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Characterization and biological stabilization of fine fraction from landfill mining

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Abstract

Landfilling has been the major method to dispose waste for the decades, thus there are thousands of landfills around the world. Landfills contain large amount of resources, which could be used as material or energy. There is an increasing interest for landfill mining which means excavation and processing of waste materials mined from landfills. While previous landfill composition studies have focused especially on metal recovery and combustible materials, they have shown that landfills contain significant amounts of soil type material with small particle size, referred as fine fraction (FF). As redisposal of FF after landfill mining is expensive and causes emissions for decades, FF should be treated to increase value for reuse. The aim of this thesis was to assess in details the characteristics of the FF and to evaluate the effects of different biological treatment methods on stability and characteristics of FF. In this study, FF was sampled from two landfills representing different eras of material consumption and waste management practices: Kuopio, landfilled 2001–2011, and Lohja, landfilled 1967–1989.

The Kuopio landfill was found to contain 38–54 % of FF (< 20 mm) and the Lohja landfill 40–74%. FF contains in various amounts of organic matter (VS 6–27% of TS), nutrients (1.4–8 kg N/t TS, 1–1.5 kg P/t TS) and soluble organic compounds (e.g. 0.5–4.6 kg COD/t TS). The organic matter content, biomethane potential (0.4–27 L CH_4/kg TS) and respiration activity (1.4–2.4 g O_2/kg TS) were detected to be higher in top layer of new landfill (1–5 years old) while bottom layer of new landfill (6–10 years old) was similar to old landfill (24–46 years old). Biological activity may limit the utilization of FF after landfill mining, thus FF needs to be stabilized to reduce biological activity. Furthermore, FF may also contain hazardous compounds, which needs to be assessed when evaluating the use of FF.

To reduce biological activity of FF, the anaerobic and aerobic stabilization of FF were studied in two laboratory experiments employing simultaneous four leach bed reactors operated for 173–180 days. In anaerobic stabilization, methane production was found to range from 9 to 18 m^3 CH_4/t VS for FFs from both landfills. Irrigation of FF was necessary for efficient methane production while sludge addition providing both moisture and inoculum deteriorated the characteristics of FF.

Aerobic stabilization reduced more efficiently organic matter content and biological activity from FF compared with anaerobic treatment. Ammonium nitrogen in the leachate was removed rapidly in aerobic treatment due to nitrification. Organic matter and soluble compounds were efficiently removed with continuous water adding, regardless of anaerobic and aerobic conditions, while leachate recirculation introduced those back to the reactor. The scaling up of the anaerobic and aerobic stabilization methods of FF showed that applied technology, for example aeration or irrigation method, and size of treatment area have major effects on the costs of FF treatment. However, anaerobic stabilization and aerobic stabilization with passive aeration without continuous irrigation would have similar costs in similar sites.

In conclusion, FF may need stabilization due to organic matter content and biological activity before utilization. Both anaerobic and aerobic stabilization improved the quality of FF by reducing organic matter content and biological activity. Both treatment methods can be used in full scale stabilization of FF. The treatment of FF has potential to increase the value and usability of FF. Treatment concept and technology should be further optimized in pilot and full scales.

Tiivistelmä

Jätteen loppusijoittaminen kaatopaikalle on ollut vuosikymmenien ajan yleisin jätteenkäsittelymenetelmä, minkä seurauksena esimerkiksi Euroopassa on arviolta 150 000–500 000 kaatopaikkaa. Kaatopaikkojen kaivamisesta on kiinnostuttu viime vuosina, koska kaatopaikat sisältävät hyödynnettäviä materiaaleja kuten metalleja ja polttokelpoisia muoveja. Näiden lisäksi kaatopaikat sisältävät paljon hienoainesta, joka on partikkelikooltaan kaatopaikan jakeista pienin. Hienoaines muistuttaa maata, ja on suurelta osin peräisin kaatopaikkojen välitäytöistä, mutta sisältää lisäksi alle 10 % muita hajonneita jätemateriaaleja, kuten metallia, lasia ja orgaanista ainesta. Kaatopaikkojen kaivamisen yhteydessä hienoaines on yleensä sijoitettu takaisin kaatopaikalle, mikä on kallista ja aiheuttaa ympäristöpäästöjä vuosikausia, joten on tarpeellista etsiä hienoainekselle hyötykäyttömahdollisuuksia. Tämän tutkimuksen tavoitteena oli tutkia hienoaineksen ominaisuuksia ja arvioida biologisten käsittelymenetelmien vaikutusta hienoaineksen ominaisuuksiin ja stabiilisuuteen, ja siten parantaa mahdollisuuksia hyötykäyttää kaatopaikalta kaivettu hienoaines.

Tätä tutkimusta varten otettiin näytteitä kahdelta yhdyskuntajätteen kaatopaikalta: Kuopiosta, täytetty vuosina 2001–2011 ja Lohjalta, täytetty vuosina 1967–1989. Kuopion kaatopaikasta 38–54 % ja Lohjan kaatopaikasta 40–74 % oli hienoainesta (raekoko alle 20 mm). Hienoaines sisälsi orgaanista ainetta 6–27 % kuiva-aineesta (ka), liukoista orgaanista ainetta 0.5–6.4 kg COD/t ka, typpeä 1.4–8 kg/t ka, fosforia 1–1.5 kg/t ka ja sen biologinen aktiivisuus mitattiin metaanintuottopotentiaalilla (0.4–27 m^3 CH_4/t ka) ja hapenkulutuksella (1.4–2.4 g O_2/kg ka). Orgaanisen aineksen määrä ja biologinen aktiivisuus voivat rajoittaa hienoaineksen hyötykäyttöä, sillä ne ovat korkeampia kuin vastaavilla luonnon maalajeilla. Mainittujen ominaisuuksien lisäksi hienoaines saattaa sisältää haitallisia yhdisteitä kuten raskasmetalleja ja orgaanisia haitta-aineita, jotka on analysoitava hyötykäyttöä arvioitaessa.

Hienoainesta stabilointiin orgaanisen aineksen ja biologisen aktiivisuuden vähentämiseksi kahdessa neljän laboratorioreaktorin kokeessa 173–180 päivän ajan anaerobisissa ja aerobisissa olosuhteissa. Anaerobisessa käsittelyssä metaania tuotettiin 9–18 m^3 CH_4/t orgaanista ainetta, mutta aerobinen käsittely eli ilmastus vähensi enemmän hienoaineksen orgaanisen aineen määrää ja biologista aktiivisuutta. Käsittelyssä hienoainekseen pitää riittävän kosteuspitoisuuden takaamiseksi lisätä vettä käsittelyn alussa tai vähitellen koko käsittelyn aikana. Myös kerätyn suotoveden kierrättäminen reaktorissa on mahdollista. Veden voi korvata myös lietteellä, mikä lisäsi metaanintuottoa, mutta heikensi hienoaineksen ominaisuuksia käsittelyn jälkeen. Liukoiset materiaalit, kuten orgaaninen aines ja anionit, poistuivat jatkuvassa veden lisäyksessä, kun taas suotoveden kierrätys palautti nämä liukoiset aineet takaisin reaktoriin. Metaania tuotettiin yhtä paljon reaktoreissa, joista toiseen lisättiin jatkuvasti puhdasta vettä ja toiseen kierrätettiin suotovettä. Aerobisessa käsittelyssä nitrifikaatio poisti tehokkaasti ammoniumtypen suotovedestä.

