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# Characteristics and natural attenuation of the Pääsküla landfill leachate

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**Abstract.** The leachate composition can vary throughout a landfill, and leachate production decreases with placement of the final cover. Anaerobic decomposition, which can produce a leachate with high concentrations of biogenous pollutants, occurs in the covered landfill. Pääsküla landfill (Tallinn, Estonia) was built as a temporal landfill on peat deposits in 1974. In June 2003 the landfill was closed and covered but the leachate will filter over many years. In March 2004 the leachate contained 62.6 mg/L total nitrogen ($N_{tot}$), 6.9 mg/L total phosphorus ($P_{tot}$), 668 mg $O_2$/L COD, and 160 mg $O_2$/L $BOD_7$. In the leachate 91% of the nitrogen was determined as ammonia. In March 2004 the content of pollutants was higher than the corresponding values analysed in 1971 and 1993. It is connected with methane production in the closed landfill. The effluent enters the Vääna River through the Pääsküla River and in the sampling place of the Vääna River the values of pollutants were 2.4 mg/L $N_{tot}$, 0.05 mg/L $P_{tot}$, 26 mg $O_2$/L COD, and 1.7 mg $O_2$/L $BOD_7$. The concentrations of pollutants had decreased as a result of dilution in the rivers but it is an insufficient antipollution measure.

**Key words:** landfill, leachate, natural attenuation, dilution, anaerobic decomposition.

## INTRODUCTION

Landfilling is a popular way for municipal solid waste treatment, but it takes up large areas of land and leads to the pollution of the surrounding area. For a long time only mixed landfilling was employed, which means that all kinds of wastes were dumped together in an unsystematic way [1]. Landfills have potential to pollute the three principal environmental media – the atmosphere, the lithosphere, and the hydrosphere [2, 3] by generating landfill gas and leachate by

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the anaerobic decomposition of organic wastes. Leachate is generated by excess rainwater percolating through the waste layers in the landfill [4]. Landfill leachates pose a high risk of groundwater and soil pollution because historically most landfills were built without engineered liners and leachate collection systems [1, 4]. The EU Landfill Directive [5] obliges member states to collect and treat leachate from landfill sites.

Several reviews have been published about the landfill leachate [4, 6, 7]. Leachates from the landfills have high concentrations of organic matter and toxic substances. The properties of leachate depend on the composition of deposited wastes [3, 6, 8]. There is a strong relationship between the state of refuse decomposition and its associated leachate characteristics. Depending on the age of the landfill, microbial degradation of the organic matter of the deposited waste takes place through four phases: (1) aerobic fermentation, (2) anaerobic, acid fermentation, (3) anaerobic, unstable methane fermentation, and (4) anaerobic, stable methane fermentation [4].

The aerobic phase in a landfill lasts only a few days and the leachate contains mainly water and dissolved compounds. In the second phase carboxylic acids accumulate and the pH decreases due to the domination of hydrolytic, fermentative, and acetogenic bacteria. The highest BOD and COD values in the leachate are also measured in this phase; this phase lasts about a couple of months [4]. The third or methanogenic phase starts when measurable quantities of methane are produced. This phase lasts a couple of years. During this phase the accumulated carboxylic acids are converted to methane and carbon dioxide. The COD and BOD values of leachate are decreasing and the pH is increasing as acids are consumed by bacteria. In the fourth or stable methanogenic phase the methane production rate reaches its maximum. The phase may last more than 10 years. The pH continues to increase to a steady-state value and the COD of the leachate is mainly caused by humic and fulvic acids in this phase [4].

A model of the processes in the landfill in the three anaerobic stages is described by White et al. [9]. As a result of synergetic interactions between various groups of microorganisms complex high-molecular-weight organic compounds transform to simple ones having terminal compounds $CH_4$, $CO_2$, $H_2$, $NH_3$, and $H_2S$.

Hydrogenotrophic methanogens (i.e. hydrogen-utilizing chemolithotrophs) convert hydrogen and carbon dioxide into methane [10]:

$$CO_2 + 4H_2 = CH_4 + 2H_2O.$$

The acetotrophic methanogens are able to convert acetate into methane and $CO_2$:

