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# Assessment of landfill wastewater pollutants and efficiency of different treatment methods

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**Abstract.** In Estonian landfills, in addition to waste sorting and depositing, most biodegradable waste is composted. Stormwater and snowmelt samples collected from 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 at Väätsa was redesigned and reconstructed. The treatment system consists of a landfill wastewater collection system, an equalizing tank, physical/chemical (i.e. reverse osmosis) treatment after biological activated sludge treatment and oxidation in pond, and stabilization of the pumping and distribution systems for concentrate discharge from reverse osmosis 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 an efficient and stabilized 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 fertilizing certain cultivated areas.

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

**Concepts:**
*Landfill wastewater* – water that consists of leachate, i.e. the 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 a 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 minimizing 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, an effective management of landfill wastewater is one of the major challenges.

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Landfill wastewater is any kind of water that is collected from the territory of a landfill, 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 in 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, stabilized methanogenic, and final aerobic phases. The quality and quantity of landfill wastewater vary throughout the lifetime of the landfill site. Municipal landfill wastewater is characterized 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.

As the amount of comparable data is limited, 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 minimization 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 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 in 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. In the waste deposit recirculation of the leachate as well as equalization of the flow rate and pollutant concentration of the landfill wastewater take place. The equalizing tank is used for minimizing the flow rate to the treatment plant and equalizing the top pollutant loads and concentrations.

Various biological, physical, and chemical methods are used for the treatment of landfill wastewater and leachate. Biological cotreatment of leachate and domestic wastewater is widely used because of its various technological and economic advantages (in Estonia Tallinn and Pärnu municipal wastewaters 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 required quality of the treated water, it is essential that the share of leachate in the mixture is not above 5–10% [8,9].

Numerous studies and handbooks describe 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 (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, coagulation, and flocculation).
- Aerobic biological treatment processes: suspended growth systems (aerated lagoons, activated sludge (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 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 a 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 unmetabolized by microorganisms. The aim of pre-ozonation is to improve the biodegradability of the treated wastewater, whereas the purpose of post-ozonation is advanced treatment of the wastewater [8,17,18]. After ozonation, the biodegradability of the processed water is higher, indicating the necessity of additional bio-treatment. 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. It consists of four stages: oxidation, neutralizing, 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 [15,21]. The advantages of this process include the reduced energy requirement for creating radicals, availability of cheaper and non-toxic reagents, and the process is not limited by mass exchange (homogeneous catalysis) [21]. Some of the shortcomings of this process are 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 the COD and a high variation of the pH. 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 case of a high salt content, RO is used for an additional treatment of the biologically pretreated landfill wastewater. It is also used as an independent method for treating leachate [4–6,10,23,24]. Its low operational costs and ability to remove the organic contaminants and 95–99% of inorganic salts with minimal chemical requirements makes RO an attractive technology for many applications [11].

In most cases, the landfill wastewater is treated applying a combination of different treatment methods. At Torma, Estonia, the landfill wastewater is treated using a buried sand filtration unit after the sedimentation pond treatment. Since 2010 processing in the equalizing tank and post-treatment with sand and ceramzit filters have been followed by mechanical, biological (AS), and chemical treatment. A stabilization pond and biochemical treatment with AS have been used for the Väätsa landfill wastewater, and since 2013 biological treatment (AS and oxidation pond) and RO have been followed by the equalizing tank. 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 mitigating the environmental impacts.

## 2. TREATMENT OF LANDFILL RUNOFF WATER AT VÄÄTSA AND TORMA

The first municipal waste depositing field in the Väätsa landfill of one hectare 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 to 7 m. In 2008, the third field of 2.8 ha for municipal waste was put into operation. Altogether, there were four depositing fields with a total area 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 at Väätsa was completed in 2002. This biochemical plant consisted of an AS container (aerotank 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 2000 $m^3$; it was regulated up to 1250 $m^3$. The AS plant was dimensioned for the flow rate of 70 $m^3$/d, including leachate from the 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 AS 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

**Table 1.** Characteristics of the Väätsa landfill wastewater and water from different treatment stages, May 2009 [7]

| Parameter | Väätsa landfill wastewater | After treatment with activated sludge | After stabilization pond treatment | Limit value in effluent [25] |
|---|---|---|---|---|
| $BOD_7$, $mgO_2/L$ | 250 | 110 | 22 | 25 |
| COD, mgO/L | 4000 | 3000 | 800 | 125 |
| Total P, mgP/L | 9.0 | 4.5 | 2.6 | 2.0 |
| Total N, mgN/L | 474 | 414 | 210 | 75 |
| pH | 8.5 | 8.5 | 8.2 | 6–9 |
| Suspended solids, mg/L | 260 | 108 | 40 | 35 |

was carried to the depositing field. Water from the AS plant was treated in the aerated oxidation pond.

In addition to the studies in 2007, the efficiency of the operation of the Väätsa landfill AS 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 varied in the limits from 3100 to 4800 mgO/L and the biological oxygen demand ($BOD_7$) was from 45 to 300 $mgO_2/L$. At the end of the month, the flow rate decreased and the values of COD and $BOD_7$ rose to 10500 mgO/L and 1550 $mgO_2/L$, respectively. In April and May 2009, the variation in the flow rate was greater due to the springtime snowmelt. The fluctuations of COD and $BOD_7$ in wastewater were smaller than in November; the COD values of the wastewater varied from 1000 to 3500 mgO/L and of $BOD_7$, from 50 to 350 $mgO_2/L$. The biodegradability of the wastewater (BOD/COD) was very low, 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].

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 AS. This is also reflected by the content of total nitrogen after the AS process in Table 1. 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 in 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 minimized. 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 RO 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 (Fig. 1), was designed and built during 2011–2012.

Since April 2012, when the new treatment system for landfill wastewater began operation, the effluent from the wastewater treatment plant was in compliance with the requirements in the water permit (Fig. 2 and Table 2). The average removal efficiency recorded in the time period from 2013 to 2015 was over 99% of $BOD_7$, COD, total nitrogen (TN), and total phosphorus (TP), and over 90% of suspended solids (SS) [23,27].