Tämä tutkimus osoittaa, että sekä anaerobisella että aerobisella käsittelyllä voidaan stabiloida hienoainesta ja että stabilointi voitaisiin toteuttaa täydessä mittakaavassa olemassa olevilla teknologioilla. Stabilointi parantaa aineksen laatua ja käytettävyyttä esimerkiksi rakennusmateriaalina tai maanparannusaineena, mikä nostaa hienoaineksen arvoa. Riittävää ilmastus- ja kastelumäärää on kuitenkin syytä tutkia lisää esimerkiksi pilot-mittakaavassa, jotta hienoaineksen käsittelyn kustannukset saadaan edullisemmiksi kuin kaatopaikkasijoittaminen.

Preface

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Tampere, January 2018

Tiina Mönkäre

Contents

List of Symbols and Abbreviations

BMP	Biomethane potential
CH_4	Methane
CO_2	Carbon dioxide
COD	Chemical oxygen demand
DIC	Dissolved inorganic carbon
DOC	Dissolved organic carbon
FF	Fine fraction
H_2S	Hydrogen sulfide
LBR	Leach bed reactor
MSW	Municipal solid waste
N, N_{tot}	Nitrogen, total nitrogen
$N_{soluble}$	Soluble nitrogen
NH_4^+-N	Ammonium nitrogen
NO_2^-	Nitrite
NO_3^-	Nitrate
O_2	Oxygen
P, P_{tot}	Phosphorus, total phosphorus
$P_{soluble}$	Soluble phosphorus
TKN	Total Kjehdahl nitrogen
TOC	Total organic carbon
TS	Total solids
VS	Volatile solids

List of Publications

This thesis is based on the following original publications, which are referred to in this thesis by the roman numerals I-IV. The publication are reproduced with kind permissions from the publishers.

I Mönkäre, T.J., Palmroth, M.R.T., Rintala, J.A., 2016. Characterization of fine fraction from two Finnish landfills. Waste Management. 47, 34-39.

II Mönkäre, T.J., Palmroth, M.R.T., Rintala, J.A., 2015. Stabilization of fine fraction from landfill mining in anaerobic and aerobic laboratory leach bed reactors. Waste Management. 45, 468-475.

III Mönkäre, T.J., Palmroth, M.R.T., Rintala, J.A., 2017. Screening biological methods for laboratory scale stabilization of fine fraction from landfill mining. Waste Management. 60, 739-747.

IV Mönkäre, T., Palmroth, M., Sormunen, K., Rintala, J., 2017. Treatment of fine fraction from landfill mining – concept and case study examples. Submitted for publication.

Author's Contribution

I Tiina Mönkäre performed the analyses, wrote the manuscript and is the corresponding author. Marja Palmroth and Jukka Rintala assisted in planning the experiments and interpretation of the results and commented on the manuscript.

II Tiina Mönkäre planned and performed the experimental work, wrote the manuscript and is the corresponding author. Marja Palmroth and Jukka Rintala assisted in planning the experiments and interpretation of the results and commented on the manuscript.

III Tiina Mönkäre planned and performed the experimental work, wrote the manuscript and is the corresponding author. Marja Palmroth and Jukka Rintala assisted in planning the experiments and interpretation of the results and commented on the manuscript.

IV Tiina Mönkäre designed the treatment concept, wrote the manuscript and is the corresponding author. Kai Sormunen assisted in planning the concept and commented the mass balance and cost structure calculations. Marja Palmroth commented on the manuscript. Jukka Rintala assisted in planning the concept and commented on the manuscript.

1 Introduction

Global material use has intensively increased during the 20th century due to population growth and industrialization. In 1900, the global material extraction was about 7 Gt, while in 2010 it was 70 Gt (Krausmann et al., 2009; Schaffartzik et al., 2014). At the same time, population grew from 1.7 billion to 7 billion (United Nations, 1999; 2015) and thus material use per capita doubled from 4.6 to 10.3 t/cap/a (Krausmann et al., 2009). The rate of material consumption has varied during the century, from the slight increase between 1900 and 1945 to the high increase between 1945 and 1973, when the material use was doubled (Krausmann et al., 2009). Since then the growth of global material use was slowed down after the oil price shocks, but still has been increasing, especially in Asia where growth has been rapid in the 21st century (Schaffartzik et al., 2014). In addition, composition of global material use has changed from being mainly renewable biomass (75% of total global material use in 1900 and 30% in 2010) to mineral materials (10% in 1900 and 38% in 2005) (Krausmann et al., 2009; Schaffartzik et al., 2014). At the same time with population growth and increased material use, waste generation has increased tenfold, as global solid waste production was 3.5 million tons per day (about 13 Gt per year) in 2010 (Haas et al., 2015; Hoornweg and Bhada-Tata, 2012).

Waste is determined to be something useless or unwanted substance or object, which the holder discards or intends or is required to discard (Directive 2008/98/EC). For decades, landfills and dumpsites have been the main method of disposal for the solid waste and is still the main waste disposal method in many countries (Eurostat, 2017). Landfills have been seen as the end point of material flow, final solution for waste management with minimum costs in short term perspective (Krook et al., 2012).

Nowadays waste is seen as the potential material and energy resource instead of something to be discarded, thus also the current trend of landfilling is decreasing in

Europe. In 2014 in the European Union (EU-27), 25% of the municipal solid waste (MSW) (61 million tons) was landfilled, while in 1995 in the same countries up to 64% was landfilled (144 million tons) (Eurostat, 2017). The first step of the paradigm of waste management changing from the end of the pipe treatment towards resource management was the waste hierarchy, first introduced in 1977 (Wilson, 2007). The present waste hierarchy (Figure 1) states that first waste prevention is preferred, after that re-use, recycling, material recovery and energy recovery, while disposal is least preferred. This development of the waste management paradigm has led to the increasing trend of circular economy in the 21st century (Wilson, 2007). Circular economy means that material flows are designed to circulate within the socioeconomic system by reuse and recycling or material flows are biological materials, which are returned back to ecological cycle (Haas et al., 2015).

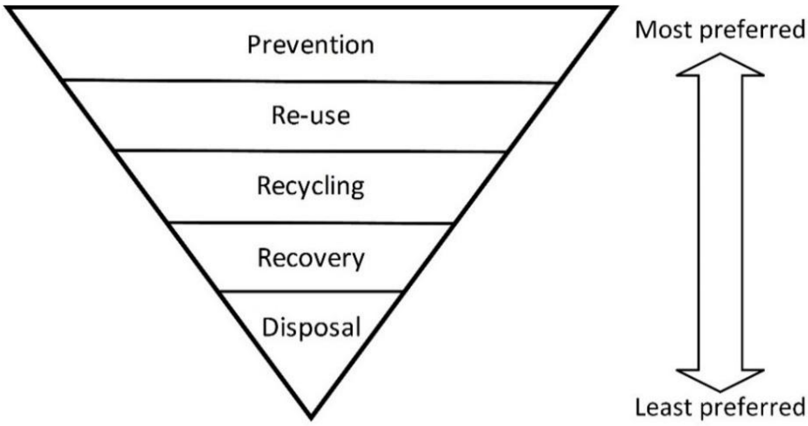

Figure 1. The waste hierarchy, which defines the steps for the most preferred waste management (based on EU Directive 2008/98/EC).