$$CH_3COOH = CH_4 + CO_2.$$

Leachate composition varies significantly among landfills [1, 4, 6], but generally landfill leachate may be characterized as a water based solution of four groups of pollutants: (1) dissolved organic matter, (2) inorganic components, (3) heavy metals, and (4) xenobiotic organic compounds [6]. The leachate can either be treated locally or transported to a municipal wastewater treatment plant. Biological, chemical, and physical methods and natural systems are used in local treatment of leachate [1, 7, 11–13]. The interest in natural systems has increased in recent years. The natural systems include planted forest, natural forest, wetlands, and special infiltration plants. The microorganisms break down organic material and plants use macro- and micronutrients in their growth, which brings about the purification of leachate [1]. Monitored natural attenuation (MNA) has received attention as a cheap alternative to other techniques that actively decontaminate polluted soil or groundwater. Natural attenuation is the use of natural processes to control the spread of the contamination from chemical spills and reduce the concentration and amounts of pollutants at contaminated sites [14]. MNA has been applied as a remediation strategy for petroleum hydrocarbon pollution [15–18]. Natural attenuation in landfill leachate plume has been also studied [6, 19, 20]. In Estonia with its cold climate the natural systems can effectively be used during the warmer part of the year.

The objective of this study was to evaluate the effects of using natural attenuation as a remedial option on landfill leachate. Pääsküla landfill was selected as the objective because the landfill is situated in a suburb of Tallinn. Following the EU Landfill Directive Pääsküla landfill was closed, but the generation of leachate will continue for a long time and the treatment of the leachate needs an optimal solution.

## MATERIALS AND METHODS

### Site description and sampling

Pääsküla landfill is situated in a suburb of Tallinn, the capital of Estonia (Fig. 1). The landfill was built as a temporary waste disposal place in 1974. Since

**Fig. 1.** Location of the Pääsküla landfill and the Pääsküla and Vääna rivers.

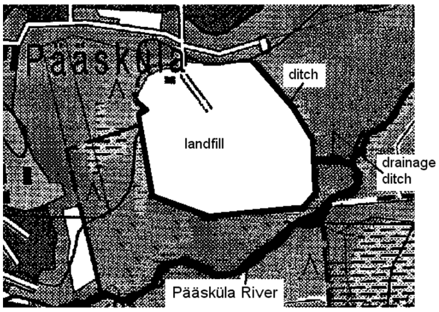

**Fig. 2.** Location of the landfill, ditches, and river in Pääsküla.

2003 it was the largest municipal and industrial landfill in the Tallinn region. The area of the landfill is 30 hectares and its relative altitude is 30 m. The amount of deposited waste is about 4 million tonnes. The landfill was built without a project into a used peat-cutting area [21]. The overburden of the area contains peat, sand, clay and moraine with a total thickness of 10–15 m. A limestone layer (thickness 22–25 m) and sandstone form the bedrock of the area.

There is no leachate treatment in this landfill. A surrounding ditch, which operates as an oxidation pond, has been dug around the landfill. The leachate flows through the additional drainage ditch and the Pääsküla River into the Vääna River (Fig. 2). The Pääsküla River is located in the southern part of the landfill. The nearest houses are situated only 200 m north of the landfill. The Vääna River flows into the Gulf of Finland. The location of the rivers is shown in Fig. 1. Willow thickets grow in the areas around the landfill, the ditches, and the Pääsküla River. The ditches and the river have become overgrown with cattail and reed.

In 1987–1997 comprehensive hydrobiological fieldwork was carried out on 249 Estonian rivers. The results are published in a monograph [22]. In 1993 the water samples of the Vääna and Pääsküla rivers were analysed during the fieldwork (the sampling points are presented in Fig. 1). In 2003 the water samples of the Vääna and Pääsküla rivers were analysed again. The samples of leachate and river water were taken from the ditch and the Pääsküla River in July. The landfill was closed in June and in July it was being covered.

## Chemical analyses

In 2003 and 2004 three samples of water were taken from the same place and mixed. At least two replicate samples were analysed. The leachate and water samples were analysed by standard methods [23, 24]. $BOD_7$ was determined by the seeded dilution method and COD was determined by the dichromate method using a LT 100 Thermostat and LASA® 20 Photometer (Dr. Lange, Germany). The pH of the leachate and water samples was measured by a pH-meter (Model 3320, Jenway, UK).

Although nitrogen and phosphorus are nutrients their high concentration is a sign of water pollution. The concentration of nitrogen and phosphorus is often used to examine the quality of the natural water. The samples were digested with persulphate to determine total nitrogen and phosphorus [24]. The concentrations of total ($N_{tot}$) and nitrate nitrogen ($NO_3^-$-N) were determined by the cadmium reduction method. The formed highly coloured azo dye was measured by a spectrophotometer at 545 nm (Model 6300, Jenway, UK) [23]. The concentration of ammoniacal nitrogen ($NH_4^+$-N) was determined by the indophenol blue method, and absorbance of the solution was measured by a spectrophotometer at 670 nm. The total ($P_{tot}$) and phosphate phosphorus ($PO_4^{3-}$-P) concentrations were determined by the ascorbic acid method, and the absorbance of the solution was measured at 880 nm (Model 6300, Jenway, UK) [24].

The concentration of sulphide was determined as total sulphide [$S^{2-}$] (including dissolved $H_2S$ and $HS^-$ ions) by the iodometric method at a certain pH [23, 25].

## RESULTS AND DISCUSSION

### Leachate in the ditch surrounding Pääsküla landfill

Samples of leachate were taken from the ditch surrounding the landfill in July 2003 and in spring 2004. The results of analyses are presented in Table 1.

In March 2004 the samples of the leachate were taken under the ice (the ditches and the river were covered with ice), and the determined characteristic parameters of the leachate were much higher (COD 4.5, $N_{tot}$ 4.8, and $P_{tot}$ 8.6 times) than in July 2003 (Table 1). In March 2004 the leachate contained 57.0 mg/L $NH_4^+$-N and 5.0 mg/L $NO_3^-$-N. In April 2004 the results of analysis showed lower values of COD, $N_{tot}$, and $P_{tot}$ but the value of BOD was higher than in March 2004 (Table 1). The lower values are connected with snowmelt water, which diluted the leachate. The higher value of BOD showed the high concentration of biodegradable compounds that had flowed with snowmelt into the surrounding ditch.

Results of the analyses of the water samples taken from the drainage ditch flowing into the Pääsküla River are presented in Table 2. The results for 1971 were presented by Roosalu [26].

**Table 1.** Characteristic parameters of the leachate from the ditch surrounding the Pääsküla landfill

| Time | pH | COD, mg $O_2$/L | $BOD_7$, mg $O_2$/L | $N_{tot}$, mg/L | $P_{tot}$, mg/L | [$S^{2-}$], mg/L |
|---|---|---|---|---|---|---|
| July 2003 | 7.09 | 150 | ND | 13.0 | 0.8 | 4.0 |
| March 2004 | 8.28 | 668 | 160 | 62.6 | 6.9 | 6.0 |
| April 2004 | 7.01 | 511 | 237 | 55.0 | 1.1 | 4.0 |

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ND – not determined.

**Table 2.** Characteristic parameters of the drainage ditch from the Pääsküla landfill

| Time | pH | COD, mg $O_2$/L | $BOD_7$, mg $O_2$/L | $NO_3^-$-N, mg/L | $NH_4^+$-N, mg/L | $PO_4^{3-}$-P, mg/L |
|---|---|---|---|---|---|---|
| June 1971 | 6.8 | 70 | 26* | 0.3 | 5.5 | ND |
| July 1971 | 7.45 | ND | 3.2 | 0.3 | 4.0 | 0.9 |
| July 1993** | ND | ND | ND | 2.8 | 4.0 | 0.08 |
| July 2003 | 6.88 | 46 | ND | 1.1 | 12.0 | 0.3 |

* $BOD_5$ was determined.
ND – not determined.
** Samples were taken 300 m downstream in comparison with 1971 and 2003.

In June 1971 the higher values of COD and $BOD_5$ of the leachate from the drainage ditch (Table 2) were connected with the location of small cattle farms near the drainage ditch. In the 1980s the small farms were closed.

The results of the analyses of the water samples taken from the mouth of the Pääsküla River flowing into the Vääna River are presented in Table 3. In 1971 $P_{tot}$ was not determined (ND) [26].

In July 2003 the samples were taken after a rainy period and the results showed characteristic parameters for a diluted leachate. The COD of the leachate from the surrounding ditch was about three times higher than in the drainage ditch (Table 2) and the Pääsküla River (Table 3). The concentrations of $N_{tot}$ were similar for the surrounding ditch (Table 3), drainage ditch (Table 2), and the Pääsküla River (Table 3), but the concentration of $P_{tot}$ was 2.6 times as high for the leachate in the surrounding ditch (0.8 mg/L, Table 1) as for water in the drainage ditch (0.3 mg/L, Table 2) and twice as high as in the river (0.4 mg/L, Table 3). This indicates that nitrogen compounds were also leached into the water in the upper course of the Pääsküla River.

In July 2003 the leachate contained 51.4 mg/L $NH_4^+$-N and 3.3 mg/L $NO_3^-$-N. The ratios of $NH_4^+$-N to $NO_3^-$-N were above 10 for both analysis. The present results accord with data published in the literature [4, 6].

Dihydrogen sulphide in the landfill is formed as a by-product of anaerobic oxidation of organic matter during bacterial sulphate reduction [9]. Dihydrogen sulphide is a very toxic gas formed on the basis of chemical equilibrium with sulphide ions. At pH = 7 the concentrations $[H_2S(aq)] = [HS^-]$ and the solution of sulphide contains a toxic concentration of dihydrogen sulphide [27].

**Table 3.** Characteristic parameters of water from the Pääsküla River mouth

| Time | COD, mg $O_2$/L | $N_{tot}$, mg/L | $P_{tot}$, mg/L |
|---|---|---|---|
| June 1971 | 40 | 5.1 | ND |
| July 1993 | 58 | 3.1 | 0.08 |
| July 2003 | 51 | 12.4 | 0.4 |

ND – not determined.

Both in July 2003 and April 2004 the concentration of total sulphide was 4 mg/L. It gives the $H_2S$ concentration about 2 mg/L, which coupled with a high concentration of ammonia caused a fish kill in the Vääna River. In March 2004 when the ditches were covered with ice and the temperature of the leachate was low, both the concentration of total sulphide and the pH of the leachate had higher values (Table 1).

## Water in the drainage ditch

Analyses of the water in the drainage ditch (Table 2) showed that nitrogen occurred mainly as $NH_4^+$-N and the landfill leachate had increased concentrations of nitrogen. The phosphorus occurred in the form of phosphate.

The ratios of the concentrations of $NH_4^+$-N to $NO_3^-$-N were above 10 both in July 1971 and in July 2003, but the concentration of $NH_4^+$-N was twice higher and that of $NO_3^-$-N up to three times higher in 2003 than in 1971. The high concentration of $NH_4^+$-N confirmed anaerobic conditions in the landfill [28]. In July 1993 the samples of water were taken 300 m downstream in comparison with 1971 and 2003, and the ratio of the concentrations of $NH_4^+$-N to $NO_3^-$-N was 1.4. This indicates that the ammonia had oxidized to nitrate in the ditch. The ratio of COD to $NH_4^+$-N was 3.8 for the leachate from the drainage ditch in July 2003 (Table 2) and the ratio of COD to $N_{tot}$ was 4.1 for the water from the Pääsküla River in July 2003 (Table 3). These ratios show significant pollution of water with nitrogen.

## Water in the mouth of the Pääsküla River

The highest COD of the Pääsküla River (Table 3) was determined in July 1993. In July 2003 it was by 12% lower (Table 3). In July 1971 (before the landfill was built) the COD of the Pääsküla River was by 31% lower than in 1993, which confirms that the landfill leachate raised the pollution load to the river. The growth of the pollution load was not very high because the distance between the sampling point on the river and the leachate ditch is 800 m. In July 2003 the concentration of $P_{tot}$ was 5.5 times higher and the concentration of $N_{tot}$ 4 times higher than in July 1993. This confirms that the pollution load by nitrogen and phosphorus grew significantly in the Pääsküla River during 10 years.

## Water in the Vääna River

The water samples of the Vääna River were analysed in July 1971 [26], 1993, and 2003. Along the Vääna River (Fig. 3a) high concentrations of ammoniacal nitrogen (12 mg/L in 2003 and 4 mg/L in 1993) were determined only in the Pääsküla sampling point (Fig. 1); so these are due to landfill leachate. The highest concentration of nitrate nitrogen was measured at Lähtse in 1971 (Fig. 3b); it was caused by intensive use of mineral fertilizers. In 2003 the highest $NO_3^-$-N concentrations were determined at Hüüru and Vääna, which are downstream from

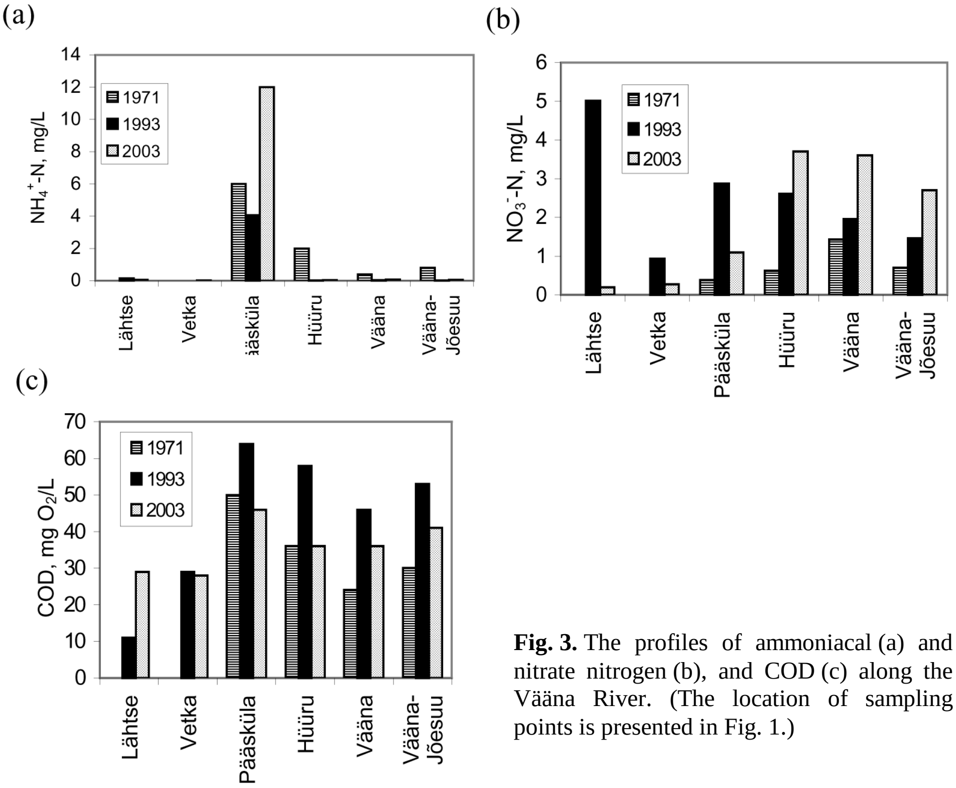

**Fig. 3.** The profiles of ammoniacal (a) and nitrate nitrogen (b), and COD (c) along the Vääna River. (The location of sampling points is presented in Fig. 1.)

the Pääsküla River, and so these can be connected with the oxidation of ammonia in the Vääna River (Fig. 3a). The very low concentration of $NH_4^+$-N at Hüüru shows an active nitrification process in the Vääna River. The high concentration of ammoniacal nitrogen in 1971 was connected with extensive agriculture (small cattle farms without manure water treatment) around the upper course of the Vääna River [26]. The high value of COD can be connected with the peat mining before the 1970s.

The highest values of COD were determined in the Pääsküla sampling point (Fig. 3c), and the values remained high as far as the mouth of the Vääna River at Vääna-Jõesuu. This testifies that landfill leachate inreases the pollution load on the Vääna River and the so-called natural attenuation through the dilution is insufficient to reduce the pollution of the Vääna River.

## CONCLUSIONS

Anaerobic degradation of biowaste in the Pääsküla landfill generates leachates with high concentrations of pollutants: COD – 668 mg $O_2$/L, $N_{tot}$ – 62.6 mg/L, and $P_{tot}$ – 6.9 mg/L in March 2004 with the ratio of the concentrations of $NH_4^+$-N to $NO_3^-$-N 11.4. In April 2004 the leachate was diluted with snowmelt and the concentrations of pollutants were lower. The concentrations of pollutants were lower in the water of the drainage ditch in comparison with the leachate of the landfill, but the ratio of the concentrations of $NH_4^+$-N to $NO_3^-$-N was 14.8.

In July 1993 the COD of the water from the Pääsküla River was by 31% (22% in July 2003) higher than in July 1971 (before the landfill was built), showing that the landfill leachate raised the pollution load on the river. Comparison of the concentrations of pollutants showed that the so-called natural attenuation of pollutants is connected mainly with dilution and it does not decrease the pollution load to the river water sufficiently.

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# Pääsküla prügila nõrgvee koostis ja looduslik isepuhastumine

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Prügilate nõrgveed on muutliku koostisega ja prügila lõpliku katmise järel nõrgvee hulk väheneb. Prügilates toimuvate anaeroobsete lagunemisprotsesside käigus tekib kõrge reoainete kontsentratsiooniga nõrgvesi. Pääsküla prügila rajati 1974. aastal ajutise prügilana endisesse turbarappa. 2003. aastal prügila suleti ja kaeti, kuid nõrgvett eraldub veel palju aastaid. 2004. aasta märtsi analüüsid näitasid Pääsküla prügila nõrgvee reostusparameetrite väärtusteks: $N_{tot}$ 62,6 mg/l, $P_{tot}$ 6,9 mg/l, KHT 668 mg $O_2$/l ja $BHT_7$ 160 mg $O_2$/l. Nõrgvees sisalduvast lämmastikust esines 91% ammoniaakaalses vormis. Pääsküla jõe sissevool Vääna jõkke sisaldas samal ajal $N_{tot}$ 2,4 mg/l, $P_{tot}$ 0,05 mg/l, KHT 26 mg $O_2$/l ja $BHT_7$ 1,7 mg $O_2$/l. Reoainete kontsentratsioon oli vähenenud lahjenemise kaudu, kuid see on ebapiisav reostustõrjemeede.