The first municipal waste depositing field in the Torma landfill 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 collected leachate and stormwater 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 a geomembrane to ensure water tightness. It was dimensioned so that the load would not exceed 90 L of 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 8000 $m^3$ (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

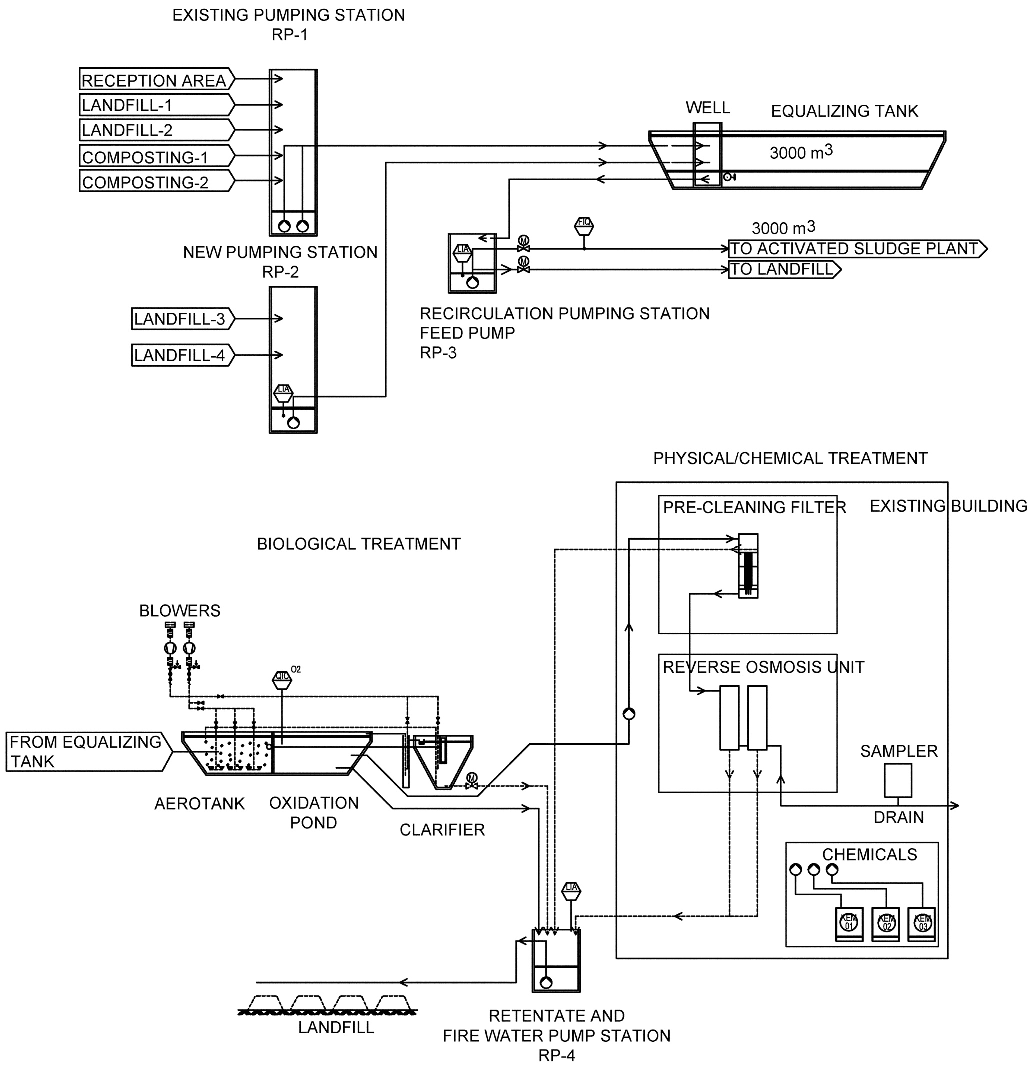


**Fig. 1.** Preliminary design of the technological scheme of the Väätsa landfill wastewater treatment plant [26].

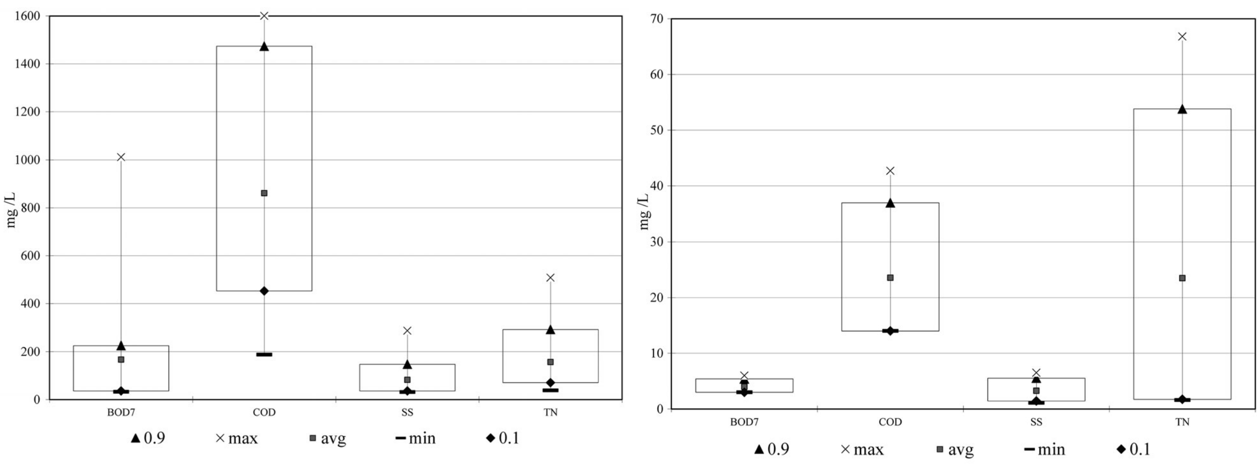


**Fig. 2.** Pollutant content in the wastewater in the Väätsa landfill during the period from 2001 until March 2012 (left) and from April 2012 to 2014 (right). SS – suspended solids.

**Table 2.** Characteristics of the Väätsa landfill wastewater effluent, 2013–2015 [27]

| Pollutant | 2013 | 2014 | 2015 | Limit value [25] |
|---|---|---|---|---|
| $BOD_7$, $mgO_2/L$ | 3 | 3 | 3 | 25 |
| COD, mgO/L | 14 | 14 | 14 | 125 |
| Suspended solids, mg/L | 6.5 | 2.25 | 2.5 | 35 |
| Total N, mgN/L | 1.5 | 2.1 | 1.6 | 75 |
| Total P, 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 |

was directed into the receiving water body. The pollutant content and treatment efficiency of the wastewater are depicted in Figs 3 and 4.

The efficiency of the buried sand filtration unit was found to be low and, after several years of use, it became even 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.

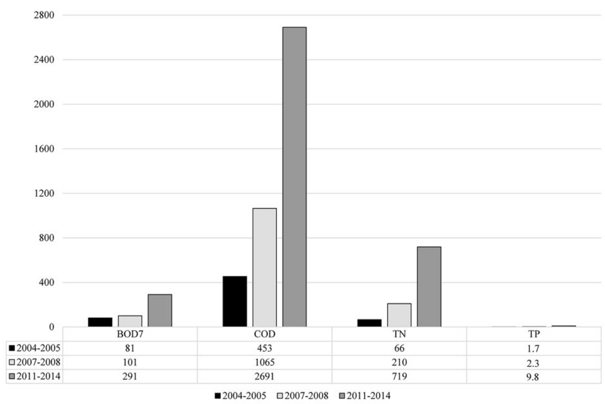


**Fig. 3.** Average pollutant content in the Torma landfill wastewater entering the treatment plant during the periods of 2004–2005, 2007–2008, and 2011–2014. TN – total nitrogen, TP – total phosphorus.

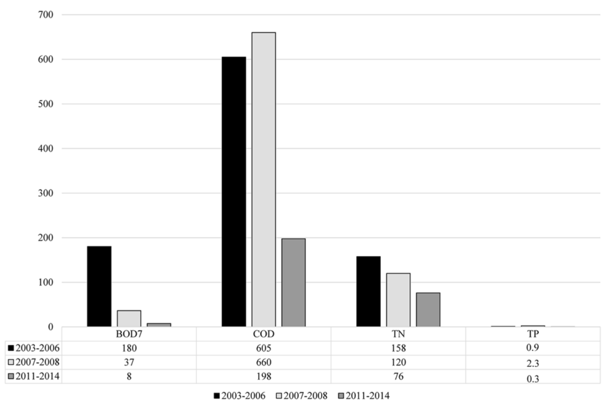


**Fig. 4.** The average pollutant content in the effluent from the Torma wastewater treatment plant falling into the final recipient during the periods of 2003–2006, 2007–2008, and 2011–2014. TN – total nitrogen, TP – total phosphorus.

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 is treated mechanically, biologically, and chemically (Fig. 5).

The treated wastewater is directed to the network of forest oxidation ponds and subsequently into the Mustvee River. To equalize the flow rate and pollution load before treatment, the existing about 1700 $m^3$ equalizing tank with the interim well and a mixer was used. The equalizing tank is 50% larger than the size found by the integral graph on the basis of annual flow rate values. 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 (Fig. 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 for TN 60%. The approximate average ($Q_{avg\ d}$) and maximum capacities ($Q_{max\ h}$) of the plant are represented as follows: $Q_{avg\ d}$ = 100 $m^3/d$ and $Q_{max\ h}$ = 5 $m^3/h$ [28].

After the new treatment system was constructed the indicators of the efficiency of the operation of the wastewater treatment plant improved for pH, SS, $BOD_7$, 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 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 to irrigate the waste and, in the last couple of years, only the polluted stormwater collected from the territory was conducted into the treatment plant. In the first quarter of 2013, no leachate was generated by stormwater in the landfill and no effluent was directed into the recipient. Thereafter, only the contaminated stormwater was collected from the territory

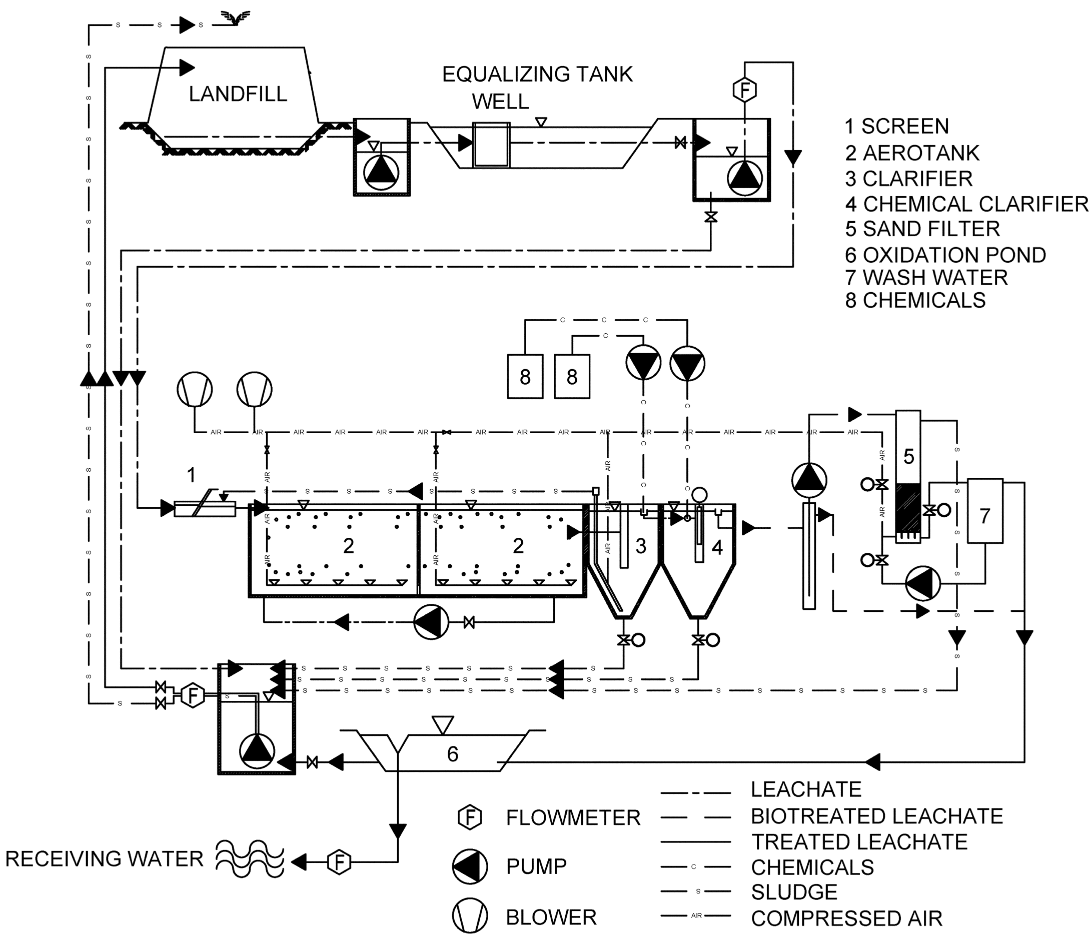


**Fig. 5.** Technological scheme of the wastewater treatment plant in the Torma landfill [29].

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

| Parameter | Entering into treatment plant | After biological treatment | After chemical treatment | Effluent | Limit value in effluent [25] |
|---|---|---|---|---|---|
| pH | 8 | 7.8 | 5.5 | 6.7 | – |
| Suspended solids, mg/L | 170 | 210 | 46 | 15 | 35 |
| $BOD_7$, $mgO_2/L$ | 57 | 40 | 4.1 | <3 | 25 |
| COD, $mgO_2/L$ | 1069 | 581 | 175 | 141 | 125 |
| Total N, mgN/L | 250 | 270 | 240 | 230 | 75 |
| Total P, mgP/L | 2.3 | 2.0 | <0.02 | <0.02 | 2 |

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: $BOD_7$ 96.4%, SS 94%, COD 93.1%, TN 92.2%, and TP 96.9% [30].

## 3. TREATMENT OF BIODEGRADABLE WASTE IN LANDFILLS

The biodegradable waste in Estonia is made up of the biodegradable portion of the 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 7970 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 these materials were composted in compost fields located at the landfills of wastewater treatment plants and in separately located composting fields. Part 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 utilized for fertilizing fields. In comparison with the prevailing situation where the main activities include the composting and depositing of waste into landfills, the impact on the environment seems to be significantly smaller.

The Landfill Directive 999/31/EC states that by 16 July 2016 the amount of biodegradable waste that is deposited in landfills should decrease by 35 mass percent compared to the total amount of deposited municipal household waste in 1995 [1]. The Estonian Waste Act stipulates 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 vital 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 [33].

The average yield of biogas obtained by methane fermentation from the biodegradable waste collected in biocontainers was 375 $m^3$/t organic dry matter (ODM), 425 $m^3$/t ODM from degradable food waste and commercial waste, and 322 to 372 $m^3$ CH4/t from wastewater sludge (residual AS) [34].

In 2010, the production of biogas in Estonia was 13.13 million $m^3$, and most of it (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) production has been estimated altogether at 22 million $m^3$. Biodegradable waste from food manufacture could yield 9 million $m^3$, biowaste 2 million $m^3$, wastewater sludge 3 million $m^3$, and biowaste from industry 8 million $m^3$. This is supplemented with 9 million $m^3$ of landfill gas [35].

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

The digestate 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 fertilizing arable land. The volumes of the digestate are more or less equal to the volume of the fermented biodegradable waste [33]. Compost made of fermented residual AS in the Tallinn wastewater treatment plant is used for fertilizing fields, green areas, and recultivated land [36].

The yield of biogas from the methane fermentation of animal waste is high, but further treatment and utilization 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 fertilizer on 7120 ha of agricultural land in place of mineral fertilizers [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 utilized as a waste fuel in cement manufacturing, and 8.5% will be deposited in landfills [31].

The present study takes into account the rapid changes in waste management (collection of sorted waste, increasing the significance of the sorting and incineration of municipal waste, decrease in depositing municipal waste, and increase in the role of composting) and the impact of the changes on the volume and pollutant content of landfill wastewater. Therefore, one of our goals was to find possibilities of decreasing and equalizing the flow rate, pollutant content, and toxicity of the landfill wastewater. Another direction of the research involved investigation of the possibilities for methane fermentation of the biodegradable waste deposited and composted in landfills, assessment of the yields of biogas generated in fermentation, assessment of the possibilities for utilizing biogas and digestate obtained by the fermentation of 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. In the case of heavy rainfall and snowmelt, we have to deal with very large hydraulic and pollutant shock loads, but 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. Depending on 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 reach the sewerage (or network).

At the 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 a 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 depend on the weather conditions, construction of sewerage, waste sorting technologies, storage 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 of the 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 the Väätsa and Jõelähtme landfills.

The stormwater samples were taken from the stormwater controlling wells in the composting fields in the Jõelähtme landfill, the stormwater flow rate $Q$ was 51 $m^3$/d, and the main filling material in the biodegradable waste composting was peat. In the Väätsa landfill, the stormwater flow rate $Q$ was 123 $m^3$/d, and the filling material in the wastewater sludge composting was chopped straw (dehydrated, thickened, and mixed with peat).

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

| Parameter | Jõelähtme | Väätsa |
|---|---|---|
| pH | 5.5 | 5.6 |
| Conductivity, μS/cm | 4400 | 4870 |
| Suspended solids, mg/L | 900 | 1130 |
| $BOD_7$, $mgO_2$/L | 3960 | 1875 |
| COD, mgO/L | 7530 | 9300 |
| Total organic C, mgC/L | 2100 | 2580 |
| $NH_4$, mgN/L | 251 | 63.5 |
| Total N, mgN/L | 700 | 179 |
| Total P, mgP/L | 42.8 | 40.1 |
| $HCO_3$, mg/L | 1000 | 2470 |
| $SO_4$, mg/L | 89 | 144 |
| Cl, mg/L | 422 | 106 |
| Monobasic phenols, mg/L | 209 | 617 |
| Dibasic phenols, mg/L | 43.5 | 71.6 |
| Hydrocarbon, mg/L | <20 | 300 |
| $Fe^{2+}$, mg/L | 3.9 | 5.5 |
| $Fe^{3+}$, mg/L | 2.9 | 6.6 |
| Hg, μg/L | <0.05 | 0.1 |
| Ag, mg/L | <0.01 | 0.01 |
| Cd, mg/L | <0.02 | <0.02 |
| Cr, mg/L | <0.02 | 0.029 |
| Mg, mg/L | 23 | 109 |
| Mn, mg/L | 0.752 | 9.86 |
| Na, mg/L | 300 | 57.5 |
| Ni, mg/L | <0.02 | 0.047 |
| Pb, mg/L | <0.04 | <0.04 |
| Zn, mg/L | <0.1 | <0.1 |
| Cu, mg/L | 0.024 | 0.027 |

The concentration of pollutants in the landfill wastewater showed a significant increase during the rainfalls. These concentrations were dependent on the intensity of precipitation and the composition and amount of the deposited biodegradable waste on the watertight fields: 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 a low concentration of TP. The levels of COD and $BOD_7$ of the leachate are high. 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 (up to 6–8 mg/L). Iron is responsible for corrosion, which may continue for up to 5–6 years leading to a blockage of the drainage.

The landfill leachate is toxic, hindering biological purification. The studies conducted in 2007 revealed a significant variation of the $BOD_7$/COD ratio in the leachate samples. 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 the Väätsa landfill leachate. This was caused by the presence of humic and fulvic acids, tannins, lignin, hazardous organic chemicals, pesticides, and herbicides in the leachate. The low $BOD_7$/COD ratio decreased the biodegradability of the wastewater.

In all the investigated landfills, the biological purification ratio of BOD : N : P (100 : 5 : 1) was out of balance. For example, in 2007, the average ratio $BOD_7$ : TN : TP in the Väätsa landfill wastewater was 46.1 : 13.5 : 1 and in the leachate it was recorded as 115 : 64.8 : 1. In the 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 $^{o}$C and 4 $^{o}$C, which significantly hindered the biological purification process. The treatment capacity of the RO equipment increased by 3% for each degree of temperature rise.

### 4.1. Results of the in vitro experiments on the possibilities of landfill wastewater treatment

In 2007–2013, experiments on the percolation and RO of landfill wastewater and on biological filter technology were conducted in the Department of Environmental Engineering and the other experiments in the Department of Chemistry of Tallinn University of Technology (TUT). The operation of the existing landfills of the wastewater treatment plants at Väätsa, Torma, Uikala, and Jõelähtme was monitored.

Aerobic biological oxidation (treatment with AS and a biological filter submerged by means of light gravel filling) 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 min, the $BOD_7$ increased by 5% and later began to fall. The reaction of the pollutants with 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 the overall efficiency remained low. The colour and odour were removed but the biodegradability of the wastewater did not change.

The coagulation process produced a decrease in the COD of wastewater by 23% and post-ozonation by a further 11%. The post-ozonation of the coagulated wastewater is not cost-effective: additional expenditures are required to purchase reagents and treat the residual sediments.

The post-ozonation of the wastewater that had been treated biologically in the AS plant decreased the COD by 13%. The efficiency of this process was also found to be low. The post-ozonation of the effluent conducted from the treatment plant into the recipient increased the BOD/COD ratio from 0.02 to 0.11 and COD decreased by 50%. The efficiency of that process was sufficient.

The post-ozonation of the wastewater that had been biologically treated with laboratory equipment increased the biodegradability (BOD/COD ratio) from 0.006 to 0.056. The efficiency of post-ozonation was recorded as 24.4% for the 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 the wastewater 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 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.

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

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

In the nanofiltration (NF) and RO (spiral filters) experiments the ULTRA-FLO PTE, UF-NF 200 equipment was used. The RO process reduced COD and $BOD_7$ by 97% and 60%, respectively. The NF process reduced the COD, $BOD_7$, and TN by 98%, 41%, and 68% of biologically treated wastewater, respectively. However, RO was ineffective in removing TN. The NF was more efficient in removing TP and TN than RO. Although the COD, $BOD_7$, 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 the filter DT 29-09 after the equalizing tank in the Uikala landfill showed that the treatment plant for wastewater operated with high efficiency and was 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 and TP.

### 4.2. Decreasing pollutant content and flow rate of the landfill wastewater for the purpose of purification

In the new landfills in Estonia, a large part of the territory is covered by composting fields with an impermeable cover. For example, the area of the Väätsa landfill is about 1.6 ha and of the Uikala landfill about 3.4 ha. The stormwater collected from the composting fields increases periodically and significantly the flow rate, pollutant content, and pollution load of the landfill wastewater, which in turn, have a great 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. 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 homogeneous mass of paper and cardboard, which 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 the treatment of the leachate more efficient.

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

### 4.3. Equalization of pollutant content and flow rate in the wastewater to be treated

The hindering factors for the aerobic biological treatment of landfill wastewater (e.g. aerated lagoons, AS, sequencing batch reactors, biofilm reactors) and physical treatment processes (e.g. RO) are the large inequalities of the flow rate and pollutant content in the wastewater, low temperature in winter (from 1 to 4 $^{o}$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 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 a 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 case of heavy rainfall (years with precipitation of up to 800 mm), the amount of rain falling on one hectare is up to 8000 $m^3$. The necessary volume of the equalizing tank and the fluctuation range of its water level are 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 the whole year round. If necessary (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.

### 4.4. Measurement of the 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 AS was determined. 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 (e.g., it was up to 974 mgN/L in the Jõelähtme landfill, up to 729 mgN/L in the Uikala landfill, and up to 332 mgN/L in the Väätsa landfill). It has been found to increase during summer due to the high pH and temperature. Of the total volume of nitrogen 90% 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 the 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. [33]. The study on the remnants revealed that the pH level and concentrations of diluted heavy metals were within the norms. The C/N ratio in the remnant was 4, which is considered to be too low. The low C/N ratio refers to the low carbon content and excessive nitrogen content. During the treatment process, carbon is first used in the fermentation; if its amount is insufficient, nitrogen will become toxic for methane bacteria [33,37]. The normal C/N ratio for biogas production is between 10 and 40 [38].

In the Väätsa landfill, the concentrate from RO is pumped into the waste deposit. A series of experiments with methane fermentation was carried out with the aim of determining the toxicity of the concentrate produced in RO, 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 RO 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 RO is depicted in Table 5, and the biomethane potential (BMP) batch experiments are depicted in Table 6.

**Table 5.** Parameters of the reverse osmosis (RO) treatment concentrate of the Väätsa landfill wastewater, October 2014

| Parameter | Level after RO |
|---|---|
| Total solids (TS) in RO treatment concentrate | 3.7% |
| Volatile solids from total solids | 43.1% of TS |
| pH | 6.9 |
| Total N | 1.2 kg/m$^3$ TS |
| $NH_4^-N$ | 0.94 kg/m$^3$ TS |
| Total P | 0.02 kg/m$^3$ TS |
| Total K | 0.47 kg/m$^3$ TS |
| Crude protein | 10.54% of TS |
| Crude fat | 0.02% of TS |
| Carbon | 5.07% of TS |
| Nitrogen | 1.27% of TS |
| Hydrogen | 1.37% of TS |
| Sulphur | 0.43% of TS |
| Zinc | 0.87 mg/kg TS |
| Copper | 0.13 mg/kg TS |
| Mercury | Not found |
| Cadmium | Not found |
| Chromium | 3.30 mg/kg TS |
| Nickel | 0.64 mg/kg TS |
| Lead | <0.01 mg/kg TS |

**Table 6.** Yield of biogas in biomethane potential tests with reverse osmosis concentrate from the Väätsa landfill (VÄ) (Figs 6–9)

| | Average yield, m$^3$ $CH_4$/tonne volatile substances | Average yield, m$^3$ $CH_4$/tonne |
|---|---|---|
| Sewage sludge 100% | 299 | 8.0 |
| Sewage sludge 90%+VÄ 10% | 131 | 3.3 |
| Sewage sludge 75%+VÄ 25% | 179 | 4.1 |
| Sewage sludge 50%+VÄ 50% | 29 | 0.6 |
| Sewage sludge 25%+VÄ 75% | −124* | −2.0* |
| VÄ 100% | −174* | −2.2* |
| VÄ 100% | −51* | −0.5* |
| Distilled water 20%+VÄ 80% | −15* | −0.1* |
| Distilled water 40%+VÄ 60% | −85* | −0.5* |
| Distilled water 60%+VÄ 40% | −144* | −0.5* |
| Distilled water 80%+VÄ 20% | −414* | −0.8* |
| Distilled water 90%+VÄ 10% | 532 | 0.5 |

* The substrate did not give higher results than the inoculum.

At first the BMP test results on the RO concentrate were promising (Figs 6–9). However, after the removal of the inoculum, the productivity became negative (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.

The additions of the RO discharging concentrate have a negative effect on the anaerobic digestion of the sewage sludge. The decline in the methane yield might be caused by the deterioration of the methanogenic bacterial activity following the treatment of the 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 the 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, intensifying the distribution of nutrients and microorganisms as well as biogas generation.

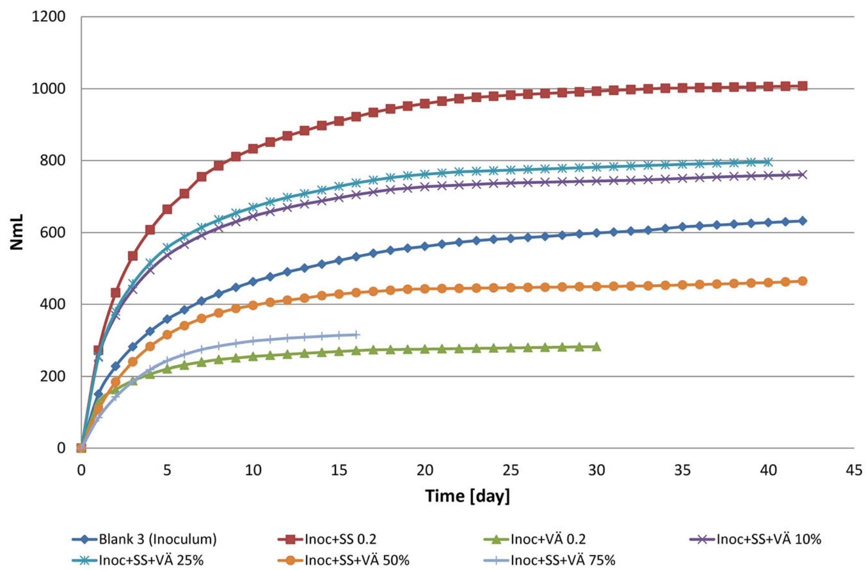


**Fig. 6.** Accumulated gas volume in the first series of tests of biomethane potential. Inoc – inoculum, VÄ – Väätsa landfill leachate, SS – sewage sludge.

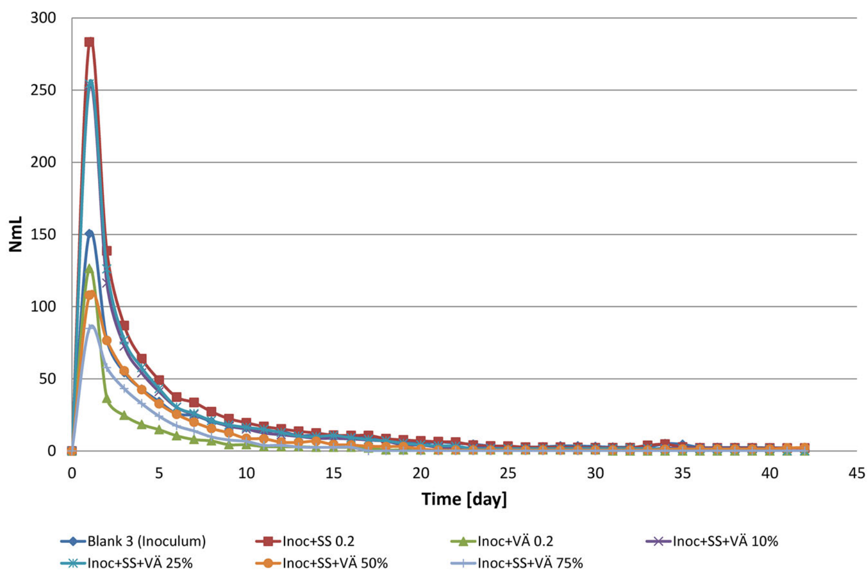


**Fig. 7.** Biomethane flow rate in the first biomethane potential test series. For abbreviations see caption of Fig. 6.

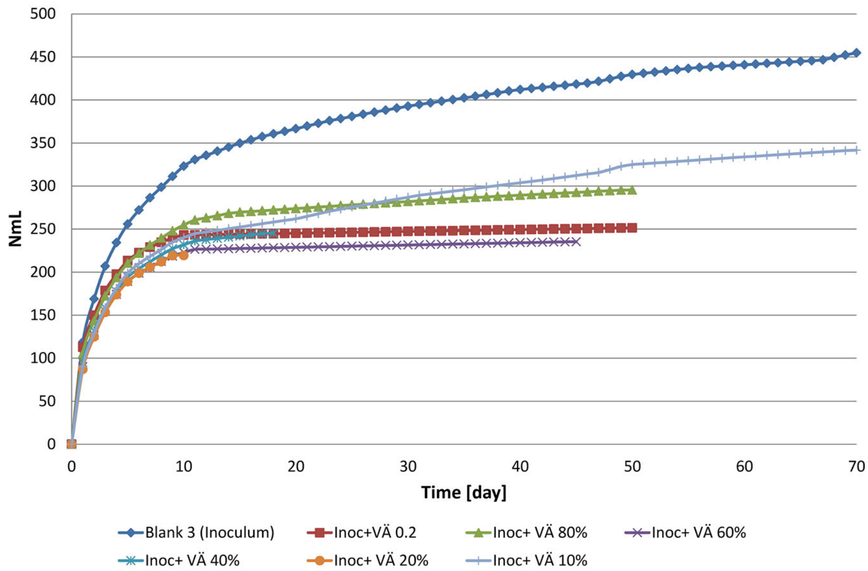


**Fig. 8.** Accumulated biomethane volume in the second series of biomethane potential tests. For abbreviations see caption of Fig. 6.

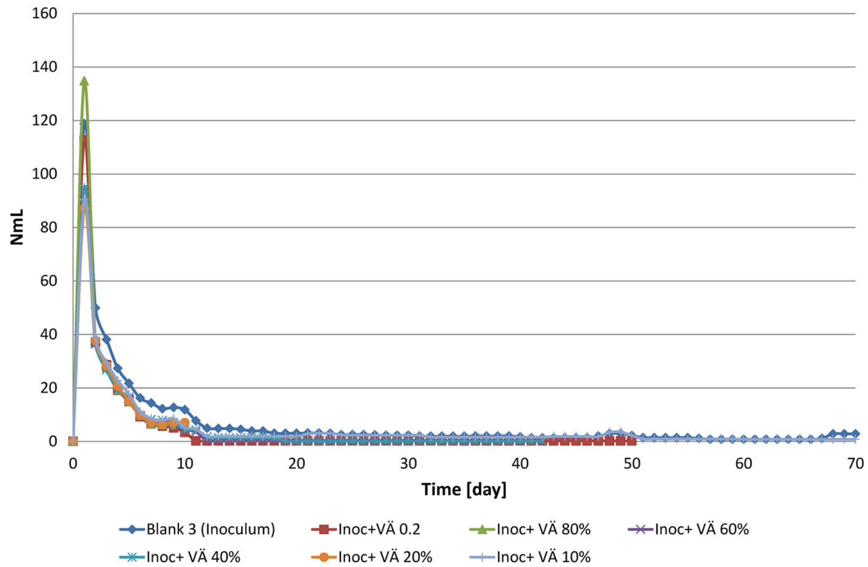


**Fig. 9.** Biomethane flow rate in the second series of BMP tests. For abbreviations see caption of Fig. 6.

### 4.5. Reducing the amount of biological waste deposited and composted in the landfills by methane fermentation

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

It should be stressed that methane fermentation followed by digestate composting is only possible from biodegradable waste that is sorted before collection and pretreated by removing unsuitable materials. Household waste (food waste from kitchens and animal wastes) should be also hygienized 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 amount of nutrients such as phosphorus and nitrogen, which can be used for fertilizing cultivable lands. For some time, the compost from wastewater treatment plant sludge and methane fermented wastewater sludge has been used for fertilizing green and recultivated areas and to a lesser extent in agriculture. In the near future, biodegradable waste should be sorted and the capacity for recycling waste by methane fermentation in landfills and in other locations where biodegradable waste is accumulated should be developed.

### 4.6. Utilization of landfill gas obtained from methane fermentation

As a result of the degradation process of the deposited and composted biodegradable waste, biogas is generated. It 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 originates in 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 the 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 is additionally generated, which causes both acidification and a bad odour. In anaerobic fermentation, ammonia emissions are much smaller.

In 2011, three landfills – Uikala, Jõelähtme, and Väätsa – practised the combustion of 100% of the biogas because the amounts produced were too small for its cost-effective utilization in the heat and energy cogeneration plants. For example, in the Väätsa landfill, 175 200–262 800 $m^3$ of biogas was annually collected from three depositing fields with a total area of 3.5 ha (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 cogeneration 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 with pilot reactors was performed in the Department of Environmental Engineering of Tallinn University of Technology (TUT) in order to find better solutions for the anaerobic digestion process and to choose suitable substrates for co-digestion. The biomethane tests were conducted in anaerobic mesophilic conditions by measuring the maximum amount of biogas or biomethane produced per gram of volatile solids (VS) contained in the organic matter used as the 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 wastes on the digestion process (co-digestion).

The substrates used in different 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),

**Table 7.** Biogas yield from biomethane potential tests

| | Average yield, $m^3$ $CH_4$/tonne volatile substances | Average yield, $m^3$ $CH_4$/tonne |
|---|---|---|
| Catering waste | 404 | 82 |
| Sewage sludge | 245 | 7 |
| Sewage sludge + catering waste 10% | 265 | 9 |
| Sewage sludge + catering waste 25% | 353 | 22 |
| Sewage sludge + catering waste 25% | 484 | 42 |
| Sewage sludge + fish residues 2.5% | 296 | 10 |
| Sewage sludge + fish residues 5% | 346 | 14 |
| Potato + gravy + salad + soup | 229 | 124 |
| Compost | 228 | 68 |
| Sewage sludge + compost 25% | 229 | 20 |
| Whey | 407 | 18 |
| K-J sewage sludge + whey 35% | 203 | 10 |
| K-J sewage sludge + whey 50% | 258 | 12 |
| K-J sewage sludge + whey 75% | 330 | 15 |
| K-J sewage sludge + whey 90% | 369 | 16 |
| Beer yeast | 831 | 98 |
| Beer yeast | 825 | 97 |
| Sewage sludge + beer yeast 35% | 624 | 34 |
| Sewage sludge + beer yeast 50% | 726 | 50 |
| Sewage sludge + beer yeast 75% | 740 | 69 |
| Sewage sludge + beer yeast 90% | 752 | 81 |

K-J – Kohtla-Järve.

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

| Substrate | Average yield, $m^3$/tonne volatile substances* | Min–Max yields, $m^3$/tonne volatile substances* | $CH_4$, % | Reference, if published |
|---|---|---|---|---|
| Sewage sludge | 110 | 68–240 | 33–62 (50) | |
| Sewage sludge + 2% fish residues | 251 | 118–565 | 52–82 (69) | [37] |
| Sewage sludge + 10% fish residues | 206 | 170–281 | 60–71 (69) | [37] |
| Sewage sludge + 36% fish residues | 205 | 198–211 | 58–70 (66) | [37] |
| Catering waste | 249 | 104–454 | 46–74 (66) | |
| Catering waste + sewage sludge | 321 | 121–659 | 55–73 (65) | |
| Compost | 50 | 80–129 | 58–76 (70) | |
| Väätsa reverse osmosis concentrate | 0 | 0–60 | 0–38 (0) | |

* Wet weight.

and RO concentrate + sewage sludge (Table 6). The BMP tests were used for the technical and economic optimization of biomethane producing plants.

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, and RO concentrate (Table 8).

The sewage sludge was obtained from the Tallinn wastewater treatment plant (WWTP). The inoculum was taken from the city of Tallinn WWTP biogas plant anaerobic digester that was operating at +38 $^oC$ with sewage sludge.

The BMP 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 makes the analysis results fully comparable with those obtained by the standard methods.

By the results of the BMP tests, the most promising substrates for biogas production were catering waste, compost, beer yeast, and their mixes with sewage sludge (Tables 7 and 8). Very good results were also achieved with the same substrates in one-stage co-digestion process tests. Unfortunately, the Väätsa RO concentrate inhibited co-digestion processes in every test in which it was added (Table 6).

On the basis of research results [7,23,24,34,37,39–47] and results of laboratory experiments (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. According to the research, the rearrangement of the composting and depositing of 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.

### 4.7. Utilization of the digestate produced from methane fermentation

The National Waste Management Plan for 2014–2020 foresees the utilization of 10% of the anaerobic fermentation digestate of municipal waste, whose amount according to the data from 2011 is 40 800 tonnes [31]. It is necessary to try to increase the amount of digestate from anaerobic fermentation that is used in agriculture.

The amount of produced digestate is equal 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 fertilizing cultivated lands. The compost that is produced in the Väätsa landfill contains 3.5 kg TN, 0.41 kg TP, and 0.54 kg total K per 1 $m^3$.

There are successful long-term results in using compost from methane fermented AS from sediments of the Tallinn WWTP in agriculture, greenery, and recultivation, and there are experiments with the forestation of abandoned less valuable arable lands and cutover peat lands [46,47].

### 4.8. Characteristics of the most efficient landfill wastewater treatment methods

In 2007–2014, different technologies for treating landfill wastewater were tested and the operation of the already existing treatment plants at Väätsa, Torma, and Uikala was observed. The conclusions drawn from the study are as follows:

1. It is necessary to equalize the flow rate and pollutant content of the wastewater in the equalizing tank before treating the wastewater. The equalizing tank should have a receiving well from where the leachate, without getting mixed with other wastewater, is pumped into the main treatment plant.

2. Only wastewater from the young landfills fulfilled the requirements set by legislation after biological treatment. Biological treatment may also be used for pre-treating wastewater if the following technological measures are applied:
- in treatment plants with AS, the hydraulic retention time for landfill wastewater should be prolonged to be twice the time used in the AS 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, the positive temperature of the leachate should be maintained with technical measures;
- part of the heat obtained from the combustion of biogas can be used for warming up the leachate in winter if excess heat is available;
- 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. In winter, the tightness of leachate may be over 1.0 t/$m^3$ due to a high content of substances and shortage of wastewater, aggravating conditions for the treatment with AS (AS 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;
- as biological treatment methods alone are not sufficient for safeguarding the required level of purification and permitted limit concentrations for pollutants (especially for nitrogen) additional measures should be applied;
- 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 on 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-ozonation of the biologically treated landfill wastewater decreases the total concentration of pollutants (COD), but the overall efficiency remains low. The colour and odour are removed, and, if the previous biological treatment has been sufficiently efficient, post-ozonation is also rather efficient and the biodegradability of the treated wastewater is improved.
4. Coagulation with lime, oil-shale ash, and other reagents is not efficient. Additional expenditures are needed to purchase the reagents and the residue has to be treated separately.
5. It is possible to use the Fenton process for profound 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 of the chemical treatment of biologically treated leachate is significantly higher.
7. We found that the most suitable purification method was RO after the equalization of the flow rate and pollutant concentration of the wastewater with the help of the equalizing tank. The RO process is conducted in two stages, either 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 RO:
- a biofilter should be able to remove high COD and nitrogen 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, $BOD_7$, 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 great influence on the efficiency of the RO equipment. Usually, the efficiency increases for about 3% per temperature rise of each 1 $^{o}$C.

8. In new landfills that are separated from the surrounding environment with a geomembrane and the leachate and/or RO concentrate and AS from biological treatment are directed back to the waste deposit for irrigation, the waste deposit can be considered to be one of the steps in the treatment process of wastewater. The aim is to bind the pollutants into the deposit and, in case of excess sludge, to perform post-treatment of the sludge in the deposit with the help of aerobic and anaerobic processes. As the 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.

## 5. CONCLUSIONS

The main results of the study are summarized below.

1. A large amount of wastewater was found to be conducted into sewerage from composting fields during rainfall and snowmelt. The flow rate, pollutant concentration, and pollution load in this wastewater are not stable. This is why equalizing tanks and high-capacity treatment plants are required.
2. Constructive and technological recommendations were compiled for collecting wastewater into equalizing tanks, channelling into treatment plants, and treating in the plant.
3. Biological treatment of landfill wastewater can only be used when the landfill is young and as a pre-treatment for the RO process.
4. On the basis of experiments and the monitoring of practical operations, two-stage RO treatment with DT filters was found to be the most efficient treatment method for landfill wastewater and it guarantees the necessary level of treatment. The other treatment methods that were studied did not provide the purification efficiency required by legislation.
5. The landfill leachate and concentrate from RO 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 a low carbon and an excessive nitrogen content. 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 should 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 composition 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 ja Enn Loigu

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

Aastatel 2007–2014 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 kanalisatsioonisüsteem ja prügila reoveepuhasti on projekteeritud ning ümberehitatud, tuginedes käesolevale uurimistööle. Kogu kanalisatsioonisüsteem koosneb prügilareovee kogumissüsteemist, ühtlustamismahutist, reovee füüsikalis-keemilisest (pöördosmoos) puhastamisest peale reovee bioloogilist puhastamist aktiivmudapuhastis ja biotiigist ning pöördosmoosi kontsentraadi prügilasse tagasipumpamise süsteemist. Alates 2012. aasta aprillist kuni praeguseni on suublasse juhitava heitvee reoainesisaldus vastavuses seadusandlusega seatud piirväärtustega.

Prügilareovee puhastamise efektiivsus on suurem ja stabiilsem, kui lõpetada prügilate territooriumil biolagunevate jäätmete komposteerimine ning komposteerimisplatsidelt reostunud sademevee juhtimine prügila reoveepuhastisse. Biolagunevatest jäätmetest on otstarbekam metaankääritamise teel toota biogaasi ja taimetoitainerikast digestaati. Biolagunevate jäätmete kompostimise ja prügilasse ladestamise ümberkorraldamine võimaldaks anaeroobset kooskääritamist kasutades toota kuni 23,1 miljonit $m^3$ biometaani aastas, millest saab toota umbes 226 000 MWh soojus- ning elektrienergiat. Metaankääritamisel tekkiv digestaat sisaldab suurel hulgal taimetoitaineid, mida saab kasutada kõlvikute väetamiseks.