The change of approach to circular economy has made waste and landfills resources. This has led to an increasing interest in urban mining, which refers to the recovery of secondary resources from technospheric stocks (Johansson et al., 2013). One form of of urban mining is landfill mining, meaning excavation and processing of landfilled materials for utilization as material and energy resource (Krook et al., 2012). Benefits of landfill mining include also the remediation of landfill and avoidance of monitoring and treatment costs during landfill aftercare, while environmental emissions in the air, soil and water are avoided and landfill area can be developed as urban or non-urban area (Johansson et al., 2017; Krook et al., 2012; Marella and Raga, 2014). Technology has been and needs to be developed to make landfill mining efficient and environmental friendly, while similarly enable re-use, recycling and recovery of materials as much as possible and avoid the redisposal of mined waste fractions (Jones et al., 2013; Krook et al., 2012).

2 Background

2.1 Landfills

Landfill is a site for waste disposal for the deposit onto or into land (Council Directive 1999/31/EC). Landfilling has been the major method to dispose of waste for the decades, even centuries, thus there are thousands of landfills around the world. Finland has an estimated 1 600 MSW landfills, Sweden 6 000 old landfills (Hogland et al., 2010), Denmark 1 500 landfills (Kjeldsen and Christophersen, 2001), the Netherlands about 3 800 landfills (Paap et al., 2011) and USA 1900 active landfills and 6270 landfills closed after 1988 (Wagner and Raymond, 2015). It has been evaluated that there are 150 000–500 000 landfills in Europe (Hogland et al., 2010). Many countries lack the statistics of the number of landfills, but there are some estimations of area or amount of waste in landfills. It has been reported that in Russia landfills cover 8000 km^2 (Kalyuzhnyi et al., 2003), China has 547 urban active landfills (Zhou et al., 2015a) and the 50 largest dumpsites located mainly in Africa and Asia contain 258–368 million tons of waste (Waste Atlas Partnership, 2014).

Different types of landfills exist from open dumps to controlled landfills and bioreactor landfills (Damgaard et al., 2011). Open dumps, which are common in developing countries, lack the environmental protection, and amount and composition of landfilled waste is not controlled (Damgaard et al., 2011; Prechthai et al., 2008; Waste Atlas Partnership, 2014). Covered dumps have a cover structure compared to open dumps but they lack gas and leachate collection and treatment systems (Damgaard et al., 2011). Open dumps and covered dumps exist also in EU and they are usually landfills build before EU legislation demanding the built bottom and top structures (Council Directive 1999/31/EC). Conventional landfills have both bottom structures and top covers, while also leachate and gases are collected. In a simple conventional landfill, landfill gas is treated by biofilters or by flares, while in an energy-recovery landfills, landfill gas is utilized in energy production. (Damgaard et al., 2011). A step further, landfills, called landfill bioreactors, are engineered to enhance the degradation and stabilization of waste, for example by leachate recirculation to ensure the supply of moisture and nutrients (Damgaard et al., 2011; Kurian et al., 2007; Manfredi and Christensen, 2009; Reinhart et al., 2002). When large amount of water is added to material (1–5 m^3 per ton of waste) and leachate collected, the process is called waste flushing to remove soluble components (Manfredi and Christensen, 2009). Landfill bioreactors could also be aerated to enhance stabilization (Heyer et al., 2005; Reinhart et al., 2002; Ritzkowski et al., 2006; Ritzkowski and Stegmann, 2007). In the future, landfills could be seen as temporary storage for waste, which will be eventually used as material or as energy (Jones et al., 2013). Current EU legislation (Council Directive 1999/31/EC) states that storage time should be limited to three years, and time over three years would be considered as disposal.

Usually, landfills contain MSW from households, commerce, trade and administration (Eurostat, 2011) but may also contain waste from industry, construction waste as well as soil (Quaghebeur et al., 2013; Kaartinen et al., 2013). MSW is mainly composed of food waste, paper and cardboard, plastics, metal and glass, while the composition of landfilled waste has changed due to the legislation and development of novel materials, products and recycling methods (Hermann et al., 2014) and is also dependent on a region and a season. On average, globally the organic fraction is the largest fraction (46%), followed by paper (17%), plastic (10%), glass (5%), metal (4%) and other (18%). However, for example income has effect on waste production, thus also the composition of waste in landfills. In high income countries, the amount of organic waste fraction (food waste) of waste production is significantly lower (28%) than in lower income countries (64%), while for example amount of paper is higher in high income countries (31 %) compared with low income countries (5 %) (Hoornweg and Bhada-Tata, 2012).

Before and during the landfilling, waste is degraded by various biological, physical and chemical processes, thus the age of landfill has significant effect on the composition of waste in landfill (Sormunen et al., 2008). Waste is degraded in landfills in four or five phases: the first, initial aerobic phase, followed by anaerobic acid phase, initial methanogenic phase and stable methanogenic phase (Kjeldsen et al., 2002; Rich et al., 2008). The fifth phase is reintroduction of aerobic phase (Rich et al., 2008). All these processes may exist simultaneously in a landfill (Rich et al., 2008).

Compositions of waste samples mined from landfills are presented in Table 1. Landfills are considered as the potential reservoirs of resources, containing for example 1–5%

metals (Table 1), from which large part is evaluated to be suitable for valorization (Quaghebeur et al., 2013). It has been evaluated that globally landfills contain about 393 million tons of copper compared to 330 million tons are currently in use in materials, while the ores contain 940 million tons of copper (Kapur and Graedel, 2006). Combustible waste is about 20–40% of landfills content, meaning plastics, textiles, paper and cardboard, which could be utilized in material or energy production. It has been evaluated that the waste landfilled after the 1960s, after the high increase in material consumption had started (Krausmann et al., 2009) and before increased source separation (in Europe since the 1990s and 2000s), would be the most suitable landfill mining as they contain high percentage of materials like plastics and metals that can be reused (Hermann et al., 2014).

MSW landfills may contain hazardous compounds, which are toxic, corrosive, flammable and reactive organic and inorganic compounds (Inglezakis and Moustakas, 2015; Slack et al., 2005). Hazardous compounds have been disposed in landfills along with household waste or from industry. However, studies have shown that hazardous compounds are less than 1% of mined landfill waste (Hogland et al., 2004; Hull et al., 2005; Jani et al., 2016),

Landfills also contain soil, amount varying from 5 % to 80 %, while fine fraction (FF) or fines consists 40–70% of the landfills content (Table 1). Fine fraction is usually defined as size fraction below 10–25.4 mm, depending on the study. Some studies consider FF as part of the soil fraction, thus there are large differences in reported compositions. Soil is used as daily and intermediate covers for example to prevent the smell (Hossain and Hague, 2009; Tchobanoglous et al., 1993), thus soil is major part of landfills content. However, landfills differ in respect of location, size and contents, but also operations such as leachate recirculation and bottom or top structure (or lack of those) (Van Passel et al., 2013), which all may affect FF and soil content in landfills.

Table 1**.** Composition as w-% of waste sampled and characterized from different landfill mining sites.

Landfill (age of landfill)	Plastics	Paper and cardboard	Textiles	Metals	Glass	Wood	Soil	Stones	Other	Fine fraction (particle size)	Reference
Kuopio, Finland (1–10 years)	24	4–7	7	3–4	-	6–7	7	-	1–2	43–47 (< 20 mm)	Kaartinen et al., 2013
Lohja, Finland (24–46 years)	20[a]			2	-	-	19	-	1	58 (< 20 mm)	Not published
Styria, Austria (24–30 years)	18.5	2.2	5.1	2	0.4	3.1	4.4	-	5	59.2 (< 12 mm)	Wolfsberger et al., 2016
Houthalen, Belgium (14–29 years)	17±10	7.5±6	6.8±6	2.8±1	1.3±0.8	6.7±5	44±12[b]	-	10±6	[b]	Quaghebeur et al., 2013
Måsalycke, Sweden (17–22 years)	2.5–8.1	5.0–21.1	0.4–4.1	1.1–2.2	0.2–0.3	5.3–12.0	40.7–65.1[b]	3.5–27.4	1.1–5.1	[b]	Hogland et al., 2004
Gladsax, Sweden (23–25 years)	0.3–4.0	1.7–2.8	0.3–1.1	1.3–1.6	0.3–1.5	1.2–2.3	64.5–78.1[b]	15.5–22.7	0–0.9	[b]	Hogland et al., 2004
Jingmen, China (8–23 years)	10.6±5.1	0.2±0.4	1.5±1.3	0.4±0.4	0.6±0.4	2.4±1.5	75.0±6.8[b]	8.3±4.1	1.0 ±1.3	[b]	Zhou et al., 2015b
Sussex, England (8–10 years)	2.6–42.3	0–51.9	0–9.6	0.2–13.6	-	0.3–8.4	-	-	0–24.2	14.2–91.1[c]	García et al., 2016
Florida, USA (3–8 years)	12.9	11.7	2.7	5.7	4.7[d]	-	-	-	3.2	59.1 (< 6.3 mm)	Jain et al., 2005
Northern Italy (5–15 years)	21.9–33.5	1.4–4.2	2.2–5.4	2.9–3.6	9.2–14.0[d]	1.4–2.2	-	-	-	44.6–55.5 (<20 mm)	Raga and Cossu, 2013
Northern Italy (25–40 years)	12.1–13.7	1.4–6.0	2.9–4.3	1.1	12.7–17[d]	1.2–1.6	-	-	-	62.3–62.7 (< 20 mm)	Raga and Cossu, 2014
Hille, Germany (8–25 years)	9.4	4.7	2.5	2.5	1.6	10.1	9.8	7.7	5.0	46.0 (< 20 mm)	Wanka et al., 2016
Istanbul, Turkey (4–20 years)	25.5±6.0	2.7±1.9	3.2±1.5	2.3±1.4	1.3±0.3	1.1±0.4	11.2±6.0	-	1.1±0.4	51.7±5.9 (< 20 mm)	Sel et al., 2016
Kudjape, Estonia (10 years)	22.4	5.1	-	3.1	4.6	4.7	-	17.5	13.4	28.7[e] (< 10 mm)	Bhatnagar et al., 2017

- = not available, ± = standard deviation, a = combustibles (plastics, textiles, paper and cardboard), b = fines included in soil fraction, c = fine organics, d = includes stones, e = includes soil

Landfills are local and global pollution source affecting also on human health (Giusti, 2009). Landfills cause pollution in air, soil, surface water and ground waters, for example in the form of polluted leachate (Giusti, 2009; Sormunen et al., 2008), especially from landfills without bottom structures and open dumps. Leachate is generated from water percolating through layers of waste in a landfill, while water comes from the waste material itself or from precipitation (Kjeldsen et al., 2002). Soluble compounds are transferred from solid waste materials to leachate during this process (Kjeldsen et al., 2002). Leachate contains dissolved organic matter, inorganic compounds, heavy metals and other compounds, and reported concentration vary, for example COD between 500 and 70 000 g/m^3 and ammonium nitrogen between 40 and 6 000 g/m^3 (Bove et al., 2015).

Waste is degraded by biological processes in the landfill and biogas is produced in anaerobic conditions. Landfill gas consists of methane CH_4 (40–50%), carbon dioxide CO_2 (25–50%), nitrogen N_2 (0–15%), oxygen O_2 (0–4%), hydrogen sulfide H_2S (0–1%), hydrogen H_2 (0–1%) and small fractions other trace gases, composition depending on the landfill (Chai et al., 2016). Methane is strong greenhouse gas, with warming potential 28–36 times higher that of CO_2 (Chai et al., 2016). In EU-28, the waste management sector produces 3% of greenhouse gas emissions, of which 71% is methane emissions from managed and unmanaged solid waste disposal sites and methane from landfills contributes 20% of anthropogenic methane emissions (EEA, 2017). The landfill gas can be collected and used for energy production or flared (Chai et al., 2016; Sormunen et al., 2008).

2.2 Landfill mining

Landfill mining means the excavation, processing, treatment and recycling of waste materials mined from landfill (Hogland et al., 2004; Krook et al., 2012) and has resource, environmental and economic aspects (Johansson et al., 2017). The aim of landfill mining is to get waste materials for use as raw materials (e.g. metals) or as an energy resource (e.g. plastics), reduce environmental effects of landfill and provide additional space (Hogland et al., 2004; Krook et al., 2012). When the focus of landfill mining is on the efficient resource recovery and valorization, landfill mining is also referred as enhanced landfill mining (Jones et al., 2013; Kieckhäfer et al., 2017). The basic process of landfill mining and recovered materials and energy are presented in Figure 2.

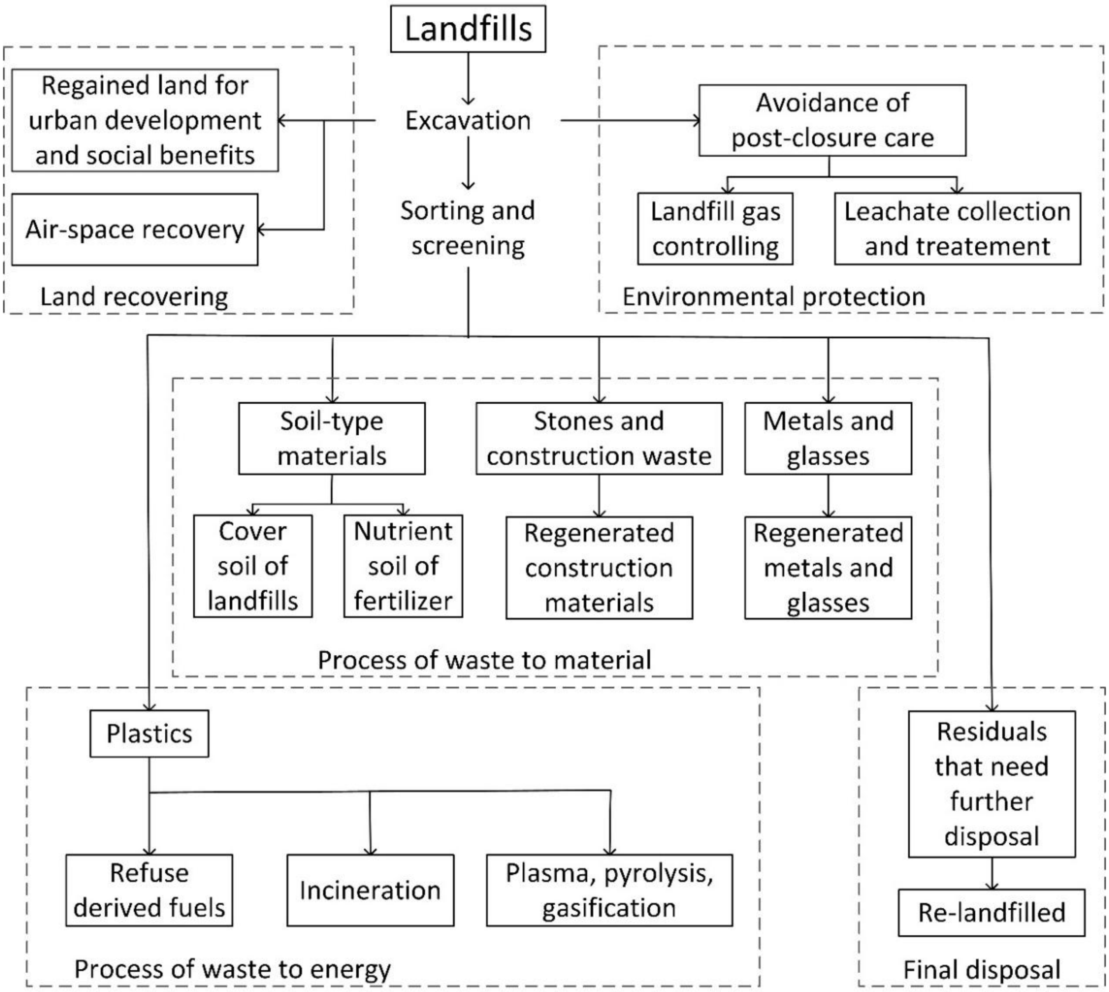

Figure 2 Process of landfill mining and recovery of materials and energy (adapted from Zhou et al., 2015a).

The first reported landfill mining action was in Tel Aviv in Israel in 1953 (Savage et al., 1993). After this in the 1970s and 1990s landfill mining projects aimed in remediation, reclamation or providing additional space in landfill. Since the end of the 1990s, material and energy recovery has increased as objective of landfill mining projects (Burlakovs et al., 2017). Many of the landfill mining projects have not been documented (Damigos et al., 2015) but at least 57 full or pilot scale projects have been reported all around the world in Europe, the USA and Asia as landfill mining projects (Burlakovs et al., 2017; Krook et al., 2012). In addition to full scale landfill mining, throughout the history in many parts of the world, the scavengers have collected part of landfilled waste materials to make profit by material that can be utilized (Burlakovs et al., 2017; Johansson et al., 2013).

Landfill mining has several uncertainties, mainly the waste composition of landfills, (Baas et al., 2010), which has been the main topic in the studies about landfill mining (Krook et al., 2012), but composition of landfill is always case specific. New and active landfills, filled under current EU legislation (Council Directive 1999/31/EC), are mostly well documented, but old disposal sites lack documentation on waste composition, thus sampling is required to explore the content of the landfill (Jones et al., 2013). Waste undergoes various biological, physical and chemical processes at different rates in landfills (Sormunen et al., 2008), thus actual composition in the landfill is different than the documented composition (Jones et al., 2013). Other uncertainties concerning landfill mining are the efficiency of materials processing technologies, markets for materials recovered from landfills and environmental and health risks from excavating landfills (Baas et al., 2010).

Main drivers for landfill mining are environmental and economic benefits, including also other benefits than material or energy resource recovery (Johansson et al., 2013). As landfills are local and global pollution source, the effects to air, soil, surface and groundwater can be decreased as landfills are mined (Danthurebandara et al., 2015; Frändegård et al., 2013; Johansson et al., 2017; Marella and Raga, 2014). Secondary resources also decrease the use of primary resources, as for example the energy utilization of mined materials replaces the need for fossil fuels (Marella and Raga, 2014). Mined landfill areas are restored as nature or developed as recreational areas or urban areas, thus landfill mining has social benefits (Marella and Raga, 2014). From an environmental point of view, landfill mining has high positive impacts on environment compared with closure and aftercare, but in addition negative impacts due to emission during the excavation and waste processing (Danthurebandara et al., 2015). Landfill mining has been shown to have an advantage over closure and aftercare scenario where landfill is not mined (Jain et al., 2014; Jones et al., 2013; Van Passel et al., 2013), while also opposite results are found (Kieckhäfer et al., 2017; Winterstetter et al., 2015). These contradictory results are due to case specific factors and assumptions, for example waste composition and the emissions during the aftercare (Laner et al., 2016).

Assessing the economic feasibility of landfill mining and evaluating the market potential of different fractions as material or as energy is necessary when deciding on landfill mining projects and has been increasingly studied (e.g. Danthurebandara et al., 2015; Frändegård et al., 2015; Kieckhäfer et al., 2017; Van Passel et al., 2013; Wolfsberger et al., 2016). Costs of landfill mining are made up of excavation, sorting and pre-treatment of mined waste material and treatment of waste material, e.g. incineration or material recycling, planning costs and personnel costs (Van Passel et al., 2013; Wolfsberger et al., 2016), and are highly case specific. Costs are dependent on the chosen technologies and the scale of the process, climate and weather during the landfill mining, logistics on the landfill mining site and as well as transportation distances and costs (Danthurebandara et al., 2015; Frändegård et al., 2013; Wolfsberger et al., 2016). Costs evaluations of landfill mining should also take in account the avoided costs from aftercare period, which include costs for leachate collection and treatment and monitoring of gaseous and liquid emissions (Laner et al., 2012; Zhou et al., 2015a). In addition, evaluated revenues and benefits are dependent on the assumptions on the prices of metal, combustibles and landfill space (Danthurebandara et al., 2015; Kieckhäfer et al., 2017; Wolfsberger et al., 2016).

2.2.1 Execution of landfill mining

Landfill mining includes several technology activities. These are removal of vegetation and top soil, possible pre-treatment of landfill to remove environmental risks concerning landfill gas production or leachates, actual waste excavation, separation processes of mined waste materials, treatment processes where material is treated for material or energy utilization and finally land reclamation (Danthurebandara et al., 2015).

Exploration of landfill content could be performed before mining especially in case composition is unknown (Jones et al., 2013). Landfills have been sampled by drilling or excavating samples from a few sampling points, thus examining the content and characteristics of landfills (Kaartinen et al., 2013; Quaghebeur et al., 2013; Wolfsberger et al., 2015). Based on these samples, landfill content can be roughly estimated and thus used in planning what scale of unit processes should be used (Frändegård et al., 2013).

Landfills produce gases and leachate even up to decades after landfill is closed (Chai et al., 2016; El-Fadel et al., 1997), thus the possible pre-treatment of landfill might be needed to avoid risks (Danthurebandara et al., 2015). Landfill can be treated for example by aeration before landfill mining, to reduce odor emission during excavation (Raga et al., 2015). Aeration has also been detected to increase biological stability measured as respiration activity, thus reducing the need for the treatment after landfill mining (Raga et al., 2015). To gain even biological stability in all landfill layers, effective leachate extraction is needed (Raga et al., 2015; Raga and Cossu, 2014).

Waste excavation is started by the removal of top soil and vegetation before landfill mining. These layers are not mixed with waste layers. Excavation is usually performed layer by layer (Raga et al., 2015). After excavation, landfill area can be used for other purposes, but may need remediation or rehabilitation (Burlakovs et al., 2017).

The pre-treatment of waste after excavation is necessary to separate different fractions so that they can be easily utilized as energy or as material (Rotheut and Quicker, 2017; Wagner and Raymond, 2015). The sorting and pre-treatment consist of different mechanical and physical processes, in which different size and material fractions are separated, and one example of the separation process is presented in Figure 3. Quality of landfill material is complex for processing due to high moisture content, irregular shapes and combined material, making the treatment of landfill waste challenging (Wagner and Raymond, 2015). Usually different types of screens are used to separate the several different size fractions and fine fraction. Shredding is often necessary to reduce the size of particles and help in separation as different types of waste may be bound to each other (Jones et al., 2013). Air separators are used to separate light fractions like paper and plastics, magnets to separate ferrous metals and eddy current to separate non-ferrous metals. (Jones et al., 2013). For the FF, also wet separation could be used to separate further e.g. small particles of plastics, paper and textiles (Jones et al., 2013; Wanka et al., 2016).

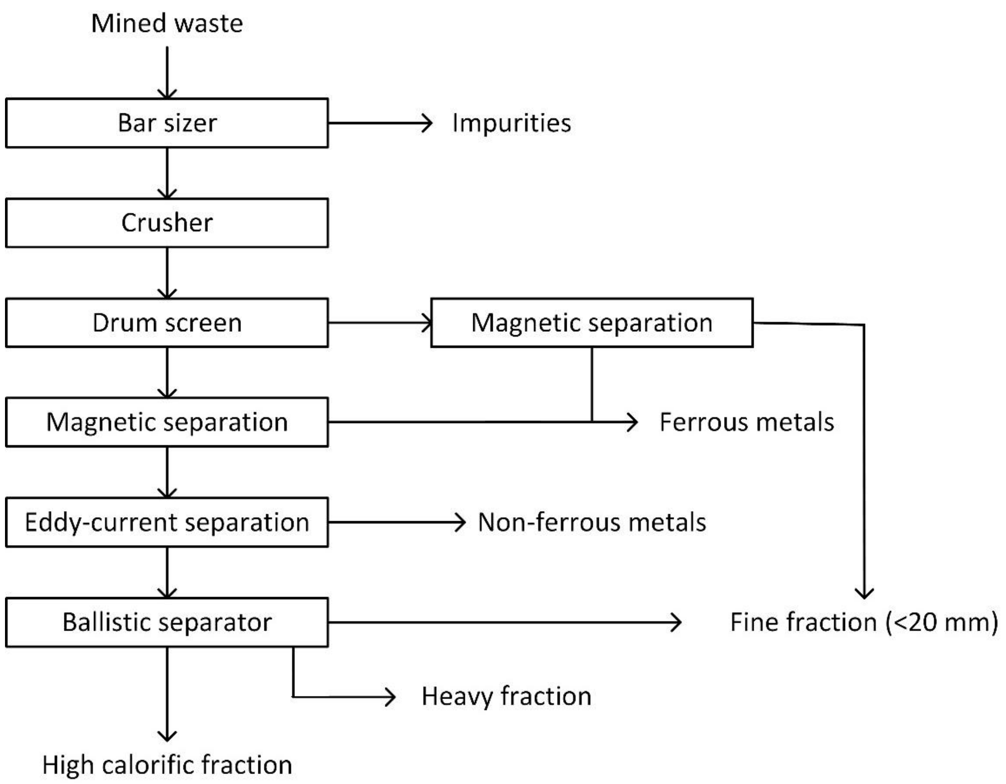

Figure 3. An example of unit processes used in sorting and separation of different waste fractions in landfill mining (adapted from Wolfsberger et al., 2016).

Used unit processes and their effectiveness can change based on what the aim of the landfill mining process is (Frändegård et al., 2013; Kieckhäfer et al., 2017). When the aim is remediation of the landfill area, and not resource recovery, mined waste could be removed to conventional landfill (Frändegård et al., 2013). However, as resource reuse and recovery should be preferred based on waste hierarchy (Wilson et al., 2007), valorization of all possible waste fractions, should be preferred to remediation (Jones et al., 2013). Enhanced landfill mining needs treatment facility with several unit processes to separate fractions (Frändegård et al., 2013; Kieckhäfer et al., 2017). Treatment facilities can be transportable or stationary treatment plants (Frändegård et al., 2013). On the other hand, material could be transported to another location for the treatment, but transportation may be a significant part of the costs of landfill mining. Based on case studies, about 33% of project costs is from excavation and separation operations, while transport costs of waste to recovery facilities are 30% of the costs (Burlakovs et al., 2017).

2.2.2 Revenues of landfill mining

Revenues of landfill mining are formed from waste that can be utilized as material or energy and from land reclamation, and the main drivers for landfill mining are the value of metals and combustibles (Kieckhäfer et al., 2017; Van Passel et al., 2013). Landfills contain many types of mineable materials and significant amounts of secondary raw materials (Jones et al., 2013; Kapur and Graedel, 2006). The total amount of material landfilled before 1995 in European landfills is estimated to be in range between 3 300–11 000 million tons, being up to 5% of material consumption of non-energy, non-food materials and minerals for the next 25 years in EU (Jones et al., 2013). Mined materials from landfill would substitute material from natural sources, thus having positive environmental effects avoiding emissions (Jones et al., 2013).

From landfill mining, metals and plastics could be recycled as materials and stone and soil-type materials could be utilized in constructions (Hogland et al., 2010; Jones et al., 2013; Kieckhäfer et al., 2017; Wagner and Raymond, 2015; Wolfsberger et al., 2016; Zhou et al., 2015a). Metals have high values, but they exist in low amount in landfills (0.5–5 % of landfills content), including ferrous and non-ferrous metals, thus they should be efficiently separated from other waste fractions. Additionally, metals are commonly part of electronics or part of other waste fraction as structural components in landfills (Wagner and Raymond, 2015). The recycling of plastics may not be beneficial as the quality of material is lower than initially, thus they are usually considered for energy utilization, which requires less treatment (Hogland et al., 2010; Kieckhäfer et al., 2017). Benefits from recycling soil-type material can be significant (Zhou et al., 2015a), but utilization is limited by contaminants as further discussed in Chapter 2.3.

The most conventional method for the utilization of combustibles, meaning plastics, paper and cardboard, wood and textiles, from landfill mining is the incineration of waste, but also thermochemical technologies like pyrolysis, gasification and plasma-based technologies have been examined (Bosmans et al., 2013). Incineration is the most commercially proven method and incineration plants have suitable robust feeding and grate technology, thus requiring less pretreatment of feed waste compared with thermochemical technologies (Bosmans et al., 2013; Rotheut and Quicker, 2017). Based on combustion performance, combustibles from landfill mining could be co-incinerated with fresh MSW or refuse derived fuel (Rotheut and Quicker, 2017). However, the incineration of landfill materials will produce high amount of ash/slag (ash content 20–33%) (Jones et al., 2013). Advantages of thermochemical technologies are the less emissions and lower volume of residues compare with incineration and in case of gasification and plasma-based technologies, material recovery is possible from slag (Bosmans et al., 2013; Danthurebandara et al., 2015). Quality of ash is affected by the quality of waste materials, which may contain heavy metals, chlorine, sulfur and other compounds (Bosmans et al., 2013). Additional treatment of ash would improve the quality and usability of ash for example as concrete aggregates or other construction material (Bosmans et al., 2013).

Landfill space in Europe is evaluated to be between 2800 and 4000 km^2 (Hogland et al., 2011), but if areas surrounding landfills are included, as their utilization may be limited due to the nearness of landfills, total area could be up to 6000 km^2 (Jones et al., 2013). Value of land varies based on location, estimated values are 3–300 €/m^2 (Van der Zee et al., 2004; Van Passel et al., 2013), being the highest near expanding cities. Space can be used as recreational area or urban area (Marella and Raga, 2014).

To increase the interest of entrepreneurs and individual companies on landfill mining, the economic benefits must outweigh the costs (Jones et al., 2013). When evaluating the cost-effectiveness of landfill mining, external benefits or costs to society (reduced global foot print, avoided land use for primary mining, sustainable material and energy production) are not usually taken into account by the private sector (Jones et al., 2013; Van Passel et al., 2013). The local authority and governments have a major role in considering these factors: subsidy schemes, taxes, allowances (e.g. EU emission trading system), permits and legislation can be used to increase the interest in the landfill mining (Johansson et al., 2017; Van Passel et al., 2013). This support could be essential for the closure and aftercare of landfill to be the least preferable option compared with landfill mining (Kieckhäfer et al., 2017).

2.3 Fine fraction

FF is the smallest size fraction in landfills, usually the size fraction smaller than 10–25.4 mm (Hull et al., 2005; Hogland et al., 2004; Jani et al., 2016; Kaartinen et al., 2013; Prechthai et al., 2008; Quaghebeur et al., 2013; Raga and Cossu, 2013; Rong et al., 2017; Zhou et al., 2015b). FF consists of soil-type materials, biodegraded waste and very little amount (less than 10 w-% in total) of plastics, paper, cardboard, textiles, metals and decomposed materials (Kaartinen et al., 2013; Quaghebeur et al., 2013). The high volume of soil is due to intermediate and daily covers, which are 15–30 cm layers e.g. of soil, clay or compost, used for example to prevent the odors in landfills, usually up to 20–30% of landfill's content (Hossain and Hague, 2009; Tchobanoglous et al., 1993). A study of a landfill in Belgium reported 1.5 million tons of sand used as intermediate covers in a landfill with 18 million tons of waste (Jones et al., 2013). While some landfill characterization studies lack the category for the finest fraction, however, usually amount of soil or soil-type fraction is high in the same studies (Table 1).

Several studies on landfills have shown that major part of landfill's content is FF, being 40–70 w-% of landfills' material (Table 2) and that is resembles soil. However, contradictory results have also been reported. In open dumpsite in Thailand 18 w-% was FF (Prechthai et al., 2008), presumably due to lack of intermediate layers. In Sweden, FF was only 15–25 w-%, while in soil-type waste was 40–65 w-% of landfill's content (Hogland et al., 2004) suggesting that size of soil particles were larger than in other studies. In addition, the volume of FF increases in landfill as the age of the waste increases, due to decomposition, as in a landfill in Shanghai, where the FF (< 15 mm) was 10 w-% of fresh waste, 18.7 w-% of 4-year-old waste and 45.3 w-% of 10-year-old waste (Zhao et al., 2007).

Characteristics of FF depend on the composition of landfilled waste, age, climate and other factors, thus they vary between landfills. Organic matter content of FF measured as volatile solids (VS) is 24.4–35% of total solids (TS) (Hull et al., 2005) or as total organic carbon (TOC) is 4.7–15.1% (Jani et al., 2016; Kaartinen et al., 2013; Quaghebeur et al., 2013; Raga and Cossu, 2013) (Table 2). FF has a low organic matter content compared with fresh MSW, showing that organic matter of landfilled material has degraded during the landfilling. Significantly higher organic matter contents have been detected in fresh MSW, for example VS content was 76% of TS in residual Danish household waste (Riber et al., 2009) and about 80% in organic fraction of MSW in the United Kingdom (Zhang and Banks, 2013). Organic matter content is dependent on the age of the landfill waste, but the most significant effect has the percentage of landfilled biodegradable waste.

Nitrogen content of FF is between 0.2–9 g N/kg TS and phosphorus content 0.1–7 g P/kg TS, being the highest in the newest landfill (Table 2). The concentrations of nitrogen are low in comparison with the fresh organic waste fraction with the nitrogen content of 30–32 g/kg TS, while the phosphorus content of fresh organic waste (3–6 g/kg TS) is higher than in FF or on similar level with the newest landfill (Tampio, 2016).

Table 2. Amount of fine fraction (FF) and its characteristics in MSW landfills presenting different ages.

Landfill	Age of landfill	Particle size	Amount of FF (w-%)	VS/TS (%)	TOC (%)	N_{tot} (g/kg TS)	P_{tot} (g/kg TS)	Reference
Kuopio, Finland	1–10 years	< 20 mm	38.0–53.9		4.7–5.6			Kaartinen et al., 2013
Houthalen, Belgium	14–29 years	< 10 mm	44 ± 12	-	7.6–12.4	3.9–6.6	-	Quaghebeur et al., 2013
New Jersey, USA	1–11 years	< 25.4 mm	50–52	24.4–35.0	-	-	-	Hull et al., 2005
Northern Italy	5–15 years	< 20 mm	45–55	-	10.7–15.1[a]	4.53–5.27	-	Raga and Cossu, 2013
Måsalycke, Sweden	17–22 years	< 18 mm	14.8–24.7	-	-	3–5	0.82–1.50	Hogland et al., 2004
Nonthaburi, Thailand	3–5 years	< 25 mm	18	-	-	9 ± 2	7 ± 2	Prechthai et al., 2008
Beijing, China	3–30 years	< 5 mm	36.7	-	5.6 ± 1.1	0.24 ± 0.04	0.075 ± 0.012	Rong et al., 2017
Jingmen, China	8–23 years	< 10 mm	75.0 ± 6.8	-	9.1 ± 1.0	1.6 ± 0.5	2.1 ± 2.4	Zhou et al., 2015b
Högbytorp, Sweden	0–5 years	< 10 mm	38	-	5.6	-	-	Jani et al., 2016

- = not available, ± = standard deviation, a = of TS

Heavy metal content varies in different landfills and inside a landfill (Table 3). For example, a landfill in Belgium contains 7–61 mg/kg of arsenic, 0.2–2 mg/kg of mercury, 170–500 mg/kg of lead and 460–800 mg/kg zinc (Quaghebeur et al., 2013), while another landfill contains 2800 mg/kg of zinc and no mercury has been detected (Masi et al., 2014). In landfills, heavy metals tend to be retained in waste as they have formed hydroxides and sulfides during anaerobic phase of landfill and are not soluble, while some heavy metals may be mobilized in aerobic landfills (Rich et al., 2008). Many countries have limit contents for heavy metals in legislation, limiting use of this kind of material e.g. in construction and soil improvement, but heavy metal content is highly case specific. In addition to heavy metals, metals and rare earth elements may be found in landfills, for example iron, magnesium and calcium have been found in higher concentrations to other metals (Bhatnagar et al., 2017; Burlakovs et al., 2016). Separation of metals from FF may be profitable depending on the value of metals and existence in ores (Bhatnagar et al., 2017; Burlakovs et al., 2016).

FF has low heating value 2.2–4.8 MJ/kg TS in a Belgian landfill (Quaghebeur et al., 2013), 0.4–0.9 MJ/kg TS in a Swedish landfill (Hogland et al., 2004) and 1.7 MJ/kg in another Swedish landfill (Jani et al., 2016). In comparison, the average calorific value of plastics from landfill is 19–28 MJ/kg TS, which is also lower than plastic that has not been landfilled (35 MJ/kg) (Quaghebeur et al., 2013).

If landfill is mined, FF should be reused to avoid redisposal. Utilization of FF is determined by its characteristics, such as organic matter content, biological activity and hazardous compounds, which should be analyzed in order to evaluate the direct usability and to assess the potential treatments that could be used to upgrade the quality of FF (Figure 4). Treatment and processing of FF could increase utilization possibilities and different options exists for the treatment of FF based on the initial characteristics (Figure 4). Biological stabilization processes integrated with adding of water or washing are potential methods for quality upgrading as they can convert biodegradable material and remove the easily leachable organic matter and compounds (Raga and Cossu, 2013; Rich et al., 2008; Šan and Onay, 2001; Sponza and Ağdağ, 2004). Utilization possibilities could be chosen based on the resemblances to soil, e.g. construction material (Quaghebeur et al., 2013), soil improvement in landscaping (Rong et al., 2017; Zhou et al., 2015b), oxidation layer in landfills (Pehme et al., 2014) or organic fertilizer in green spaces (Joseph et al., 2007). Utilization as cover soil in landfills is also suggested (Zhou et al., 2015a), but may be limited in the future as number of active landfills is decreasing. Utilization of FF as an energy resource might not be profitable due to low heating value, even incineration has been suggested as an overall method to treat all mined waste without sieving and separation, and may be economically feasible for recovery of metals from fine fraction (Rotheut and Quicker, 2017; Wagner and Raymond, 2015).

Table 3. Heavy metal content of FF from various MSW landfills.

Landfill	Age of landfill	Particle size	Unit	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Reference
Houthalen, Belgium	14–29 years	< 10 mm	mg/kg	7.2–61	3.3–8.5	113–770	107–760	0.19–2.0	46–335	172–500	463–800	Quaghebeur et al., 2013
Lavello, Italy	30–60 years	< 10 mm	mg/kg	73	54	145	1067	-	138	302	2779	Masi et al., 2014
Lavello, Italy	30–60 years	< 4 mm	mg/kg	68	55	117	538	-	89	292	1096	Masi et al., 2014
Nonthaburi, Thailand	3–5 years	< 25 mm	mg/kg	-	4.2	166.6	2 245	-	47.8	132.0	1497	Prechthai et al., 2008
New Jersey, USA	1–11 years	< 25.4 mm	mg/kg TS	9.1	1.2	26	-	0.4	-	55	487	Hull et al., 2005
Florida, USA	3–8 years	< 6.3 mm	mg/kg TS	1.4–6.5	0.4–2.4	9.9–86.4	17.8–308	0.17–1.44	9.0–65.3	10.1–51.7	175–898	Jain et al., 2005
Måsalycke, Sweden	17–22 years	< 18 mm	mg/kg TS	< 0.4	0.9–1.2	47–78	34–36	0.2–0.3	14–15	110–270	180–230	Hogland et al., 2004
Högbytorp, Sweden	0–5 years	< 10 mm	mg/kg TS	5.1	2.1	254	1460	0.7	111.4	240	1848	Jani et al., 2016

- = not available

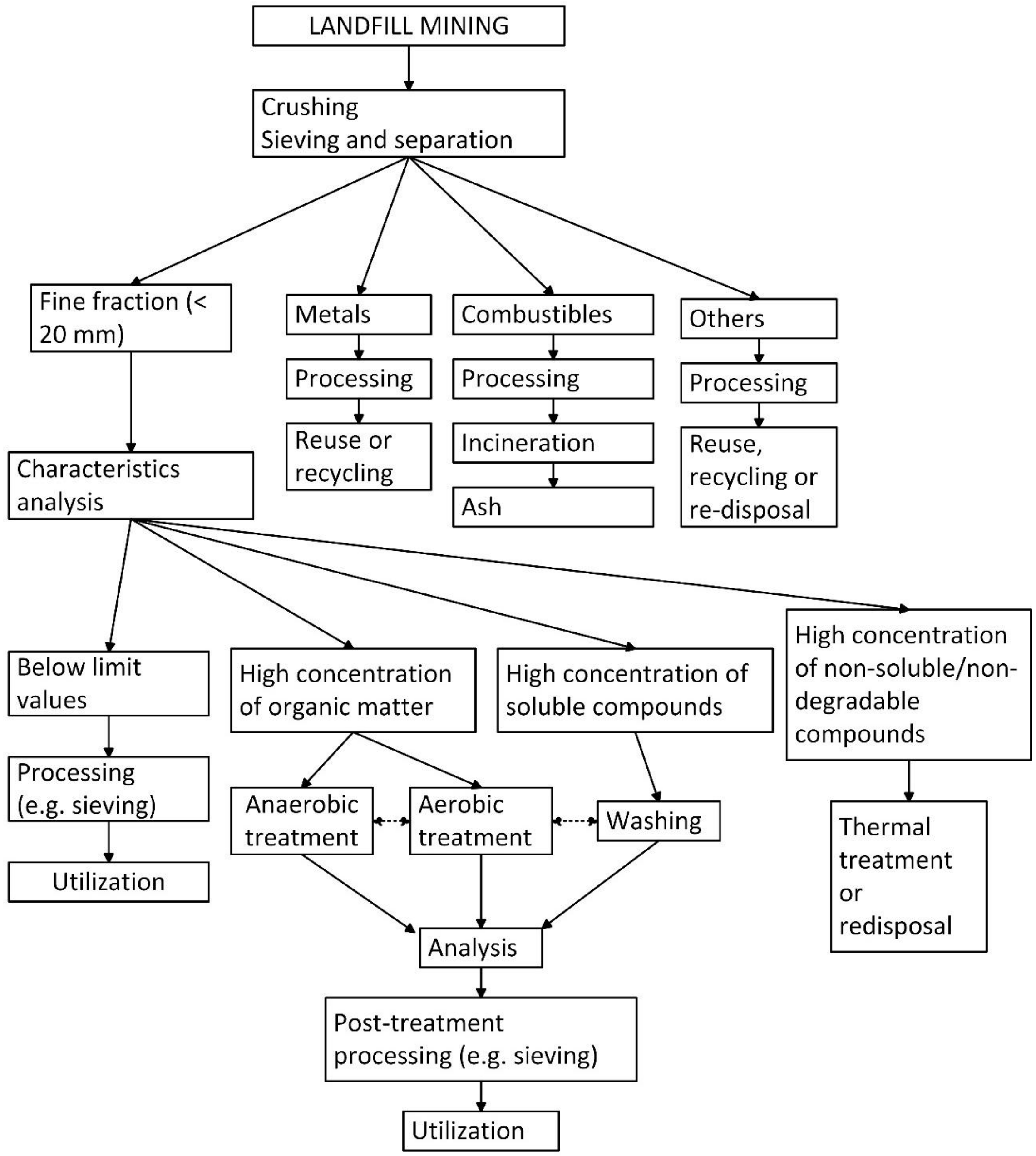

Figure 4. Landfill mining process to separate different waste fractions and options for FF treatment. The selection of the treatment is based on the characteristics of FF.

2.4 Biological stabilization methods

Stabilization improves the quality of the waste, reducing organic and inorganic pollutants (Hrad et al., 2013). The aim of stabilization is to reduce and eliminate the risk for the environment and human health (Rich et al., 2008). Stabilization can be done after the waste is excavated or it can be part of the conditioning of the landfill before the mining process (Raga and Cossu, 2014). The same stabilization methods can be modified for both cases. If stabilization is performed before landfill mining, it may help the actual mining process and reduce emission during the excavation (Raga et al., 2015). However, stabilization after landfill mining enables that stabilization conditions are better optimized for FF.

The stabilization of waste can be achieved using biotechnological anaerobic and aerobic methods, which reduce the organic matter content and biological activity of FF by microbial activity. These methods have been used for the stabilization of fresh MSW or landfill waste (e.g. Erses et al., 2008; Grilli et al., 2012; Morello et al., 2017; Rich et al., 2008) and for the stabilization of FF from landfill (Brandstätter et al., 2015a; 2015b; Raga and Cossu, 2013) as well as for the remediation of contaminated soil (Khan et al., 2004; Rhykerd et al., 1999). Anaerobic and aerobic treatment can be also combined to enhance the degradation of organic matter (Berge et al., 2009; