^3/d, Q_{avg} = 5–15 m^3/d (1.0 to 2.9 m^3/ha), Q_{max} = 20–30 m^3/d (3.9–5.8 m^3/ha). In the case of heavy rainfall and snowmelt, one must expect very big hydraulic and pollutant shock loads, and the flow rate of LWW may be close or even equal to zero during long drought periods. The results of measurments show that the big problem is the low temperature of rain and snowmelt water in winter (from 1 to 4 ºC) i.e. flowing from composting areas and the landfill territory (Studies, 2010; Studies, 2008) (Paper I).

Pollutant content, concentration and pollution load of landfill wastewater

The study results show that the pollutant content and pollution load of landfill wastewater mainly depends on weather conditions, the construction of wastewater facilities, the size and intensity of exploitation of composting areas, the content of biodegradable waste and filling materials used for composting, the size and loading of depositing areas and less on the sorting and storing technologies of different types of waste, the washing technologies of machinery and containers, the number of workers, etc. In winter and during longer drought periods, only LLE together with domestic wastewater from the workers or only domestic wastewater from the workers should generally be treated (Studies, 2010; Studies, 2008).

Municipal solid waste LLE is mainly polluted by organic substances and nitrogen, mainly in the form of ammonium nitrogen (NH_4-N), the content and concentration of which may fluctuate within a wide range, depending on precipitation (Table 4.1) (Studies, 2010). COD values can be very high, especially of the leachate from recently landfilled waste. In many cases, nitrogen may be the most important risk to the environment (Christensen, 2010).

Table 4.1. Fluctuation of the concentration of pollutants in the leachate and LWW measured at the Väätsa, Uikala and Jõelähtme landfills (Studies, 2010)

Parameter	Unit	Väätsa	Uikala	Jõelähtme
LWW				
BOD$_7$	mgO$_2$/l	366–1,663	541–1,851	650–3,126
COD	mgO/l	988–8,730	2,870–4,840	2,220–6,120
NH$_4$-N	mgN/l	68–120	439–684	237–834
TN	mgN/l	200–394	720–920	372–864
Leachate				
BOD$_7$	mgO$_2$/l	300–950	231–1,750	3,051–5,451
COD	mgO/l	583–2,390	1,230–4,240	7,200–9,100
NH$_4$-N	mgN/l	50–330	427–852	726–974
TN	mgN/l	230–469	564–1,567	820–1,335

Leachate contains natural organic substances in the form of suspended or colloidal particles, macro polymers or simple low molecular substances. A part of these organic substances remains dissolved. Dissolved organic carbon is a fraction of TOC. The high concentration of TOC mainly includes humic substances that consist of humic acids, fulvic acid and humins. In addition to the large number of organic substances, domestic landfill leachates tend to have large concentration of salts, mainly NaCl and NH_4-N, along with heavy metals (Sumanaweera, 2010).

In Estonia the heavy metal content of LWW does not exceed concentration limits (Studies, 2010). The pollutant content of the water that percolates through the body of a landfill is heavily influenced by the waste content and the processes taking place inside the landfill body.

Processes or landfill phases are influenced by the decomposition phases of the waste, i.e. the aerobic, anaerobic acid, intermediate methanogenic, stabilised methanogenic and final aerobic phases (Figure 4.1). Generally speaking, the various landfill parts are in different phases of degradation. The flow rate and pollutant content of LWW is significantly influenced by the closing of old landfill areas and the opening of new ones.
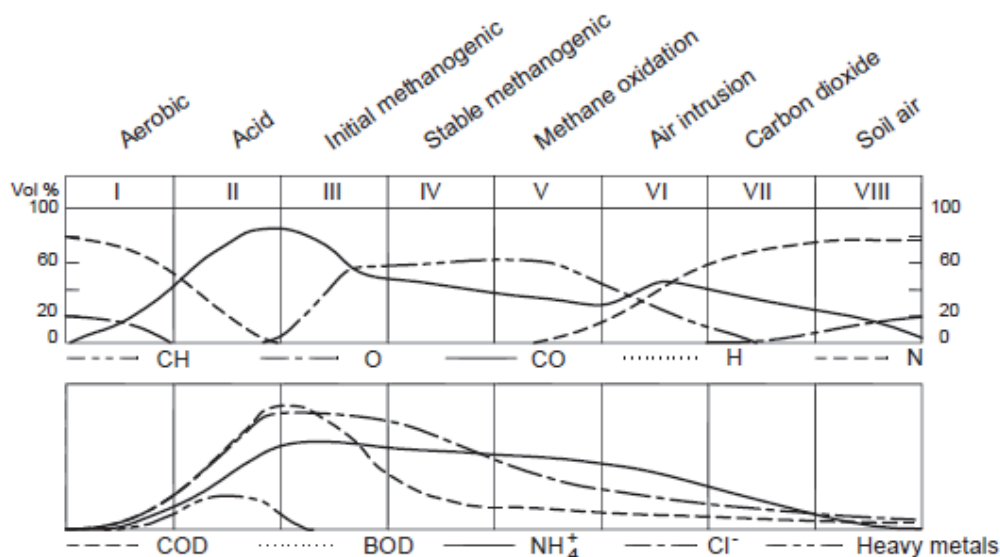
Figure 4.1. The lifetime of a landfill showing general trends in gas and leachate quality development (Kjeldsen, 2002)

In each decomposition phase of waste, leachate of different composition is generated. Although it is usually balanced in a balancing tank, its pollutant content that changes over time, depending on the decomposition phase, has a significant effect on the treatment technology of leachate (Sumanaweera, 2010; Studies, 2010; Baun et al., 2003) (Paper I):

Aerobic phase.
Degradation initially starts under aerobic conditions. In the beginning of the aerobic phase, oxygen is quickly used up and this results in a release of CO_2 and a rise in waste temperature (the exothermic stage). The compression of waste releases moisture, which creates leachate along with stormwater. When the oxygen sources run out, the environment in the waste layer becomes anaerobic and this contributes to the generation of fermentation. In this phase, proteins are degraded to amino acids and then into CO_2, water, nitrates and sulphates. Carbohydrates are converted to carbon dioxide and water. Fats are hydrolysed to fatty acids and glycerol and are then further degraded into simple compounds through the formation of volatile acids and alkalis. Cellulose, which is the main organic fraction of the waste, is degraded by extracellular enzymes into glucose, which is subsequently converted into carbon dioxide and water by bacteria.

Anaerobic acid phase
In the second phase, hydrolytic and acetogenic bacteria dominate due to the accumulation of carboxylic acids, and pH decreases. The highest concentrations of BOD and COD can be measured in this phase. If the pH is acidic, the leachate of the acidic phase is chemically aggressive and can increase the solubility of many components. Metals are more soluble at this stage. Leachate in this phase is characterised by high BOD values usually greater than 10,000 mgO_2/l, high BOD:COD ratios generally greater than 0.7, acidic pH values from 5 to 6 (pH values from 4.7 to 7.7 and ammonia, 500–1,000 mgN/l due to the hydrolysis of proteinaceous compounds (Worell and Vesilind, 2010).

Intermediate methanogenic phase
This phase starts with the slow growth of methanogenic bacteria, and it may be inhibited by excess organic volatile acids, which are toxic to methanogenic bacteria. The CH_4 concentration in

17

the gas increases while the hydrogen, CO_2 and volatile fatty acid (VFA) concentration decrease. This is also the phase when cellulose and hemicellulose begin to decompose. The COD and BOD begin to decrease and the pH increases when the acids have been exhausted. The BOD:COD ratio can also increase when the carboxylic acids have been exhausted. Conversion of fatty acids increases the pH and alkalinity making calcium, iron, manganese and heavy metals less soluble. These metals precipitate as sulphides. Ammonia is released without conversion in the anaerobic environment.

Stabilised methanogenic phase
The CH_4 production speed can reach its maximum and slow down when the supply of soluble substrate (carboxylic acid) decreases. In this phase, the CH_4 production speed depends on the speed of cellulose and hemicellulose hydrolysis. The pH range for methanogenic bacteria is 6 to 8. Low volatile acids and low total dissolved solids indicate that the solubilisation of most of the organic components has decreased at this stage. Therefore, the leachates at this stage are characterised by low BOD values and low BOD:COD ratios. In this phase, the BOD:COD ratio can drop down to 0.1, because carboxylic acid is being exhausted as fast as it is generated.

The rate of degradation and the leachate content of pollutants are mostly dependent on the conditions within the landfill. An important factor is the moisture content of the waste. It is normal to send leachate or concentrate formed during leachate treatment back to the landfill, where the accumulation of hazardous substances takes place.

At the Väätsa and Uikala landfills, the RO concentrate is conducted to the upper layers of the landfill.

Final aerobic phase
Waste continues to decompose until CH_4 production is very low and the atmospheric air starts to diffuse into the landfill – then the landfill becomes aerobic.

Biological degradation methods comprise of aerobic and anaerobic degradation techniques. In the published research, the decomposition of municipal solid waste (MSW) in landfills was carried out in five phases as shown in Figure 4.2. The decomposition phases were: I) initial adjustment, II) transition phase, III) acid phase, IV) methane formation phase and V) maturation phase (Tchobanoglous et al., 1993).
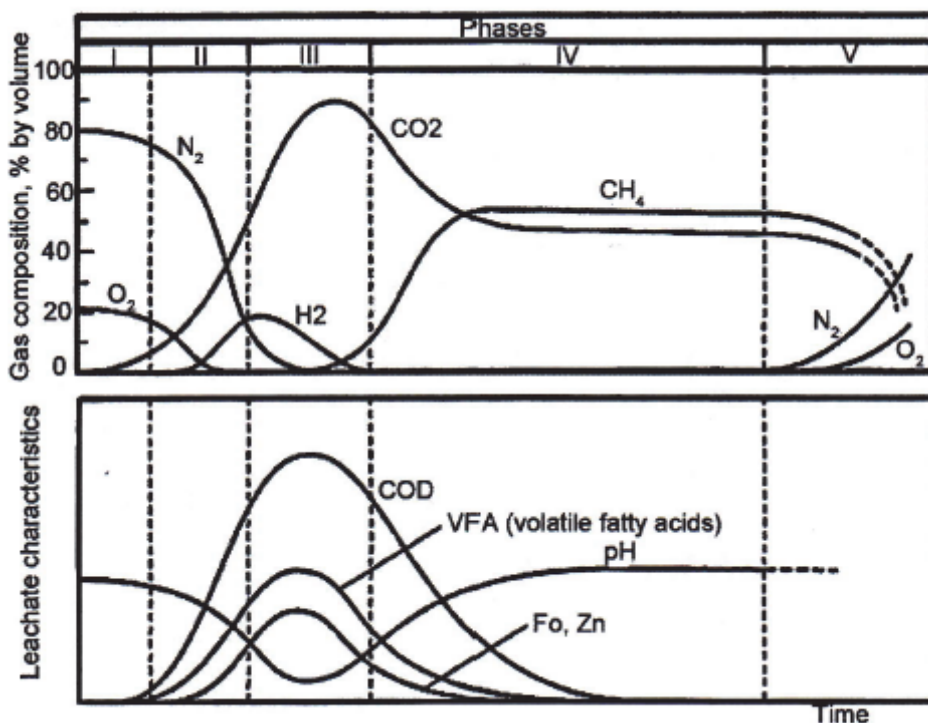
Figure 4.2. Gas composition and LLE characteristics at different phases of decomposition (Tchobanoglous and Kreith, 2002)

Pollutants in domestic waste LLE can be divided into four groups (Studies, 2010; Tengrui et al., 2007):

- dissolved organic matter COD or TOC, VFA, humic substances, which consist of humic acids, fulvic acid and humans;
- inorganic compounds (e.g., calcium, potassium, sodium, ammonium, magnesium, iron, manganese, sulphates, sodium, bicarbonate and chlorides);
- heavy metals (e.g., nickel, lead, copper, chromium, cadmium, iron, and zinc);
- xenobiotic organic materials from households or industrial chemicals (aromatic bicarbonates, phenols, pesticides, etc.).

Aerobic and anaerobic decomposition of the organic materials results in both liquid (leachate) and gaseous end products (Weiner and Matthews, 2003). The produced leachate could contain a large volume of pollutants measured as COD, BOD_5, NH_3-N, SS, heavy metals, phenols, P, etc. (Tables 4.2 and 4.3) (Uygur and Kargi, 2004; Renou et al., 2008; Foul et al., 2009; Aziz et al., 2010).

Table 4.2 shows that it is difficult to associate the composition of LLE with the standard characteristics of LWW presented in different publications, and with the landfill age classification. The reason for this in Estonian landfills, compared to other European countries, is that the composition of waste is different in each landfll and the waste is treated differently (in Estonia, the importance of waste separate collection, sorting, recycling and waste incineration is increasing, and the importance of waste landfilling is decreasing, etc.).

Table 4.2. Typical LLE characteristics

Parameter	Unit	Landfills in Estonia (2007 medium data) (Studies, 2010)			Type of LLE (Alvarez-Vazques et al., 2004; Chian and DeWalle, 1976)		
		Jõelähtme (4 years old)	Uikala (6 years old)	Väätsa (7 years old)	Young (< 5 years)	Intermediate (5–10 years)	Stabilised (> 10 years)
pH		7.7	7.5	7.7	< 6.5	6.5–7.5	> 7.5
COD	mgO/l	8,160	2,538	1,366	> 10,000	4,000–10,000	< 4,000
BOD_5/COD		0.57[d]	0.34[d]	0.39[d]	0.5–1.0	0.1–0.5	< 0.1
NH_3-N	mgN/l	NH_4-N 879	NH_4-N 592	NH_4-N 198	< 400	NA[c]	> 400
TOC/COD		0.26	0.27	0.27	< 0.3	0.3–0.5	> 0.5
Kjeldahl nitrogen	gN/l	TN - 1.1	TN - 1.07	TN - 0.3	0.1–0.2	NA[c]	NA[c]
Heavy metals	mg/l	Low	Low	Low	Low to medium	Low	Low
Biodegrada-bility		Medium	Medium	Medium	Important	Medium	Low

[a] VFA, [b] humic and fulvic acids, [c] not available, [d] BOD_7:COD ratio

If the BOD_t:COD ratio is ≥ 0.5, biological treatment can be used; if < 0.5, wastewater is to be treated using other methods (Kuusik, 1995).

Stabilised leachate contains much more refractory organics than young leachate. In this respect, young LLE (age < 5 years) is typically characterised by high biodegradable organic compounds such as VFA, BOD_5 and COD concentrations, as well as quite a high volume of NH_3-N and a high BOD_5:COD ratio. In contrast, stabilised LLE (age > 10 years) normally contains a high volume of NH_3-N, moderately high COD and a low BOD_5:COD ratio (less than 0.1), and the substance is generally not readily degradable organic matter (Tables 4.2 and 4.3) (Aziz, 2016; Alvarez-Vazquez et al., 2004; Chian and DeWalle, 1976).

In Estonia, the average conductivity of landfill leachate is high: Jõelähtme 14,977 µS/cm, Uikala 15,072 µS/cm and Väätsa 8,090 µS/cm) and the leachate is slightly alkaline (Table 4.2) (Studies, 2010). It is difficult to classify Estonian landfills according to the criteria in Tables 4.2 and 4.3 because new landfills are successively established and this significantly affects the pollutant content and pH of leachate.

Table 4.3. LLE constituent content ranges as a function of the degree of landfill stabilisation (Worell and Vesilind, 2010)

Parameter	Phases			
	Transition	Acid Formation	Methane Formation	Maturation
COD (mgO/l)	480–18,000	1,500–71,000	580–9,760	31–900
Total volatile acids (mg/l as acetic acid)	10–3,000	3,000–18,800	250–4,000	0
NH_3-N (mgN/l)	120–125	2–1,030	6–430	6–430
pH	6.7	4.7–7.7	6.3–8.8	7.1–8.8
Conductivity µS/cm	2,450–3,310	1,600–17,100	2,900–7,700	1,400–4,500

Very high concentrations of pollutants (and hence the pollution load) were measured for rainwater collected at the composting area in 2007: at the Jõelähtme landfill BOD_7 3,960 mgO_2/l, COD 7,530 mgO/l and at the Väätsa landfill BOD_7 1,875 mgO_2/l, COD 9,300 mgO/l. At Jõelähtme the measured LWW and leachate medium concentrations of pollutants in 2007 were: BOD_7 1,885 mgO_2/l, COD 4,170 mgO/l for the LWW and BOD_7 4,668 mgO_2/l and COD 8,160 mgO/l for the leachate. The medium values of Uikala and Väätsa LWW and leachate, according to our investigations, are presented in Table 4.4 (Paper I).

Solid waste LLE is mainly polluted by a high concentration of organic substances and nitrogen that fluctuate to a great extent depending on precipitation (Table 4.1) (Studies, 2010).

Table 4.4. Measured LWW and average pollutant concent of leachate in 2007 at the Väätsa and Uikala landfills (Studies, 2010) (Paper I)

Parameter	Unit	Väätsa landfill (medium)		Uikala landfill (medium)	
		Wastewater	Leachate	Wastewater	Leachate
pH		6.75	7.7	7.9	7.5
conductivity	µS/cm	5,545	8,090	14,010	14,553
SS	mg/l	650	400	411	298
BOD_7	mgO_2/l	1,015	529	1,196	697
COD	mgO/l	4,859	1,366	3,855	2,538
TOC	mgC/l	1,313	375	1,073	718
NH_4-N	mgN/l	94	198	562	592
TN	mgN/l	297	298	820	1,072
TP	mgP/l	22.0	4.6	4.5	6.1
HCO_3	mg/l	2,800	3,805	6,470	7,260
SO_4	mg/l	290	588	401	172
Cl	mg/l	107	439	1,593	1,553
monobasic phenols	mg/l	235	49	2,590	1,510
dibasic phenols	mg/l	23	140	1,878	1,005
hydrocarbon	mg/l	< 20	29	< 20	60
Fe^{2+}	mg/l	7.4	2.0	1.2	0.6
Fe^{3+}	mg/l	5.3	8.0	14.7	21.4
Hg	µg/l	0.05	0.15	< 0.05	< 0.05
Ag	mg/l	0.01	< 0.01	< 0.01	< 0.01
Cd	mg/l	< 0.02	< 0.02	< 0.02	< 0.02
Cr	mg/l	0.032	0.187	0.286	0.260
Mg	mg/l	105	108	293	283
Mn	mg/l	0.260	0.273	0.367	0.557
Na	mg/l	58	675	1,400	1,425
Ni	mg/l	0.041	0.090	0.061	0.080
Pb	mg/l	< 0.04	< 0.04	< 0.04	< 0.04
Zn	mg/l	< 0.1	< 0.1	< 0.1	< 0.1
Cu	mg/l	0.04	0.09	< 0.02	0.023

Table 4.4 shows that at the Väätsa and Uikala landfills the the SS, BOD_7, COD and TOC of the LWW (together with leachate, polluted rainwater runoff from composting areas and wastewater collected from the landfill territory) were significantly higher than in the leachate. This was mainly purposed by to the high level of pollutants in the rainwater runoff from composting areas. The content of TN and TP was variable, but the concentration of TN was slightly higher in the leachate, and the concentration of TP higher in the LWW. The metal content was low (see Table

4.3), which can be due to a very high concentration of sulphates and a high conductivity, which creates favourable conditions for the metals to precipitate.

Study results show that (Studies, 2010) (Papers I and III):
- the pollutant content of the LWW increased significantly during rainfalls, depending on the intensity of precipitation and composition and amount of biodegradable waste, deposited on watertight areas: for up to 40 % for SS, 50 % for BOD_7 and 70 % for COD. The pollution load of TN increased by 20 % and by up to 40 % for TP;
- LLE has a very high TN concentration and low concentration of TP. The leachate contains high levels of COD and BOD_7;
- the metal content of LWW and leachate is low due a high pH;
- dumping of waste tyres in the base lift leads to an increase in the concentration of iron in the leachate (up to 6–8 mg/l). Iron is responsible for corrosion that may continue for up to 5–6 years leading to a blockage of the drainage. As a result of a fire on Paikre landfill, the iron concentration of iron in LWW increased twofold (17 mg/l);
- LLE may be toxic and hinder biological treatment;
- in 2007, the BOD_7:COD ratio values in different samples were as follows: Väätsa 0.3–0.5, Jõelähtme 0.2–0.7, Uikala 0.2–0.6. In 2010, however, the BOD_7:COD ratio in the Väätsa LLE was less than 0.1. This was caused by the presence of humic and fulvic acids, tannins, lignin and hazardous organic chemicals, pesticides and herbicides, which decreased the biodegradability of the wastewater;
- at all of the investigated landfills, the optimal ratio of BOD:N:P for biological treatment (100:5:1) was out of balance, indicating a deficit of phosphorus. For example, in 2007, in the Väätsa LWW, the ratio BOD_7:TN:TP was in average 46:14:1 and in the LLE 115:65:1. In the Uikala LWW, this ratio was 267:183:1 and in the LLE 115:177:1;
- the temperature of LWW, including leachate, was from October to April between 1 °C and 4 °C, which significantly hindered the biological treatment process. The degree of purification of RO increased 3 % for each degree of temperature rise.

Since the goal is to significantly decrease the volume of biodegradable domestic waste in landfills, changes in the pollutant content of leachate are expected. The TOC, KHT and BHT of the leachate abviously decrease. The concentration of heavy metals will not change significantly, due to the slightly alkaline environment and the high concentration of sulphates that contribute to the precipitation of metals from the leachate. The toxicity of the leachate will somewhat decrease due to fewer sources of nitrogen being dumped in the landfills. It is difficult to predict the behaviour of other substances and compounds, due to the lack of studies undertaken on this matter. All this should be taken into consideration when planning new WWTP-s for landfills (Papers I and III).

4.2. Environmental standards of landfills

In the last decade, the recovery of waste materials has increased in Estonia, and this is related to the increase in the importance of the separate collection and sorting of waste. The landfilling of domestic waste has decreased slowly but surely, which relates to a growing trend of waste incineration for energy production. The economic recession of 2007–2009 affected the generation of waste significantly. The mentioned trends in generating and treating waste significantly affect the amount and pollutant content of wastewater generated at landfills. At the landfills and their surrounding areas, the environment is endangered by LWW (including leachate) as well as by volatile gases and pollutants emitted from landfilled waste, compost and in the waste sorting

process. The waste treatment process as a whole causes unpleasant odours. The waste treatment taking place at waste generators and in landfills must be approached in tandem with the caused environmental impact. Reducing the environmental impact begins at the generators of waste in the form of separate collection and recovery of waste, and continues at waste management centres when sorting waste, landfilling unrecoverable waste, collecting landfill gas, treating LWW (including leachate) and/or using it for moistening landfilled waste, producing biogas by digesting biodegradable waste, and producing electricity and thermal energy from biogas in combined heat and power plants (Environmental, 2004; Environmental, 2008; Strategic, 2009).

The developed countries of the world (including Estonia) have veered towards integrated solid waste management (ISWM), which aims to dispose of the ever growing volume of waste in an environmentally and health-friendly way while keeping the costs to a minimum. ISWM is a comprehensive waste prevention, recycling, composting and disposal programme. An effective ISWM system considers how to prevent, recycle and manage solid waste in ways that most effectively protect human health and the environment. The major ISWM activities are waste prevention, recycling and composting, combustion and disposal in properly designed, constructed and managed landfills.

In accordance with the end-of-waste criteria (EoW), waste ceases to be waste and begins to be a product again after it has undergone recovery operations, including recycling, and meets certain criteria. The aim is to set quality criteria for certain waste streams that, when met, would allow these wastes to be considered a (new) product either as a) a fertiliser or soil improving substance or its component; b) a building material or its component; or c) soil (End of waste criteria, 2008).

According to the Estonian National Waste Management Plan 2014–2020, it is of primary importance to decrease the entire volume of deposited waste (National, 2014). In Estonia, the requirements for producing compost from biodegradable waste and the quality and safety characteristics of compost (on the basis of the criteria that allow waste to cease being waste) are imposed by Regulation No. 7 of the Minister of the Environment from 08.04.2013 entitled "Requirements for compost production from biodegradable waste" (RT I, 10.04.2013, 1). In order to protect surface water, groundwater, soil, fauna, flora and human beings from the negative impact of sewage sludge from the most common biodegradable waste in Estonia, the use of sewage sludge in agriculture, landscaping and recultivation is regulated with Regulation No. 78 of the Minister of the Environment from 30.12.2002 entitled "Requirements for using sewage sludge in agriculture, landscaping and recultivation" (RTL 2003, 5, 48).

Another issue is the treatment of biodegradable animal waste. If the aim is to use animal by-products as one of raw materials for producing digestate and biogas, in Estonia one must follow the Infectious Animal Disease Control Act (RT I 1999, 57, 598), which is based on EC Regulation No. 1774/2002 and lays down the health rules concerning animal by-products not intended for human consumption (EC Regulation No. 1774/2002), and its amendment EC Regulation No. 208/2006 (EC Regulation No. 208/2006), which establish strict health rules for the use of animal by-products, so as to ensure a high level of health and safety. The waste treatment of animal by-products in biodegradable operations is to adhere to this Directive.

The Waste Framework Directive 2008/98/EC established a legal framework for the treatment of waste in the EU. It sets out the basic concepts and definitions related to waste treatment and lays down waste treatment principles for all other EU legislation related to waste, such as the "polluter pays principle" and "waste hierarchy". The Directive introduces a five-step European waste hierarchy (EC Directive 2008/98/EC):

- prevention – preventing and reducing waste generation;
- reuse and preparation for reuse – giving the products a second life before they become waste;
- recycle – any recovery operation by which waste materials are reprocessed into products, materials or substances whether for their original or other purpose. It includes composting and it does not include incineration;
- recovery – some waste incineration based on a political non-scientific formula that upgrades the less inefficient incinerators;
- disposal – processes to dispose of waste be it landfilling, incineration, pyrolysis, gasification and other finalist solutions.

Council Directive on the landfill of waste obliges Member States to minimise biodegradable waste to landfills to 75 % by 2006, 50 % by 2009 and 35 % by 2016 (mass percents compared to the total volume of deposited domestic waste in 1995), and to treat it before disposal (EC Directive 1999/31/EC). The Estonian Waste Act has stipulated limits for depositing biodegradable waste into landfills. By the year 2020, the share of biodegradable waste in the deposited domestic waste must not exceed 20 % (RT I, 23.03.2015, 204). According to the Estonian National Waste Management Plan 2014-2020, 30 % of waste will be recycled as organic material, 3 % as compost and 10 % as anaerobic fermentation digestate, 40 % will be incinerated, 8.5 % will be used in the cement industry as refuse derived fuel and 8.5 % will be landfilled by 2020 (National, 2014).

According to Regulation No. 38 of the Minister of the Environment from 15.08.2004 (Landfill construction, operation and closure requirements), a landfill with biodegradable waste must be equipped with gas capture and collection equipment in order to avoid uncontrolled collection and outflow of biogas (RTL 2004, 56, 938). In the Estonian National Waste Management Plan 2014–2020, it is taken into account that a compliant collecting of landfill gas will take place in 2020. On average, 50 % of landfill gas will be collected and part of it will be used in electricity production (National, 2014).

In most cases, LWW is treated by a combination of different treatment methods. The treatment technologies that have been used and are in use today as well as the pollutant levels in the effluent from some landfills have not been able to stay below the levels prescribed by law.

In Estonia, LWW treatment is regulated by the Estonian Water Act (RT I, 30.06.2015, 4) and related legislative acts. LWW is treated on-site or directed into the closest suitable WWTP according to the requirements of Regulation No. 38 of the Minister of the Environment from 15.08.2004 (Landfill construction, operation and closure requirements) (RTL 2004, 56, 938). The requirements for LWW treatment are the same as for those for domestic wastewater. The requirements for the treatment of wastewater (including LWW) and for discharging effluent and stormwater to recipients, the limit values of indicators of pollution of effluent and stormwater and the actions for controlling the compliance with the requirements can be found in Government Regulation No. 99 from 29.11.2012 (RT I, 04.12.2012, 1), see also Table 4.6. The requirements established by the regulation for the TP and TN content of LWW apply only to effluent discharged to recipients through an outlet structure located separately from the public sewerage system. The limit value for the TP content of LWW is 2 mg P/l and the degree of purification of wastewater is 80 %, while the TN content limit value is 75 mg N/l and the degree of purification 75 % (RT I, 04.12.2012, 1). In comparison, the Waste Framework Directive 2008/98/EC states that appropriate measures shall be taken, with respect to the characteristics of the landfill and the

meteorological conditions, in order to treat contaminated water and leachate collected from the landfill according to the appropriate standard required for their discharge.

The objective of the Urban Wastewater Treatment EC Directive 91/271/EEC is to protect the environment from the adverse effects of urban wastewater discharges as well as certain industrial sectors, to protect the environment from the adverse effects of the treated wastewater effluent, secondary or tertiary treatment in less sensitive areas. LWW must be treated biologically according to Directive articles 4 and 5 and in Table 4.5.

Table 4.5. Requirements for discharges from urban WWTP-s subject to Articles 4 and 5 of the Directive. The values for the percentage of reduction shall apply (EC Directive 91/271/EEC)

Parameter	Concentration	Minimum percentage of reduction	Comment
BOD_5 at 20 ºC without nitrification*	25 mg O_2/l	70–90**	* The parameter can be replaced by another parameter: TOC if a relationship can be established between BOD_5 and the substitute parameter. ** Reduction in relation to the load of the influent.
COD	125 mg O/l	75	
Total SS	35 mg/l*** 35 (more than 10,000 p.e.) 60 (2,000–10,000 p.e.)	90 90 (more than 10,000 p.e.) 70 (2,000–10,000 p.e.)	*** This requirement is optional. Under Article 4 (2).
TP	2 mg P/l (10,000–100,000 p.e.)	80****	**** Reduction in relation to the load of the influent.
TN*****	15 mg N/l (i.e. 10,000–100,000)	70–80	***** Total nitrogen means: sum of total Kjeldahl-nitrogen (organic N + NH_3), nitrate (NO_3) and nitrites (NO_2) nitrogen. This requirement refers to a water temperature of 12 °C or more during the operation of the biological reactor of the WWTP.

Based on the Urban Wastewater Treatment EC Directive 91/271/EEC, the requirements for discharges from LWW treatment plants are established in the EU countries. The limit values of pollutants are also established for LWW that is discharged into the public sewerage system. Table 4.6 shows the limit values of pollutants for wastewater or effluent if discharged into the public sewerage system or surface water in Germany, the Netherlands, Austria, Switzerland and Estonia that have to be taken into account when choosing the treatment technology for LWW.

Table 4.6 shows that different EU countries have different limit values and partially do not correspond to the data in Table 4.5, as well as with the data in Directive 2008/105/EC from 16 December 2008 of the European Parliament and of the Council. Estonian legislation on wastewater discharges to the environment is in adjustment. The differences result from the differences in the flow rate and the ecological condition of watercourses (as recipients of effluent) in the member countries, which also result in differences in environmental policy. In Estonia, the limit values for metals and hazardous substances meet the limit values established in Directive 2008/105/EC from 16 December 2008 of the European Parliament and of the Council, and are significantly stricter than in other EU countries (Table 4.6).

In Finland, LWW is widely discharged into the public sewerage system. In Lithuania, Sweden and Germany, LWW is generally treated on-site (Giržadas, 2009; Ehring et al., 1988; Analysen, 1988; Ehring, 1989; Johansen and Carlson, 1976).

Table 4.6. The limit values of pollutants for LWW or effluent discharged into the public sewerage system or surface water (Kaartinen et al., 2009, RT I, 04.12.2012, 1; RTL 2003, 110, 1736; RT I, 30.06.2015, 4; RT I, 08.01.2016, 10)

Parameter	Unit	Germany to surface water / to public sewerage system	Netherlands to surface water / to public sewerage system	Austria to surface water (Christensen, 2010) / to public sewerage system	Switzer-land to public sewerage system	Estonia to surface water (RT I, 04.12.2012, 1) / to public sewerage system (RTL 2003, 110, 1736)
SS	mg/l	-/-	-/-	-/-	-	25/500 [c]
COD	mgO/l	200/400	-/-	50/75–90 max	b	125/750 [c]
BOD_5	mgO_2/l	20/-	7–20/ 300–400	10/-	-	BOD_7 15/ BOD_7 375 [c]
TN	mgN/l	70 [a]/-	8–15 [b]/300	-/-	-	75/125 [c]
NH_4-N	mgN/l	-/-	4–8 [b]/200	NH_4^+ 10/[b]	b	-/ c)
NO_2-N	mgN/l	2/-	1–4 [b]/-	-/1.5	0.3	-/ c)
Hydrocarbons	mg/l	10/-	-/-	-/-	10	-/ c)
TP	mgP/l	3/-	-/-	-/-	-	2/15, [c]
Hg	mg/l	0.05/0.05	0.0005/ 0.002	0.01/0.01	0.01	0.00007 [f]/ 0.05
Cd	mg/l	0.1/0.1	0.003/0.01	0.1/0.1	0.1	≤ 0.00045–0.0015 [f]/0.2
Cr (tot)	mg/l	0.5/0.5	-/-	0.5/2.1	2.1	0.005 [d]/0.5
Cr (-VI)	mg/l	0.1/0.1	0.075/0.375	-/0.1	0.1	0.005 [d]/ 0.1
Ni	mg/l	1/1.0	0.1/0.17	0.5/2	2	0.034 [f]/1.0
Pb	mg/l	0.5/0.5	0.05/0.2	0.5/1	0.5	0.014 [f]/0.5
Cu	mg/l	0.5/0.5	0.05/0.25	0.5/1	0.5	0.015 [d]/ 2.0
Zn	mg/l	2/2	0.2/1	0.5/3	2	0.01 [d]/2.0
As	mg/l	0.1/-	-/-	-/-	-	0.01 [d]/0.2
Cyanide	mg/l	0.2/0.2	-/-	-/-	-	[e]/0.2
Sulphide	mg/l	1/1.0	-/-	0.5/-	-	[e]/-
AOX	mg/l	0.5/0.5	-/-	0.5/-	-	[e]/1.0
Sulphate	mg/l	-/-	500/200	-/[b]	300	[e]/-
Chloride	mg/l	-/-	-/-	-/[b]	b	[e]/-

[a] up to 100 mg N/l is allowed if the degree of purification is > 75 %;
[b] depends on the configuration of the treatment equipment of the public sewerage system and the conditions of the water body;
[c] according to the conditions established by the local authorities (Tallinn City Council, pollution group RG-3, limit values (RT IV, 07.08.2013, 17));
[d] environmental quality limit values for river basin specific pollutants in surface water and in effluent and stormwater discharged to a water body and ground (RT I, 30.06.2015, 4; RT I, 04.12.2012, 1; RT I, 08.01.2016, 10);
[e] in accordance with the Water Act (RT I, 30.06.2015, 4) and the requirements of the permit for the special use of water;
[f] the highest environmental quality limits value for inland surface water. The limit value for Cd concentration depends on the hardness class of the water (RT I, 30.06.2015, 4; RT I, 04.12.2012, 1; RT I, 08.01.2016, 10).

The tendency of LWW treatment on-site is related to the ever-tightening requirements for the pollutant concentration and toxicity of wastewater discharged into the public sewerage system, the increasing costs of the treatment service, and lastly the arising issues with heavy metals, waste incineration and ash disposal (Kaartinen et al., 2009).

The biological co-treatment of leachate and domestic wastewater is widely used because of its various technological and economic advantages (Tallinn and Pärnu domestic WWTP-s as well as the LWW from the landfills Jõelähtme and Paikre are co-treated) (Paper III).

At the Jõelähtme and Paikre landfills, regulations approved by the Tallinn and Pärnu city councils (such as Regulation No. 53 by Tallinn City Council from 14.06.2006 "Wastewater discharge fee differentiation guidelines" (RT IV, 07.08.2013, 17)) should be taken into account when discharging LWW into the public sewerage system. The regulation is used for differentiating between the fee for discharging LWW into the public sewerage system according to the pollutant concentration in the wastewater (see also Table 4.6). In order to reduce the fine concerning the above standard pollution, LWW or leachate must be treated on-site before it can be discharged into the public sewerage system. However, to avoid the hindering influence of leachate in the treatment processes and to guarantee the quality of the treated water, it is essential that the share of leachate in the mixture does not increase above 5–10 % (Di Iaconi et al., 2006; Ried and Mielcke, 1999).

Our research results have shown that efficient treatment of LWW (including leachate) is complicated by the significantly variable pollutant concentration and flow rate of LWW (which depend on the landfill waste layer age and intensity of precipitation), the low temperature of LWW in winter and early spring, very high values of certain parameters (BOD, COD, TN) and the high toxicity of LWW. At present, the concentration of metals and other hazardous substances (see Table 4.4) in LWW and leachate exceeds the limit values for effluent established by new legislation (see Table 4.6). Achieving such a degree of purification of LWW which is established by Estonian legislation (which has been harmonised with the Directive 2008/105/EC of the European Parliament and of the Council), is technologically very complicated, costly and economically unjustified.

5. RESULTS AND DISCUSSION

5.1 Decreasing the amount, equalisation and treatment of landfill wastewater

The rain and snowmelt water runoff from paved areas (including composting areas) is quickly directed into an equalising tank. The retention time of rainwater in the waste deposit depends on the thickness and density of waste layers. The flow rate, pollutant content and pollution load of the LWW have a big impact on the efficiency and economy of LWW treatment. The hindering factors for the aerobic biological treatment of LWW (for example aerated lagoons, AS, sequencing batch reactors, biofilm reactors) and physical treatment processes (for example RO) are the large fluctuation of the flow rate and pollutant content of the LWW, low temperature in winter (from 1 to 4 ºC) and the toxicity of the leachate. In the case of heavy rainfall and snowmelt, one has to deal with very big hydraulic and pollutant shock loads (Table 4.1). During a longer drought period, the flow rate of the LWW may be close or even equal to zero. LWW contains chemical substances that are difficult to degrade and, as a rule, is heavily polluted by organic and inorganic compounds (Tables 4.2 and 4.4) (Paper I). LWW or leachate treatment faces three specific problems, usually not met in sewage treatment: large variations of flow, changes in composition over time and changes in composition for new landfill sections (Christensen, 2010; Studies, 2010).

To decrease the pollution load, flow rate and toxicity of the wastewater requiring treatment, composting of biodegradable waste in landfills and conduction of runoff from composting areas into equalisation tanks should be terminated. Biogas and nutrient-rich digestate are produced when applying methane fermentation to biodegradable waste.

In the waste deposit recirculation of leachate as well as equalisation of the flow rate and pollutant concentration of the LWW take place. The LWW, including leachate, is considered to be toxic and hinders the microbiological processes in the biological treatment process. The LWW toxicity should be taken into account while choosing the technology for LWW treatment. Equalising tanks are used for minimising the flow rate and toxicity of LWW to the treatment plant as well as equalising the pollutant load and concentrations (Paper III).

Unpolluted rainwater can be collected separately and used for irrigating the waste deposits to promote biogas production or to dilute the leachate during drought periods to promote its treatment. Polluted rainwater and leachate are to be conducted to an equalising tank. In dimensioning equalising tanks, it should be taken into account that, according to a previous study, the share of rainwater that flows from a landfill is up to 20 % for old landfills and up to 60 % for new landfills (Studies, 2010). The volume of the equalising tank and fluctuation range of its water level is determined using an integral graph compiled on the basis of annual rows of runoff values, so that the buffering capacity of the equalising tank would be maintained for whole year round. The water from the equalising tank can be used for putting out possible fires in the waste deposit as has happened at the Torma and Paikre landfills (Paper III).

Many studies and handbooks have described different methods for the treatment of LWW and leachate. Examples of leachate treatment activities are, as follows (Christensen, 2010; Sumanaweera, 2010; Studies, 2010; Aziz and Amr, 2016; Guidance, 2006; Wang, 2007; Wang, 2006; Cervantes, 2009):

- *physical treatment processes*: air stripping (CH_4 stripping, removal of ammonia-N, and stripping of other volatile pollutants); RO; removal of solids (sedimentation and settlement, sand filtration and dissolved air flotation); activated carbon adsorption (powdered and granular activated carbon); ion exchange and evaporation/concentration;
- *chemical treatment processes*: chemical oxidation processes (ozonation and hydrogen peroxide) and precipitation/coagulation/flocculation (chemical precipitation of metals, and coagulation and flocculation);
- *aerobic biological treatment processes*: suspended growth systems (aerated lagoons, AS, sequencing batch reactors and membrane bioreactors), attached growth systems (percolating filters, rotating biological contactors, biological aerated filters/submerged biological aerated filters and biofilm reactors);
- *aerobic/anaerobic biological treatment processes*: engineered wetlands (horizontal flow reed beds, vertical flow reed beds and wetland ponds).

Traditionally, aerobic biological oxidation is the most widely used treatment method (treatment with AS and biofilms), but the results of this treatment are not satisfactory due to the specifics of generation and contents of the LWW (Boyle and Ham, 1974). Combinations of aerobic and anaerobic biological oxidation have been used for the treatment of LLE (Kettunen, 1997). Aerobic/anaerobic biological treatment processes are engineered wetlands (horizontal flow reed beds, vertical flow reed beds and wetland ponds) (Boyle and Ham, 1974; Mahlum and Stalnacke, 1999; Mahlum, 1988; Mander and Mauring, 1977; Jenssen et al., 1996). Wetland treatment may function better in warmer climates that allow the vegetation to flourish for a greater portion of the year (Townsend et al., 2015).

In Norway, there are 250 domestic landfills, with 30 of those using aerated ponds, wetlands and land filtration with vertical or horizontal flow (widely used, even in regions with colder climates) for leachate treatment. A general requirement is that water's hydraulic retention period in the system should be at least 20 days (Johansen and Carlson, 1976). The created wetlands successfully purify leachate from organic compounds and SS, but there are issues with the removal of N and P, as well as high concentrations of BOD and COD (Mahlum, 1988; Mander and Mauring, 1977; Jenssen et al., 1996). Groundwater pollution must be precluded. Biological treatment technologies are largely efficient in the case of young domestic landfills, if the BOD:COD ratio is greater than 0.5 (Studies, 2010) (Paper III).

According to published research data, the most tested and used physicochemical processes are ion-exchange, ammonia stripping, struvite precipitation, membrane separation (RO), Fenton process, ozonation and oxidation processes (Boyle and Ham, 1974; Giržadas, 2009; Goi et al., 2008; Christensen, 2010; Sumanaweera, 2010; Aziz and Amr, 2016; Wang, 2007; Wang, 2006; Cervantes, 2009).

Coagulation/flocculation and active carbon adsorption are the most commonly used physical-chemical treatment methods (Wang et al., 2007). Struvite precipitation is recommended for removing ammonia. Both membrane reactors and struvite precipitation may be used for the treatment of wastewater from young landfills after anaerobic pretreatment (Wang et al., 2007).

Ozonation in combination with biological treatment decreases the toxicity of wastewater, the volume of the required oxidant and financial costs, and increases the biodegradability of the wastewater (Gottschalk et al., 2002).

Chemical oxidation including ozonation is the only process for decomposing organic matter that is unmetabolised by microorganisms. The aim of preozonation is to improve the biodegradability of the treated wastewater, whereas post-ozonation aims at the advanced treatment of wastewater (Di Iaconi et al., 2006; Beaman et al., 1998; Huang et al., 1993). After ozonation, the biodegradability of the processed water is increased, indicating the requirement of additional biotreatment (Ried and Mielcke, 1999; Kamenev et al., 2008). The main areas for using ozone are disinfection, oxidation of organic substances and compounds, removal of taste, smell and colour, and increasing biodegradability (Gottschalk et al., 2002).

The Fenton-process consists of four stages: oxidation, neutralising, coagulation/flocculation and separation of the solid and liquid phases (Di Iaconi et al., 2006; Lopez et al., 2004; Gau and Chang, 1996). Under optimal conditions the Fenton-process can decrease the COD by 70 % (Goi et al., 2008). The Fenton-process may be used for both pretreatment of LWW before biological treatment as well as for posttreatment (Di Iaconi et al., 2006; Lopez et al., 2004; Gau and Chang, 1996). In this process, the toxicity of treated wastewater decreases with the increase in its biodegradability (Lopez et al., 2004; Goi et al., 2008). In practice, the Fenton-process is used in the posttreatment stage, but it has also been recommended for processing LWW before biological treatment with the aim of increasing its biodegradability (Di Iaconi et al., 2006; Lopez et al., 2004).

In many countries, reverse osmosis, a very efficient treatment technology, is widely used for treating leachate and LWW at landfills of different age (Christensen, 2010; Sumanaweera, 2010; Studies, 2010; Aziz and Amr, 2016; Wang, 2007; Wang, 2006; Cervantes, 2009).

Lately, biological processes such as nitrification-denitrification (SBR – sequencing batch reactors, lagoons, membrane bioreactors, denitrification processes), nitrification-denitrification processes and nitrification-Anammox processes have become used for LWW treatment (Cervantes, 2009).

Generally, biological treatment methods are effective for leachate from young (< 5 years) landfills or freshly landfilled waste, but ineffective for leachate from older (> 10 years) landfills. In contrast, physical-chemical techniques that are not favoured for young leachate treatment are advised for older leachate treatment (Studies, 2010; Aziz, 2013; Aziz, 2014). A combination of physical-chemical and biological methods (such as adsorption and aerobic processes) is efficient in removal of hazardous pollutants from mature LLE (Aziz and Amr, 2016; Aziz, 2013). The high efficiency of leachate treatment methods versus leachate age can be seen in young (< 5 years) and stabilised (> 10 years) leachate treatment with NF and RO, while ion exchange has also produced good results (Studies, 2010; Abbas et al., 2009; Aziz, 2013).

With more restrictive effluent standards, and a focus on additional parameters, leachate treatment is becoming more complex, and today they are a combination of different treatment steps in many cases leachate treatments plants (Christensen, 2010). Figure 5.1 presents a flow sheet of frequently used leachate treatment options.
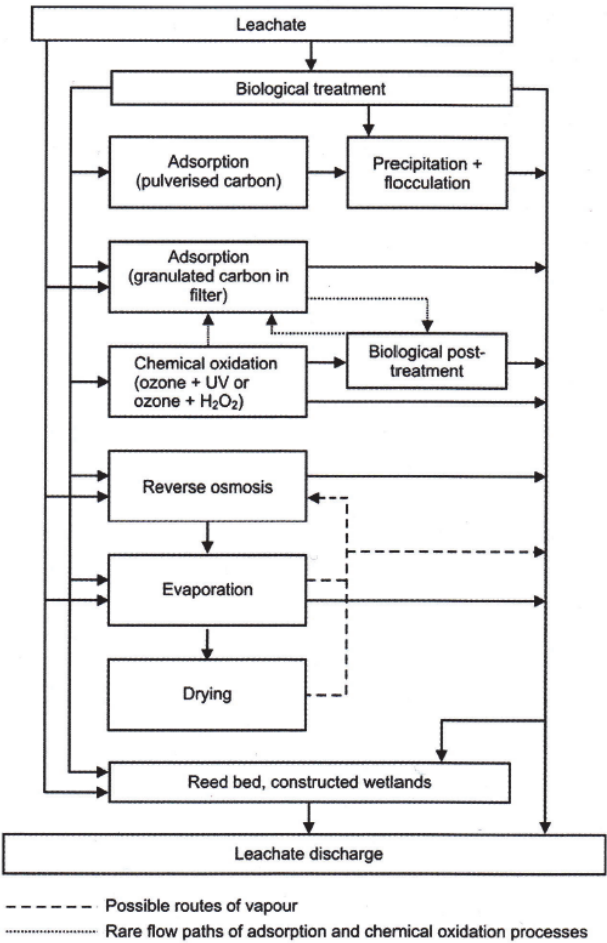

Figure 5.1. Flow scheme of possible leachate treatment systems (Christensen, 2010)

Publications and practical experience has shown that the best treatment methods for leachate and LWW from the opening until closing of the landfill are physicochemical processes with membrane separation (RO and NF), ion exchange process and different combinations of biological and physicochemical processes (Christensen, 2010; Aziz and Amr, 2016).

5.1.1. Results of experiments on landfill wastewater treatment methods

In the TUT Department of Environmental Engineering laboratory (LWW treatment with AS and biological filter with light gravel submerged support media) and in the Department of Chemistry laboratory (the rest of tests), different technologies for treating LWW were tested in the years 2007–2014. The results of laboratory studies have been presented as follows (Studies, 2010) (Paper III):

- aerobic biological oxidation (percolation, treatment with AS and submerged biological filter) of wastewater from the Väätsa landfill (which was opened at the end of 2000) was performed in 2009. The COD values of the LWW varied from 1,000 to 3,500 mgO/l and BOD_7 from 50 to 350 mgO_2/l. The biodegradability (BOD/COD) of the LWW was very small (below 0.1), and this allowed to achieve a decrease of the COD of LWW by only 30–50 %. Conclusion: aerobic biological oxidation alone does not guarantee a sufficient degree of purification if the biodegradability of LWW is low;
- the ozonation reactor allowed to achieve a 9 % decrease of the COD of LWW. In the first 20 minutes, the BOD increased by 5 % and later began to fall. The reaction of pollutants with ozone was very slow and depended on specific conditions. Therefore, the ozonation of LWW was not considered to be an efficient treatment method. The COD somewhat decreased but the overall efficiency remained low. The colour and odour were removed but the biodegradability of the LWW did not change;
- the coagulation process allowed to decrease the COD of LWW by 23 % and post-ozonation by a further 11 %. Post-ozonation of the coagulated LWW was not efficient. Additional costs are required for the purchase of reagents and treatment of the residual sediments;
- post-ozonation of the LWW that had been biologically treated in the AS plant decreased the COD by 13 %. The efficiency of this process was found to be low;
- post-ozonation of the effluent discharged from the treatment plant into the recipient increased the biodegradability of BOD/COD from 0.02 to 0.11 and COD decreased by 50 %. The efficiency of that process was sufficient.
- post-ozonation of LWW that had been biologically treated in laboratory equipment increased the biodegradability of BOD/COD from 0.006 to 0.056. The efficiency of post-ozonation was recorded as 24.4 % for COD. Biological treatment of LWW plus post-ozonation decreased the COD by 55 %. In the case of stable operation of the bioreactor, it is possible to use ozonation for increasing the biodegradability (BOD/COD ratio) of LWW that is being treated in the recirculation cycle;
- lime coagulation (10 % lime milk and 3.8% aluminium hydroxychloride were used) and post-ozonation of LWW increased the biodegradability (BOD/COD ratio) of the treated LWW from 0.038 to 0.540. COD decreased by 23–27 %. The efficiency of the process was considered to be low;
- coagulation with oil-shale ash plus post-ozonation of LWW increased the biodegradability of untreated LWW less than post-ozonation of LWW that had been coagulated with lime milk. It is not practical to use oil-shale ash for coagulation, as the required amount of ash is large and lengthy intensive mechanical mixing is needed. The

biodegradability of LWW treated with oil-shale ash increased less than in the case of ozonising untreated wastewater. The decrease of COD in post-ozonation was small;

- the Fenton process constituted the first series of experiments, where pH = 3, the overall content of pollutants (COD) was decreased by up to 70 % with a low H_2O_2:COD ratio (0.5/1), colour and odour were removed and biodegradability (BOD:COD) was increased to 0.1;

- in the second series of experiments, in which pH was not regulated, the Fenton process decreased the COD of LWW by up to 37 % with the highest H_2O_2:COD ratio 2/1. There was no change in the biodegradability (BOD:COD) ratio. Treating LWW with the Fenton process at a pH value of 8 was (unlike literature (Goi et al., 2008) data) less effective than processes in acidic medium. The best result, 37 %, was achieved at the H_2O_2:COD ratio of 2:1. The biodegradability, in this case, did not change significantly.

During the years 2007–2010 the TUT Department of Environmental Engineering conducted NF and RO treatment tests for treating the LWW of the Väätsa landfill. Physicochemical treatment processes such as RO and NF proved to be most promising and effective in the cases of a low concentration of organic matter. NF has a place between ultrafiltration and RO in the removal of recalcitrant organic compounds and heavy metals from leachate (Marttinen et al., 2002). In general, NF has a high rejection for sulphate ions and dissolved organic matter but very low for chloride and sodium (Chaudhari and Murthy, 2010). NF treatment allows to achieve a more than 85 % efficiency of total SS, heavy metals, conductivity and COD removal. However, the removal of NO_3^- and NH_3 was much lower (45.5 % and 20.5 %, respectively) (Mohammad et al., 2004). The overall process can be improved if combined with conventional biological-physicochemical treatment (Aziz and Amr, 2016). Unlike RO, NF technology has a looser membrane structure enabling higher fluxes and lower operating pressure for the treatment of leachate (Kurniawan, 2006). The main problem of NF technology is membrane fouling (Aziz and Amr, 2016).

In the case of high salt content, RO is used for additional treatment of biologically pretreated LWW. RO is also used as an independent method for treating leachate (Sumanaweera, 2010; Christensen, 2010; Guidance, 2006; Wang et al., 2007; Cervantes, 2009). Low operational costs and the ability to remove the organic pollutants and 95–99 % of inorganic salts with minimal chemical requirements make the RO an attractive technology for many applications (Wang et al., 2006). In 1988, Pall-Exekia developed a disc-tube-module to allow the system to be cleaned from scaling, fouling and bio-fouling in an effective manner (Peters, 1996). RO operated at a high pressure and is able to diminish heavy metal concentrations at high efficiency. RO technology was reported as most effective in COD removal among different physicochemical processes evaluated (Aziz and Amr, 2016).

Our ULTRA-FLO PTE, UF-NF 200 pilot plant of low pressure RO (Figure 5.2) was purchased from the Ultra-FLO Pte Ltd. in Singapore. The system consists of two low pressure RO membrane (4040 – spiral membranes) cartridges and one BT420 UF cartridge, one 5 μm guard-filter, one feed pump, and control valves, pressure gauges and flow meters (Paper II).

Figure 5.2. Experimental set-up of RO and NF (views from front and rear)

Biologically treated Väätsa LWW (Figure 5.3) was passed through a cloth filter and two microfilter cartridges BB 5 mm. Finally, it was conducted through either a low pressure (pressure no more than 6 bar) RO spiral membrane or a NF spiral wound NE 4040-90 membrane. During the exhaustion test, two successive 5 mm microfilter BB Cartridge Filters, or two new filters of spiral wound PP 5 μm, and two consecutive RO spiral wound NF 2-4040 or NF spiral wound NE 4040-90 were used to test the lifetime of these membranes in treating biologically treated LWW (Paper II).

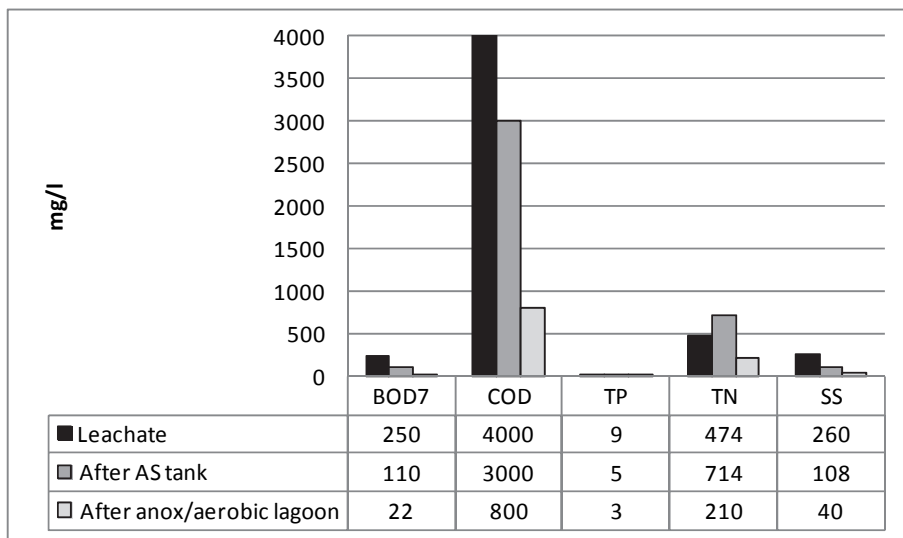|	BOD7	COD	TP	TN	SS
■ Leachate	250	4000	9	474	260
■ After AS tank	110	3000	5	714	108
□ After anox/aerobic lagoon	22	800	3	210	40

Figure 5.3. Treatment results at the Väätsa LWW treatment plant

Figure 5.4 shows that the treatment efficiency of BOD_7 was significantly less than that of COD.
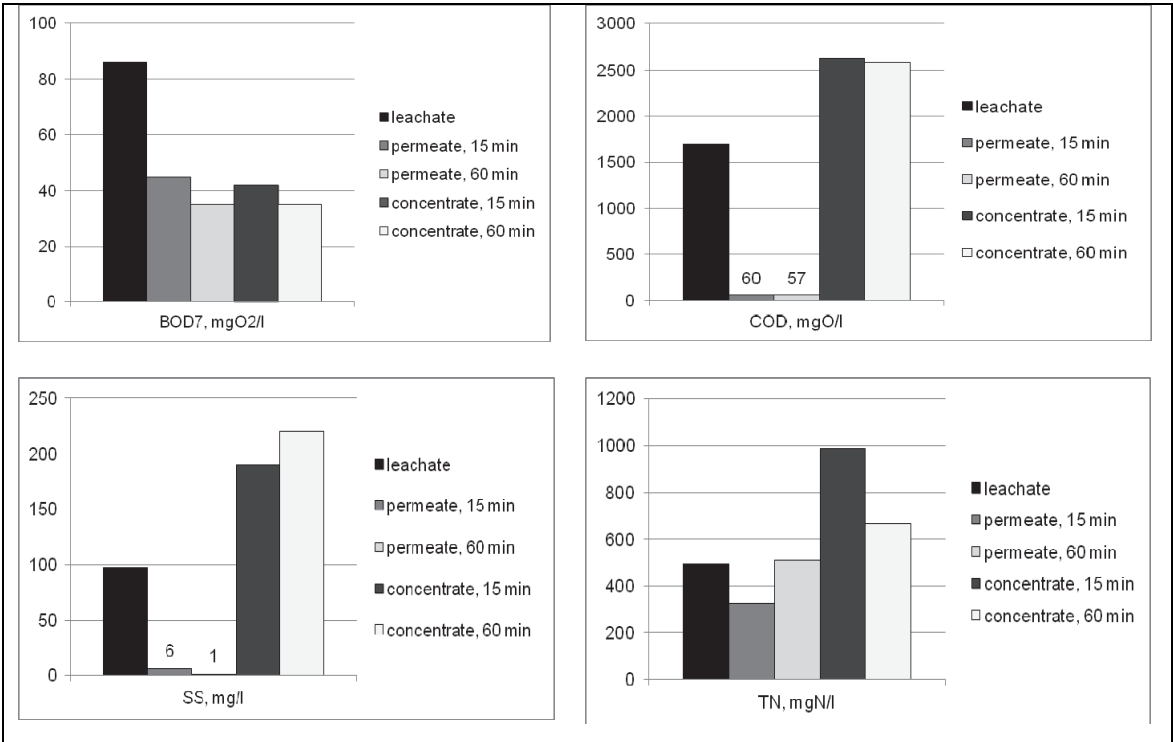

Figure 5.4. Treatment efficiency of RO

For example, the average removal efficiency of BOD_7 was only 60 % while COD removal could be as high as 98 % for either 15 or 60 min filtration. This result suggests that soluble organic compounds may pass the membrane and enter the permeate. For SS, excellent percent removal has been achieved. After 60 min of RO treatment, the purified permeate reached the following quality: COD 57 mgO/l, BOD_7 35 mgO$_2$/l, and SS 1 mg/l, which meet all discharge standards. However, the total N remained as high as 512 mgN/l.

The indicators of purified water deteriorated during experimental runs due to either organic compounds, adsorbed on the membrane surface or clogging of the filter module by SS. Since the RO membrane is a spiral filter, it was easily clogged by the SS. In addition, a significant portion of organic matter in leachate is humic substance, which is a refractory organic chemical. The fulvic and humic compound content in the leachate increase with landfill aging (Paper II).

Figure 5.5 indicates that NF reduced the COD, BOD and TN of LWW by 98 %, 41 % and 68 %, respectively. After one hour of NF, the effluent reached the COD 32 mgO/l, BOD_7 17 mgO$_2$/l, TN 202 mgN/l, and SS 3 mg/l. Initial test results showed that RO and NF allowed to achieve a good efficiency in reducing COD, BOD and SS. However, they were ineffective in reducing TN (Paper II).

Figure 5.5. Treatment efficiency of NF
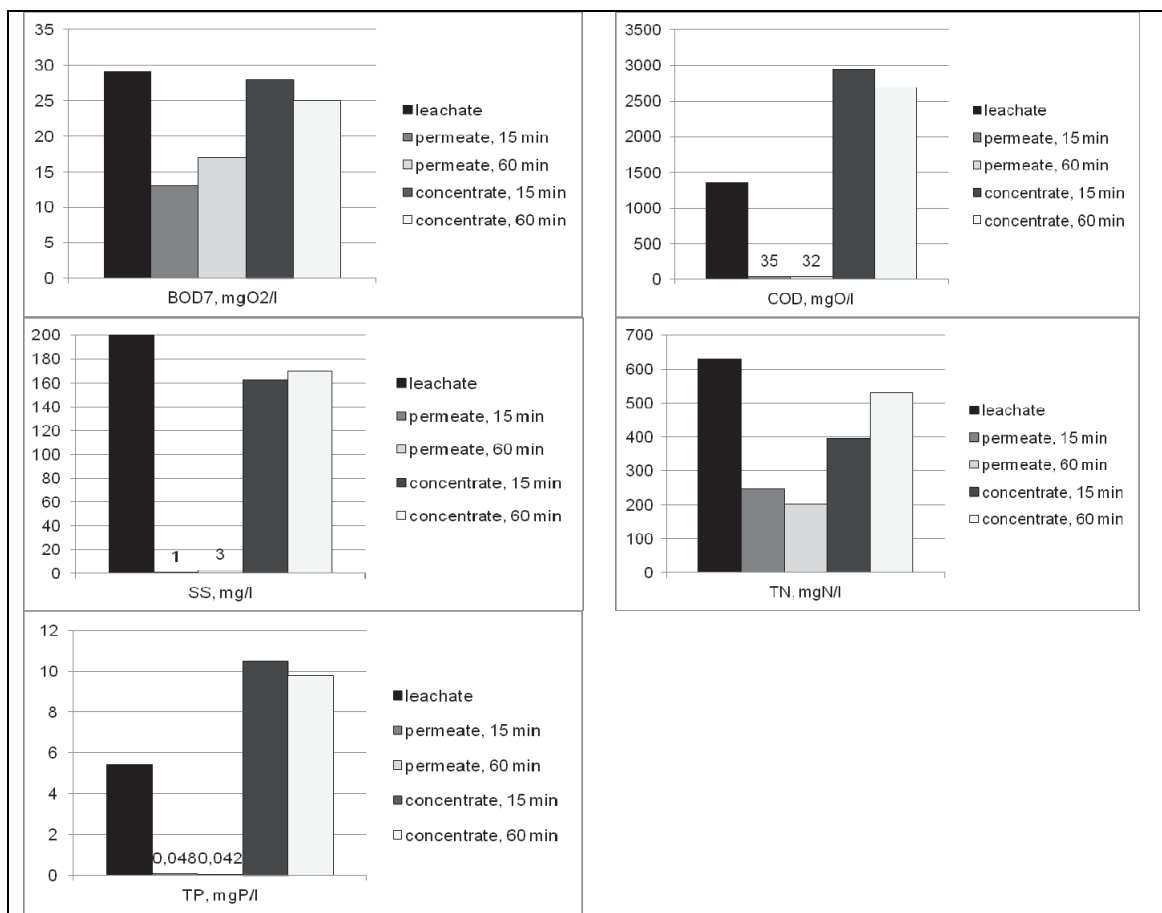
Figure 5.6 quantifies these advantages by comparing the treatment efficiency of biologically treated LWW with that of RO. It is interesting to note that NF gave better results in terms of reduction of conductivity, TN, TP and SS, while RO and NF were equally effective in removing BOD_7 and COD, with removal efficiencies greater than 95 %.
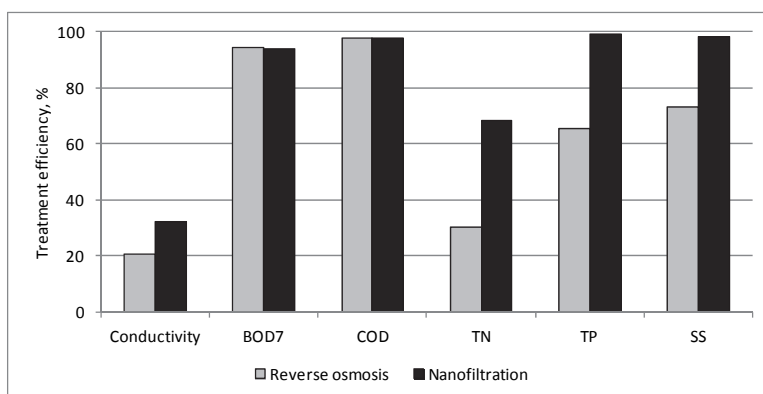

Figure 5.6. Efficiency of RO and NF treatment

The RO process reduced COD and BOD by 97 % and 60 %, respectively. NF reduced the COD, BOD and TN of biologically treated LWW by 98 %, 41 % and 68 %, respectively. However, RO was ineffective in removing TN. NF was more efficient in removing TP and TN than RO. Although the resulting COD, BOD, SS and TP may meet current legislative requirements, neither NF nor RO could bring the TN below the discharge limit of 75 mgN/l (RT I, 04.12.2012, 1). In addition, successful application of membrane filtration technologies requires efficient control of membrane fouling, especially when spiral membranes are used (Studies, 2010) (Paper II).

5.1.2. Toxicity of landfill wastewater and reverse osmosis concentrate

The toxicity of LWW was measured by ecotoxicological tests on the basis of the impact on *Protozoa*; subsequently, the impact on the bacteria in the AS was concluded. The wastewater from landfills is considered to be toxic and to hinder microbiological processes in the biological treatment process. Toxicity is caused by the high content of NH_4-N (for example, it was up to 974 mgN/l at the Jõelähtme landfill, up to 852 mgN/l at the Uikala landfill and up to 330 mgN/l at the Väätsa landfill – see Table 4.1) (Studies, 2010). It was found to increase in summer due to high pH and temperature. 90 % of total nitrogen was in the form of NH_4-N, with a big share of the latter being in the form of ammonia, which is toxic for water organisms and hinders biological treatment. In addition, the toxicity of the aquatic environment can be influenced by pH, conductivity, concentration of chlorides and the content of copper and zinc. The xenobiotic organic compounds contained in the leachate can be utterly toxic. All these determinants should be taken into account when choosing a technology for LWW treatment (Studies, 2010; Studies, 2007; Studies, 2008) (Paper III).

The LLE can become toxic also due to many other factors, such as excessive content of diluted heavy metals, a too low or high pH level, an unfavourable carbon to nitrogen ratio (C:N), etc. (Handreichung, 2006). The pH and concentration of diluted heavy metals were within norms. The C:N ratio was 4, which is considered to be too low. The low C:N ratio refers to a low carbon and excessive nitrogen content. During the process, carbon is first used in fermentation, and if its amount is insufficient, nitrogen becomes toxic for methane bacteria (Handreichung, 2006; Pitk, 2014). The normal C:N ratio for biogas production is 10–40 (Manikandan and Viruthagiri, 2010).

A series of experiments with methane fermentation were carried out with the aim of determining the toxicity of the RO concentrate produced at the Väätsa landfill and its influence on the different phases of degradation in the waste deposit as well as the degradation of organic substances during fermentation. The RO concentrate was co-digested anaerobically in a mixture with Tallinn WWTP sewage sludge to evaluate the degradability and CH_4 productivity in various mixing ratios.

Characteristics of the RO concentrate from Väätsa LWW are presented in table 5.1 and the biomethane potential (BMP) batch experiments depicted in Figures 5.7–5.10.

For comparison, Tallinn WWTP compost was anaerobically co-digested in a mixture with Tallinn WWTP sewage sludge to evaluate its degradability and CH_4 productivity (Figures 5.7 and 5.8). As can be seen, the BMP of the compost (8.9–67.7 m^3CH_4/m^3, see Figures 5.7–5.8) and one stage digestion tests gave promising results compared with those of the Väätsa LWW RO concentrate (0–4.1 m^3CH_4/m^3, see Figures 5.9–5.10) and even with those of Tallinn WWTP sewage sludge (2.6–8.0 m^3CH_4/m^3, see Figures 5.7–5.10). The Väätsa LWW RO concentrate BMP test showed that adding a LWW RO concentrate to a substrate in co-digestion, the biogas flow rate and accumulated biogas amount will drop but one can increase the biogas production by adding

compost to the co-digested substrate. What is more important is that compost can be digested alone without the addition of sewage sludge or some other easily fermentable material. Unforunately, the Väätsa LWW RO concentrate could not be digested alone due to lack of carbon as well as nitrogen inhibition (Paper III). According to the graphs, we can say that the optimum hydraulic retention time for the digestion of compost and co-digestion of compost with sewage sludge is 7 to 10 days. By these 7 to 10 days, most of the biogas has been produced, and further biogas production is minimal and not ~~as~~ significant.

Table 5.1. Väätsa LWW RO treatment concentrate in October 2014 (Paper III)

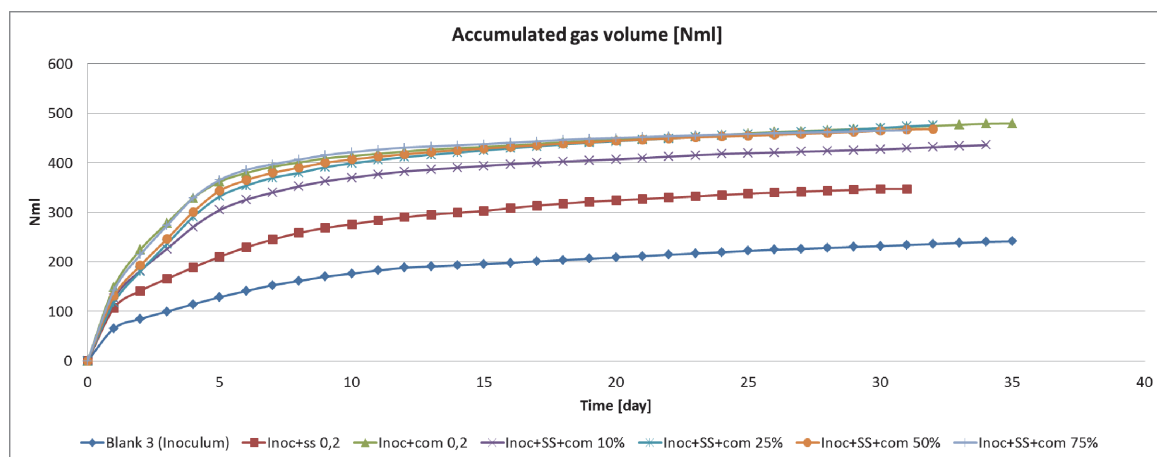| Parameter	Result
TS in RO treatment concentrate	3.7 %
VS*	43.1 % TS
pH	6.9
TN	1.2 kg/m^3 TS
NH$_4$-N	0.94 kg/m^3 TS
TP	0.02 kg/m^3 TS
total potassium (TK)	0.47 kg/m^3 TS
crude protein	10.54 % TS
crude fat	0.02 % TS
C	5.07 % TS
N	1.27 % TS
hydrogen H	1.37 % TS
S	0.43 % TS
Zn	0.87 mg/kg TS
Cu	0.13 mg/kg TS
Hg	not found
Cd	not found
Cr	3.30 mg/kg TS
Ni	0.64 mg/kg TS
Pb	< 0.01 mg/kg TS

* Volatile solids in TS

Figure 5.7. Accumulated gas volume in BMP test series. Anaerobic co-digestion of Jõelähtme landfill compost in a mixture with Tallinn WWTP sewage sludge
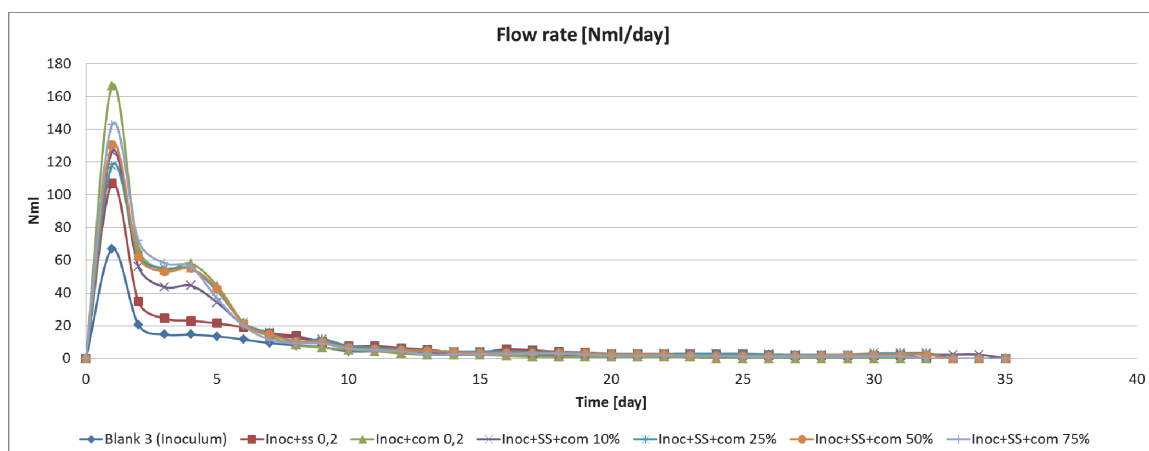
Figure 5.8. Biomethane flow rate in the BMP test series. Anaerobic co-digestion of Jõelähtme landfill compost in a mixture with Tallinn WWTP sewage sludge

At first, the Väätsa LWW RO concentrate BMP test results were promising (Figures 5.9 and 5.10). However, after the removal of inoculum (sewage sludge in distilled water), the productivity revealed a negative outcome. The RO concentrate had a negative effect on the AD process with or without sewage sludge. Even diluting the RO concentrate with distilled water did not give a positive result (Paper III).

Figure 5.9. Accumulated gas volume in the first BMP test series. Co-digestion of Väätsa LWW RO concentrate in a mixture with Tallinn WWTP sewage sludge

The addition of RO concentrate has a negative effect on the anaerobic digestion of sewage sludge. The decline in the CH_4 yield might be caused by deterioration of methanogenic bacterial activity following the treatment of discharged RO concentrate.

Since degradation processes similar to methane fermentation take place in the landfill, directing RO concentrate to the landfill will slow down the decay processes of biodegradable waste. The concentrate should be directed to a closed section of the landfill, where the extraction of biogas is in the final stage, as it is toxic and does not support biological decomposition processes.

Unclosed landfills can be moistened with rainwater from the landfill territory as well as LWW (Paper III).
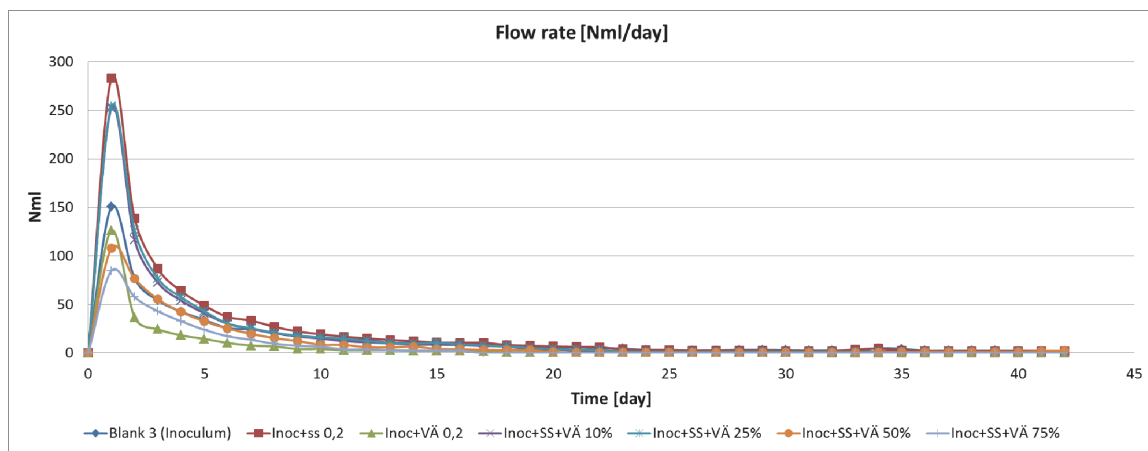
Figure 5.10. Biomethane flow rate in the first BMP test series. Väätsa LWW RO concentrate co-digestion in a mixture with Tallinn WWTP sewage sludge

5.1.3. Treatment of landfill wastewater at the Väätsa, Torma and Uikala landfills

In Estonia, biological treatment methods (such as sand filtration, oxidation pond and AS) have been used for treating LWW in young landfills. These methods have had to be replaced with more efficient treatment methods and their combinations at the landfills have aged and the content and concentration of pollutants in the LWW have increased.

The Väätsa and Torma landfills were reconstructed and the Uikala landfill designed after 2000 in accordance with the EU environmental requirements.

For treating the **Väätsa landfill** wastewater a stabilisation pond (combination of an usual and aerated oxidation pond) and biological treatment with activated sludge (AS) were previously used. In addition to the studies carried out in 2007 (Studies, 2010), the efficiency of operation of the AS plant was monitored during the rainy autumn 2008 and the snowmelt period in spring 2009. The BOD_7:COD ratio dropped to less than 0.1, which indicates the inefficiency of the biological processes (see Table 5.2). The TN concentration could not be reduced to the required 75 mg/l. The high content of ammonia-nitrogen seemed to be toxic to the microorganisms taking part in the AS process. Consequently, the effluent was dark and contained solids in high concentrations. According to the results of the current study, during rainfall and snowmelt, a significant part of the flow and pollution load of the LWW originates from the composting areas of biodegradable waste, which must be decreased substantially. A tank for equalising the flow rate, pollution load and toxicity of the LWW should be constructed. During winter, the decrease of the temperature of the leachate directed into the treatment plant should be minimised. The removal of fat and oil prior to biological treatment is very important. It was understood that biological methods were insufficient for treating the LWW to meet the requirements, and RO should be added. In 2009, tests on different methods for the treatment of LWW leachate were conducted. By the end of the year, a preliminary project for reconstructing the Väätsa LWW treatment plant was completed (Preliminary, 2009).

The LWW treatment plant at Väätsa was completed in 2002. This biochemical plant consisted of an AS container (aeration tank and lamella clarifier) together with an oxidation pond and an

aerated sludge stabilisation tank. The first section of the oxidation pond was aerated. Before directing wastewater into the treatment plant, its chemical composition was regulated, when necessary, by adding phosphoric acid to avoid P deficit. P removal was achieved by dosing iron sulphate into the AS plant. The excess sludge that was generated during the treatment process was directed into an aerated stabilisation tank, and the clarified water recycled into the treatment process. The stabilised sludge was directed to a depositing area. Water from the AS plant was treated in an aerated oxidation pond. In addition to the studies in 2007, the efficiency of the Väätsa landfill AS plant was monitored during the rainy period in autumn 2008 and snowmelt period in spring 2009 (Studies, 2010). The samples taken in May 2009 indicated the inability of the treatment plant to operate according to the requirements (Table 5.2) (Studies, 2010) (Paper III).

Table 5.2. Content of pollutants in the Väätsa LWW and wastewater from different treatment stages in May 2009 (Studies, 2010)

Pollutant	Unit	Väätsa LWW	Pollutant content after treatment with AS	Pollutant content after stabilisation pond treatment	Permitted limit values in effluent (RT I, 04.12.2012, 1)
BOD_7	mgO_2/l	250	110	22	25
COD	mgO/l	4,000	3,000	800	125
TP	mgP/l	9.0	4.5	2.6	2.0
TN	mgN/l	474	414	210	75
pH		8.52	8.53	8.21	6–9
SS	mg/l	260	108	40	35

The first one hectare large phase of the Väätsa landfill was put into operation in November 2000, and the second (1.34 ha) in November 2005. The planned height of the phase was 6–7 m. In 2008 the third landfill phase (2.8 ha) for landfilling domestic waste was completed. Altogether, there are four phases, covering 8.8 ha. The first composting area of 0.268 ha was ready in November 2003 and the second (1.34 ha) was put into operation in July 2008.

A new LWW collection system and treatment plant was designed and built during 2011/2012. It is a WWTP that consists of an equalising tank, physicochemical treatment (RO) unit following biological treatment (AS) and stabilisation in a pond treatment system. Since April 2012, when the new treatment system was put into operation, the effluent from the WWTP has been in compliance with the water permit requirements (see Table 5.3). The average treatment efficiency in the period between 2013 and 2015 was recorded as over 99 % for BOD_7, COD, TN and TP and over 90 % for SS (Paper III).

Table 5.3. Average pollutant content in the Väätsa LWW effluent according to the Väätsa landfill annual water use report

Parameter	2013	2014	2015	Limit value (RT I, 04.12.2012, 1)
BOD_7, mgO_2/l	3	3	3	25
COD, mgO/l	14	14	14	125
SS mg/l	6.5	2.25	2.5	35
TN, mgN/l	1.5	2.1	1.6	75
TP, mgP/l	0.04	0.025	0.02	2
monobasic phenols, mg/l	0.0054	0.0021	0.0003	0.1
dibasic phenols, mg/l	0.01	0.0065	0	15

The first phase (0.65 ha) for domestic waste of the **Torma landfill** was completed in June 2001. The designed average height of the phase was 6 m. By the end of 2007, a second phase of 1.58 ha was in operation. Here, the planned height was 7 m. It is forseen to create a third phase (0.85 ha) having a height of 6 m (Paper III).

LWW was previously treated using a buried sand filtration unit followed by sedimentation pond treatment. After several years of usage the efficiency of the buried sand filtration unit became very low, probably because treatment capacity of the filter bed body became exhausted. For improving the situation, a new landfill was designed, based on results of tests conducted during design work. Test results made clear that mere mechanical-chemical treatment (coagulation with ferrosulphate and flocculation which decreased the COD and BOD only by 10 %) or activated sludge treatment (decrease of COD and BOD up to 10–30 %) would not enable to reach the necessary efficiency. Nevertheless, AS treatment provided necessary conditions for further chemical treatment, but this needed further investigation. The adaptation of activated sludge is a prolonged process ond one must consider adding fresh sludge from some other AS plant. Special treatment (possibly PO) is necessary for removing nitrogen. Excess P is removed by chemical treatment. Large doses of chemicals are needed and a large amount of floating sludge is produced what should be taken into account whwn designing contact chambers and clarifiers with surface sludge scrapers. Chemicals (metal salts) lower the pH and raise the sulphate ion content. At Torma, in summer part of the LWW is pumped to the landfill to compensate evaporation. In winter the leachate is conducted to the purification plant from the collection well, were it is somewhat warmer. According to test results it recommendable to dimension the equalisation basin on the basis of the annual precipitation (in Estonia about 800 mm, i.e. 8000 $m^3/(a \cdot ha)$) plus the average omount of leachate, multiplied by 0.5.

On the basis of the test results in 2009 a new WWTP was designed and put into operation in 2010. The plant consists of screen, an AS plant with long detention time (at least 6 days), a chemical treatment (coagulation with ferrosulphate and flocculation) unit, active sand filter and oxidation pond. For equalising the flow and pollutant content the volume of the existind sedimentation pond was enlarged and equipped with a mixing device. The chemical sludge is pumped to the landfill. The efficiency indicators pH, SS, BOD and COD as well as TP improved after the new treatment system was put into operation, but COD and TN remained problematic (Figures 5.11 and 5.12). (Tõnisberg, 2011) (Paper III.

In 2012, the conservation and closing activities of the landfill were started as the amount of deposited waste had significantly decreased. Most of the LWW is pumped back to the landfill for moistening the waste and, in the last years, only polluted rainwater collected from the landfill territory is conducted to the treatment plant. In the first quarter of 2013, no rainwater runoff occurred from the landfill and no effluent was directed to the recipient. Thereafter, only contaminated rainwater from the landfill territory was treated in the WWTP; the leachate was pumped back into the waste deposit. In 2014, only rainwater from open areas was treated and the leachate was pumped back to the landfill. The treatment efficiency was high: BOD_7 96.4 %, SS 94 %, COD 93.1 %, TN 92.2 % and TP 96.9 %.

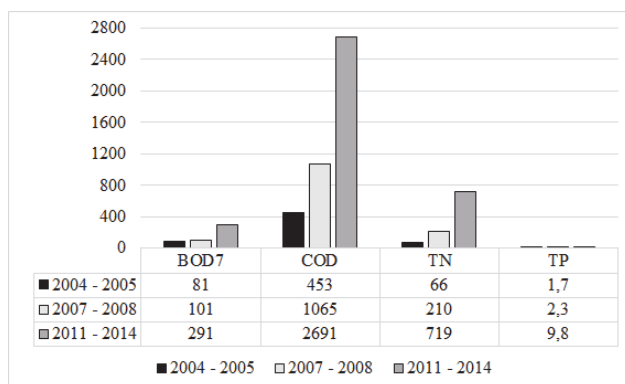|	BOD7	COD	TN	TP
■ 2004 - 2005	81	453	66	1,7
□ 2007 - 2008	101	1065	210	2,3
■ 2011 - 2014	291	2691	719	9,8

■ 2004 - 2005   □ 2007 - 2008   ■ 2011 - 2014

Figure 5.11. Average pollutant content of wastewater treated at the Torma LWW treatment plant during the years 2004–2005, 2007–2008 and 2011–2014 according to Torma landfill annual water use report

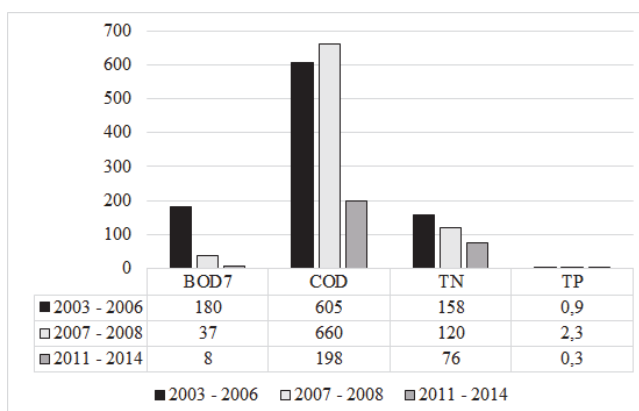|	BOD7	COD	TN	TP
■ 2003 - 2006	180	605	158	0,9
□ 2007 - 2008	37	660	120	2,3
■ 2011 - 2014	8	198	76	0,3

■ 2003 - 2006   □ 2007 - 2008   ■ 2011 - 2014

Figure 5.12. Average pollutant content of the Torma WWTP effluent discharged to the recipient during the years 2003–2006, 2007–2008 and 2011–2014 according to the Torma Landfill annual water use report

The first phase (2.2 ha) of the **Uikala landfill** was put into operation in January 2000, and in November 2006 the second phase (1.9 ha) was completed (total landfill area 8.8 ha). The designed average height of the landfill is 23 m. The composting area is about 3.4 ha (Studies, 2010). For treating LWW, RO was applied in 2005, and an equalising pond was constructed. The survey of the operation of the container RO treatment plant with filters DT 29-09 following an equalising tank showed that the treatment plant for LWW operates with high and stable efficiency. In the fourth quarter of 2013 the medium treatment efficiency was 98.3 % for BOD_7, 98.7 % for SS and over 99 % for both TN and TP (Paper II).

5.2 Treatment of biodegradable wastes

The biodegradable waste in Estonia consists of the biodegradable part of domestic waste, park waste, yard waste, agricultural waste, commercial waste, industrial waste, sewage sludge and animal waste. In 2011, the amount of biodegradable waste generated in Estonia was 1,196,670 tonnes: 158,900 tonnes (13 % of total amount) of sewage sludge, 123,100 tonnes (10 %) of biodegradable domestic waste (kitchen, catering, park and yard waste) and 7,970 tonnes (1 %) of other biodegradable waste. Altogether 289,970 tonnes (24 % of the total amount) were considered

to be suitable for a fermentation process leading to biogas production. Other biodegradable waste included wood, paper and cardboard (a total of 906,700 tonnes) (National, 2014).

Most of the aforementioned materials were composted on composting areas located at the landfills or on separately located composting areas. A part of biodegradable waste was deposited in landfills. In the WWTP-s in Tallinn, Narva and Kuressaare sewage sludge is treated by methane fermentation before composting. At the Tartu treatment plant, methane fermentation is still being adjusted. The produced biogas is used for energy production, and the digestate utilised for fertilising fields, landscaping and recultivation.

In 2010, 13.13 million m^3 of biogas was produced in Estonia, and most of it (9.3 million m^3) at landfills. A total of 3 million m^3 of biogas was produced from sewage sludge (Applicability, 2014).

In 2013 sewage sludge from WWTP-s with a treatment capacity of over 2,000 p.e. was used in landscaping (54 %), agriculture (16 %), recultivation (13 %) or in other ways, such as transportation (6 %) (Elaboration, 2015).

The annual potential of biomethane (excluding agricultural biomethane) has been recorded as follows: biodegradable waste from food manufacturing 9 million m^3, biowaste 2 million m^3, sewage sludge 3 million m^3, biowaste from industry 8 million m^3 – altogether 22 million m^3. This is supplemented with landfill biogas of 9.3 million m^3 (Applicability, 2014).

In comparison with the present situation where the main activities are composting and depositing of waste into landfills, the impact of methane fermentation on the environment seems to be significantly smaller (Paper III).

According to the Estonian National Waste Action Plan, it is of primary importance to decrease the entire volume of deposited waste by 2014–2020. The utilisation of biological waste should be increased significantly and anaerobic fermentation preferred over composting. The fermentation residue (digestate) should be used in agriculture as much as possible. The optimal plan for managing domestic waste by 2020 with the least environmental impact stipulates that 30 % of waste will be recycled as secondary raw material, 3 % as compost and 10 % as anaerobic fermentation digestate, 40 % will be incinerated, 8.5% utilised as waste fuel in cement manufacturing and only 8.5 % deposited in landfills (National, 2014) (Paper III).

The digestate that is generated in methane fermentation of biodegradable waste contains a large amount of plant nutrients, such as P and N; therefore, it can be used for fertilising arable land. The amount of digestate is more or less equal to that of fermented biodegradable waste (Handreichung, 2006). Compost produced at the Tallinn WWTP from fermented sewage sludge is used for fertilising fields and green areas as well for recultivation purposes (Kuusik, Aare, 2003) (Paper III).

5.2.1 Decreasing the volume of biodegradable waste composted and deposited at landfills

Composting is a microbial aerobic transformation and stabilisation of heterogeneous organic matter in aerobic conditions and in solid state. The first phase of the composting process is mesophilic and starts the aerobic decomposition of easily degradable organic matter. In the

thermophilic phase without control, the temperature can easily reach and exceed 70 ºC and serve for reducing the number of pathogenic agents present in the waste. In the third phase mineralisation of slowly degradable molecules and humification of lignocellulosic compounds takes place. The composting process leads to the production of CO_2, water, minerals and biologically stabilised organic matter (Christensen, 2010; Turovskiy and Mathai, 2006).

The composting technology covers a broad spectrum of approaches, as open, enclosed and reactor technology. The open technology includes aerated pile, naturally vented pile, aerated windrows and naturally vented windrows technologies. Enclosed technologies include aerated pile, the Brikollare process and channel technology. The reactor technology includes box reactor, container reactor, tunnel reactor, tower reactor and rotating drum technology (Christensen, 2010; Wang et al., 2008; Wang et al., 2007b; Turovskiy and Mathai, 2006).

In a composting facility that produces a marketable product, the purpose of the feedstock recovery and preparation step is to remove noncompostable materials to improve the final compost quality (reduction of the visible inert material and chemical pollutant content) and to prepare the waste to adjust its biological, chemical and physical properties (Christensen, 2010; Turovskiy and Mathai, 2006).

When open technologies are used, the exhaust gas of the composting process in most cases escapes to the surrounding environment without deodorisation. Enclosed technologies and the reactor technology enable the treatment of the exhaust gas. In a reactor, the free air space above the compost is minimal, which lowers the volume of exhaust gas compared to that in an enclosed building system (Christensen, 2010; Studies, 2010; Wang et al., 2007b).

Windrow composting is currently the most widely used treatment technology for sewage sludge and other separately collected and sorted biodegradable waste. In Estonia, sewage sludge is the most commonly composted biodegradable waste on landfill territories. The most important pollutants that are generated in landfills by composting biodegradable waste are emitted into the air are NH_3 and volatile organic compounds (VOC). Also, CH_4 and hydrogen sulphide (H_2S) might be emitted. During the microbiological degradation of organic nitrogen compounds nitrous oxide (N_2O) is generated. H_2S forms during anaerobic degradation. In an anaerobic environment, if the microbiological degradation process is balanced, VOC breaks down into CH_4 and CO_2. CH_4 emerges when organic substances break down in anaerobic conditions. Environmental issues are mainly related to the risk of surface and ground water pollution as well as unpleasant odours. The emissions not only cause acid rain (NH_3, SO_2, NO_X) but also ozone layer depletion (methyl bromide CH_3Br), greenhouse gases (CO_2, CH_4, N_2O) and heavy metal diffuse pollution (Environmental, 2004; Environmental, 2008; Strategic, 2009).

Greenhouse gases consist of carbon dioxide (76.7 % CO_2), methane (14.37 % CH_4), nitrous oxide (7.9 % N_2O) and fluorinated gases (Aziz and Amr, 2016; Applicability, 2014). Some of the important research related to greenhouse gases, global warming and recycling indicate that recycling is an effective method for decreasing CO_2 gas emission and hence reduce global warming. Thus green technology in waste disposal can be followed, which saves us from greenhouse gases (Kothari et al., 2010; Nakata et al., 2010).

The use of low quality compost in agriculture can cause soil contamination (Elaboration, 2013).
During composting, environmental control may be necessary to appropriately handle contaminated liquids (condensate from active aeration, leachate, and rainwater runoff) and

emissions into the atmosphere (odour, bioaerosols, dust) and noise (Christensen, 2010; Wang et al., 2007b).

Before discharging into water bodies, the polluted rainwater from composting fields must be treated for removing coarse SS in straw or hay bale filters and a settlement tank.

Leachate, and to a lesser extent condensate, contain high concentrations of dissolved organic matter and nutrients, which should not reach surface water bodies or groundwater. Leachate should either be recycled to the landfill as a source of moisture or treated prior to discharge.

Odour emission during composting can be a nuisance to neighbours and has resulted in the closure of several composting facilities (Christensen, 2010; Wang et al., 2008). Bioaerosols (colloidal particles with bacteria, fungi, actinomycetes or viruses attached) can be a potential health hazard to composting facility workers and neighbours (Millner et al., 1994).

In this study the possibility of decreasing the volume of biodegradable waste deposited and composted in landfills and the potential for methane fermentation of sorted biodegradable waste at landfills was analysed. The study results show that methane fermentation followed by digestate composting is only possible in the case of biodegradable waste that is sorted before collection and pretreated by removing nonsuitable materials and is especially suitable for kitchen, food and animal waste. The volume of digestate generated in methane fermentation of biodegradable waste is almost equal to the volume of biowaste used as a raw material for this process. The digestate contains a large amount of nutrients, such as P and N, which can be used for fertilising arable lands. Compost from WWTP sludge and methane fermented sewage sludge has been used for fertilising green areas and recultivation purposes, and to a lesser extent in agriculture (Paper III).

In Estonia in 2002–2009, on average 18.1 million tons of waste was generated per year, of which 13.8 % (2.5 million tons per year) was biodegradable waste (1.5 to 4.1 million tons per year) (Elaboration, 2013). In 2011, the amount of biodegradable waste was 1,196,670 tonnes, of which 158,900 tonnes (13 % of total amount) was sewage sludge (National, 2014). The volume of biodegradable waste varies significantly from year to year. In the last decade, the general production of waste, including biodegradable waste, has decreased (Table 5.4).

Table 5.4. Production, landfilling and recovery of waste in Estonia in 2008–2014 (million tons) (National, 2014)

Year	Production of waste	Landfilling	Recovery	Production of domestic waste	Landfilling of domestic waste
2008	19.3	11.7	6.0	0.5	0.33
2009	15.6	8.5	4.5	0.44	0.29
2010	19.5	11.7	7.3	0.41	0.27
2011	21.7	9.3	12.1	0.42	0.24
2012	22.2	8.2	13.1	0.38	0.13
2013	22.5	10.7	11.7	0.38	0.05
2014	22.1	-	8.1	-	-

In Europe, at larger WWTP-s methane fermentation is the main method for treating sewage sludge, compared to composting in smaller plants. In Estonia, the sludge treatment technology depends on the average load of the plant: in the case of loads over 2000 population equivalents the practice in 2013 was as follows: outdoor windrow composting 65 %, methane fermentation 25 %, in-vessel composting (8 %) and other treatment methods (e.g., transportation to other

treatment plants) 2 % (Elaboration, 2015). Since one of the goals of the EU energy policy is to increase energy independence, methane fermentation of sewage sludge and usage of biogas for energy production of are widely used. In order to increase digester profitability, sewage sludge is with other industrial and domestic biowaste. In the Nordic countries, biogas obtained by methane fermentation in enriched form is used in public transportation vehicles. In Central Europe, drying and incineration of sewage sludge is becoming more commonplace. When setting perspectives for sludge use, the main option is to use the nutrient-rich sludge in landscaping and agriculture (Elaboration, 2015).

The present study takes into account the quick changes in waste management (collection of sorted waste, increasing the significance of sorting and incineration of domestic waste, decrease in depositing of domestic waste in landfills and increase in the role of composting) and the impact of the mentioned changes on the amount and pollutant content of the LWW. Therefore our research involved investigation of the possibilities to significantly decrease composting of biodegradable waste at landfills and replace it with methane fermentation as well as finding possibilities for decreasing and equalising the flow rate, pollutant content and toxicity of the LWW, methane fermentation of biodegradable waste, assessment of the yield of biogas and possibilities for utilising biogas and digestate obtained by fermentation of the biodegradable waste or collected from landfills (Paper III).

5.2.2 Methane fermentation of biodegradable waste in landfills

Methane fermentation of biodegradable waste in Estonia is presently taking place in landfill waste layers. As a result of the process, landfill gas and leachate are generated.

Anaeribic digestion (AD) is defined as a biological process that produces gas (biogas) containing CH_4 and CO_2 as its primary constituents through the concerted action of mixed microbial population under conditions of oxygen deficiency. AD has been the process most often used to stabilise wastewater and biosolids (Pitk, 2014).

AD takes place at 37–40 °C (mesophilic digestion) or 50–55 °C (termophilic digestion) within 20–30 days. In this process pathogens are reduced The generated biogas is mainly used for maintaining digester temperature or in electricity production (Elaboration, 2013). In Estonia, single step anaerobic mesophilic digestion is mainly used. During the digestion process, the following changes take place in the biomass (Environmental, 2004; Environmental, 2008; Strategic, 2009; Handreichung, 2006; Wang et al., 2007b):
- the content of organic dry matter (oDM) of the parent material decreases up to 80 %;
- a large part of carbon compounds in the oDM are degraded to CH_4 and CO_2;
- the aromatic substance and organic acid content of the substrate decreases significantly;
- the NH_4 content increases and the TN content does not decrease. The P, K, Ca and Mg content do not change;
- a part of phosphorous is transformed into inorganic form, which is more easily accessable for plants;
- the digestate pH rises to 8–8.5;
- the sulphur content might decrease because sulphur becomes gaseous hydrogen sulphide and is discharged from the process together with the other gases;
- the concentration of heavy metals that are not subject to biological processes increases;
- during the digestion process pathogens are eliminated;
- animal substrates must be treated thermally at 70 °C or sterilised;

- in mesophilic single step fermentation chambers, plant seeds are destroyed within a few days;
- up to 90 % of pathogenic bacteria (such as salmonella) are eliminated in mesophilic conditions within a few days;
- the infectious capacity of parasite eggs and larvae is eliminated in mesophilic conditions within a few days.

The production of renewable energy from organic waste streams is definitely one of the important aspects in the concept of sustainable development (De Vrieze et al., 2012). At the same time, biogas is also an energy source that helps to reduce anthropogenic greenhouse gas emission if used as a replacement for fossil fuels, and it also has many additional positive environmental aspects (Börjesson and Mattiasson, 2007; Weinland, 2010; Amani et al., 2010; Plugge et al., 2010), such as reduction of acid rain and the global warming potential (Chynoweth et al., 2001), and the amount of waste as well as inactivation of pathogens (Chen et al., 2008). A further important aspect is recirculation of stabilised digestate back to the soil as an organic fertiliser and soil improver with a concurrent reduction in the requirement of mineral fertilisers (Salminen and Rintala, 2002; Demirel and Scherer, 2008; Luste et al., 2009).

Lately, more and more such large landfills are built, the waste in which is fully isolated from the environment (called reactor landfills) are being utilised (Christensen, 2010). Reactor landfills are engineered landfills that receive waste containing untreated or partly treated organic matter and therefore produce gas as well as a contaminated leachate. A distinction is made between aerobic (Hudgins and March, 1998) and anaerobic reactor landfills (Tchobanoglous and Kreith, 2002).
In an aerobic reactor landfill, hydroinsulation of the ground is needed, as well as aeration of the landfill body through installed piping. The organic compounds degrade in the landfill body and are aerobically stabilised (as when composting) in a much shorter time than in an ordinary landfill, and practically no leachate is formed. From the viewpoint of environmental protection, this is probably the best way to landfill (Studies, 2010).

Every landfill having underground hydroinsulation that keeps leachate from diffusing into the environment can be transformed into an anaerobic reactor landfill. The landfill body is irrigated or moistened, especially in the dry season, and this intensifies the methanisation of biodegradable waste during warm weather. The irrigation water is usually leachate collected from the landfill, and LWW is used if it runs out. As a result, the landfill's acute environmental hazardousness period is shorter and the production of biogas increases, which also means an increase in the use of a potential energy resource (Studies, 2010).

Biogas begins to form in a landfill when the thickness of the landfill body is 4–6 m. Biodegradable waste produces landfill gas, which mainly consists of CH_4. In addition to CH_4, the gas also contains CO_2, non-VOC and small volumes of N_2O, NO_x, CO (Eggleston et al., 2006). Gas collected from the edge areas of the landfill can also contain atmosphere gases such as oxygen and nitrogen. The production of landfill gas is usually between 60–250 litres per 1 kg degradable organic compound, which theoretically could even be 750–940 l/kg (Christensen, 2010). When calculating the shielding of thermal radiation of landfill gases, it is considered that a ton of domestic waste produces 0.085 tons of CH_4 and 0.193 tons of CO_2 in emissions that would be equivalent to about 220 l/kg. Incinerating pure CH_4 produces energy of about 55.6 MJ/kg, in normal gaseous state conditions, it is about 40 MJ/m^3. In the case of natural biogas, this is 20–25 MJ/m^3 (Christensen, 2010; Studies, 2010).

Landfill biogas (Christensen, 2010; Studies, 2010):

- is collected for as long as possible and is incinerated in order to decrease the effect of the shielding of thermal radiation to Earth (the greenhouse effect) because the thermal radiation shielding capability of CH_4 (the component of biogas) is 21 times higher than that of CO_2;
- is incinerated and its heat is used for warming the water or leachate that is used for landfill watering. Warm water accelerates biological degradation processes in the landfill body and the landfill environmental hazardous period becomes shorter;
- is purified from CO_2 (the content of which is 35–50 %) and, as a result, the calorific value of the gas for use in combustion engines increases. Nowadays, biogas is purified by filtration, which also removes NH_3 and H_2S that would otherwise damage combustion engines. Gas that has been treated in such a way can be compressed into tanks as a fuel or used on-site for electricity production.

Estonian landfills are small and the amount of collected landfill gas is often too small to make it economically viable to build a combined heat and power plant that would produce electricity and thermal energy. The first combined heat and power plants are installed at the larger closed Ääsmäe, Jõelähtme and Uikala landfills. Smaller landfills do not produce sufficient biogas for such utilisation and at the Väätsa landfill, for example, the gas is incinerated. In the future, it would be practical to equip landfills with digesters that could co-digest separately collected and sorted biodegradable domestic waste. This would increase the volume of biogas produced at landfills, and establishing combined heat and power plants would become profitable.

Methane fermentation is considered to be the most effective method of biodegradable waste treatment that generates biogas as a by-product. The digestate that is generated in methane fermentation of biodegradable waste contains a large volume of plant nutrients, such as P and N; therefore, it can be used for fertilising arable land.

5.2.3. Utilisation of landfill gas, digester gas and digestate

In our study the yield of landfill gas and digester gas was determined. Also, the nutrient content of digestate and its potential for were analysed.

The study results show that the CH_4 content of landfill gas is 50–55 %. When composting biowaste a large amount of sulphur compounds is generated, while a smaller amount is produced in the anaerobic fermentation process. Emission of sulphur and nitrogen compounds (SO_2, NO_x, HCl and NH_3) into the ambient air during decomposition of the biodegradable substances cause acidification of soil and water bodies. In the case of open composting, a large amount of ammonia (NH_3) was emitted, causing both acidification as well as a bad odour. Ammonia emissions from anaerobic fermentation are much smaller. Most of Estonian landfills are small and the landfill gas production too small for cost efficient generation of heat and energy, and the gas is combusted. For example, at Väätsa, 175,200–262,800 m^3 (20–30 m^3/h) of landfill gas was annually collected from three depositing phases with a total area of 3.5 ha (Paper III).
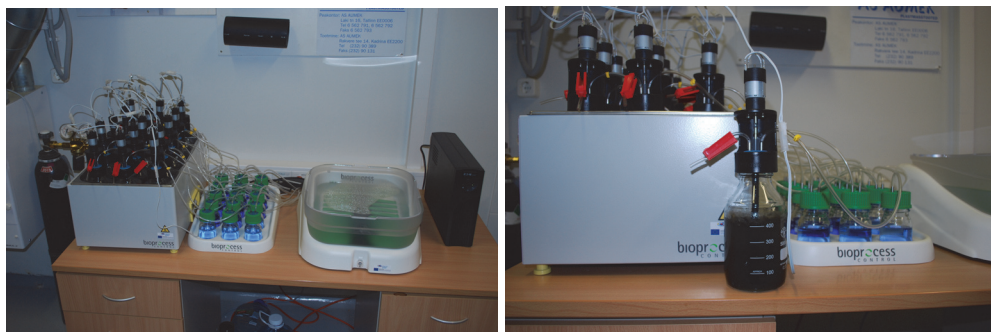
Biologically treated (composted or anaerobically digested) organic waste or unprocessed organic waste from households, gardens, commerce and industry can be recycled to agriculture and forestry (Christensen, 2010; Petersen, 1996; Poulsen, 2009). Typical characteristics of waste applied to agricultural land are presented in Table 5.5.

Table 5.5. Typical characteristics of waste applied to agricultural land based on numerous sources (including Christensen, 2010)

Parameter	Anaerobic digestate (domestic waste)	Sewage sludge/ biosolids	Compost (organic domestic waste)	Compost (green/garden waste)	Pig slurry	Tallinn WWTP anaerobic digestate (Kuusik, Argo, 2014)
TS, % ww	1–2	2–30	40–75	50–75	2.5–6.5	2.4
VS, % TS	65	40–80	35–70	20–40	80–90	60
TN, % TS	3.5	2–5	1.0–2.5	0.7–1.6	7.5–15.0	2.6
NH_4-N, % TS	1.7	0.01	<0.2	-	5.6–11.3	-
NO_3-N, % TS	0.2–1.0	0.005	-	-	0	-
N-organic, % TS	1.8	2–5	-	-	1.9–3.8	-
TP, % TS	0.9	0.5–3.0	0.2–1.0	0.15–0.7	1.8–2.8	0.03 kg/m^3
K, % TS	3.7	0.1–1.0	0.5–2.0	0.8–1.9	3.8–6.5	0.04 kg/m^3
C, % TS	39	-	20–45	10–30	38	26
H, % TS	5	-	-	-	nd	3.7
S, % TS	0.5	0.4–2.0	0.06–0.3	0.1–0.15	1	0.01
Cl, % TS	2.9	-	0.3	0.001–0.1	1.3	-
C/N total ratio	11	5–20	12–20	10–30	3–5	-
C/N organic ratio	-	nd	-	-	10–20	-
Mg, % TS	0.8–1.1	0.1–0.5	0.3–1.6	0.3–0.4	0.8	-
Ca, % TS	2–5	1–5	3–7	2.2–3.0	2.5	-
Pb, mg/kg TS	10–60	15–100	25–110	20–55	3.3	0.18
Cd, mg/kg TS	0.3–0.7	0.5–5.0	0.3–1.0	0.2–0.5	0.5	0.03
Cr, mg/kg TS	-	10–70	15–40	15–35	10.3	0.35
Cu, mg/kg TS	45–125	100–1,000	28–47	28–50	325	5.02
Ni, mg/kg TS	8–28	7–100	8–25	6.3–8.3	13.8	<0.3
Hg, mg/kg TS	-	0.2–3.0	0.1–0.3	0.05–0.1	nd	nd
Zn, mg/kg TS	150–300	400–1,000	120–270	100–200	750	5.80

TS – total solids, ww – wet weight

An experimental study under laboratory conditions and in pilot reactors was performed at the Department of Environmental Engineering of TUT to elaborating better solutions for AD process and for choosing suitable substrates for co-digestion. The biomethane potential tests were conducted in anaerobic mesophilic conditions by measuring the maximum amount of biogas or biomethane produced per gram of VS contained in the organic matter used as a substrate in the AD process. These tests were conducted using either pure substrates or a mixture of two substrates in order to investigate the effect of combining different organic waste on the co-digestion process. BMP tests were carried out using the Automatic Methane Potential Test System II (AMPTS II – Figure 5.13).

Figure 5.13. Automatic Methane Potential Test System II

The AMPTS II follows the same measuring principles as practised in conventional CH_4 potential tests, which make the analysis results fully comparable with standard methods. For the technical and economical optimisation of biomethane producing plants batch methane potential (BMP) tests were used. The most promising substrates for biogas production according to BMP tests are catering waste, compost, beer yeast and their mixtures with sewage sludge (see Table 5.5). Excellent results were also achieved with the same substrates in one-stage co-digestion process tests.

On the basis of results of our studies and laboratory experiments, it can be considered that the average yield of biomethane produced from biodegradable waste deposited in landfills in Estonia was 451.5 m^3CH_4/t VS. Moving, in Estonia, from composting and landfilling biodegradable waste to anaerobic co-digestion would, according to our research results, enable to annually produce up to 23.1 million m^3 of biomethane, which could be converted into 226 thousand MWh of heat and electric energy (Paper III).

Although the production of biogas and its energy has been rather modest in Estonia, the deployment of this energy source has been quite apparent in the last twenty years in terms of LWW treatment, landfilling biodegradable waste and the treatment of dung. An environmental impact evaluation of biogas plants showed that the digestate of methane fermentation is environmentally friendlier and its nutrients more easily available for cultivated plants. It is predicted that biogas production and its use for energy production will increase and spread throughout Estonia. In Europe, the growth of green energy production is about 20 % per year. When energy is produced by incinerating biogas, only CO_2, which is 21 times less hazardous than CH_4, is emitted to the atmosphere (Paper III).

Research, conducted by the Estonian Environmental Research Centre and the TUT Department of Environmental Engineering on sewage sludge showed that the following (Elaboration, 2013):
- the average nitrogen content was 49.4 ± 17.7 g/kgTS, and TP content 10.6 ± 6.6 g/kgTS;
- limit values of heavy metals in sewage sludge were exceeded only at one WWTP;
- composting does not guarantee hygienic safety of sewage sludge. *Salmonella* spp were found in 38 % samples of treated sludge, and the number of *Escherichia coli* exceeded the limit values in 70.2 % of the samples. *Clostridium perfringensis* was found in only two samples.

The National Waste Management Plan for 2014–2020 foresees the utilisation of 10 % of domestic waste anaerobic fermentation digestate according to the data from 2011 (40,800 tonnes of digestate) (National, 2014). There is a need to try to increase the amount of anaerobic fermentation digestate that is used in agriculture.

The sludge digested and solidified at the Tallinn Waste Water Treatment Plant of AS Tallinna Vesi (dry matter concentration 31.7 %, pH 7.6) on 26 August 2010 had the following parameters: total nitrogen 8.9 kg/t; total phosphorus 6.7 kg/t; total potassium 0.80 kg/t. The sample was analysed in the Agrochemical Laboratory of Agricultural Tests.

The amount of produced digestate equates to the amount of digested biodegradable waste. The digestate from methane fermentation is rich in TP (0.4–1.8 kg P/m^3) and TN (3.5–4.5 kg N/m^3) that can be used for fertilising cultivated land. 1 m^3 of compost that is produced at the Väätsa landfill contains 3.5 kg TN, 0.41 kg TP and 0.54 kg TK. Successful long-term results were obtained when compost from methane fermented sludge of the Tallinn activated sludge WWTP

was used in agriculture, greenery and recultivation. Experiments in afforestation of abandoned less valuable arable land and cutover peatlands have given promisable results (Paper III).

In terms of water conservation, it is recommendable to increase fertilising areas with unprotected or poorly protected ground water with digestate instead of mineral fertilisers. For protecting waters and better plant nutrition it is recommendable to spread the digestate mainly in spring before ploughing (Environmental, 2004; Environmental, 2008; Strategic, 2009).

A study "Elaboration of methodology for experiments on deforestation and peatland renovation with the sludge from Tallinn domestic wastewater treatment by the Tallinn Water Utility" was conducted in 2002–2007 with the aim of elucidating the influence of the treatment of alvar soil and peat with different doses of sewage sludge on different seedlings under experimental conditions. The treatment of soils with sewage sludge considerably changed the physical properties of the soils and the concentration of nutrients in them: the pH of the soils rose and concentration of organic matter and nutrients increased.

Sewage sludge compost proved to be more efficient on bog soils, contributing to high productivity forest growth. This creates opportunities for cutover peatlands to be re-arranged and taken into use as woodland, which has so far been problematic. Sewage sludge compost also improves the properties of alvar soils for forest cultivation and creates a basis for areas to be afforested.

Systematic ground water monitoring was conducted prior to, during and after the tests. Although the upper aquifer in the monitored area (Liikva village) is unprotected from surface pollution, there were during and after the tests no signs of changes in the water quality of the monitoring wells that could be related to the use of sludge compost. At the same time, the spreading quantities of sludge recommended in the study should be followed when spreading the sludge, and ground water background studies should be conducted on each prospective land area beforehand. The study was conducted in accordance with Regulation No. 78 of the Minister for Environment from 30.12.2002 "Requirements for using sewage sludge in agriculture, landscaping and recultivation" (RTL 2003, 5, 48).

6. CONCLUSIONS AND RECOMMENDATIONS

One of the most important aspects when designing a landfill is reduction of landfill emissions, including contaminated precipitation water, wastewater and leachate (all together named as LWW), that harm the environment. In other words – planning and operating LWW treatment plants (WWTP) and collection and handling landfill gas.

The flow rate of LWW is high in spring and autumn rainfall and snowmelt periods. Our conducted measurements show that the landfill runoff varies significantly, depending on the intensity of precipitation. This was particularly apparent at the Jõelähtme and Väätsa landfills, where the difference was tenfold.

At Väätsa, the landfill wastewater flow rate depended on the intensity of rainfall and snowmelt as follows: Q_{min} = 0–2 m³/d, Q_{max} = 50–95 m³/d (7.1 to 13.6 m³/ha, in some cases 150 m³/d, i.e. 21.4 m³/ha). Leachate flow fluctuations were smaller: Q_{min} = 0–2 m³/d, Q_{max} = 20–30 m³/d (3.89–5.84 m³/ha). The study demonstrated that 60 % to 80 % of the rainwater falling on watertight areas and 10 % to 30 % of the rainwater falling onto the landfill waste lifts reaches sewers (or drainage network), depending on the duration of rainfall and on the size of watertight areas. The dynamics

and flow rate of leachate is significantly affected by winter conditions and drought periods in summer (Paper I).

Our research results indicate that:
- very high concentrations of pollutants (and hence the pollution load) were detected in leachate and rainwater from composting areas (e.g., at the Jõelähtme landfill BOD_7 3,960 mgO_2/l and COD 7,530 mgO/l, and at the Väätsa landfill BOD_7 1,875 mgO_2/l, COD 9,300 mgO/l);
- the amount and pollutant content of leachate is directly dependent on rainfall intensity. The pollutant concentration of LWW increased significally during rainfalls: SS up to 40 %, BOD_7 50 %, and COD 70 %. The pollution load of TN increased 20 % and TP up to 40 %;
- landfill leachates have a very high TN concentration and low concentration of TP;
- at all investigated landfills, the ratio BOD:N:P (recommendable for biological treatment 100:5:1) in LWW was out of balance. For example, in 2007 at Väätsa the ratio BOD_7:TN:TP for LWW was 46.1:13.5:1 and for leachate 115:64.8:1, and at Uikala LWW, the ratio was 267:183:1 and 115:177.2:1, respectively;
- values of the BOD_7:COD ratio in different samples were as follows: Väätsa 0.3–0.5, Jõelähtme 0.2–0.7, Uikala 0.2–0.6, and Paikre 0.2–0.5. At Väätsa, the BOD_7:COD ratio of the leachate was in 2010 less than 0.1, what was caused by the presence of humic and fulvic acids, tannins, lignin and hazardous organic chemicals, pesticides and herbicides, and which decreased its biodegradability;
- it is incorrect to use waste tyres in the landfill basements, as it causes a high iron concentration (6-8 mg/l) in the leachate, because iron corrodes in 5–6 years and blocks drainage systems;
- the heavy metal content of LWW and leachate does not exceed the limits;
- leachate is toxic and hinders biological treatment;
- the temperature of LWW was in October to April 1 °C to 4 °C, which significantly hindered the biological treatment processes. For example, the efficiency of reverse osmosis increases 3 % for each degree of temperature rise while the efficiency of biological nitrogen removal in AS plants decreases.

Before treating LWW, its flow rate, pollutant concentration and toxicity must be balanced and this is why equalising tanks (or ponds) and treatment plants with high capacity are necessary.

To decrease the pollution load and flow rate of the wastewater that requires treatment, composting of biodegradable waste at landfills and conducting rainwater from composting areas to equalising tanks should be terminated. Clean rainwater from composting areas and other parts of the landfill may be discharged to a recipient as well as used for irrigating waste deposits for intensifying landfill gas production or diluting the highly polluted leachate during drought periods, thereby making treatment of the latter more efficient.

When dimensioning equalising tanks, it should be taken into account that, according to previous studies, the rainwater flow from old landfills is up to 20 % and from new landfills up to 60 % of the amount of precipitation (Papers I and III).

In 2007–2014, different technologies for treating LWW were tested and the operation of the already existing treatment plants at Väätsa, Torma and Uikala observed. The study results show that the only wastewater from the young landfills provided the expected results after biological treatment. When the of non-biodegradable organic matter and ammonia content increases to a critical level, the AS system will simply close down (Paper III).

Biological treatment is acceptable for pretreating LWW, when the following technological means are applied:

- in activated sludge plants the hydraulic retention time for LWW should be prolonged two-fold in comparison to the time necessary for treating domestic wastewater;
- the content of dissolved oxygen in the aeration chamber should be brought up to at least 5–7 mg O_2/l;
- in winter conditions, it is recommended to maintain the positive temperature of leachate with technical measures. In the equalising tank, there should be a receiving well from where the leachate, without getting mixed with other wastewater, is pumped into the main treatment plant. Heat obtained from combustion of biogas can be used for warming up the leachate;
- it is essential to remove grease and oil prior to biological treatment;
- lack of incoming wastewater may become a problem for AS plants during longer drought periods or winters without snowmelt periods. In winter, the density of leachate may be over 1.0 t/m^3, due to the lack of wastewater and high content of substances, making the treatment with AS more difficult (bulking AS is carried out from the sedimentation tank). It is recommended to recycle the effluent into the equalising tank to avoid such problems and breakdown of the treatment process;
- additional treatment measures should be applied if biological methods are insufficient to meet the required degree of purification and allowed limits of pollutant (especially nitrogen) concentration in the effluent;
- biological co-treatment of leachate and domestic wastewater has certain technological and economic advantages. However, for avoiding the hindering impact of the leachate on the treatment process and safeguarding meeting the quality limits of treated wastewater, it is essential to ensure that the percentage of leachate would not exceed 10 %.

During our studies it was found that after equalisation of the flow rate and pollutant concentration of the LWW in an equalising tank, the most suitable treatment method is two-stage reverse osmosis (RO), wich may be followed by biological treatment. Other treatment methods, that were studied, included the aerobic biological oxidation process, ozonation process, coagulation process, post-ozonation of the coagulated LWW, post-ozonation of treatment plant effluent, post-ozonation of LWW that had been biologically treated in a AS plant, lime coagulation and post-ozonation of LWW, coagulation with oil-shale ash and the Fenton process, did not provide a degree of purification required by legislation.

In the first stage of RO treatment of LWW, about 95 % of nitrogen was removed, and in the second stage 99 % of the remainder. When choosing filters for RO it is recommended to keep to the following principles (Papers II and III):

- the RO-filter should be able to remove most of the COD and nitrogen content from LWW. In the case of two-stage RO, DT disk module membranes were used and it was possible to achieve the permitted limit values for COD, BOD, SS, TP and TN;
- before applying RO, the content of SS in water shoud be decreased. A sand filter enabling fast flow should be used ahead of DT filters.

In new landfills that are separated from the surrounding environment with geomembrane, the practice of directing leachate and/or RO concentrate as well as excess activated sludge from biological treatment to the waste deposit for irrigation, may be considered to be one of the steps of the wastewater treatment process. The aim is to accumulate the pollutants in the deposit and, in the case of excess sludge, to perform post-treatment of the latter in the deposit with the help of aerobic and anaerobic processes.

Landfill leachate and concentrate from RO treatment are toxic and can only be used for watering closed depositing fields, where the extraction of biogas is in the final stage. The low C:N ratio in the concentrate LLE refers to its low carbon (C) and excessive nitrogen content. During the fermentation process, at first carbon is used, and nitrogen becomes toxic for the methanogenic bacteria when the carbon content is insufficient. Therefore, LLE can only be fermented with carbon-rich co-substrate and in small volumes (Paper III).

Based on the TUT Environmental Engineering Institute previous research at Väätsa, a new LWW collection and treatment system was designed and put into service in 2011/2012. The system consists of an equalising tank, a physicochemical treatment (RO) unit following biological treatment (AS), and a stabilisation pond system. Since April 2012, when the new LWW treatment system began to function, the effluent from the WWTP was in compliance with the water permit requirements. The average treatment efficiency in the period between 2013 and 2015 was recorded at over 99 % of BOD_7, COD, TN and TP and over 90 % of SS (Paper III).

Composting biodegradable waste that is suitable for methane fermentation should be substituted with fermentation. It should be taken into account that methane fermentation followed by digestate composting is possible only for biodegradable waste that sorted before collection and pretreated by removing undigestible foreign matter. In the process of fermentation kitchen, food and animal waste are hygienised. In the near future, sorted biodegradable waste should be collected and the capacity of reusing them by methane fermentation in landfills and in other locations where biodegradable waste is accumulated should be developed (Paper III).

On the basis of the results of our studies and laboratory experiments, the estimated average yield of biomethane produced from biodegradable waste deposited in landfills in Estonia was 451.5 m^3CH_4/tVS. Substituting composting and depositing of biodegradable waste by anaerobic co-digestion according to the research would allow for the production of up to 23.07 million m^3 of biomethane annually, which could be converted into 226 thousand MWh of heat and electric energy (Paper III).

The amount of biogas collected now from waste deposits of Estonian landfills is too small for running combined heat and power plants for the production of heat and electricity. If plants for methane fermentation of biodegradable waste were constructed at landfills, the amount of landfill gas could be significantly increased and cogeneration of heat and electricity on the basis of landfill gas would become cost-effective (Paper III).

Methane fermentation is considered to be the most efficient way of biodegradable waste treatment, as biogas is generated. The digestate is rich in nutrients: TP (0.4–1.8 kg P/m^3) and TN (3.5–4.5 kg N/m^3), which can be used for fertilising cultivated land. 1 m^3 of compost that is produced at the Väätsa landfill contains 3.5 kg TN, 0.41 kg TP and 0.54 kg TK. The digestate from fermenting biodegradable waste can, after adjusting its composition to meet legal requirements, be used in agriculture, greenery and recultivation as well as at forest plantations. Long-term research demonstrates that compost from methane fermented sludge from the Tallinn AS WWTP may successfully be used in greenery and recultivation, and in afforestation of abandoned less valuable arable land and cutover peatlands (Paper III).

In the near future, LWW and leachate collection and treatment at landfills must be organised so that only landfill leachate, polluted rainwater from paved areas, domestic wastewater and wastewater from washing containers and other inventory are treated and only waste unsuitable for

recovery or energy production is landfilled. This would allow controlling environmentally hazardous emissions and can be achieved by:

- reducing or ending composting of biodegradable waste which would signaficantly decrease the flow rate and pollution load of LWW, and cut the necessary capacity of LWW collection and treatment facilities. If LWW is treated by RO, diluted RO concentrate is to be discharged into closed landfills or landfills that are in the state of closing. Wastewater treatment effluent that meets limit values is discharged to recipients;
- using unpolluted rainwater from paved areas for moistening landfill waste layers in order to increase the volume of landfill gas. It can also be collected in holding ponds and used for extinguishing landfill fires and for keeping biological WWTP-s in operation during drought periods;
- treating at landfills (waste management centres) separately collected solid waste for recovery (as material or producing new products) or preparing them for energy production;
- landfilling only unsuitable for recovery waste that has been sorted out of domestic waste or incinerated it to produce electricity and thermal energy. Biodegradable waste that has been obtained by sorting and separate collection is methane-fermented on site;
- replacing composting of biodegradable waste with fermentation on site. The collected from landfill waste layers and treated landfill gas is used for producing electricity and thermal energy in combined heat and power plants together with the biogas that is produced in biodegradable waste digesters.

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ABSTRACT

The continuously increasing requirements for environmental protection create problems in many countries, including Estonia, in treating LWW. In the present paper, the results of studies on the formation, chemical composition and treatment possibilities of LWW (including leachate) at five Estonian landfills are presented.

LWW flow is high in the periods of spring and autumn rainfall and winter snow melts. At the Väätsa landfill, the different flow rates were measured: Q_{min} = 0–2 m^3/d, Q_{med} = 10–20 m^3/d (1.4 to 2.9 m^3/ha), Q_{max} = 50–95 m^3/d (7.1 to 13.6 m^3/ha), in some cases even up to 150 m^3/d (21.4 m^3/ha).

At first (since 2002) the LWW was treated in an equalisation pond and somewhat later in an oxidation pond, a part of which was aerated, followed by an activated sludge unit. However, after six years of operation, the biodegradability index (BOD$_7$:COD ratio) dropped below 0.1 and the TN concentration rose to 400 mg/l in 2008. As a result, the AS system became totally inefficient.

In the years 2007 to 2013 at the TUT Environmental Engineering Institute, different technologies for LWW treatment were tested, and operation problems of several treatment plants at Estonian landfills was observed. It was found that after equalisation of the flow rate and pollutant concentration of the LWW in an equalising tank, the most suitable treatment method proved to be two-stage RO (with DT filters), which may be preceded by biological treatment. Other treatment methods did not provide the degree of purification that is required by legislation. The RO spiral membrane process was able to reduce the COD and BOD of biologically treated leachate by 97.9 % and 93.2 %, respectively, even after 328 and 586 hours of running the test unit, but only 39.0 % and 21.7 % reductions in TP and TN were achieved. Neither RO (spiral membranes process) nor NF was able to reduce TN to the required discharge limit of 75 mgN/l.

Based on the results of the research in the years 2012–2013, a new system for treating the Väätsa LWW was designed and constructed that consists of the LWW collection system, equalisation tank, biological treatment unit, RO with DT filters, as well pumping and distribution system for discharging concentrate from the RO to the landfill. In the fourth quarter of 2013, treatment efficiency by BOD$_7$ was 97.7 %, by SS 96.2 % and over 99 % for COD, N_{tot} and P_{tot}.

At Estonian landfills, in addition to sorting and land filling of wastes, biodegradable waste is composted. The pollutant content and flow of rain and snowmelt water from composting areas are highly variable. This has a significant influence on the possibilities and efficiency of the treatment methods. A possibility for solving of problems connected with LWW is to move from open composting of biodegradable waste to reactor composting or methane fermentation – an efficient means of treatment, in which useable biogas is produced. The digestate from methane fermentation is rich in nutrients: TP (0.4-1.8 kg P/m^3) and TN (3.5-4.5 kg N/m^3). Compost that is produced at the Väätsa landfill contains 3.5 kg TN, 0.41 kg TP and 0.54 kg TK per cubic metre. The digestate from fermenting biodegradable waste, after unifying its contents with legal requirements, can be used in agriculture, greenery and recultivation. Successful long-term results have been obtained when using compost obtained by methane fermentation of the Tallinn wastewater treatment plant sludge.

On the basis of the results of studies and laboratory experiments, the average yield of biomethane produced from biodegradable waste deposited in landfills in Estonia was calculated to be 451.5 m^{3}CH$_4$/tVS. The rearrangement of composting and landfilling biodegradable waste and

substituting it with anaerobic co-digestion would enable to produce up to 23.1 million m^3 of biomethane annually, which could be converted into 226 thousand MWh of heat and electric energy.

At landfills, waste management and the collection and treatment of LWW (including leachate) should be organised so that only leachate, polluted rainwater from paved areas, wastewater from washing containers and other inventory, and domestic wastewater is treated, and solely waste unsuitable for recovery or energy production is landfilled. Environmentally hazardous emissions should be controlled as much as possible.

KOKKUVÕTE

Üha karmistuvad keskkonnanõuded tekitavad paljudes riikides probleeme prügilareovee puhastamisel, nõnda ka Eestis.

Käesolev töö võtab kokku viie Eesti prügila reo- ja nõrgvee keemilise koostisele ning prügilareovee puhastamisvõimalustele pühendatud uuringute tulemused.

Prügilareovee vooluhulk on suur kevadiste ja sügiseste sajuperioodide ja lume sulamise ajal. Väätsa prügilas eri intensiivsusega sadude ja lumesulamise ajal mõõdetud prügilareovee vooluhulgad olid Q_{min} = 0–2 m^3/d, Q_{keskm} = 10–20 m^3/d (ööpäevas 1,4 –2,9 m^3/ha) ning Q_{max} = 50–95 m^3/d (7,1–13,6 m^3/ha), mõnikord isegi üle 150 m^3/d (21,4 m^3/ha). Algul (alates aastast 2002) puhastati prügilareovett Väätsal ühtlustustiigis ja veidi hiljem biotiigis, millest pool oli õhustatav, ning aktiivmudapuhastis. Kuue aasta jooksul kahanes reovee biolagundatavus BHT_7/KHT alla 0,1 ning $N_{üld}$ kontsentratsioon tõusis 2008. aastal üle 400 mgN/l, mistõttu aktiivmudapuhastuse tõhusus oluliselt vähenes.

Aastatel 2007–2013 uuriti Tallinna Tehnikaülikooli keskkonnatehnika instituudis mitmesuguseid prügilareovee puhastamise meetodeid ning vaadati üle Eesti prügilate reoveepuhastite käitamisel tekkinud probleemid. Selgus, et pärast prügilareovee vooluhulga ja reoainesisalduse ühtlustamist ühtlustusmahutis osutus sobivaimaks puhastusmeetodiks kaheastmeline DT-filtritega pöördosmoospuhastus kas eelneva bioloogilise puhastusega või ilma. Ülejäänud testitud puhastusmeetodid ei andnud õigusaktides nõutavaid tulemusi. Spiraalfiltritega pöördosmoospuhastus vähendas küll bioloogiliselt puhastatud prügilareovee BHT-d ja KHT-d vastavalt 97,9 ja 93,2 % isegi pärast katseseadme 328- ja 586-tunnist töötamist, kuid $P_{üld}$ ja $N_{üld}$ sisaldus vähenes vaid 39,0 ja 21,7 %. Spiraalfiltritega PO- ja NF-protsessi katseseade ei suutnud saavutada õigusaktides nõutavat $N_{üld}$ piirnormi 75 mg N/l.

Tuginedes uurimistöö tulemustele projekteeriti ja ehitati aastatel 2012 ja 2013 Väätsale uus prügilareovee puhastussüsteem, mis koosneb kogumismahutist, töötlemisest DT-filtritega pöördosmoosseadmes, millele eelnes reovee ühtlustamine ja bioloogiline puhastus, ning pöördosmoosis tekkiva kontsentraadi prügilademesse pumpamis- ja jaotussüsteemist. 2013. aasta neljandas kvartalis oli uue süsteemi tõhusus väga hea: BHT_7 97,7 %, heljumisisaldus 96,2 % ning KHT, $N_{üld}$ ja $P_{üld}$ üle 99 %.

Eesti prügilates toimub lisaks jäätmete sortimisele ja ladestamisele biolagunevate jäätmete kompostimine. Kompostimisväljakutelt voolava sademe- ja lumesulamisvee reostusaste ja vooluhulk muutuvad suurtes piirides. See mõjutab suuresti reovee puhastamisviisi valikut ja tõhusust. Üks võimalusi reoveega seotud probleemide lahendamiseks on oluliselt vähendada biolagunevate jäätmete kompostimist avaväljakutel ning üle minna reaktorkompostimisele või metaankääritamisele. Metaankääritamine on tõhus biolagunevate jäätmete käitlusviis, mis annab ka kasulikku biogaasi. Metaankääritamisel üle jääv digestaat sisaldab rohkesti peamisi taimetoitaineid fosforit (0,4–1,8 kg P/m^3) ja lämmastikku (3,5–4,5 kg N/m^3). Väätsa prügilas toodetud komposti kuupmeeter sisaldab 3,5 kg $N_{üld}$, 0,41 kg $P_{üld}$ ja 0,54 kg $K_{üld}$. Õigusaktide nõuetele vastavat komposti ja metaankääriti digestaati saab kasutada põllumajanduses, haljastamisel ja rekultiveerimisel. Selles suhtes on pikaajalisi positiivseid kogemusi on selles Tallinna linna reoveepuhastusjaamal.

Tuginedes sooritatud uuringute ja laboratoorsete katsete tulemustele, arvutati Eestis prügilatesse viidavate biolagunevate jäätmete keskmine biometaanisaagis – 451,5 m^3CH_4/tVS. Biolagunevate

jäätmete kompostimise ja prügilasse ladestamise ümberkorraldamine võimaldaks anaeroobse kääritamisega saada kuni 23,1 miljonit m^3 biometaani aastas, millest saaks toota umbes 226 tuhat MWh soojus- ja elektrienergiat.

Prügilates tuleb jäätmekäitlus ning prügilate reovee, sh nõrgvee, kogumine ja puhastamine korraldada nõnda, et puhastatakse ainult prügilademete nõrgvett, sillutatud aladelt voolavat reostunud sademevett, konteinerite ja muu inventari pesemisel tekkivat reovett ja prügila olmereovett ning ladestatakse üksnes taaskasutamiseks ja energia tootmiseks kõlbmatuid jäätmeid. Keskkonna saastamine tuleb viia miinimumini.

APPENDIX I ORIGINAL PUBLICATIONS

PAPER I

Kuusik, Aare, Pachel, K., Kuusik, Argo, Loigu, E. (2014). Landfill runoff water and landfill leachate discharge and treatment. In: 9th International Conference Environmental Engineering: Water Engineering: Selected Papers (1-6). Vilnius, Lithuania: VGTU Press "Technika".

Section: Water Engineering

Landfill runoff water and landfill leachate discharge and treatment

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Abstract

The continuously increasing requirements for environmental protection create problems for many countries in treating landfill runoff water. In the present paper, the creation, chemical composition and treatment possibilities for leachate in 5 Estonian landfills are studied. Generally, the younger the landfill, the higher concentrations of pollutants. In the Väätsa landfill, the landfill runoff water flow rate has been measured up to 150 m^3/d, average flow ($Q_{average}$) = 10–20 m^3/d. The composition of pollutants in the leachate depends on the character of the deposited waste material and on the age of the landfill. In Väätsa landfill, the leachate biochemical oxygen demand (BOD$_7$) is in the limits of 300–960 mgO$_2$/l, in Uikala landfill the leachate BOD$_7$ is in the limits of 231–1750 mgO$_2$/l, but in Tallinn landfill it can reach up to 5500 mgO$_2$/l, chemical oxygen demand (COD) is 580–2390 mgO/l and 9100 mgO/l correspondingly. Ammonia nitrogen in Väätsa is between 50–330 mgN/l, in Uikala it is between 427–729 mgN/l, but it can reach up to 970 mgN/l in Tallinn. Total nitrogen (N$_{tot}$) in Väätsa is 130–470 mgN/l and 1335 mgN/l correspondingly, and in Uikala 564–1567 mgN/l. The content of heavy metals is relatively low, caused by the high content of sulphates (500 – 700 mg/l), causing rapid sedimentation of heavy metals from the leachate. In the older landfills, the efficiency of nanofiltration (NF) and reverse osmosis (RO) were investigated, and the rather good results were obtained for BOD$_7$ and COD. After RO treatment, the results were as follows: COD of 57 mgO/l, BOD$_7$ of 35 mgO$_2$/l. RO was able to reduce COD and BOD$_7$ of biologically treated leachate by 97.9% and 93.2%. NF reduced 98 and 41% of leachate COD and BOD$_7$. Neither RO nor NF was able to reduce the total nitrogen to the required discharge limit of 15 mg/l.

Keywords: landfill, landfill runoff water, landfill leachate, leachate characteristics, leachate treatment.

Concepts: Landfill runoff water – liquid from a landfill or waste site containing dissolved and toxic materials; Landfill leachate – any liquid percolating through the deposited waste and emitted from or contained within a landfill.

1. Introduction

More than 500,000 tons of municipal solid waste (MSW) are being disposed annually at the five new landfills in Estonia – Väätsa, Uikala, Torma, Paikre and Tallinn. The effective management of landfill runoff water and leachate, as a by-product of landfills, is one of the major challenges in meeting strict EU discharge standards [1]. Väätsa landfill runoff water including leachate is treated by reverse osmosis (RO) after biological treatment and stabilisation pond, Uikala landfill runoff water is treated by RO after stabilisation pond, Torma landfill leachate is treated by activated sludge treatment (AS) after stabilisation pond and is directly discharged into the nearest small-scale rivers. Due to small flow rate and insufficient dilution, the discharged landfill runoff water may impose a significant impact on the water quality if the discharge standards were not met [2]. The quantity and pollutant content of landfill runoff water depends on the characteristics of the site, the climatic and meteorological conditions of the site and composting sites on the density of wastewater systems, and on the physical characteristics of the waste.

Estonia lies in the northern part of the temperate climate zone, in the transition zone between maritime and continental climate, which is heavily influenced by the North Atlantic Stream, Baltic Sea, and geographical location of the country [3].

The flow rate of landfill runoff water in Estonian landfills is directly related to the intensity of rainfall and melting of snow. The diurnal, weekly and annual variations of the flow rates of landfill runoff water and leachate have large variations.

The long-term average annual rainfall for all of Estonia is 750 mm. Rainfall variability by territory or temporally may be considerable. Figure 1 shows that the well-annual rainfall distribution has undergone major changes, particularly in the last analysed period [4].
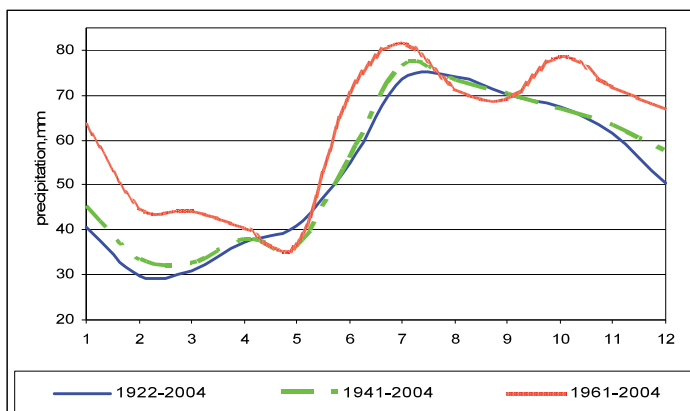

Fig. 1. Precipitation variability by Pärnu Meteorological station data [4]

2. Results and Discussion

Studies show that the quantity of some components of water from landfill (leachate, water from loading and collection sites, compost fields water, equipment and container's wash- and disinfection water) depending on the climatic and meteorological conditions of the site, but also on the activities in the landfill could be in the short term as well as long-term zero or negligible.

Municipal wastewater flow depends on the number of workers. However, landfill water and leachate flows are at high flow rates in the periods of spring and autumn rainfall and in the period when the snows melt. Some rainfall is absorbed in to the landfill sites, while more evaporates, with the intensity of the evaporation depending on temperature and rainfall. A mere 1-2 mm of rainfall evaporates completely in most cases. The impact of the leachate amount and duration of the discharge is directly dependent on the age of the dumps and the dump density. The flow is achieved approximately 3-5 hours after the start of the period of the rain or melt, depending on the obesity increase of the intensive dumps. High flow rate duration is also directly related to the duration of rainfall or the snow melt period. In case of a new deposition site and the compost field, before waste landfilling and the introduction of the material to be composted, approximately 90% of the stormwater is quickly collected from watertight underlain and discharged to sewer. Water is substantially pure and does not need to be treated. In accordance with the waste layer thickening, amount of landfill water begins to decline - some rainwater will retain a layer of waste and compost, and the share of evaporation increases. With waste layer obesity the leachate flow dynamics starts to stabilise. In new dump sites, storm water pass quickly through the waste and ends up in sewer taking along a large part of the easily washable pollutants. This evaporation is low. Research has shown that the quantity of leachate drained from old dumps is up to 20% of rainwater and from new dumps is on average 60% of rainwater. The older the landfill and the better compacted then the greater is the intensity of evaporation and the smaller is the amount of leachate. Stormwater residence time and amount of leached pollutants will increase. Landfill water as well leachate flow and dynamics are significantly affected by summer droughts and wintertime. A very large fluctuation is in the min, max as well as in average flow rates. For example, in Väätsa landfill different intensities of rain and snow melt period landfill water flow fluctuations were measured: Qmin = 0–2 m³/d; Qmedium = 10–20 m³/d (day 1.4 to 2.9 m³/ha), Qmax = 50–95 m³/d (7.1 to 13.6 m³/ha), in some cases even up to 150 m³/d (21.4 m³/ha). Leachate flow fluctuations are smaller Qmin = 0–2 m³/d; Qmedium = 5–15 m³/s (0.97 to 2.92 m³/ha), Qmax = 20–30 m³/d (3.89–5.84 m³/ha).

The refuse rate of degradation and the leachate content of pollutants are mostly dependent on the conditions within the landfill. An important factor is the moisture content of the waste. In dry environments, the decomposition of the waste and the methane gas released are substantially lower than in areas where the annual infiltration into waste is 50 to 100 cm [6]. It is widespread to send leachate or concentrate formed during the leachate treatment back to the landfill. At Väätsa and Uikala landfills, reverse osmosis concentrate is sent in the upper layers of landfill for this purpose. This increases the amount of leachate (for example, up to 10% in Väätsa and in Uikala up to 12%), and in that the concentration of pollutants. Through recirculation, the waste moisture content will usually increase from 15–20% to 40–50% [6]. In addition, this improves the spread of nutrients, substrate and bacteria with leachate recirculation.

At the stable methanogenic phase, the methane production rate will reach its maximum, and decrease thereafter, as a soluble substrate stock (carboxylic acids) decreases. In this phase, the methane production rate depends on the rate of hydrolysis of cellulose and hemicellulose. The pH continues to rise in the permanent status that comes when the concentration is only a few mg/l. In this phase, the BOD and COD ratio may fall below 0.1, because the carboxylic acids are

consumed as soon as they occur [4]. The BOD_7/COD ratio significantly fluctuated quite according to studies in 2007 in landfill leachates: Väätsa 0.3–0.5; Tallinn 0.2–0.7; Uikala 0.2–0.6; Paikre 0.2–0.5. In 2010, the landfill leachate BOD_7/COD ratio was less than 0.1 in Väätsa [5].

Landfill water and leachate pollutant content is directly dependent on the rainfall intensity and the on-going activities on the territory of landfill: garbage sorting, technology of tipping and use of depositing site, etc. Research has shown that the highest concentrations of pollutants are in leachate and in the storm water collected from composting site. For example, the Väätsa landfill composting site sewage BOD_7 was 201–1875 mgO_2/l and in Tallinn landfill it was 850–2825 mgO_2/l. The leachate content of pollutant is strongly influenced by the composition of the waste deposited in the landfill and the processes in the body of landfill. As a result of physical, chemical and microbiological processes, pollutants from waste are transported to leachate. Estonian landfills contain mixed construction waste, mixed municipal and industrial waste; there are no significant quantities of chemical waste and landfill leachate can be characterised by the four types of pollutants in an aqueous solution: dissolved organic matter, inorganic macro components, xenobiotic heavy metals and organic compounds.

Garbage will go through at least four phases of decay in landfills: 1) the initial aerobic phase, 2) the acid phase of the anaerobic, 3) the initial methanogenic phase, and 4) the stabilising methanogenic phase. It is quite common that different decay phases are occurring simultaneously in different parts of the landfill. Moreover, each phase will affect the different composition of the leachate, leachate treatment technology selection and the changes taking place over time, depending on the degradation phase [7].

Väätsa landfill is 14 years old, Uikala landfill is 12 years old, Torma landfil is 13 years old, Tallinn landfill is 11 years old and Paikuse landfill is 8 years old. All of the landfills first disposal areas are in transition state from anaerobic acid phase to initial methaogenic phase. The new disposal sites of the landfills are in the initial aerobic phase. During the study period pollutant content of wastewater of different sources and treated landfill water of Estonia landfills was monitored. The landfill and leachate samples were analysed by the Environmental Research Centre laboratory and the hydrochemistry laboratory of the Institute of Environmental Engineering at Tallinn University of Technology. Dioxins were studied in Czech Republic.

Table 1 shows that in Uikala and Väätsa landfills, concentrations of pollutants have large variations in leachate and. landfill runoff water.

Table 1. Measured landfill runoff water and leachate average concentrations of pollutants in Väätsa and Uikala landfills 2007 [5].

Parameter	Unit	Väätsa landfill (medium)		Uikala landfill (medium)	
		Runoff water	Leachate	Runoff water	Leachate
pH		6.75	7.7	7.9	7.5
conductivity	µS/cm	5545	8090	14010	14553
SS	mg/l	650	400	411	298
BOD_7	mgO_2/l	1015	529	1196	697
COD	mgO/l	4859	1366	3855	2538
TOC	mgC/l	1313	375	1073	718
NH_4	mgN/l	94	198	562	592
N_{tot}	mgN/l	297	298	820	1072
P_{tot}	mgP/l	22	4.6	4.48	6.05
HCO_3	mg/l	2800	3805	6470	7260
SO_4	mg/l	290	588	401	172
Cl	mg/l	107	439	1593	1553
monobasic phenols	mg/l	235	49.1	2590	1510
dibasic phenols	mg/l	22.6	140	1878	1005
petroleum product	mg/l	14	29	<20	60
Fe^{2+}	mg/l	7.4	1.95	1.2	0.62
Fe^{3+}	mg/l	5.3	7.95	14.7	21.4
Hg	µg/l	0.05	0.148	<0.05	<0.05
Ag	mg/l	0.01	<0.01	<0.01	<0.01
Cd	mg/l	<0.02	<0.02	<0.02	<0.02
Cr	mg/l	0.032	0.187	0.286	0.260
Mg	mg/l	105	108	293	283
Mn	mg/l	10	0.273	0.367	0.557
Na	mg/l	57.5	675	1400	1425
Ni	mg/l	0.041	0.090	0.061	0.080
Pb	mg/l	<0.04	<0.04	<0.04	<0.04
Zn	mg/l	<0.1	<0.1	<0.01	<0.01
Cu	mg/l	0.040	0.094	<0.02	0.023

Landfill runoff water including leachate from the Väätsa landfill is treated by RO after biological treatment and stabilisation pond and Uikala landfill runoff water including leachate is treated by RO after stabilisation pond. Wastewater treatment plant efficiency indicators in the third and fourth quarters of 2013 are shown in Table 2.

Table 2. Content of wastewater discharged to the treatment plant and treated water effluent pollutant content of the wastewater treatment plants of Väätsa and Uikala landfills in 2013 [8], [9].

Parameter	Unit	Väätsa landfill			Uikala landfill		
		Inflow	III quarter	IV quarter	Inflow*	III quarter	IV quarter
pH		8.52**			7.5	6.7	6.6
SS	mg/l	260**	4	10	215		2.7
BOD$_7$	mgO$_2$/l	130	<3	<3	235	3	4
COD	mgO/l	3500	<14	<14	1370		
TOC	mgC/l	1500					
NH$_4$	mgN/l				790	8.3	1.5
N$_{tot}$	mgN/l	474**	3.6	1.5	820	8.3	3.7
P$_{tot}$	mgP/l	9.0**	0.03	0.09	5.8	0.017	0.022
SO$_4$	mg/l				490		5
Cl	mg/l	1500					
monobasic phenols	µg/l	15	7.3	<2	150	0.625	
dibasic phenols	µg/l	<10		<10	2200	2.2	
petroleum product	mg/l	0.02	<0.02	<0.02	0.08		

* inflow 2012 average data
** inflow data 14. 05. 2009

After six years of the successful operation of Väätsa landfill wastewater treatment plant from 2002 to 2008, the activated sludge (AS) treatment process was not working according to its design conditions. As the landfill ages, ammonia concentration increased significantly from 50 to 330 mgN/l. As a result, ammonia-nitrogen concentration could not be reduced to the required 15 mg/l without additional treatment. The toxicity of ammonia to microorganisms in the activated sludge tank also made the sedimentation tank unoperable, as designed. Consequently, the effluent is dark and contains high concentration solids. Since AS was not primarily designed to remove non-biodegradable organic chemicals, the removal of non-seeing as BOD$_7$/COD ratio dropped less than 0.18 in 2011, rendering the biological processes ineffective. To meet the discharge standards, membrane technologies such as reverse osmosis can be used either as a main step in a landfill leachate treatment chain or as a single post-treatment step. Membranes such as reverse osmosis (RO) and nanofiltration (NF) have pore sizes that are sufficient in retaining non-biodegradable organic pollutants and are very effective in the physical separation of a variety of large non-biodegradable compounds from water [10]. Therefore, the objectives of this study are to assess the treatment efficiency of RO and nanofiltration of the biologically treated leachate, so that the effluent qualities meet the discharge standards. In addition, the treatment efficiency by both RO and nanofiltration are also compared to select the best membrane for full scale implementation. In the present study, RO and NF were investigated to determine the treatment efficiency of biologically treated leachate at the Väätsa landfill [11].

The ineffectiveness of AS is confirmed by the percentage reduction of pollutants before the treated leachate was discharged into the receiving river from 2001 to 2012. The AS system completely lost its treatment efficiency because both BOD$_7$ and COD reduction decreased from more than 80% in 2003 to about 60% after the AS system in 2011. In 2012, the BOD$_7$ and COD reduction data reflected the new treatment with the RO disc tube type module as recommended by this study (Table 2).

During the research, it was decided in parallel to Väätsa landfill RO device testing to monitor the effectiveness of the full-scale RO treatment process in Uikala landfill. Polluted water from Uikala landfill territory, including leachate is collected into the equalising pond. From the equalising pond 4.5 to 5.0 m^3 of wastewater per hour is discharged to the wastewater RO container treatment plant with filter DT 29-09. Efficiency of RO is considered 99.6%. About 2–3 m^3/ h of treated wastewater is discharged into the recipient and the rest of the concentrate is pumped back into the landfill. Before the process of entering, the landfill runoff water is filtered in sand filter following cartridge filter. A sand filter captures particles greater than 50 µm and the cartridge filter separates suspended solids, which are $\geq$ 10 µm. If the loss of pressure in the sand filter is between 2-2.5 bar the filter will be washed with water-air mixture. If cartridge is clogged, then it is not regenerated; it is changed. Reverse osmosis will take place in a designated DT filter module, which is located in the disc-shaped membranes. The process is operated with a plunger pump, which is capable of operating at a pressure from 0.5 to 65 bar, the normal operating pressure is 60 bar.

A treatment plant works in difficult conditions, which can be seen in Tables 1 and 2. Characteristics of the efficiency of the wastewater treatment plant are in Table 3. In September 2009, in the RO equipment modules the disk membranes were replaced. There has not been an emergency, however, technical failures of individual nodes (e.g., pumps) has been reported.

Table 3. Pollution content in treated landfill runoff water at Uikala landfill wastewater treatment plant [5], [9].

Time	BOD$_7$ mgO$_2$/l	COD mgO/l	SS mg/l	N$_{tot}$ mgN/l	P$_{tot}$ mgP/l
21/02/2005	6	73	6	0.87	0.11
16/11/2005	2.1	40	2	2.8	0.054
09/01/2006	1.9	30	16	2.1	0.047
05/04/2006	11	75	12	2.8	0.088
12/09/2006	11	160	26	1.7	0.056
13/11/2006	11	90	6.0	4.8	0.029
15/01/2007	5.0	60	13	1.9	0.068
04/04/2007	17	130	9.0	0.71	0.04
08/10/2007	73	375	4	79	0.187
22/10/2007	13	30.6	5	16	0.029
2008	16.75	146.25	16	32.25	0.17
15/01/2009	<3	45	<2	6.4	0.024
07/04/2009	15	110	2	20	0.027
12/08/2009	3.6	40	2.7	25	0.015
30/11/2009	<3	36	6.6	3.9	0.012
2010	7.5	55.25	2.225	15.425	0.079
2011	2.25	26.25	1.1	4.375	0.04475
2012	3.0	30.0	2.0	5.8	0.02
II quarter 2013	6.7			8.9	0.02
III quarter 2013	3			8.3	0.017
IV quarter 2013	4		2.7	3,7	0,022

The Väätsa landfill Ultra-FLO pilot plant of low pressure reverse osmosis (LRO) was purchased from the Ultra-FLO Pte Ltd in Singapore. The Ultra-FLO pilot plant was operated in either RO or NF modes to test the treatment efficiency of either RO or NF. Nanofiltration testing spiral filtration uses NE 4040-90 membrane. Biologically treated landfill leachate passed through a cloth filter and two micro filter cartridge BB 5 mm. Finally, it passed through either a RO membrane or nanofiltration spiral wound NE 4040-90 membrane. Samples were taken from the permeate and the concentrate for analysis of different chemical parameters after a specific time of filtration [11].

Compared to reverse osmosis membranes, nanofiltration membranes have many advantages, such as lower operational pressure, high flux, high rejection of polyvalent ions, relatively low investment, and operational and maintenance costs [10], [12]. It is interesting to note that nanofiltration gave better results in terms of conductivity, N$_{tot}$, P$_{tot}$, and SS, while nanofiltration and RO were equally effective in removing BOD$_7$ and COD, which have a removal efficient greater than 95% [11].

During the fatigue test, RO reduced the COD and BOD$_7$ of biologically treated leachate by 97.9% and 93.2%, respectively, after 328 hours. However, only 39.0% and 21.7% reductions of P$_{tot}$ and N$_{tot}$ were achieved. After 586 hours, COD and BOD$_7$ reduction of the biologically treated leachate reduced slightly to 95.2% and 91.7%, respectively. After 328 hours of the nanofiltration process, there was a reduction in the biologically treated leachate for COD, BOD$_7$, P$_{tot}$ and N$_{tot}$ 97.5%, 93.8%, 99.2% and 68.6% respectively. During the test, fouling problems occurred and the spiral membranes needed to be washed, but finally were fully replaced [11].

3. Conclusions

At Väätsa landfill different intensities of rain and snow melt period landfill water flow fluctuations were measured: Qmin = 0-2 m^3/d; Qmedium = 10–20 m^3/d (day 1.4 to 2.9 m^3/ha), Qmax = 50–95 m^3/d (7.1 to 13.6 m^3/ha), in some cases even up to 150 m^3/d (21.4 m^3/ha). Leachate flow fluctuations are smaller Qmin = 0–2 m^3/d; Qmedium = 5–15 m^3/s (0.97 to 2.92 m^3/ha), Qmax = 20–30 m^3/d (3.89–5.84 m^3/ha).

When non-biodegradable organic and ammonia increased to critical level, the AS system will simply stop working. Even worse, the AS biomass may have decayed and release N$_{tot}$ and COD into the treated leachate. To meet the discharge standards, reverse osmosis and nanofiltration were investigated in treating the biologically treated leachate. The reduction efficiencies of COD, BOD, dissolved inorganic as expressed as conductivity and suspended solids were all excellent after ineffective biological processes such as AS and biological lagoon. For example, the reverse osmosis process reduced COD or BOD$_7$ by 97% and 60%, respectively. Nanofiltration reduced 98%, 41%, and 68 % of biologically treated leachate COD, BOD$_7$, and N$_{tot}$, respectively. However, reverse osmosis was ineffective in removing N$_{tot}$. Nanofiltration has better results in removing P$_{tot}$ and N$_{tot}$ than RO. Although the COD, BOD$_7$, suspended solids, and P$_{tot}$ can meet current legislation requirements, neither NF nor RO could bring the N$_{tot}$ below the discharge limit of 15 mg/l. However, successful application of membrane filtration technologies require the efficient control of membrane fouling, especially when spiral membranes are used [11].

Based on the Tallinn University of Technology Environmental Engineering Institute previous research on wastewater formation and pollutant content evolution in Väätsa and Uikala landfills the Väätsa landfill wastewater collection and treatment system is designed and built in the years 2012/2013. The whole system consist from the landfill runoff water collection system, RO after biological treatment and stabilization in pond, as well pumping and distribution system for discharging concentrate from the RO back to the landfill. In the third and fourth quarters of 2013, the figures for the effluent were respectively: BOD_7 <3 mgO₂/l and <3 mgO₂/l, COD <14 mgO/l and <14 mgO/l, SS 4 mg/l and 10 mg/l, N_{tot} 3.6 mgN/l and 1.5 mgN/l, P_{tot} 0.03 mgP/l and 0.09 mgP/l, monobasic phenols 7.3 mg/l and <2 mg/l, dibasic phenols <10 mg/l (see Table 2). Treatment efficiency was in the fourth quarter by BOD_7 was97.7% and SS 96.2% and by COD, N_{tot} and P_{tot} treatment efficiency was more than 99%.

Uikala landfill sewage treatment plant works under difficult conditions, which is apparent in Tables 1 and 2. Treatment plant operational efficiencies are visible in Table 3. In September 2009, they replaced the RO equipment disk module membranes. There has not been an emergency. There have been technical failures of individual nodes (e.g., pumps). In 2011, the treatment efficiency was by BOD_7 was 98.5%, COD 97.1%, P_{tot} 98.8% and by SS and N_{tot} more than 99%. In 2013, in the fourth quarter, efficiency by BOD_7 was 98.3%, SS 98.7%, N_{tot} as well P_{tot} more than 99%.

The research showed:

- Very high concentrations of pollutants (and hence the pollution load) is collected from leachate and rainwater collected at the compost site;
- Leachate quantity and concentration of pollutants is directly dependent on rainfall intensity and dump obesity;
- Landfill leachates have a very high N_{tot} concentration and low concentration of P_{tot};
- Leachate contains high amounts of CODs (Uikala 1600 mg/l) and BOD_7;
- It is incorrect to use old car tyres dumps in the basecoat: iron concentration in leachate 6–8 mg/l, of iron corrodes 5–6 years and drainage blockages;
- Leachate contains very large amounts of phenols – in Uikala landfill leachate 3000–4000 mg/l;
- Leachate contain high amounts of magnesium and sodium. Uikkala landfill leachate at 300 and 1600 mg/l;
- The heavy metal content in landfill water does not exceed the limits.

Acknowledgments

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Reverse osmosis and nanofiltration of biologically treated leachate

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Experiments of nano-filtration (NF) and reverse osmosis (RO) were conducted to remove most pollutants from the biological treated leachate. For example, the purified permeate after reverse osmosis treatment with spiral membranes reached effluent water quality as follows: COD of 57 mg O_2/l, BOD_7 of 35 mg O_2/l, and suspended solid of 1 mg/l which satisfies the discharge standards in Estonia. For both RO and NF, conductivity can be reduced by 91% from 6.06 to 0.371 mS/cm by RO and 99% from 200 to 1 mS/cm by NF. To test the service life of the RO spiral membranes, the process was able to reduce chemical oxygen demand (COD) and biological oxygen demand (BOD) of biologically treated leachate by 97.9% and 93.2% even after 328 and 586 hours, respectively. However, only 39.0% and 21.7% reductions of P_{tot} and N_{tot} were achieved. As a result, neither RO (spiral membranes process) nor NF was able to reduce the total nitrogen (TN) to the required discharge limit of 15 mg/l.

Keywords: landfill leachate; leachate characteristics; pre-treatment; biological treatment; reverse osmosis; nanofiltration

1. Introduction

More than 500 thousand tons of municipal solid waste are being disposed annually at five landfills in Estonia. Effective management of landfill leachate is one of the major challenges to meet strict European Union discharge standards at these landfills.[1] For example, leachate treated by activated sludge (AS) from the Väätsa landfill is directly discharged into a small river. Due to the small flow rate, the discharged leachate could impose significant impact on the water quality due to insufficient dilution if the discharge standards were not met.[2] The quantity of leachate depends not only on the characteristics of climatic and meteorological conditions of the site, but also on the physical characteristics of waste. The flow rate of landfill leachate in Väätsa site is directly related to the intensity of rainfall and melting of snow. Diurnal, weekly and annual flow rates of landfill leachate have large variations. For example, the leachate flow rate in Väätsa landfill ranged from 2 to 130 m³/day in the past. Annual mean flow rate was in the range from 10 to 20 m³/day. Calculated specific leachate runoff was 8 l/s/ha. The leachate flow rate from Väätsa landfill was 5500 m³/yr in the rainy year of 2008 and only 2000 m³/yr in the draught year of 2006. Since the leachate flow rate changes significantly, mean leachate quality, as given in Table 1, shows a low BOD_7/COD ratio of 0.38 and high ammonia concentration of 198 mg/l.

After six years of successful operation from 2002 to 2008, AS treatment process was not working at its design conditions. As the landfill ages, more and more non-biodegradable organic compounds, such as humic and fulvic acids, are produced.[3] In addition, ammonia concentration increased significantly from 50 to 164 mg/l. As a result, ammonia–nitrogen concentration could not be reduced to the discharge standard of 15 mg/l without additional treatment. Ammonia toxicity to microorganisms in the AS tank also made the sedimentation tank not working as it was designed. Consequently, the effluent was dark and contained high concentration solids. Since AS was not primarily designed to remove non-biodegradable organic chemicals, removal of non-biodegradable after AS is critical to meet the discharge standards especially after the BOD_7/COD ratio dropped to less than 0.18 in 2011, which prompted this study by using reverse osmosis (RO) and nanofiltration (NF) to improve the discharge quality. In the literature, Mohammad et al. [4] investigated NF of leachate. Turan et al. [5] compared RO and NF performance in treating diary wastewater.

To meet the Estonia discharge standards, membrane technologies, such as RO, can be used either as a main step in a landfill leachate treatment chain or as single post-treatment step. Membranes such as RO and NF have pore sizes that are sufficient to retain non-biodegradable organic pollutants and are very effective in the physical separation of a variety of large non-biodegradable compounds from water.[6] Mohammad et al. [4] reported that NF removed more than 85% total suspended solids, heavy metals,

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Table 1. Leachate water quality.

Parameter	Unit	Concentration
BOD$_7$	mg O$_2$/l	529
COD	mg O/l	1366
TN	mg N/l	298
NH$_4$	mg N/l	198
TP	mg P/l	4.6
Conductivity	μS/cm	8090
SO$_4$	mg/l	588
Cl$^-$	mg/l	439

conductivity and Chemical Oxygen Demand (COD). However, only 45.4% and 20.5% removal was achieved for nitrate and ammonia–nitrogen, respectively. Therefore, the objectives of this study are to assess the treatment efficiencies of RO and NF of the biologically treated leachate so that the effluent qualities can meet the Estonia discharge standards. In addition, after treatment efficiencies by both RO and NF were compared, the best membrane for full-scale implementation was selected. This study reports the treatment efficiencies of the biologically treated leachate at the Väätsa landfill by pilot-scale RO and NF.

2. Experimental design

2.1. *Biological leachate treatment systems*

The biological treatment systems as shown in Figure 1 include a biological lagoon and an AS which were built in 2002. Väätsa biological treatment process of leachate has two components. One is biological lagoon which has been further divided into two processes, aerobic vs. anoxic and the other is AS. All the data were taken after either biological lagoon or AS treatment of the effluent from biological lagoon.
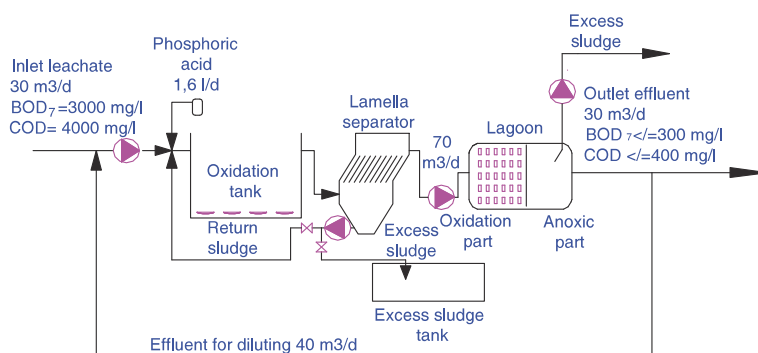
2.2. *Experimental set-up of RO and NF*

The flow diagram for the experimental set-up of RO and NF is shown in Figure 2. The Ultra-FLO pilot plant of low-pressure reverse osmosis (LRO) was purchased from the Ultra-FLO Pte Ltd. in Singapore. The system consists of two LRO membrane (4040 − spiral membranes) cartridges and one BT420 UF cartridge, one 5 μm guard-filter, one feed pump, as well as control valves, pressure gauges and flow meters. The system was connected with either Polyvinyl Chloride or stainless steel pipes. According to the manufacturer's recommendation, the flow rates for RO were kept at the following ranges: permeate at 0.2 m^3/h, concentrate at 0.3 m^3/h, while the feedback flow rate was kept from 0.9 to 1.2 m^3/h. When NF-filter was tested, the flow rate through the NF filter is about 0.3 m^3/h, while the feedback flow rate was operated from 0.9 to 1.2 m^3/h.

Figure 3 shows the photos of experimental set-up from both the front and side views. The front two cartridges are pre-treated to remove suspended particles. The RO or NF membranes can be seen from the side view of the two stainless steel columns.

2.3. *Experimental procedure*

The Ultra-FLO pilot plant was operated in either RO or NF mode to test the treatment efficiency of either RO or NF. NF testing spiral filtration uses NE 4040-90 membrane. The permeate expense ratio (P/F) was kept at 1.7. Biologically treated landfill leachate passed through a cloth filter and two microfilter cartridges BB 5 mm. Finally, it passed through either RO membrane or NF spiral wound NE 4040-90 membrane.

During the exhaustion test, two successive 5 mm microfilter BB Cartridge Filters, or two new filters of spiral wound PP 5 μm, and two consecutive PO spiral wound NF 2-4040 or NF spiral wound NE 4040-90 were used to test the lifetime of these membranes in treating the biologically

Figure 1. Väätsa landfill biological treatment process.

Figure 2. Flow diagram for overall experimental set-up.

Figure 3. Experimental set-up of RO and NF (views from front and back).

treated leachate. Samples were taken from both the permeate and concentrate for analysis of different chemical parameters after a specific time of filtration, respectively. All the chemical analysis was conducted according to the United States Environmental Protection Agency (USEPA) standard methods.

3. Results and discussion

3.1. *Biological treatment*

Figure 4 plots the cumulative COD concentration of inlet and outlet from AS during the heavy rain in November 2008 and melting period in April 2009. First, the raining season COD was much less than the melting season COD. Second, during the heavy rainy season in November 2008, there was certain percentage reduction in COD. However,

there was no COD reduction in April 2009. For example, the COD concentration of both inlet and outlet from the AS was the same at about 1000 mg/l. Therefore, the biological systems became totally ineffective in 2009, as the effluent COD reached the same level as the influent COD.

The ineffectiveness of AS was confirmed by the percentage reduction of pollutants before the treated leachate was discharged into the receiving river from 2001 to 2012. Figure 5 reflects that AS system almost completely lost its treatment efficiency because both BOD_7 and COD reduction decreased from more than 80% in 2003 to about 60% after the AS system in 2011, respectively. In 2012, the BOD_7 and COD reduction data reflected the new treatment with RO disc tube-type module as recommended by this study.

Figure 6 shows the treatment results by using biological lagoon, which was effective under significant variations
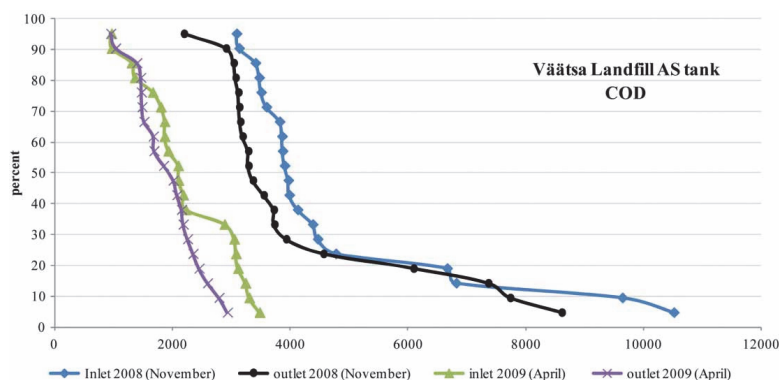
Figure 4.   COD inlet and outlet during heavy rain (November 2008) and melting period (April 2009).
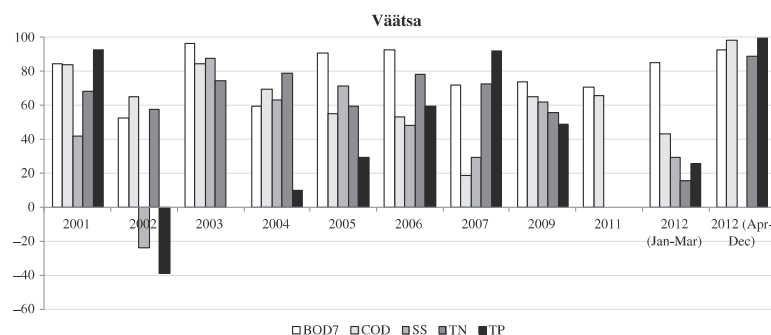

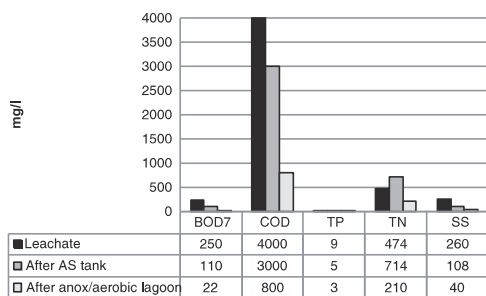Figure 5.   Leachate treatment efficiency at Väätsa landfill from 2001 to 2012.

Figure 6.   Treatment results at the Väätsa landfill leachate treatment plant.

of concentration and strength by serving as an equalization tank. Organic compounds, nitrogen, phosphorous, suspended solids and pathogenic micro-organisms all can be removed at low operational and maintenance costs.[7,8] However, biological lagoon alone could not meet the discharge standards at the Väätsa landfill site. When the reduction efficiency by AS was compared with that by biological lagoon in Figure 4, the latter was more efficient than

the former because it removed about 91.2% and 80% of BOD_7 and COD, respectively, while AS only removed 56% BOD_7 and 25% COD. Total nitrogen (TN) increased by 73.5 after AS compared with the inlet TN, which may be contributed by the addition of biomass from AS. Naturally, the removal efficiency will be impacted by the biodegradability of leachate. For example, Christensen and Nielsen [9] reported that aerated-lagoons-removed COD increased from 35% to 95% as the BOD_7/COD ratio increased from 0.05 to 0.40, respectively.

As landfill ages, more and more non-biodegradable organic compounds were produced with high NH_4 content.[10] Figure 7 shows a typical 90% confidence interval of BOD_7 varying from 40 mg/l to a maximal of 250 mg/l. At the same time, the mean COD was 1350 mg/l with 95% confidence interval ranging from 1050 to 1550 mg/l. As a result, the BOD_7/COD has a mean value of 0.08 with 95% confidence interval changing from 0.021 to 0.18. Since this ratio was significantly less than 0.5, poor performance of the AS system was expected and observed. In addition, ammonia also accumulated to extremely toxic level as shown, as total nitrogen as high as the mean of 450 mg/l with 95% varies from 220 to
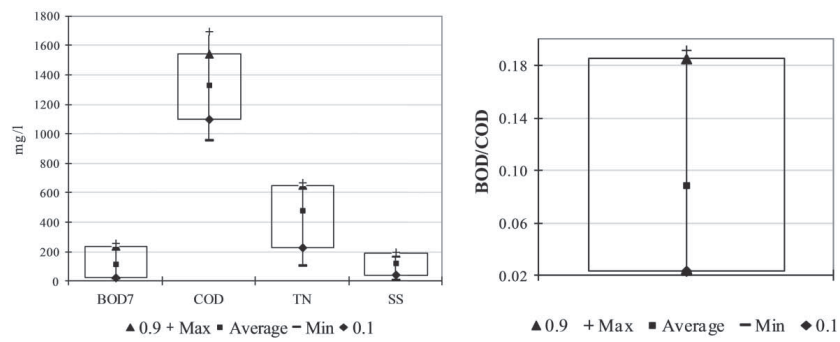
Figure 7. Box plots of pollutant concentrations and BOD_7/COD ratio in leachate before the AS biological treatment in 2009.
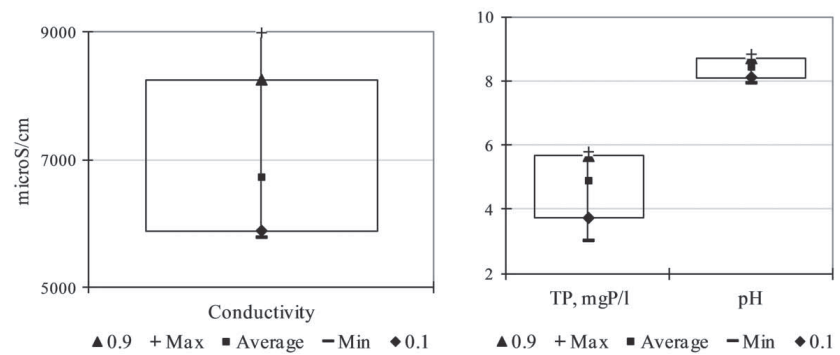

Figure 8. Box plots of conductivity, total phosphorus (TP) and pH in leachate before the AS biological treatment in 2009.
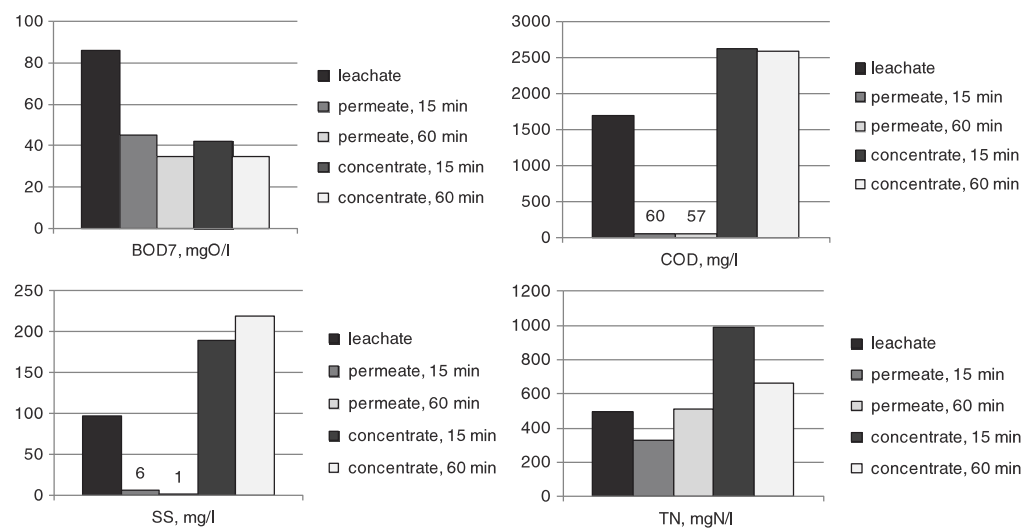

Figure 9. Treatment efficiency of RO.

630 mg/l. The toxicity of ammonia completely upset the AS system and rendered the biological treatment system ineffective (Figure 8).

3.2. *Reverse osmosis*

Since biological treatment could not meet the local discharge standards, RO and NF were used to treat the leachate after biological lagoon. Many research reported that RO separate pollutants from landfill leachate well at both laboratory and industrial scales.[11,12] Rejection coefficients of COD and heavy metal could be higher than 98% and 99%, respectively.

Our results contradict to the results reported by Chan and Dudeney.[13] When RO was directly applied to treat the stabilized leachate of high COD (8000 mg/l) and NH_3-N (2620 mg/l), lower than the local effluent limits for COD of 200 mg/l and NH_3-N of 5 mg/l were obtained.[13] In this study, TN reductions were no more than 33% after 15 min filtration, although the RO reduced conductivity of biologically treated leachate from 6.06 to 0.375 mS/cm. This

may be very likely due to different RO systems used in two different studies because the RO used in this study was a low-pressure RO system with operating pressure no more than 6 bar.

Figure 9 shows that treatment efficiency of BOD_7 was significantly less than that of COD. For example, the average removal efficiency of BOD_7 was only 60% while COD removal could achieve as high as 98% from 1700 to 60 mg/l for either 15 or 60 min filtration. This result suggests that soluble organic compounds may pass the membrane and entered into the permeate. For suspended solid (SS), excellent percent removal has been achieved. After 60 min of RO treatment, purified permeate reached effluent water quality as follows: COD of 57 mg O_2/l, BOD_7 of 35 mg O_2/l and suspended solid of 1 mg/l, which meet all the discharge standards. However, total N remained as high as 512 mg N/l. The permeate expense ratio between the permeate and concentrate (P/F) decreased from 0.5 to 0.43 during the operation of the unit. The filters were clogging the purified water indicators, which deteriorated during the experimental runs due to either organic compounds
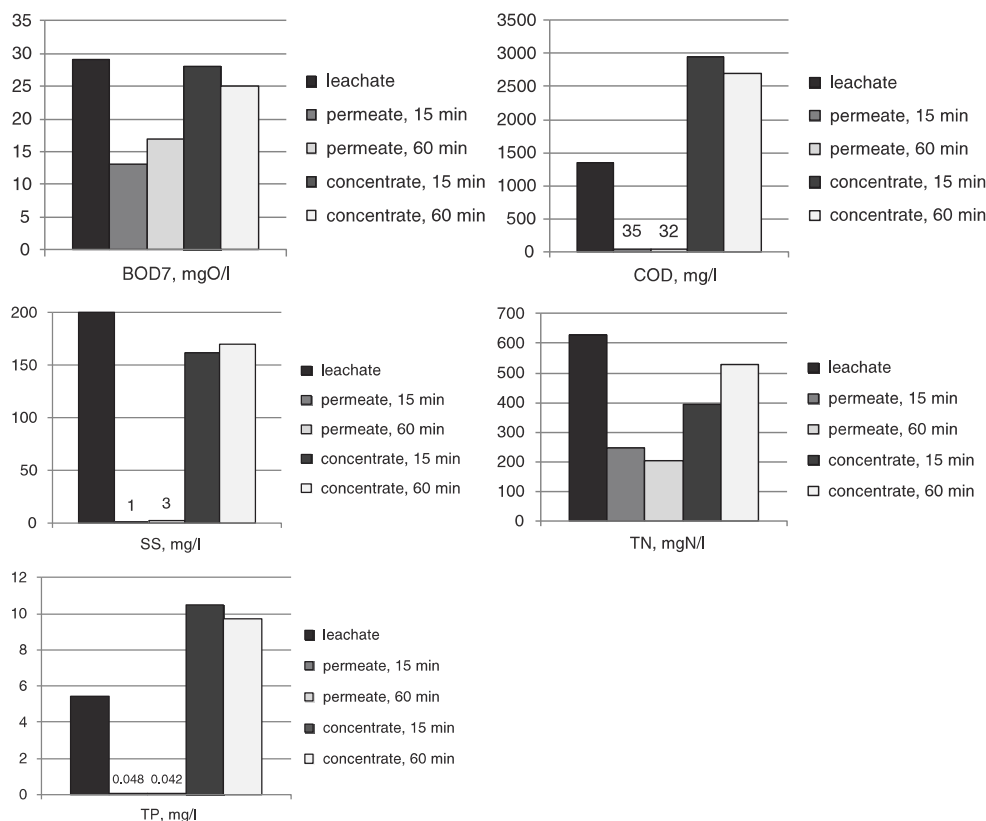

Figure 10.   Treatment efficiency of NF.

adsorbed on the membrane surface or clogging of the filter module by the SS. Since RO membrane was a spiral filter, it could be easily clogged by the suspended solids. In addition, a significant portion of organic matter in leachate is humic substance which is refractory organic chemical. Fulvic and humic compounds increases with landfill age. Therefore, it is not surprising that Biological Oxygen Demand (BOD) of permeate did not change with the filtration time.

3.3. Nanofiltration

After biological lagoon, leachate still contained a significant amount of inorganic salts, as reflected in its conductivity of 6.06 mS/cm. In NF, the substances in the feed are separated according to two principles. Neutral species are separated according to their sizes, e.g. molecules larger

than 200–300 g/mol, and the ions are separated by electrostatic interaction between ions and the membrane.[14] Due to their unique properties among the other membranes, RO and NF have many advantages of removing recalcitrant organic compounds and heavy metals from the leachate.[15]

Figure 10 shows that NF reduced 98%, 41% and 68% of leachate COD, BOD and total N, respectively. After 1 h of NF, the effluent reached COD of 32 mg O_2/l, BOD_5 of 17 mg O_2/l, N_{tot} of 202 mg N/l, and suspended solid of 3 mg/l.

All the experimental results after RO and NF are listed in Table 2 as follows:

Initial test results showed that RO and NF achieved good efficiency in reducing COD, BOD and suspended solids. However, they were ineffective in reducing total nitrogen.

Table 2. Flow rate and water qualities of raw, permeate, and concentrate.

Time (min)	Parameter	Unit	Pressure (atm) P1	P2	P3	Leachate	Permeate	Concentrate	Feed back flow rate
Spiral RO									
0	Pressure		2.7	2.7	5				
	Flow rate	m³/h					0.3	0.3	1.65
15	Pressure		2.9	2.9	5.3				
	Flow rate	m³/h					0.25	0.3	1.65
	pH					8.85	9.7	9	
	Conductivity	mS/cm				6.06	0.375	10.2	
	BOD_7	mg O_2/l				86	45	42	
	COD	mg O_2/l				1696	60	2626	
	N_{tot}	mg N/l				495	328	988	
	SS	mg/l				97	6.0	190	
60			3.8	3.8	6.1				
	Flow rate	m³/h					0.23	0.3	1.65
	pH					8.85	8.9	8.8	
	Conductivity	mS/cm				6.06	0.371	10.07	
	BOD_7	mg O_2/l				86	35	35	
	COD	mg O_2/l				1696	57	2588	
	N_{tot}	mg N/l				495	512	665	
	SS	mg/l				97	1.0	220	
NF									
0	Pressure		3.6	3.6	5				
	Flow rate	m³/h					0.5	0.3	1.4
15	Pressure		3.4	3.4	6				
	pH					8.6	8.75	8.45	
	Conductivity	mS/cm				6.05	3.83	8.68	
	BOD_5	mg O_2/l				29	13	28	
	COD	mg O_2/l				1350	35	2940	
	N_{tot}	mg N/l				628	247	395	
	SS	mg P/l				5.41	0.048	10.5	
	Conductivity	mg/l				200	1.0	162	
60	Pressure		3.4	3.4	6				
	Flow rate	m³/h					0.5	0.3	1.4
	pH					8.6	8.7	8.45	
	Conductivity	mS/cm				6.05	4.01	7.95	
	BOD_7	mg O_2/l				29	17	25	
	COD	mg O_2/l				1350	32	2690	
	N_{tot}	mg N/l				628	202	529	
	SS	mg P/l				5.41	0.042	9.75	
	Conductivity	mg/l				200	3.0	170	

Table 3. Biologically treated leachate quality.

	COD (mg/l)	BOD$_7$ (mg O/l)	TN (mg N/l)	TP (mg P/l)	SS (mg/l)
Leachate	2560	190	610	8.1	149
After AS tank	2900	160	642	7.6	100
After bio-lagoon	960	25	111	12	3.05
After experimental equipment	150	19	39.9	3	0.43

Table 4. Water quality after treatment by either RO or NF membrane.

	Date	pH	Conductivity (mS/cm)	BOD$_7$ (mg O$_2$/l)	COD (mg O$_2$/l)	NH$_4$ (mg N/l)	TN (mg N/l)	TP (mg P/l)	SS (mg/l)
Spiral wound NF 2-4040 RO									
Inlet	07/28/09	8.5	6.06	222	1240.0		350	4.8	80
Outlet	07/28/09	8.1	4.71	15	26		274	0.293	40
Concentrate	07/28/09	8.5	5.91	189	1740		820	6.24	87
Efficiency (%)			22.3	93.2	97.9		21.7	39	50
Spiral NE 4040-90 NF									
Inlet	08/28/09	8.5	6	260	1355	187	637	5.48	188
Outlet	08/28/09	8.2	4.05	16	34		200	0.044	3
Concentrate	08/28/09	8.5	7.95	250	2700		532	9.81	160
Efficiency (%)			32.3	93.8	97.5		68.6	99.2	98.4
Spiral NF 2-4040 RO									
AS container outlet	09/23/09	8.3	10.47	90	1390		671	5.81	170
Inlet	09/23/09	8.1	6.07	251	1540		338	6.39	117
Outlet	09/23/09	7.9	4.86	12	39		205	0.53	4
Concentrate	09/23/09	7.85	5.86	241	2790		368	11.3	115
Efficiency (%)			19.1	95.2	97.5		39.3	91.7	96.6

The initial results lead to the following long-term test by using different types of RO or NF membranes.

3.4. Comparison of RO and NF

Table 3 lists the biologically treated leachate quality after either AS or biological lagoon, which were taken on 6 August 2009.

Table 4 presents the water quality after treatment by either RO or NF membranes.

During the experimental period from July to September 2009, the water quality of the biologically treated leachate changed significantly. To reflect these changes, box plots are presented in Figure 11. The box plots reflect the maximal, upper 90%, mean, and lower 10%, and minimal values of each corresponding water quality parameters. Again, the major point here is that the COD increased after the AS treatment and had a mean value of 2200 mg/l and a minimal of 1410 and a maximal of 2900 mg/l due to added COD by the AS biomass.

Compared to RO membranes, NF membranes have many advantages such as low operational pressure, high flux, high rejection of polyvalent ions, and relatively low investment, operational and maintenance costs.[6,16]
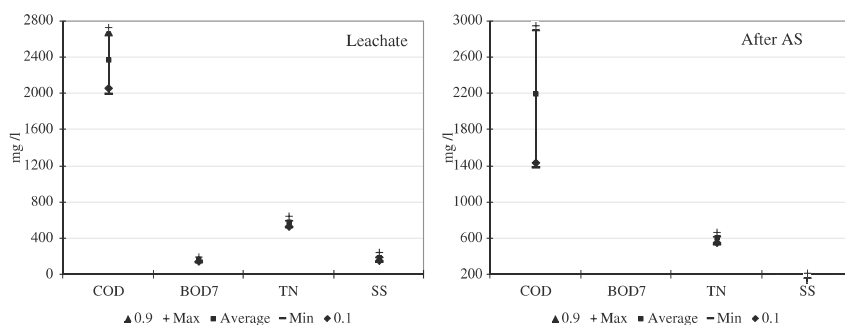

Figure 11. Box plots of COD, BOD$_7$, TN and SS in the leachate (left) and after AS tank (right) from Väätsa landfill during the period of experiments (from July to October 2009).
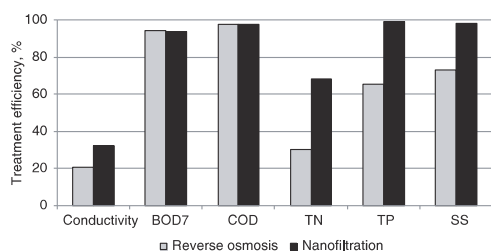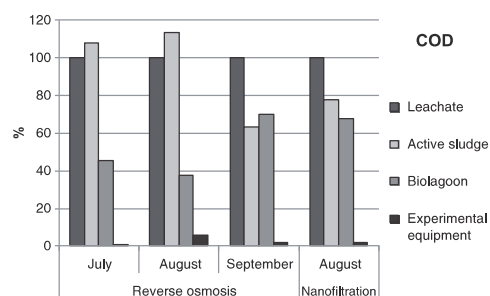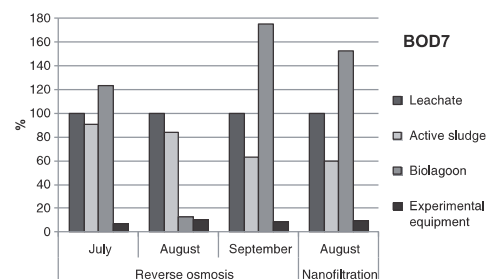
Figure 12. RO and NF efficiencies.

Figure 13. Treatment efficiency (leachate = 100%).

Figure 12 quantifies these advantages by comparing the treatment efficiency of biologically treated leachate with those from OR. It is interesting to note that NF gave better results in terms of reduction of conductivity, TN, TP and SS while RO and NF were equally effective in removing BOD_7 and COD, which have removal efficiencies of greater than 95%.

Figure 13 clearly shows that biological treatments, such as AS and biological lagoon, only achieve marginal percentage removal. For AS, COD effluent was even higher than the original leachate. One explanation is that the biomass from some AS contributed to the COD of the treated effluent due to the toxicity of ammonia. For biological lagoon, more

Figure 14. Treatment efficiency of BOD_7 (leachate = 100%).

than 40% COD removal can be achieved. For all the non-biodegradable COD, RO or NF will result in more than 96% COD removal, which can meet the regulator's standards.

Biological lagoon effectively serves as a good hydrolysis basin, as shown in Figure 14. Three out of four samples, BOD_7 increased from 20% to 75%. In other words, biodegradability of the leachate increased significantly after biological lagoon. The increase in BOD_7 may also be the reason that the removal efficiencies of RO and NF decreased from 96% to 90%, because soluble organic compounds were dominant species, which could pass through the membrane pores. Both RO and NF were very effective in removing SS, as shown in Figure 15. More than 93% SS removal was
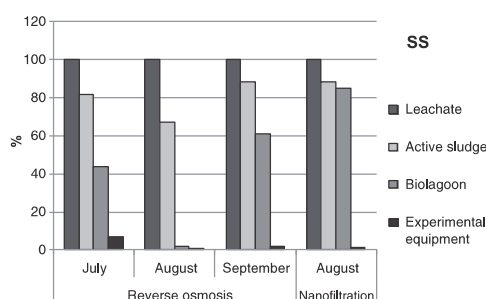

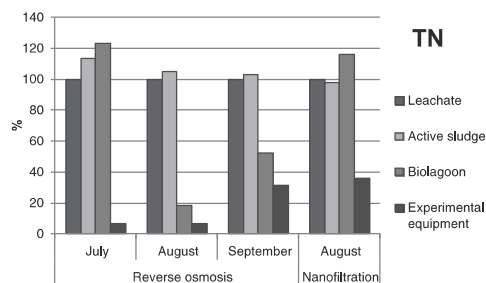Figure 15. Treatment efficiency of SS (leachate = 100%).

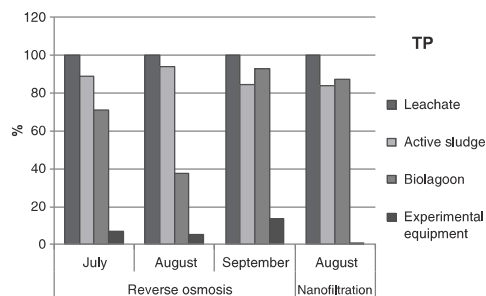Figure 16. Treatment efficiency of TN (leachate = 100%).

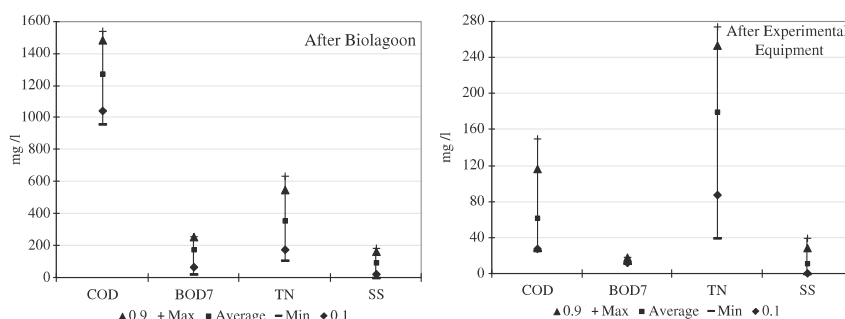Figure 17. Treatment efficiency of TP (leachate = 100%).

Figure 18.   Box plots of pollutants in the outlet of biolagoon (left) and after experimental equipment (right) from Väätsa landfill (experimental IV–X 2009).

obtained by using RO while more than 98% SS removal was accomplished by NF.

Figure 16 shows the removal efficiency of TN. Only 60% of removal of TN was achieved by either RO or NF. Therefore, de-nitrification should be recommended if TN is to be reduced below the discharge standards.

Figure 17 presents the removal efficient of TP. Since the concentration of TP in raw leachate is relatively low, the removal percentage was greater than 85%.

Figure 18 clearly shows that all the water quality parameters, such as COD, SS and BOD_7, were well below the Estonia discharge standards after either RO or NF. Therefore, these processes will be implemented in the full-scale leachate treatment plant at the Väätsa landfill site. However, if TN needs to be reduced, de-nitrification process is recommended.

During the exhaustion test, RO reduced COD and BOD of the biologically treated leachate by 97.9% and 93.2%, respectively, after 328 h. However, only 39.0% and 21.7% reduction in P_{tot} and N_{tot} were achieved. After 586 h, COD and BOD of the biologically treated leachate was reduced slightly to 95.2% and 91.7%, respectively. NF process after 328 h of work fell biologically treated leachate COD, BOD, and 97.5%, 93.8% and 99.2% total P and total N 68.6%. During the test, fouling problems occurred and spiral membranes needed to be washed and finally were fully replaced.

4.   Conclusions

After non-biodegradable organic and ammonia in the leachate at the Väätsa landfill site increased to a critical level, the AS system would simply become ineffective. Even worse, the AS biomass decayed and the TN and COD released to the treated leachate so that the effluent COD exceeded the influent COD. To meet the Estonia discharge standards, RO and NF were investigated to treat the biologically treated leachate. The reduction efficiencies of COD, BOD, dissolved inorganic expressed as conductivity and suspended solids were all excellent after ineffective

biological processes, such as AS and biological lagoon. For example, the RO process reduced 97% and 60% for COD or BOD, respectively. NF reduced 98%, 41% and 68% of biologically treated leachate COD, BOD and total N, respectively. However, RO was ineffective in removing TN. NF has a better result in removing P_{tot} and N_{tot} than RO. Although the COD, BOD, suspended solids and P_{tot} can meet current legislation requirements, neither NF nor RO could bring the TN below the discharge limit of 15 mg/l. In addition, successful application of membrane filtration technologies requires efficient control of membrane fouling, especially when spiral membranes are used.

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PAPER III

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Assessment of landfill wastewater pollutants and efficiency of different treatment methods

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Abstract
In Estonian landfills, the majority of biodegradable waste is composted, in addition to waste sorting and depositing. Stormwater and snowmelt water samples collected from the compost fields have shown a high content of pollutants. Furthermore, the flow rate of landfill wastewater can vary greatly. This has a significant influence on the options for and efficiency of treatment methods.
Different technologies for landfill wastewater treatment were tested, and the operation of several treatment plants was observed from 2007 to 2014. On the basis of the present research, the wastewater treatment system in Väätsa was redesigned and reconstructed. The treatment system consists of the landfill wastewater collection system, equalizing tank, physical/chemical, i.e. reverse osmosis (RO), treatment after biological activated sludge (AS) treatment and oxidation in pond, and stabilization of the pumping and distribution systems for concentrate discharge from the RO back to the landfill. Since April 2012, the parameters in the effluent from the treatment plant have been in compliance with the permitted limit values.
The composting of biodegradable waste needs to cease for the efficient and stabilised treatment of landfill wastewater. Methane fermentation is considered to be the most effective method for biodegradable waste treatment, and it generates biogas as a by-product.
The rearrangement of composting and depositing of biodegradable waste in combination with anaerobic fermentation would facilitate the production of up to 23.1 million m^3 of biomethane per year, which is equal to about 226 MWh heat and electric energy. The digestate that is produced during methane fermentation contains a significant amount of plant nutrients, which could be used for fertilising certain cultivated areas.

Keywords: *landfill wastewater treatment, landfill wastewater characteristics, landfill wastewater, landfill, biodegradable waste, anaerobic treatment, biogas.*

Concepts:
Landfill wastewater – Water that consists of leachate, i.e., liquid that moves through or drains from a landfill, precipitation that passes over the landfill site, vehicle washing water, and water drained from sanitation devices.

Landfill leachate – Water that has percolated through the contaminated material, e.g., tipped refuse.

1. INTRODUCTION

The EU Landfill Directive 1999/31/EC on waste and landfills provides the technical requirements for waste treatment during the landfill's life cycle, thereby minimising the negative environmental impacts on the surrounding environment, including surface and ground water, soil, ambient air and human health [1]. As per the Directive 2000/60/EC by the European Parliament and the Council and Estonian Water Act, the good environmental status of all water bodies should be achieved by 2015 [2, 3]. Therefore, the effective management of landfill wastewater is one of the major challenges.

Landfill wastewater is any kind of water that is collected from the landfill territory, including stormwater and leachate. The amount and content of pollutants in the landfill wastewater are directly related to the activities carried out in the area (sorting of waste, ways of depositing, usage of the composting fields, cleaning of machinery and tanks, etc.). The pollutant content in the water running through the landfill body and the leachate released from the decomposition of the waste is influenced by the composition of the deposited waste and decomposition processes taking place in the landfill. These factors are influenced by the age of the landfill and decomposition phases of the waste layer, i.e. aerobic, anaerobic acid, intermediate methanogenic, stabilised methanogenic and final aerobic phases. The quality and quantity of landfill wastewater varies throughout the

lifetime of the landfill site. Municipal landfill wastewater is characterised by a high concentration of organic matter, salts (mainly NaCl), nitrogen (NH_4-N) and toxic elements [4-6].

The landfills in Väätsa, Torma, Uikala, Jõelähtme and Paikre (in Estonia) were constructed after the year 2000 in accordance with EU environmental requirements.

Due to the limited amount of comparable data, a long-term study was performed to specify the pollutant content and flow rates of landfill wastewater and leachate.

The main condition for designing and operating a landfill is the minimisation of harmful emissions, i.e. employing an effectively operating treatment plant and system for landfill wastewater and setting up a collection and treatment system for the landfill gas. The efficiency of landfill wastewater treatment should be in compliance with the legislative requirements, and costs for the construction and maintenance of the landfill should be optimal. The selected treatment process should stand inconsistencies in the landfill wastewater flow rate as well as changes in the concentration of the pollutants and their chemical composition, major fluctuations in the temperature, and toxicity and high nitrogen content in the leachate water.

Stormwater and leachate, water from washing machinery and tanks, and domestic wastewater are collected from the landfill territory. Stormwater originating from the new unused, watertight areas of the landfill and composting fields is directed to an equalizing tank. The stormwater from depositing areas runs through the waste deposit and reaches the drainage system. The retention time depends on the thickness and density of the waste layer and could be a day, week, month or even a longer period [7]. The polluted stormwater collected from the composting fields contains a large amount of pollutants in high concentrations, making it difficult to choose a suitable treatment method. The recirculation of leachate occurs in the waste deposit as well as the equalization of the flow rate and pollutant concentration of the landfill wastewater. The equalizing tank is used for minimising the flow rate to the treatment plant and equalization the top pollutant loads and concentrations.

Different biological, physical and chemical methods are used for the treatment of landfill wastewater and leachate. The biological cotreatment of leachate and domestic wastewater is widely used because of its various technological and economic advantages (Tallinn and Pärnu municipal wastewater treatment plants as well as the landfill wastewater from the landfills in Jõelähtme and Paikre are cotreated). However, to avoid the hindering influence of leachate in the treatment processes and to guarantee the quality of the treated water, it is essential that the share of leachate in the mixture does not increase above 5-10% [8, 9].

Many studies and handbooks have described different methods for the treatment of landfill wastewater and leachate [4-6, 10,11]. Examples of leachate treatment activities are as follows:
Physical treatment processes: air stripping (methane stripping, removal of ammonia-N and stripping of other volatile contaminants); reverse osmosis; removal of solids (sedimentation and settlement, sand filtration and dissolved air flotation); activated carbon adsorption (powdered and granular activated carbon); ion exchange; and evaporation/concentration.
Chemical treatment processes: chemical oxidation processes (ozonation and hydrogen peroxide) and precipitation/coagulation/flocculation (chemical precipitation of metals, and coagulation and flocculation).
Aerobic biological treatment processes: suspended growth systems (aerated lagoons, activated sludge, sequencing batch reactors and membrane bioreactors), attached growth systems (percolating filters, rotating biological contactors, biological aerated filters/submerged biological aerated filters and biofilm reactors).
Aerobic/anaerobic biological treatment processes: engineered wetlands (horizontal flow reed beds, vertical flow reed beds and wetland ponds).

Traditionally, aerobic biological oxidation is the most widely used treatment method (treatment with active sludge and biofilms), but the results of this treatment are not satisfactory due to the specifics of generation and the content of the landfill wastewater [12].

Combinations of aerobic and anaerobic biological oxidation have been used for the treatment of landfill leachate [13]. The water emanating from the biological treatment plant requires additional processing and, along with the biological treatment, either physical-chemical or chemical methods should be applied to achieve the required treatment level [14-16].

Coagulation/flocculation and active carbon adsorption are the most commonly used physical-chemical treatment methods. Humic substances can be removed from the biologically-treated landfill wastewater by flotation or with the use of bioflocculants. Struvite precipitation is recommended for removing ammonia. Both membrane

reactors and struvite precipitation may be used following anaerobic pre-treatment for the treatment of wastewater from young landfills [10].

Chemical oxidation including ozonation is the only process for decomposing organic matter that is unmetabolised by microorganisms. The aim of pre-ozonation is to improve the biodegradability of the treated wastewater, whereas the purpose of post-ozonation is the advanced treatment of the wastewater [8, 17, 18]. After ozonation, the biodegradability of the processed water is increased, indicating the requirement for additional biotreatment. Therefore, special attention needs to be given to the ozonation technique in the recirculation cycle [9, 19]. The main areas for using ozone are disinfection, oxidation of organic substances and compounds, removal of taste, smell, and colour, and increasing biodegradability [20].

The Fenton process has been used in treating landfill wastewater, and it consists of four stages: oxidation, neutralising, coagulation/flocculation and separation of the solid and liquid phases [8, 21, 22]. Under optimal conditions, this treatment process can decrease the chemical oxygen demand (COD) by 70% [15]. The Fenton process may be used for both the pre-treatment of landfill wastewater before biological treatment and for post-treatment [8, 22, 23]. In this process, the toxicity of the treated wastewater decreases while there is an increase in its biodegradability [21, 15]. The advantage of this process includes the reduced energy requirement for creating radicals, availability of cheaper and nontoxic reagents, and the process is not limited by mass exchange (homogenous catalysis) [21]. However, some of the shortcomings of this process include the generation of sediments and the need to regulate the dosage of reagents according to the COD and pH for safeguarding the optimal conditions, as there is the problem in the treatment of landfill wastewater due to COD and pH high variation.
In practice, the Fenton process is used in the post-treatment stage, but it has also been recommended for processing landfill wastewater prior to biological treatment with the aim of increasing its biodegradability [8, 21].

Ozonation in combination with biological treatment decreases the toxicity of the wastewater, the required oxidant amount and financial costs, and it increases the biodegradability of the wastewater [20].

In the case of high salt content, reverse osmosis (RO) is used for the additional treatment of the biologically pretreated landfill wastewater. RO is also used as an independent method for treating leachate [4-6, 10, 23, 24]. The low operational costs and the ability to remove the organic contaminants and 95-99% of inorganic salts with minimal chemical requirements makes the RO an attractive technology for many applications [11].

In most cases, the landfill wastewater is treated through a combination of different treatment methods. In Torma, Estonia, the landfill wastewater is treated using a buried sand filtration unit after the sedimentation pond treatment. Subsequently, processing in the equalizing tank and post-treatment with sand and ceramzit filters have been followed by mechanical, biological (AS) and chemical treatment methods since 2010. A stabilization pond and biochemical treatment with activated sludge have been used for the Väätsa landfill wastewater, and biological treatment (AS and oxidation pond) and RO treatment has been followed by the equalizing tank since 2013. In the Uikala landfill, RO was applied after the equalizing pond. During the period from 2007 to 2014, detailed studies on the operation of the sewage systems were conducted in two problematic landfills, i.e. Väätsa and Torma. In the Uikala and Jõelähtme landfills, the emissions generated in the sewage systems were investigated with the aim of determining the technical and technological solutions for decreasing the environmental impacts.

2. TREATMENT OF LANDFILL RUNOFF WATER IN VÄÄTSA AND TORMA

The first municipal waste depositing field in Väätsa landfill at a size of one ha was ready in November 2000. In November 2005, the second depositing field of 1.34 ha was completed. The planned height of the waste layers ranged from 6-7 m. In 2008, the third field of 2.8 ha for municipal waste was completed. Altogether, there were four depositing fields with a total territory of 8.8 ha. The first composting field of 0.268 ha was ready in November 2003 and the second one-sized 1.34 ha began operation in July 2008.

The landfill wastewater treatment plant in Väätsa was completed in 2002. This biochemical plant consisted of an activated sludge container (aero tank and lamella clarifier) together with an oxidation pond and a sludge stabilization tank with aeration. The first part of the oxidation pond was aerated. Before directing wastewater into the treatment plant, the chemical composition of the water was regulated, where necessary, by adding phosphoric acid to avoid phosphorus deficit. The leachate was diluted with treated wastewater before being directed into the treatment process. The aquatic part of the oxidation pond amounted to about 2,000 m^3; it was regulated up to 1,250 m^3. The activated sludge plant was dimensioned for the flow rate of 70 m^3/d, including

leachate from depositing fields (30 m^3/d) and dilution water from the aerated oxidation pond (40 m^3/d). Phosphorus removal was achieved by dosing iron sulphate into the activated sludge plant. The excess sludge that was generated during the treatment process was directed into the stabilization tank with aeration, and the clarified water was conducted back into the treatment process. The stabilized sludge was carried to the depositing field. Water from the activated sludge plant was treated in the aerated oxidation pond.
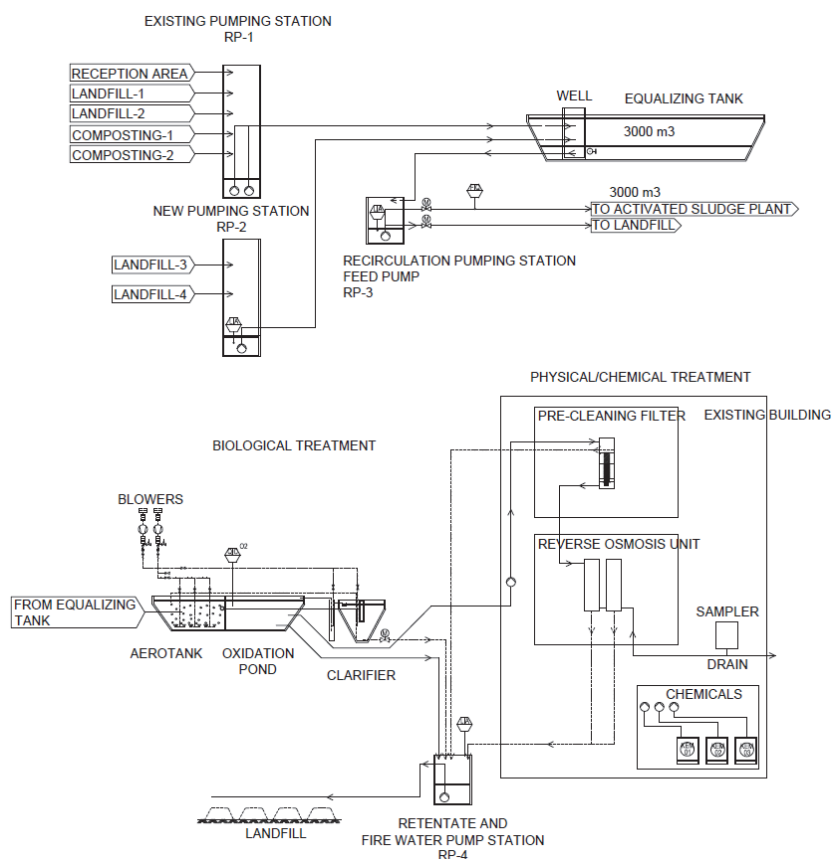
In addition to the studies in 2007, the efficiency of the operation by the Väätsa landfill activated sludge plant was monitored during the precipitation period in autumn 2008 and snowmelt period in spring 2009 [7]. In November 2008, the COD of the wastewater changed in the limits from 3100 to 4800 mgO/l and the biological oxygen demand (BOD$_7$) changed from 45 to 300 mgO$_2$/l. At the end of the month, the flow rate decreased and the values of COD and BOD were increased to 10500 mgO/l and 1550 mgO$_2$/l, respectively. In April and May 2009, the changes in the flow rate were greater due to the springtime snowmelt. The fluctuations of COD and BOD in wastewater were smaller than in November; the COD values of the wastewater varied from 1000 to 3500 mgO/l and BOD$_7$ varied from 50 to 350 mgO$_2$/l. The biodegradability of the wastewater (BOD/COD) was very small, i.e. below 0.1. The samples taken in May 2009 indicated the inability of the treatment plant to operate according to the requirements (Table 1) [7].

Table 1. Content of pollutants in the Väätsa landfill wastewater and water from different treatment stages in May 2009 [7].
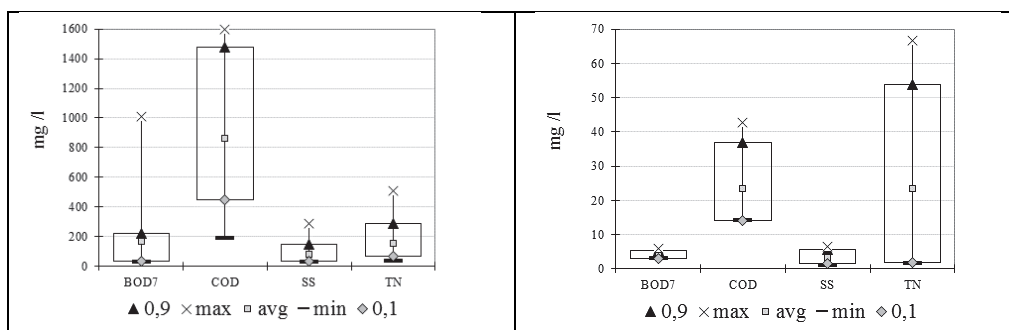
Pollutants	Unit	Väätsa landfill wastewater	Pollutant content after treatment with activated sludge	Pollutant content after stabilization pond treatment	Permitted limit values in effluent [25]
BOD$_7$	mgO$_2$/l	250	110	22	25
COD	mgO/l	4000	3000	800	125
TP	mgP/l	9.0	4.5	2.6	2.0
TN	mgN/l	474	414	210	75
pH		8.5	8.5	8.2	6-9
SS	mg/l	260	108	40	35

The ammonia-nitrogen concentration could not be reduced to the required 75 mg/l without any additional treatment. The high content of ammonia-nitrogen seemed to be toxic to the microorganisms taking part in the activated sludge process, and this is also reflected in the data in Table 1 that shows the efficiency of the activated sludge process. Consequently, the effluent was found to be dark and contained solids in high concentrations. The BOD$_7$/COD ratio dropped to less than 0.1 during 2008-2011, which indicates the ineffectiveness of the biological processes. According to the results of the current study, during rainfall and snowmelt, a significant part of the flow rate and pollution load of the wastewater originates from the composting fields of biodegradable waste, which must be decreased substantially. An equalizing tank for flow rate, pollution load and toxicity of the wastewater should be constructed. During the winter, the decrease of the temperature of the leachate directed into the treatment plant should be minimised. The removal of fat and oil prior to the biological treatment is very important. However, the biological methods are not enough for treating the landfill wastewater up to the requirements, and reverse osmosis should also be applied in the treatment protocol.
In 2009, the tests on different methods for the treatment of seepage landfill wastewater were conducted. By the end of the year, the preliminary project for reconstructing the Väätsa landfill wastewater treatment plant was completed [26]. The Väätsa landfill wastewater collection system – consisting of an equalizing tank, physical/chemical treatment (RO) after biological treatment (AS) and stabilization in pond treatment system (Figure 1) – was designed and built during 2011/2012.

Figure 1. Preliminary design of the technological scheme of Väätsa landfill wastewater treatment plant [26].

Figure 2. Pollutant content in the wastewater in Väätsa landfill during the period from 2001 until March 2012 (left) and from April 2012 to 2014 (right).

Since April 2012, when the new treatment system for landfill wastewater began, the effluent from the wastewater treatment plant was in compliance with the requirements in the water permit (Figure 2 and Table 2). The average treatment efficiency in the time period between 2013 and 2015 was recorded at over 99% of BOD_7 COD, TN and TP and over 90% of SS [23, 27].
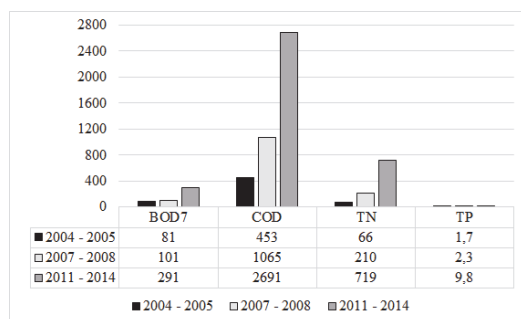
Table 2. Average pollutant content in the Väätsa landfill wastewater effluent, 2013-2015 [27]

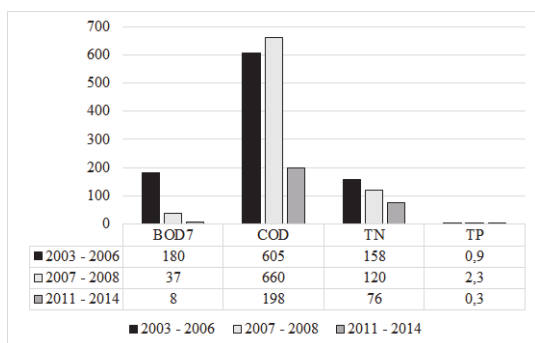| Parameter	2013	2014	2015	Limit value [25]
BOD$_7$, mgO$_2$/l	3	3	3	25
COD, mgO/l	14	14	14	125
SS mg/l	6.5	2.25	2.5	35
TN, mgN/l	1.5	2.1	1.6	75
TP, mgP/l	0.04	0.03	0.02	2
monobasic phenols, mg/l	0.005	0.002	0.0003	0.1
dibasic phenols, mg/l	0.01	0.01	0	15

The first municipal waste depositing field in the Torma landfill at a size of 0.65 ha was completed in June 2001. The designed average height of the deposit was 6 m. By the end of 2007, a second depositing field of 1.58 ha began operation. Here, the planned height of the deposits was 7 m. There is a plan to create a third depositing field with an area of 0.85 ha and a deposit height of 6 m.

During the first years of operation, the leachate and stormwater collected from the territory were conducted into the sedimentation pond, where the pollutant content in the water was equalized and primary treatment (mainly sedimentation) was performed. In the following step, the wastewater was directed into the pumping station and then into the buried sand filtration unit of 0.07 ha. The sand filtration unit was lined with geomembrane to ensure water tightness and it was dimensioned such that the load would not exceed 90 l wastewater per 1 m of distribution pipelines per 24 h. It was estimated that the annual load of wastewater directed into the treatment plant was 8,000 m^3 (average flow rate being 0.25 l/s). To avoid overloading the sand filtration unit, a cylindrical extension was placed at the end of the pipe running from the sedimentation pond to a pump well, which safeguarded the stable flow of wastewater into the pump well. The wastewater treated in the buried sand filtration unit was collected in the pumping station, from where it was directed into the receiving waterbody. The pollutant content and treatment efficiency of the wastewater are depicted in Figures 3 and 4.

The efficiency of the buried sand filtration unit was found to be low and, after several years of usage, it became lower during 2007 and 2008. The probable reason was exhaustion of the treatment capacity of the filter bed body. In order to clarify the operational failures, the sand filtration unit should be dug open. To treat wastewater from the Torma landfill, a new wastewater treatment plant was designed in 2009, which was ready for use at the beginning of 2010. The landfill wastewater was treated mechanically, biologically and chemically (Figure 5).

	BOD7	COD	TN	TP
2004 - 2005	81	453	66	1,7
2007 - 2008	101	1065	210	2,3
2011 - 2014	291	2691	719	9,8

Figure 3. Average pollutant content in Torma landfill wastewater entering the treatment plant during the period of 2004-2005, 2007-2008 and 2011-2014.

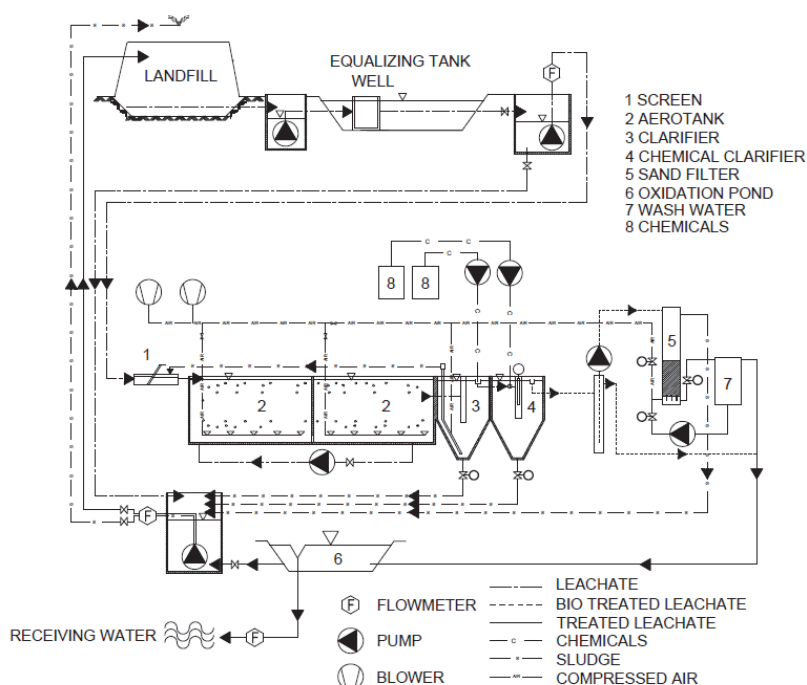|	BOD7	COD	TN	TP
■ 2003 - 2006	180	605	158	0,9
□ 2007 - 2008	37	660	120	2,3
▤ 2011 - 2014	8	198	76	0,3

■ 2003 - 2006   □ 2007 - 2008   ▤ 2011 - 2014

Figure 4. The average pollutant content in the effluent from Torma wastewater treatment plant falling into the final recipient during the period of 2003-2006, 2007-2008 and 2011-2014.

The treated wastewater is directed to the network of forest oxidation ponds and subsequently into Mustvee river. For equalization of the flow rate and pollution load before treatment, the existing interim well and equalizing tank with a mixer amounting to about 1,700 m^3 was used. The equalizing tank is 50% larger than the number found by the integral graph on the basis of annual flow rate values and this facilitates maintenance at a depth suitable for the mixer as well as a buffering capacity for pollutants throughout the year. During the winter period, water is taken directly from the well located in the equalizing tank (Figure 5), where it has arrived from the depositing fields at a relatively higher temperature. The designed decrease for COD for this complex is about 80% and 60% for TN. The approximate average ($Q_{avg\ d}$) and maximum capacities ($Q_{max\ h}$) of the plant are represented as: capacity $Q_{avg\ d}$ = 100 m^3/d and $Q_{max\ h}$ = 5 m^3/h [34]

The indicators for the efficiency of the wastewater treatment plant operation improved after the new treatment system was constructed for pH, SS, BOD and COD as well as for TP, but COD and TN remained problematic (Table 3). The limit values stipulated in the permit have been exceeded for both COD as well as TN. In 2012, the conservation and closing activities of the landfill were started. The amount of deposited waste has decreased. Most of the wastewater is pumped back into the landfill for irrigation of the waste and, in the last number of years, only the polluted stormwater collected from the territory is conducted into the treatment plant. In the first quarter of 2013, no leachate was generated by the stormwater in the landfill and no effluent was directed into the recipient. Thereafter, only the contaminated stormwater was collected from the territory of the landfill and treated in the plant; the leachate was pumped back into the waste deposit. In 2014, only the stormwater collected from the open fields was treated; the leachate was pumped back into the landfill. The treatment efficiency was high with values such as BOD_7 = 96.4%, SS = 94%, COD = 93.1%, TN = 92.2% and TP = 96.9% [29].

Figure 5. Technological scheme of the wastewater treatment plant in Torma landfill [30].

Table 3. Results of the Torma landfill wastewater analysis from different stages of treatment and effluent directed to the final recipient on 16 September 2010 [30].

Pollutants	Unit	Entering into the treatment plant	After biological treatment	After chemical treatment	Effluent	Limited values in effluent [25]
pH		8	7.8	5.5	6.7	-
SS	mg/l	170	210	46	15	35
BOD$_7$	mgO$_2$/l	57	40	4,1	< 3	25
COD	mgO/l	1069	581	175	141	125
TN	mgN/l	250	270	240	230	75
TP	mgP/l	2.3	2.0	< 0.02	< 0.02	2

3. TREATMENT OF BIODEGRADABLE WASTE IN THE LANDFILL

Biodegradable waste in Estonia is made up of the biodegradable portion of municipal waste, park waste, yard waste, agricultural waste, commercial waste, industrial waste, wastewater sludge and animal waste.

In 2011, the amount of biodegradable waste generated in Estonia was 1,196,670 tonnes. Of the 158,900 tonnes of wastewater sludge, 123,100 tonnes of biodegradable waste from municipal waste (kitchen waste, catering waste, park waste and yard waste) and 7,970 tonnes of other types of biodegradable waste (altogether 289,970 tonnes) were considered as suitable raw materials for the fermentation process leading to the production of biogas [31]. Most of the aforementioned materials were composted in compost fields located at the landfills in wastewater treatment plants and in separately located composting fields. A portion of the biodegradable waste was deposited in landfills. Before composting, the wastewater sludge is treated with methane fermentation in the wastewater treatment plants in Tallinn, Narva and Kuressaare. In the Tartu treatment plant, methane

fermentation is still being adjusted. In these plants, the biodegradable waste is anaerobically fermented. The produced biogas is collected from the landfills and used as an energy source, and the digestate is utilised for fertilising fields. In comparison with the present situation where the main activities include the composting and depositing of waste into landfills, the impact on the environment seems to be significantly smaller.

In the Landfill Directive 999/31/EC, it has been stated that by 16 July 2016, the amount of biodegradable waste that is deposited in landfills should decrease for 35 mass percent compared to the total amount of deposited municipal household waste in 1995 [1].

The Estonian Waste Act has stipulated limits for depositing biodegradable waste in landfills. By 2020, the share of biodegradable waste in the deposited municipal household waste must not exceed 20% [32].

According to the national waste management plan, it is of primary importance to decrease the entire volume of deposited waste by 2014-2020. The reusage of biological waste should be increased significantly and anaerobic fermentation should be preferred over composting. The fermentation residue (digestate) should be used in agriculture as much as possible [31].

The average yield of biogas obtained by methane fermentation from the biodegradable waste collected with biocontainers was 375 m^3/t ODM (organic dry matter), 425 m^3/t ODM from degradable food waste and commercial waste [39] and 322 to 372 m^3CH_4/t (CH_4 - methane) from wastewater sludge (residual activated sludge) [34].

In 2010, the production of biogas in Estonia was 13.13 million m^3, and most of that (9.3 million m^3) originated from landfills. A total of 3 million m^3 of biogas was produced from wastewater sludge.

The annual potential of biomethane (excluding agricultural biomethane) has been recorded as follows: biodegradable waste from food manufacture 9 million m^3, biowaste 2 million m^3, wastewater sludge 3 million m^3, biowaste from industry 8 million m^3 – altogether 22 million m^3. This is supplemented with landfill gas of 9 million m^3 [35].

It is expected that by 2020 landfill gas will be collected from all landfills according to the requirements and about 50% of the collected landfill gas will partly be used for producing electric energy [31].

The digestate that is generated in the methane fermentation of biodegradable waste contains a large amount of plant nutrients, such as phosphorus and nitrogen; therefore, it can be used for fertilising arable land. The volumes of the digestate are more or less equal to the volume of the fermented biodegradable waste [33]. In Tallinn wastewater treatment plant, compost made up of the fermented residual activated sludge is used in fertilising fields, green areas and recultivation [36].

The yield of biogas from the methane fermentation of animal waste is high, but further treatment and utilisation of that digestate have been problematic. For example, to further increase the interest of stakeholders, the primary energy production potential for annually produced Category 2 and 3 solid slaughterhouse waste in Estonia was evaluated. It was found that a maximum of 5.5 million litres of the petrol equivalent of biomethane should be produced and used as a transportation fuel derived from the local renewable resource. The digestate, which is rich in nitrogen and other nutrients, could be used as an organic fertiliser on 7,120 ha of agricultural land in place of mineral fertilisers [37].

According to the national waste management plan for 2014-2020, in the optimal plan for managing municipal waste with the least environmental impact, by 2020 30% of waste will be recycled as a secondary raw material, 3% will be recycled as compost and 10% as a digestate from anaerobic fermentation, 40% will be incinerated, 8.5% will be utilised as a waste fuel in cement manufacturing and 8.5% will be deposited in landfills [31].

The present study takes into account the quick changes in waste management (collection of sorted waste, increasing the significance of the sorting and incineration of municipal waste, decrease in depositing of municipal waste and increase in the role of composting) and the impact of the mentioned changes on the volume and pollutant content of the landfill wastewater. Therefore, accordingly, one of the goals was to find the possibilities for decreasing and equalizing the flow rate, pollutant content and toxicity of the landfill wastewater. The second direction of the research involved investigation of the possibilities for methane fermentation of the biodegradable waste deposited and composted in the landfill, assessment of the yields of biogas generated in

fermentation, assessment of the possibilities for utilising biogas and digestate obtained by fermentation of the biodegradable waste, and its collection from the landfills.

4. RESULTS AND DISCUSSION

The studies on the landfill wastewater and leachate generated in Estonian landfills were performed in 2007-2013. The volume of the leachate and stormwater collected from the landfill depends mostly on the weather conditions, rainfall and evaporation. In the case of heavy rainfall and snowmelt, we have to deal with very big hydraulic and pollutant shock loads and, during a longer drought period, the flow rate of the wastewater may be close or even equal to zero. Stormwater from the unused watertight waste deposition areas of the landfill and composting fields is quickly directed into the equalizing tank and subsequently to the treatment plant for landfill wastewater treatment.

According to the duration of rainfall and the purpose of the watertight fields, 60 to 80% of the stormwater falling on the watertight fields and 10 to 30% of the stormwater falling onto the landfill waste lifts reaches the sewerage (or network).
At Väätsa landfill, the wastewater flow rate fluctuations were measured at periods with different intensity of precipitation and snowmelt: $Q_{min} = 0$-2 m^3/d; $Q_{avg} = 10$-20 m^3/d (1.4 to 2.9 m^3/ha in day), $Q_{max} = 50$-95 m^3/d (7.1 to 13.6 m^3/ha); in some cases, Q_{max} increased up to 150 m^3/d (21.4 m^3/ha). The leachate flow rate fluctuations were smaller $Q_{min} = 0$-2 m^3/d; $Q_{avg} = 5$-15 m^3/s (0.97 to 2.92 m^3/ha), $Q_{max} = 20$-30 m^3/d (3.89-5.84 m^3/ha) [23].
The pollutant content and its concentration in landfill wastewater directly depends on the weather conditions, construction of sewerage, waste sorting technologies, store technologies of different types of waste, size and loading of depositing fields, size and intensity of exploitation of composting fields, contents of the biodegradable waste and filling materials used for composting, washing technologies for machinery and containers, etc. The pollutant content in the landfill leachate depends on the weather conditions, construction of the systems for leachate collection, size and loading of depositing fields, depositing technologies (thickening, amount and contents of irrigation water, materials for interim layers, etc.), contents of deposited waste, degradation processes that depend on the age of the waste deposit, mixing of the leachate from waste lifts of different age, etc.
Very high concentrations of pollutants (and hence the pollution load) were measured in the leachate and stormwater collected at the compost site in Jõelähtme landfill. Table 4 shows the average physicochemical parameters and the content of hazardous substances in the stormwater collected in 2007 from the composting field of biodegradable waste in Väätsa and Jõelähtme landfills.
In the Jõelähtme landfill, the samples were taken from the stormwater controlling well in the composting field. The stormwater flow rate Q was 51 m^3/d, and the main filling material in the biodegradable waste composting was peat.
In Väätsa landfill, samples were taken from the stormwater controlling well in the composting field. The stormwater flow rate Q was 123 m^3/d, and the filling material in the (dehydrated, thickened and mixed with peat) wastewater sludge composting was chopped straw.

Table 4. Average physicochemical parameters and hazardous substances content in stormwater samples collected from the composting fields of biodegradable waste in 2007 [7]

Parameter	Unit	Jõelähtme	Väätsa	Parameter	Unit	Jõelähtme	Väätsa
pH		5.5	5.6	Hydrocarbon	mg/l	<20	300
Conductivity	μS/cm	4400	4870	Fe^{2+}	mg/l	3.9	5.5
SS	mg/l	900	1130	Fe^{3+}	mg/l	2.9	6.6
BOD$_7$	mgO$_2$/l	3960	1875	Hg	μg/l	<0.05	0.1
COD	mgO/l	7530	9300	Ag	mg/l	<0.01	0.01
TOC	mgC/l	2100	2580	Cd	mg/l	<0.02	<0.02
NH$_4$	mgN/l	251	63.5	Cr	mg/l	<0.02	0.029
TN	mgN/l	700	179	Mg	mg/l	23	109
TP	mgP/l	42.8	40.1	Mn	mg/l	0.752	9.86
HCO$_3$	mg/l	1000	2470	Na	mg/l	300	57.5
SO$_4$	mg/l	89	144	Ni	mg/l	<0.,02	0.047
Cl	mg/l	422	106	Pb	mg/l	<0.04	<0.04
monobasic phenols	mg/l	209	617	Zn	mg/l	<0.1	<0.1
dibasic phenols	mg/l	43.5	71.6	Cu	mg/l	0.024	0.027

The concentration of the landfill wastewater pollutants showed a significant increase during the rainfalls, and these were dependent on the intensity of precipitation and the composition and amount of the deposited biodegradable waste on the watertight fields: for up to 40% for SS, 50% for BOD_7 and 70% for COD. The pollution load of TN increased by 20% and of TP by up to 40%.

Landfill leachate has a very high TN concentration and low concentration of TP. The leachate contains high levels of COD and BOD_7. The Uikala landfill leachate contained very large amounts of phenols (3000-4000 mg/l), magnesium (300 mg/l) and sodium (1600 mg/l). The heavy metal content in landfill wastewater did not exceed the permitted limits. The dumping of old car tyres in the base lift leads to an increase in the concentration of iron (heavy metal) in the leachate, i.e., up to 6-8 mg/l. Iron is responsible for the corrosion that may continue for up to 5-6 years leading to a blockage of the drainage.

The landfill leachate is toxic, hindering biological purification. The BOD_7/COD ratio in the leachate samples fluctuated significantly according to the studies conducted in 2007. The values in the different samples were as follows: Väätsa, 0.3-0.5; Jõelähtme, 0.2-0.7; Uikala, 0.2-0.6; and Paikre, 0.2-0.5. In 2010, the BOD_7/COD ratio was less than 0.1 in Väätsa landfill leachate. This was caused by the presence of humic and fulvic acids, tannins, lignin and hazardous organic chemicals, pesticides and herbicides in the leachate that decreased the biodegradability of water.

In all the investigated landfills, the biological purification ratio of BOD : N : P (100 : 5 : 1) was out of balance. For example, in 2007, this ratio in Väätsa landfill wastewater according to average measurement resulted in the BOD_7 : TN : TP ratio to be 46.1 : 13.5 : 1 and in leachate it was recorded as 115 : 64.8 : 1. In Uikala landfill wastewater, the ratio was 267 : 183 : 1 and in the leachate it was 115 : 177.2 : 1.

The temperature of the wastewater from October to April was between 1°C and 4°C, which significantly hindered the biological purification process. The treatment capacity of the RO equipment increased by 3% for each degree of temperature rise.

5.   RESULTS OF THE *IN VITRO* EXPERIMENTS ON THE POSSIBILITIES OF LANDFILL WASTEWATER TREATMENT

From 2007-2013, experiments on the percolation and RO of landfill wastewater and on biological filter technology, submerged by means of light gravel filling, were conducted in the Department of Environmental Engineering and the other experiments in the Department of Chemistry Tallinn University of Technology (TUT).
The operation of the wastewater treatment plants existing landfills in Väätsa, Torma, Uikala and Jõelähtme was supervised.
Aerobic biological oxidation (treatment with activated sludge and submerged biological filter) was performed in the laboratory in order to achieve a decrease in the COD of landfill wastewater by 35%.
The ozonation reactor achieved a 9% decrease of the COD in the wastewater. In the first 20 minutes, the BOD increased by 5% and later began to fall. The reaction of the pollutants with the ozone was very slow and it depended on specific conditions. Therefore, the ozonation of landfill wastewater was not considered to be an efficient treatment method. There was a decrease in the COD but overall efficiency remained low. The colour and odour were removed but the biodegradability of the wastewater did not change.
The coagulation process showed a decrease in the COD of wastewater by 23% and post-ozonation by a further 11%. Post-ozonation of the coagulated wastewater is not efficient. Additional costs are required for the purchase of reagents and treatment of the residual sediments.
The post-ozonation of the wastewater that had been treated biologically in the activated sludge plant decreased the COD by 13%. The efficiency of this process was found to be low.
The post-ozonation of the effluent conducted from the treatment plant into the recipient increased biodegradability BOD/COD from 0.02 to 0.11 and COD decreased by 50%. The efficiency of that process was sufficient.
The post-ozonation of wastewater that had been biologically treated with laboratory equipment increased the biodegradability BOD/COD from 0.006 to 0.056. The efficiency of post-ozonation was recorded as 24.4% for COD. The biological treatment of wastewater and post-ozonation together decreased the COD by 55%. In the case of stable operation of the bioreactor, it is possible to use ozonation for increasing the biodegradability (BOD/COD ratio) of wastewater that is being treated in the recirculation cycle.
The lime coagulation and post-ozonation of wastewater (10% lime milk and 3.8% aluminium hydroxychloride were used) increased the biodegradability (BOD/COD ratio) of the treated wastewater from 0.038 to 0.540. The COD decreased by 23-27%. The efficiency of this process was considered to be low.

The coagulation with oil-shale ash and post-ozonation of wastewater increased the biodegradability of untreated wastewater less than the post-ozonation of water that had been coagulated with lime milk. It is not practical to use oil-shale ash for coagulation as the amounts of ash required are large and lengthy intensive mechanical mixing is needed. The biodegradability of wastewater treated with oil-shale ash increased less than in the case of ozonating untreated wastewater. The decrease in the COD in post-ozonation was small.

The Fenton process constituted the first series of experiments, where pH=3, the overall content of pollutants (COD) was decreased by up to 70% with low H_2O_2/COD ratio (0.5/1), colour and odour were removed and biodegradability (BOD/COD) was increased to 0.1.

In the second series of experiments, where pH was not regulated, the Fenton process decreased the COD of wastewater by up to 37% with the highest H_2O_2/COD ratio 2/1. There was no change in the biodegradability (BOD/COD) ratio. Treating landfill wastewater with the Fenton process at a pH value of 8 was (unlike the literature data [24]) less effective than processes in acidic medium. The best result, 37%, was achieved at the H_2O_2/COD ratio of 2/1. The biodegradability, in this case, did not change significantly.

The nanofiltration (NF) and RO (spiral filters) experiments used the equipment named ULTRA-FLO PTE, UF-NF 200. The RO process reduced COD and BOD by 97% and 60%, respectively. NF reduced the COD, BOD and TN, by 98%, 41% and 68% of biologically treated wastewater, respectively. However, RO was ineffective in removing TN. NF was more efficient in removing TP and TN than RO. Although the COD, BOD, suspended solids and TP can meet current legislative requirements, neither NF nor RO could bring the TN below the discharge limit of 15 mg/l. In addition, the successful application of membrane filtration technologies requires efficient control of membrane fouling, especially when spiral membranes are used [7].

The survey of the operation of the RO container treatment plant with filter DT 29-09 after the equalizing tank in the Uikala landfill showed that the treatment plant for wastewater operates with high efficiency and is stable. The medium treatment efficiency in the fourth quarter of 2013 was 98.3% for BOD_7, 98.7% for suspended solids and over 99% for both TN as well as for TP.

6. DECREASING POLLUTANT CONTENT AND FLOW RATE OF THE LANDFILL WASTEWATER FOR PURIFICATION PURPOSE

In the new landfills in Estonia, a large part of the territory is covered by composting fields with an impermeable cover. For example, Väätsa landfill is about 1.6 ha and Uikala landfill about 3.4 ha. The storm and snowmelt water collected from the composting fields periodically and significantly increases the flow rate, pollutant content and pollution load of the landfill wastewater, and that, in turn, has a big impact on the possibilities, efficiency and costs of wastewater treatment. To decrease the pollution load and flow rate of the wastewater requiring treatment, composting of the biodegradable waste in the landfills and channelling stormwater from composting fields into equalizing tanks should be terminated. The biogas and nutrient-rich digestate can be produced from the biodegradable waste by applying methane fermentation. Stormwater collected from the composting fields and other parts of the landfill that are clean from pollution does not require additional treatment. This stormwater can be directed into a recipient or it can be partially collected and used for irrigating the waste deposits in the process of biogas production. In recent decades, the homogenous mass of paper and cardboard that was binding a lot of water has been substituted by plastic waste mixture that is pressed together, layer by layer, and the amount of biodegradable waste deposited has decreased significantly. In the case of highly intensive irrigation, wastewater starts to drip out from the sides of the waste deposit.

The collected stormwater can be used for diluting leachate with a high concentration of pollutants during drought periods, thereby making treatment of the latter more efficient.

If the above-listed measures for handling landfill wastewater are applied, smaller infrastructures (equalizing tank and wastewater treatment plant) are required and the treatment process is made more stable and efficient.

7. EQUALIZATION OF POLLUTANT CONTENT AND FLOW RATE IN THE WASTEWATER TO BE TREATED

The hindering factors for the aerobic biological treatment of landfill wastewater (for example aerated lagoons, activated sludge, sequencing batch reactors, biofilm reactors) and physical treatment processes (for example reverse osmosis) are the big inequalities of the flow rate and pollutant content in the wastewater, low temperature in winter (from 1 to 4°C) and toxicity of the leachate. The landfill wastewater contains chemical substances that are difficult to degrade and, as a rule, it is heavily polluted with organic and inorganic compounds.

The inequalities of the flow rate, pollutant content and pollution load of the wastewater can be decreased if the composting is terminated, composting fields are cleared and the clean stormwater is conducted directly into the recipient water body. The remainder of the polluted stormwater and leachate has to be collected into the

equalizing tank with a regulated volume. In dimensioning equalizing tanks, it should be taken into account that, according to the previous study, the share of stormwater that can be removed from the depositing field is up to 20% for old landfills and up to 60% for new landfills [7]. In the case of heavy rainfall (years with high precipitation of up to 800 mm), the amount of rain falling on one hectare is up to 8,000 m^3. The necessary volume of the equalizing tank and fluctuation range of its water level is determined with the help of an integral graph compiled on the bases of annual rows of runoff values, so that the buffering capacity of the equalizing tank would be maintained for the whole year round. In case of necessity (during periods of drought), the tank can receive additional water from the wastewater treatment plant as effluent, or from the deposit of previously collected clean stormwater. The water from the equalizing tank can be used for putting out possible fires in the waste deposit as has happened at the Torma and Paikre landfills.

In the winter periods, when the temperature of landfill wastewater is very low, leachate is taken immediately from the interim well located in the equalizing tank, where the water is relatively warmer, arriving directly from the waste deposit. In the summer, wastewater is collected from the tank.

8. CLARIFICATION OF TOXICITY OF LANDFILL LEACHATE AND CONCENTRATE FROM REVERSE OSMOSIS

The toxicity of landfill wastewater was measured with the help of ecotoxicological tests on the basis of the impact on *Protozoa*; subsequently, the impact on the bacteria in the activated sludge was concluded. The wastewater from landfills is considered to be toxic and hinders the microbiological processes in the biological treatment process. Toxicity is caused by the high content of ammoniacal nitrogen (for example, it was up to 974 mgN/l in Jõelähtme landfill, up to 729 mgN/l in Uikala landfill and up to 332 mgN/l in Väätsa landfill). It has been found to increase during the summertime due to high pH and temperature. 90% of the total volume of nitrogen was found to be in the form of ammoniacal nitrogen, with a big share of the latter being found in the form of ammonia, which is toxic for water organisms. In addition, the toxicity of the aquatic environment can be influenced by pH, conductivity, concentration of chlorides and the content of copper and zinc. The xenobiotic organic compounds contained in leachate can be utterly toxic. All these determinants should be taken into account when choosing the correct technology for the wastewater treatment.

The remnant of landfill leachate can also become toxic due to many other factors, such as the excessive content of diluted heavy metals, a pH level that is too low or too high, an unfavourable carbon and nitrogen (C/N) ratio, etc. [38].

The study on the remnants revealed that the pH level and measures of diluted heavy metals were within the norms. The C/N ratio in the case of the remnant was 4, which is considered to be too low. The low C/N ratio refers to the low carbon (C) content and excessive nitrogen content. During the process, carbon is first used in the fermentation, and if its amount is insufficient nitrogen then becomes toxic for methane bacteria [38, 39].

The normal C/N ratio for biogas production is between 10-40 [40].

In Väätsa landfill, the concentrate from reverse osmosis is pumped into the waste deposit. A series of experiments with methane fermentation were carried out with the aim of determining the toxicity of the concentrate produced in reverse osmosis and its influence on the different phases of degradation in the waste deposit as well as the degradation of organic substances during fermentation. The concentrate discharged from reverse osmosis was co-digested anaerobically in a mixture with Tallinn wastewater treatment plant sewage sludge to evaluate the degradability and methane productivity in various mixing ratios. The content of substances in the concentrate from reverse osmosis is depicted in Table 5 and the biomethane potential (BMP) batch experiments are depicted in Table 6.

Table 5. Väätsa landfill wastewater RO treatment concentrate October 2014

Parameters	Result
Total solids (TS) in RO treatment concentrate	3.7%
Volatile solids (VS)[*)]	43.1% TS
pH	6.9
Total nitrogen (TN)	1.2 kg/m^3 TS
Ammonium nitrogen (NH$_4^+$-N)	0.94 kg/m^3 TS
Total phosphorus (TP)	0.02 kg/m^3 TS
Total potassium (TK)	0.47 kg/m^3 TS
Crude protein	10.54% TS
Crude fat	0.02% TS
Carbon C	5.07% TS
Nitrogen N	1.27% TS
Hydrogen H	1.37% TS
Sulphur (S)	0.43% TS

Zinc (Zn)	0.87 mg/kg TS
Copper (Cu)	0.13 mg/kg TS
Mercury (Hg)	not found
Cadmium (Cd)	not found
Chromium (Cr)	3.30 mg/kg TS
Nickel (Ni)	0.64 mg/kg TS
Lead (Pb)	<0.01 mg/kg TS

*) Volatile Solids from Total Solids

Table 6. Yield of biogas in BMP tests with reverse osmosis concentrate (Figures 6 – 9)

No	Substrate S/I=0,2 *)	Average yield of biomethane, m^3 CH_4 / tonnes VS (volatile substance)	Average yield of biomethane, m^3 CH_4 / m^3
1	SS 100%	299	8.0
2	SS 90%+VÄ 10%	131	3.3
3	SS 75%+VÄ 25%	179	4.1
4	SS 50%+VÄ 50%	29	0.6
5	SS 25%+VÄ 75%	−124**)	−2.0**)
6	VÄ 100%	−174**)	−2.2**)
7	VÄ 100%	−51**)	−0.5**)
8	DW20%+VÄ 80%	−15**)	−0.1**)
9	DW40%+VÄ 60%	−85**)	−0.5**)
10	DW60%+VÄ 40%	−144**)	−0.5**)
11	DW80%+VÄ 20%	−414**)	−0.8**)
12	DW90%+VÄ 10%	532	0.5

*) S/I = 0.2 - Substrate/Inoculum rate 0,2; DW - distilled water; VÄ - Väätsa leachate concentrate; SS - sewage sludge
**) The substrate did not give higher results than the inoculum

At first RO concentrate BMP test results were promising (Figure 6 – 9). However, after removal of inoculum productivity from total result revealed negative outcome (Table 6). RO concentrate had negative effect on anaerobic digestion process with or without sewage sludge. Even RO concentrate dilution with distilled water did not give a positive result.

Figure 6. Accumulated gas volume in the first BMP test series

Figure 7. Biomethane flow rate in the first BMP test series

Figure 8. Accumulated biomethane volume in the second series of BMP tests

Figure 9. Biomethane flow rate in the second series of BMP tests

The RO discharging concentrate additions have a negative effect on the anaerobic digestion of the sewage sludge. This decline in methane yield might be caused by the deterioration of methanogenic bacterial activity following treatment of RO discharging concentrate. Leachate reject should be directed to the closed site, where the extraction of biogas is in the final stage as it is toxic and does not support the biological decomposition processes.

The impact of RO concentrate on the processes taking place in the closed landfills in the anaerobic acidific phase and methane fermentation phase needs further study. When the treated wastewater or collected stormwater is pumped back into the waste deposit, the humidity of the waste increases significantly, bringing along a more intensive distribution of nutrients and microorganisms and more intensive biogas generation.

9. DECREASING THE AMOUNT OF BIOLOGICAL WASTE DEPOSITED AND COMPOSTED IN THE LANDFILLS BY METHANE FERMENTATION

158,900 tonnes of wastewater sludge were considered as a suitable raw material for the fermentation process leading to the production of biogas in 2011 [31]. Almost the entire amount of wastewater sediments that is generated will be reused, mainly composted and stabilized and, to a lesser extent, methane fermented and composted. The collection and reusage of sorted biodegradable waste in municipal waste is still in its initial phase (out of 123,100 tons of biowaste, collected in 2011, 85% was biowaste, which was contained in the municipal mixed waste and 15% of biowaste was collected separately), and most of the biowaste is deposited with mixed municipal waste in waste deposits or is incinerated [31].

It should be taken into account that methane fermentation followed by digestate composting is only possible from the biodegradable waste that is sorted before collection and pretreated by removing unsuitable materials. In the case of household waste (food waste from kitchens and animal wastes), it should be also hygienised in case of need. The amount of digestate generated in the methane fermentation of biodegradable waste is almost equal to the volume of biowaste used as a raw material for this process. The digestate that is generated from the fermentation process contains a large volume of nutrients such as phosphorus and nitrogen, which can be used for fertilising cultivable lands. For some time, the compost from wastewater treatment plant sludge and methane fermented wastewater sludge has been used for fertilising green areas and recultivation, and to a lesser extent for agricultural use. In the near future, the sorted biodegradable waste should be collected and the capacity for reusing those by methane fermentation in landfills and in other locations where biodegradable waste is accumulated should be developed.

10. UTILISATION OF LANDFILL GAS OBTAINED FROM METHANE FERMENTATION

As the result of degradation process of the deposited and composted biodegradable waste, biogas is generated, which consists mainly of carbon dioxide (CO_2) and methane (CH_4), whereas the content of methane in landfill gas remains in the limits of 50-55%. A large amount of sulphur compounds is generated in the composting biowaste and a smaller amount is originated from the anaerobic fermentation process. The emissions from the sulphur and nitrogen compounds (SO_2, NO_x, HCl and NH_3) into the ambient air due to decomposition of the biodegradable substances into biogas, cause acidification of soil and water bodies. In the case of the open composting of biodegradable waste, a large amount of ammonia (NH_3) emissions was additionally generated, which caused both acidification and a bad odour. In anaerobic fermentation, ammonia emissions are much smaller. In 2011, three landfills – Uikala, Jõelähtme and Väätsa – practice the combustion of 100% of biogas, because the amounts produced are too small for cost beneficial utilisation in the heat and energy cogeneration plants. For example, in Väätsa landfill, 175,200-262,800 m^3 biogas was collected from three depositing fields with a total area of 3.5 ha, annually (20-30 m^3/h). In order to increase the yield of biogas produced in the landfills and to start the exploitation of heat and energy co-generation plants, the composting of biodegradable waste in landfills should be substituted with the mesophilic methane fermentation of biowaste.

An experimental study under laboratory conditions and pilot reactors was performed at the Tallinn University of Technology (TUT) in order to find better solutions for anaerobic digestion (AD) process and to choose suitable substrates and co-digestion. Such experiments were carried out in the TUT Department of Environmental Engineering. The potential biomethane tests were conducted in anaerobic mesophilic conditions by measuring the maximum amount of biogas or bio-methane produced per gram of volatile solids (VS) contained in the organic matter used as a substrate for the anaerobic digestion process. These tests were conducted using either pure substrates or a mixture of two substrates in order to investigate the effect of the combination of different organic waste on the digestion process (co-digestion).

The substrates used in different batch methane potential (BMP) tests were as follows: sewage sludge, catering waste, sewage sludge + catering waste, sewage sludge + fishing industry waste, compost, sewage sludge + compost, whey, whey + sewage sludge, beer yeast, beer yeast + sludge (Table 7); RO concentrate + sewage sludge (Table 6). The BMP tests were used for the technical and economical optimisation of bio-methane producing plants.

Table 7 shows the yield of biogas from BMP tests and Table 8 depicts the yield of biogas from the one-stage co-digestion process.

Table 7. Biogas yield from BMP tests

No	Substrate /I =0.2 [*)]	Average yield of biomethane (CH$_4$), m^3/tonnes VS (volatile substance)	Average yield of biomethane (CH$_4$), m^3
1	Catering waste (CW) S/I 0.2	404	82
2	Sewage sludge (SS) S/I 0.2	245	7
3	SS+CW 10% S/I 0.2	265	9
4	SS+CW 25% S/I 0.2	353	22
5	SS+CW 25% S/I 0.2	484	42
6	SS+fish residues 2.5% S/I 0.2	296	10
7	SS+fish residues 5% S/I 0.2	346	14
8	Potato+gravy+salad+soup S/I 0.5	229	124
9	Compost (com) S/I 0.2	228	68
10	SS+com 25% S/I 0.2	229	20
11	Whey (W) 0.5	407	18
12	K-JSS+W 35% 0.5	203	10
13	K-JSS+W 50% 0.5	258	12
14	K-JSS+W 75% 0.5	330	15
15	K-JSS+W 90% 0.5	369	16
16	Beer yeast (BY) 0.2	831	98
17	Beer yeast 0.5	825	97
18	SS+BY 35%	624	34
19	SS+BY 50%	726	50
20	SS+BY 75%	740	69
21	SS+BY 90%	752	81

[*)] S/I = 0.2 - Substrate/Inoculum rate 0.2; K-JSS – Kohtla-Järve sewage sludge

The following substrates were tested in one-stage co-digestion process: sewage sludge, sewage sludge + fishing industry waste, catering waste, sewage sludge + catering waste, compost, RO concentrate (Table 8).
The sewage sludge originated from the Tallinn wastewater treatment plant (WWTP). The inoculum (Inoc) was taken from the city of Tallinn WWTP biogas plant anaerobic digester that was operating at +38°C with sewage sludge.

Table 8. Biogas yield from one-stage co-digestion process tests

No	Substrate	Average yield of biogas [m^3/tonnes VS]	Min and Max yields of biogas [m^3/tonnes VS]	CH$_4$ [%]	Reference, if published
1	Sewage sludge (SS)	110	68-240	33-62 (50)	
2	SS + 2% fish residues (F)	251	118-565	52-82 (69)	[37]
3	SS + 10% fish residues	206	170-281	60-71 (69)	[37]
4	SS + 36% F	205	198-211	58-70 (66)	[37]
5	Catering waste (C)	249	104-454	46-74 (66)	
6	C + SS	321	121-659	55-73 (65)	
7	Compost	50	80-129	58-76 (70)	
8	Väätsa RO concentrate	0	0-60	0-38 (0)	

Biomethane potential tests were done with Automatic Methane Potential Test System II (AMPTS II). The AMPTS II follows the same measuring principles as conventional methane potential tests, which make the analysis results fully comparable with the standard methods.

As can be seen from Tables 7 and 8, the most promising substrates for biogas production by BMP tests are catering waste, compost, beer yeast and their mixes with sewage sludge. Also, very good results were achieved with the same substrates in one-stage co-digestion process tests. Unfortunately, Väätsa RO concentrate inhibited co-digestion processes in every test in which it was added, as can be seen in Table 6.

On the basis of the results of the studies [7, 23, 24, 34, 37, 41-49] and results of laboratory experiments in Tables 7 and 8, it can be calculated that the average yield of biomethane produced from biodegradable waste deposited in landfills in Estonia was 451.5 m^3CH_4/t VS. The rearrangement of the composting and depositing of biodegradable waste and substituting it with anaerobic co-digestion according to the research would enable the production of up to 23.1 million m^3 of biomethane annually, which could be converted into 226 thousand MWh of heat and electric energy.

11. UTILISATION OF THE DIGESTATE PRODUCED FROM METHANE FERMENTATION

The National Waste Management Plan for 2014-2020 foresees the utilisation of 10% of municipal waste anaerobic fermentation digestate according to the data from 2011, i.e. 40,800 tonnes of digestate [31]. There is a need to try to increase the amount of digestate from anaerobic fermentation that is used in agriculture.

The amount of produced digestate equates to the volume of biodegradable waste used for digestion. The digestate from methane fermentation is rich in nutrients TP (about 0.4-1.8 kg P/m^3) and TN (about 3.5-4.5 kg N/m^3), which can be used for fertilising cultivated lands. 1 m^3 of compost that is produced in Väätsa landfill contains 3.5 kg TN, 0.41 kg TP and 0.54 kg TK.
There are successful long-term results in using compost from methane fermented activated sludge in sediments from Tallinn wastewater treatment plant in agriculture, greenery and recultivation, and there are experiments with the forestation of abandoned less valuable arable land and cutover peat lands [48, 49].

12. CLARIFICATION OF THE MOST EFFICIENT LANDFILL WASTEWATER TREATMENT METHODS.

From 2007-2014, different technologies for treating landfill wastewater were tested and the operation of the already existing treatment plants in Väätsa, Torma and Uikala were observed.
The study results have been presented as follows:
1.   The equalization of the flow rate and pollutant content of the wastewater in the equalizing tank is necessary before treating the wastewater.
2.   Taking into account the efficiency of the treatment required by legislation, only wastewater from the young landfills provided the expected results after biological treatment. Biological treatment is also possible for pre-treating wastewater, if the following technological means are applied:
-       In treatment plants with activated sludge, the hydraulic retention time for landfill wastewater should be prolonged by twice the time used in the activated sludge treatment process for municipal wastewater;
-       The content of dissolved oxygen in the aeration chamber should be brought up to at least 5-7 mg O_2/l;
-       In winter conditions, it is recommended to maintain the positive temperature of leachate with technical measures. In the equalizing tank, there should be a receiving well from where the leachate, without getting mixed with other wastewater, is pumped into the main treatment plant. Part of the heat obtained from the combustion of biogas can be used for warming up the leachate in winter, in the case of the availability of excess heat;
-       It is essential to remove grease and oil prior to the biological treatment;
-       Lack of incoming wastewater may become a problem for activated sludge plants during the longer drought periods or winters without snowmelt weather. In winter, the tightness of leachate may be over 1.0 t/m^3 due to the lack of wastewater and high content of substances, making treatment with activated sludge more difficult (activated sludge is carried out from the sedimentation tank). It is recommended to direct the effluent water back into the equalizing tank to avoid such problems and a breaking up of the treatment process.
-       Additional measures for treatment should be applied for biological methods that are not sufficient for safeguarding the required level of purification and permitted limit concentrations for pollutants (especially for nitrogen);
-       Biological co-treatment for leachate and municipal wastewater has certain technological and economic advantages. However, in order to avoid the hindering impact from the leachate to the treatment process and to safeguard the quality of treated wastewater, it is essential to ensure that the share of leachate would not exceed 10%.
3.   Reactions between the pollutants and ozone are slow and specific. This is why ozonating landfill wastewater is expensive and not very efficient. Post-ozonating the biologically treated landfill wastewater decreases the overall concentration of pollutants (COD) but the overall efficiency remains low. The colour

and odour are removed, and, if the previous biological treatment has been more efficient, the post-ozonation is also more efficient and the biodegradability of the treated wastewater is improved.

4.   Coagulation with lime, oil-shale ash and other reagents is not efficient. Additional costs are needed to purchase additional reagents and the residue has to be treated separately.

5.   It is possible to use the Fenton process for deep post-treatment following biological treatment in order to decrease the colour, odour and overall content of pollutants or prior to biological treatment to decrease the content of pollution in the wastewater. Apart from this, certain other factors also require attention, such as the conditions of the unstable chemical composition of landfill wastewater, the optimal conditions for treatment, that is, the optimal ratio of COD of the wastewater and reagent doses ($KHT/H_2O_2/Fe^{2+}$) that need to be maintained.

6.   The efficiency of immediate chemical sedimentation for removing pollutants from leachate is low. The efficiency is significantly higher in terms of the chemical treatment of biologically treated leachate.

7.   It was found during the current study that the most suitable purification method is reverse osmosis after the equalisation of the flow rate and pollutant concentration of the wastewater with the help of equalizing tank; this reverse osmosis is conducted in two stages, either by following biological treatment or without it. About 95% of nitrogen is removed from the wastewater in the first stage and 99% in the second stage. The following principles are recommended to be followed in selecting suitable filters for reverse osmosis:
-     The use of a biofilter should be able to remove high COD and nitrogen content from the wastewater. For two-stage RO, DT disk module membranes were used, and it was possible to achieve the permitted limit values for the following pollutants: COD, BOD, suspended solids, TP and TN;
-     Before the application of RO, the content of suspended solids in water needs to be decreased. A sand filter that enables quick flow should be used before DT filters;
-     The temperature of wastewater can have a big influence on the efficiency of the reverse osmosis equipment. Usually, the efficiency increases for about 3% per temperature rise of each $1^{\circ}C$.

8.   In new landfills that are separated from the surrounding environment with geomembrane and where leachate and/or RO concentrate and activated sludge from biological treatment are directed back to the waste deposit for irrigation, waste deposit can be considered to be one of the steps in the treatment process for wastewater. The aim is to bind the pollutants into the deposit and, in case of excess sludge, to perform after treatment of the sludge in the deposit with the help of aerobic and anaerobic processes. As leachate and RO concentrate are toxic, it is recommended to pump the concentrate only into closed waste deposits, where biogas extraction has almost stopped. It should be taken into account that the water absorption capacity of the deposit consisting mainly of plastic and films is low, and wastewater will drip out from the sides of the waste deposit of unsuitable hydraulic loads and will not penetrate the lower waste layers.

13. CONCLUSIONS

The results of the study have been summarised below:

1.   It was found that a large amount of wastewater is conducted into sewerage from composting fields during rainfall and snowmelt, and the flow rate, pollutant concentration and pollution load in this wastewater are not stable. This is why equalizing tanks and treatment plants with high capacity are required.

2.   Constructive and technological recommendations for collecting wastewater into equalizing tanks, channelling into treatment plants and treating in the plant were compiled.

3.   It was found that the biological treatment of landfill wastewater can only be used when the landfills are young and as a pre-treatment for the reverse osmosis process.

4.   On the basis of experiments and the monitoring of practical operations, it was concluded that the most efficient treatment method for landfill wastewater guarantees the necessary level of treatment and consists of two-stage reverse osmosis treatment with DT filters. The other treatment methods that were studied did not provide the purification efficiency that is required by legislation.

5.   The landfill leachate and concentrate from reverse osmosis are toxic and can only be used for irrigating closed depositing fields, where the extraction of biogas is in the final stage.

6.   The low C/N ratio in the remnant landfill leachate refers to the low carbon (C) and excessive nitrogen contents. During the fermentation process, carbon is used first, and if the amount of carbon is insufficient, nitrogen becomes toxic for the methanogenic bacteria. Therefore, the landfill leachate remnant can only be fermented with carbon-rich co-substrate and in small amounts.

7.   The composting of biodegradable waste that is suitable for methane fermentation should be substituted with fermentation.

8.   The amount of biogas collected from the waste deposits of Estonian landfills is too small for starting cogeneration plants for the production of heat and electricity. In the case of methane fermentation, plants for biodegradable waste would be constructed in the landfills; then the amount of biogas produced would

increase significantly and the cogeneration of heat and electricity from the biogas collected and produced would become cost-effective.

9. The digestate from fermenting biodegradable waste – after unifying its contents with legal requirements – can be used in agriculture, greenery and recultivation, including forest plantations.

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Prügilate reovee reoainesisalduse ja erinevate puhastusmeetodite hindamine

Aare Kuusik, Karin Pachel, Argo Kuusik, Enn Loigu

Eestis paiknevates prügilates toimub lisaks jäätmete sorteerimisele ja ladestamisele sageli ka biolagunevate jäätmete kompostimine. Kompostimisväljakutelt kogutav sademevesi ja lumesulamisvesi on enamasti kõrge reoainete kontsentratsiooniga ja suurtes piirides kõikuva vooluhulgaga, mis oluliselt mõjutab prügilareovee reoainete sisaldust ja kontsentratsiooni ning selle puhastamise võimalusi ja efektiivsust. Prügilates tekkiv prügilareovesi koosneb territooriumilt kogutud reostunud sademete (vihma ja lumesulamise) veest, tööliste olmereoveest, konteinerite ja masinate pesuveest ning prügilademetes tekkivast ja lademetest läbi imbuvast veest ehk nõrgveest.

Aastatel 2007 kuni 2014 katsetati erinevaid prügilareovee puhastustehnoloogiaid ja jälgiti mitme prügila reoveepuhasti tööd. Prügilareovee edukaks seadusandlusega seatud piirnormide kohaseks puhastamiseks tuleb enne puhastamist kasutada prügilareovee vooluhulga ja reoainete kontsentratsiooni ning nõrgvee toksilisuse ühtlustamist ühtlustamismahutis ja bioloogilisele puhastusele järgnevat või ilma selleta kaheastmelist ketasmembraanidega pöördosmoosi. Prügilareovee puhastamise teised katsetatud puhastusmeetodid ei andnud vajalikku tulemust.

Väätsa prügila kanalistsioonisüsteem ja prügila reoveepuhasti on projekteeritud ja ümberehitatud tuginedes käesolevale uurimistööle. Kogu kanalisatsioonisüsteem koosneb prügilareovee kogumissüsteemist, ühtlustamismahutist, reovee füüsikalis/keemilisest (pöördosmoos) puhastamisest peale reovee bioloogilist puhastamist aktiivmudapuhastis ja biotiigis ja pöördosmoosi kontsentraadi prügilasse tagasipumpamise süsteemist. Alates 2012 aasta aprilli kuust kuni tänaseni on suublasse juhitava heitvee reoainete sisaldus vastavuses seadusandlusega seatud piirväärtustega.

Prügilareovee puhastamise efektiivsus on suurem ja stabiilsem, kui lõpetada prügilate territooriumil biolagunevate jäätmete komposteerimine ja komposteerimisplatsidelt reostunud sademevee juhtimine prügila reoveepuhastisse. Biolagunevatest jäätmetest on otstarbekam metaankääritamise teel toota biogaasi ja taimetoitaineterikast digestaati. Biolagunevate jäätmete kompostimise ja prügilasse ladestamise ümberkorraldamine võimaldaks anaeroobset kooskääritamist kasutades toota kuni 23,1 miljonit m^3 biometaani aastas, millest saab toota ca 226 tuhat MWh soojus- ja elektrienergiat. Metaankääritamisel tekkiv digestaat sisaldab suurel hulgal taimetoitaineid, mida saab kasutada kõlvikute väetamiseks.

APPENDIX II CURRICULUM VITAE

1. Personal data

First name	AARE
Last name	KUUSIK

Date and place of birth	11/10/1958, Tartu, Estonia
Estonian ID code	35810110217
Nationality	Estonian

2. Contact information

Address	Vainu, Pudisoo, 74626, Harjumaa, Estonia
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3. Education

Educational institution	Graduation year	Education (field of study/degree)
Tallinn University of Technology	1996	Master degree (Diploma CB 000071), speciality: Master of Engineering
Estonian Agricultural Academy	1982	Engineering degree (Diploma 3B no. 857074), speciality: Engineer of hydrotechnics
Kehra Secondary School	1977	Secondary education

Official title Hydraulic Engineer, M.Sc (Eng.)

4. Language competence/skills

Language	Level
Estonian	Native language, fluent
Finnish	Intermediate
Russian	Advanced
German	Intermediate
English	Intermediate

5. Professional experience

Date from – date to	Location	Company	Position	Description
Since March 1996	Pudisoo, Harjumaa, Estonia	VETEPERE Ltd.	Manager	Managing and realisation of water supply, sewerage, wastewater treatment and environmental projects. Environmental impact assessment.
01.09.2011–29.07.2012	Tallinn, Estonia	Tallinn University of Technology, Faculty of Civil Engineering	Engineer	Managing and realisation of water supply, sewerage, wastewater treatment and environmental projects.
01.09.2008–31.08.2011	Tallinn, Estonia	Tallinn University of Technology, Faculty of Civil Engineering, Department of Environmental Engineering	Extraordinary researcher	Managing and realisation of water supply, wastewater treatment, sewerage and environmental projects.
January 1995–April 1997	Tallinn, Estonia	FIXTEC Ltd.	Sales manager	Selling of sewage and water treatment technologies and facilities
January 1993–January 1995	Tallinn, Estonia	Ministry of Environment of Estonia	Chief specialist	Water and wastewater legislation
January 1992–January 1993	Tallinn, Estonia	Estonian Union of Rationalisation and Invention	Consulting engineer	Realisation of water supply, wastewater treatment and sewerage rationalisation and invention problems
September 1984–December 1991	Tallinn, Estonia	State Project Institute "Land Reclamation Project"	Chief engineer	Managing and realisation of water supply, wastewater treatment, sewerage and environmental projects
July 1984–September 1984	Tallinn, Estonia	Water Management Inspection of Estonia	Chief engineer	Inspection of water and wastewater legislation implementation
1982–1984	–	Service in the Soviet Army	–	–
August 1982–October 1982	Tallinn, Estonia	Committee of Land Reclamation and Water Management.	Chief specialist	Water and wastewater legislation

6. Special Courses

16.08-20.08.1993. SWEDEN. Norrkoping. Swedish Water and Wastewater Works Association (VAV). Wastewater treatment for minor urban areas.

27.09-17.12.1993. TEMPUS JEP – 4925 - 93/2 preliminary course. Tallinn Technical University. Open University. Centre for Continuing Education.
01.03-31.05.1994. TEMPUS JEP – 4925/93 - 2 fundamental course. FINLAND. Tampere University of Technology.
12.02-05.03.1993. Denmark. Silkeborg. The Freshwater Centre. Basic course on environmental administration and Special course on surface water and wastewater.
18.05-29.05.1998. C&E Consulting und Engineering GmbH. Chemnitz. Germany. Preventive Protection of Water Resources.

7. Defended theses

Master theses: Kuusik, A. Small wastewater treatment plants in Estonia. 1996. 247 p.

Engineering theses: Kuusik, A. Improving water quality in small artificial lakes in the interests of fish farming. 1982. 91 p.

8. Main areas of scientific work / current research topics

Study and implementation of new LWW (including leachate) and biodegradable waste treatment technologies in Estonia. Decreasing and balancing of LWW and increasing its degree of purification, including everything related to the treatment of leachate and other polluted water collected from the landfill on site or in a domestic WWTP as well as other activities related to the treatment of LWW, sewage sludge and biodegradable waste in landfills. Fermentation of biodegradable waste in landfills. The use of landfill gas and biogas obtained from the fermentation of biodegradable waste. The use of methane fermentation digestate.

9. Membership of professional bodies

1993−... Estonian Water Association

10. Scientific work

Kuusik, Aare, Pachel, K., Kuusik, Argo, Loigu, E. (2016). Assessment of landfill wastewater pollutants and efficiency of different treatment methods. Proceeding of the Estonian Academy of Security Sciences, 65 (4), 1-23.

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11. Excerpt from published books, projects and expertise:

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2. Kuusik, A. Biogenics in wastewater. Tallinn, 1996. 39 p.
3. Kuusik, A. Large and small wastewater treatment plant effluent effect on receiving waters. Tallinn Universiy of Technology. Department of Environmental Engineering. Tallinn, 1999. 32 p.
4. Kuusik, A. Inventory management of sewage sludge in Estonia. Vetepere Ltd. Tallinn, 2001. 62 p.
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9. Kuusik, A. Feasibility study on the utilisation of pig slurry, sewage sludge and biowaste in Saaremaa. Vetepere Ltd. Tallinn, 2004.

10. Kuusik, A. Preliminary project for AS Horizon Kehra paper mill sludge treatment. Vetepere Ltd. Tallinn, 2006.

11. Environmental impact assessment of construction and exploitation Valjala pig slurry methane digestion plant. Vetepere Ltd. Lead expert: Aare Kuusik. Pudisoo, 2004.

12. Environmental impact assessment of construction by Oisu Biogaas OÜ of a plant in Mõisavalla, Väljaotsa village, Türi municipality, for producing biogas by methane digestion of pig and cattle slurry and for the co-generation of heat and power from biogas and, based on the production process, the construction and utilisation of a manure lagoon in Sikevälja lot in Tänassilma village. Vetepere Ltd. Lead expert: Aare Kuusik. Pudisoo, 2008. 12-56.

13. Strategic assessment of the biogas plant planned to be built in Vinni. OÜ Vetepere. Lead expert: Aare Kuusik. Vetepere Ltd. Pudisoo, 2009. 21-32.

14. Kuusik, A. Preliminary project for reconstructing the wastewater treatment of Väätsa landfill. Vetepere Ltd. Pudisoo, 2009. 25 p.

15. Kuusik, A., Malmre, T. Possibilities for production and consumption of electricity and thermal energy in Ambla area by using biogas obtained by anaerobic digestion of agricultural products and waste. Vetepere Ltd. Pudisoo, 2009.

16. Studies on Landfill Leachate and analyses of different treatment methods: Elaboration of a treatment methodology suitable for Estonian conditions. Tallinn Universiy of Technology. Department of Environmental Engineering. Project No 100. Final Report. Team leader: Aare Kuusik. Tallinn, 2010, 9-211.

17. Kuusik, A. Expert analysis of the work project of wastewater treatment plant aftertreatment and necessary infrastructure of Valio Eesti AS Laeva Meierei. Vetepere Ltd. Pudisoo, 2013.

18. Kuusik, A. Planning and reconstruction of a public water supply and sewerage for Ahula, Kaalepi and Seidla villages, stage II. Technological design. Vetepere Ltd. Pudisoo, 2015.

LISA III ELULOOKIRJELDUS

1. Isikuandmed

Eesnimi   **AARE**
Perekonnanimi   **KUUSIK**

Sünniaeg ja -koht   11/10/1958, Tartu, Eesti
Isikukood   35810110217
Kodakondsus   Eesti

2. Kontaktandmed

Aadress   Vainu, Pudisoo, 74626, Harjumaa, Eesti
Telefon   +372 5162476
E-post   aare@vetepere.ee

3. Hariduskäik

Õppeasutus	Lõpetamise aeg	Haridus (eriala/kraad)
Tallinna tehnikaülikool	1996	Ehitus (Diplom CB 000071), tehnikamagister
Eesti põllumajanduse akadeemia	1982	Maaparandus (Diplom 3B nr. 857074), hüdrotehnikainsener
Kehra keskkool	1977	Keskharidus

Ametlik nimetus   Hüdrotehnikainsener, M.Sc (Eng)

4. Keeleoskus

Keel	Tase
eesti keel	Emakeel, kõrgtase
soome keel	kesktase
vene keel	kesktase
saksa keel	kesktase
Inglise keel	kesktase

5. Erialane töö

Töötamisaeg	Asukoht	Tööandja nimetus	Ametikoht	Töö kirjeldus
Alates märtsist 1996	Pudisoo, Harjumaa	VETEPERE OÜ	Juhataja	Veevarustus- ja kanalisatsioonisüsteemide projekteerimine, ekspertiis ja omanikujärelevalve. Keskkonnaspetsialistide ja inseneride koolitamine. Keskkonnamõju hindamine.
01.09.2011–29.07.2012	Tallinn	Tallinna Tehnikaülikooli keskkonnatehnika instituut	Insener	Veevarustus- ja kanalisatsioonisüsteemide projekteerimine. Õppetöö.
01.09.2008–31.08.2011	Tallinn	Tallinna Tehnikaülikooli keskkonnatehnika instituut	Erakorraline teadur	Veevarustus- ja kanalisatsioonisüsteemide projekteerimine. Õppetöö.
Jaanuar 1995–aprill 1997	Tallinn	FIXTEC AS	Müügijuht	Vee- ja reoveepuhastite müük
Jaanuar 1993–jaanuar 1995	Tallinn	Eesti Keskkonna-ministeeriumi veeosakond	Peaspetsialist	Vee- ja reoveealased õigusaktid
Jaanuar 1992–jaanuar 1993	Tallinn	Eesti Põllumajanduse leiutajate ja ratsionaliseerijate liit	Insener-konsultant	Veevarustus- ja kanalisat-siooonisüsteemide alase ratsionaliseerimise ja leiutamisega seonduv
September 1984–detsember 1991	Tallinn	RPUI Eesti Maaparandus-projekt	Juhtiv projektiinsener	Veevarustus- ja kanalisatsioonisüsteemide ning veekeskkonnaga seotud projektid.
Juuli 1984–september 1984	Tallinn	Eesti Veemajandus-inspektsioon	Vaneminsener	Vee- ja reoveealaste õigusaktide nõuete täitmise kontrollimine
1982–1984	-	Teenistus nõukogude armees	-	-
August 1982–oktoober 1982	Tallinn	Veemajandus- ja maaparandus-komitee	Peaspetsialist	Vee- ja reoveealased õigusaktid

6. Täiendusõpingud

16.08-20.08.1993. SWEDEN. Norrkoping. Swedish Water and Wastewater Works Association (VAV). Wastewater treatment for minor urban areas.
27.09-17.12.1993. TEMPUS JEP – 4925 - 93/2 preliminary course. Tallinn Technical University. Open University. Centre for Continuing Education.
01.03-31.05.1994. TEMPUS JEP – 4925/93 - 2 fundamental course. FINLAND. Tampere University of Technology.
12.02-05.03.1993. Denmark. Silkeborg. The Freshwater Centre. Basic course on environmental administration and Special course on surface water and wastewater.

18.05-29.05.1998. C&E Consulting und Engineering GmbH. Chemnitz. Germany. Preventive Protection of Water Resources.

7. Kaitstud lõputööd

Inseneritöö: Väikeste tehisjärvede vee kvaliteedi parandamine kalakasvatuse huvides, 1982. 91 lk

Magistritöö: Reoveeväikepuhastid Eestis, 1996. 247 lk.

8. Teadustöö põhisuunad

Uute prügilareovee (sh nõrgvee) ja biolagunevate jäätmete käitlemistehnoloogiate uurimine ja juurutamine Eestis. Prügilates tekkiva prügilareovee vähendamine, ühtlustamine ja puhastusefektiivsuse suurendamine, st nõrgvee ja muu prügilas kogutava reostunud vee kohapeal või asula reoveepuhastis puhastamisega seonduv ning muud tegevused, mis on seotud prügilareovee, reoveesette ja biolagunevate jäätmete käitlemisega prügilates. Biolagunevate jäätmete kääritamine prügilates. Prügilagaasi ja biolagunevate jäätmete kääritamisel saadava biogaasi kasutamine. Metaankääritamise digestaadi kasutamine.

9. Osalemine eralastes organisatsioonides

1993−... Eesti Veeühing

10. Teadusartiklid

Kuusik, Aare, Pachel, K., Kuusik, Argo, Loigu, E. (2016). Assessment of landfill wastewater pollutants and efficiency of different treatment methods. Proceeding of the Estonian Academy of Security Sciences, 65 (4), 1-23.

Kuusik, Aare, Pachel, K., Kuusik, Argo, Loigu, E., Tang, W. Z (2014). Reverse osmosis and nanofiltration of biologically treated leachate. Environmental Technology, 1−11, 10.1080/09593330.2014.908241.

Kuusik, Aare, Pachel, K., Kuusik, Argo, Loigu, E. (2014). Landfill runoff water and landfill leachate discharge and treatment. In: 9th International Conference Environmental Engineering: Water Engineering: Selected Papers (1−6). Vilnius, Lithuania: VGTU Press "Technika".

Kuusik, Argo, Pachel, K., Kuusik, Aare, Loigu, E. (2014). Anaerobic co-digestion of sewage sludge with fish farming waste. In: 9th International Conference on Environmental Engineering: Water Engineering (1−8). Vilnius, Lithuania: VGTU Press "Technika".

Kuusik, Argo, Kuusik, Aare, Pachel, K., Loigu, E., Sokk, O. (2013). Generalised Integration of Solid Waste Treatment Practices to Enhance Methane Productivity, Generate Suspension Fertiliser and Upgrade Biogas. European Scientific Journal, 9 (36), 14−30.

Kuusik, Argo, Kuusik, Aare, Loigu, E., Sokk, O., Pachel, K. (2013). Selection of Most Promising Substrates for Biogas Production. International Journal of Energy and Environment, 7 (3), 115−124.

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Kuusik, Aare, Sokk, O. (2005). Sewage sludge management and its possible trends in Estonia. *In: Proceedings of the Water Environment Federation. Technology 2005. 2nd Joint Specialty Conference for Sustainable Management of Water Quality Systems for the 21st Century: Working to Protect Public Health. San Francisco, California, USA. 28-31 August 2005.* San Francisco, California, USA. Publisher: Water Environment Federation, 962−967.

Kuusik, Aare, Sokk, O. (2004). Sewage sludge management and its trends in Estonia. *In: EWA Conference. Nutrient Management European Experiences and Perspectives. EWA Conference. Amsterdam. 28-29 September 2004.* Amsterdam. EWA European Water Association, 131−138.

11. Väljavõte publikatsioonidest, ekspertiisidest ja projektidest

1. Kuusik, A. Reoveeväikepuhastid Eestis. Tallinn, 1995. 247 lk.
2. Kuusik, A. Biogeenid reovees. Tallinn, 1996. 39 lk.
3. Kuusik, A. Suurte ja väikeste reoveepuhastite heitvee mõju suublale. Tallinna Tehnikaülikool. Keskkonnatehnika Instituut. Tallinn, 1999. 32 lk.
4. Kuusik, A. Reoveesette käitluse inventuur Eestis. OÜ Vetepere. Tallinn, 2001. 62 lk.
5. Alasi, K., Heinsaar, Ü., Kriipsalu, M., Kuusik, A., Metsur, M. Omaveevärk ja omakanalisatsioon. Ehitame kirjastus. Tallinn, 2001. 141 lk.
6. Kuusik, A. Reoveesette käitlemise, kasutamise ja kahjutustamise parima võimaliku tehnika väljaselgitamine. OÜ Vetepere. Tallinn, 2002. 65 lk.
7. Kuusik, A. Eksperdi arvamus reovee väikepuhastites tekkiva sette käitlemisvõimaluste kohta. OÜ Vetepere. Tallinn, 2002.
8. Kuusik, A. Tallinna reoveepuhastusjaamas stabiliseeritud ja kuivatatud reoveesette ja sellest valmistatud kasvumulla haljastuses, põllumajanduses ja rekultiveerimisel või muudes valdkondades kasutamise võimaluste uuring. OÜ Vetepere. Tallinn, 2003.
9. Kuusik, A. Tasuvusuuring sealäga, reoveesette ja biojäätmete utiliseerimiseks Saaremaal. OÜ Vetepere. Tallinn, 2004.
10. Kuusik, A. AS Horizon Kehra paberitööstuse sette käitlemise eelprojekt. OÜ Vetepere. Tallinn, 2006.
11. Valjala sealäga metaankääritusjaama ehitamise ja kasutamise keskkonnamõju hindamine.. OÜ Vetepere. Juhtekspert: Aare Kuusik. Pudisoo, 2004.

12. Oisu Biogaas OÜ poolt Türi valda Väljaotsa külla Mõisavalla maaüksusele sea- ja veiselägast metaankääritamise teel biogaasi ning sellest elektri- ja soojusenergia tootmisjaama ning tootmisprotsessist tulenevalt Tänassilma külla Sikevälja maaüksusele sõnnikulaguuni ehitamise ja kasutamise keskkonnamõju hindamine. OÜ Vetepere. Juhtekspert: Aare Kuusik. Pudisoo, 2008.

13. Vinni asulasse planeeritava biogaasitehase keskkonnamõju strateegiline hindamine. OÜ Vetepere. Juhtekspert: Aare Kuusik. Pudisoo, 2009.

14. Kuusik, A. Väätsa prügila reoveepuhastusjaama rekonstrueerimise eelprojekt. Seletuskiri ja joonised. Töö nr. OÜ Vetepere. Pudisoo, 2009.

15. Kuusik, A., Malmre, T. Põllumajanduslikest toodetest ja jääkproduktidest anaeroobse kääritamise teel saadud biogaasist elektri- ja soojusenergia tootmise ja tarbimise võimalused Ambla piirkonnas. OÜ Vetepere. Pudisoo, 2009.

16. Prügilavee uuringud ja erinevate puhastustehnoloogiate analüüs: Eesti oludesse sobiva puhastustehnoloogia väljatöötamine. Projekt nr 100. Lõpparuanne. Tallinna Tehnikaülikool. Keskkonnatehnika Instituut. Töögrupi juht: Aare Kuusik. Tallinn, 2010.

17. Kuusik, A. Valio Eesti AS Laeva Meierei reoveepuhastusjaama järelpuhastuse ja vajalike taristute tööprojekti ekspertiis. OÜ Vetepere. Pudisoo, 2013. a.

18. Kuusik, A. Ahula, Kaalepi ja Seidla asulate ühisveevärgi ja -kanalisatsiooni projekteerimise ja rekonstrueerimise II etapp. Tehnoloogiline projekt. OÜ Vetepere. Pudisoo, 2015. a.

DISSERTATIONS DEFENDED AT
TALLINN UNIVERSITY OF TECHNOLOGY ON
CIVIL ENGINEERING

1. **Heino Mölder**. Cycle of Investigations to Improve the Efficiency and Reliability of Activated Sludge Process in Sewage Treatment Plants. 1992.

2. **Stellian Grabko**. Structure and Properties of Oil-Shale Portland Cement Concrete. 1993.

3. **Kent Arvidsson**. Analysis of Interacting Systems of Shear Walls, Coupled Shear Walls and Frames in Multi-Storey Buildings. 1996.

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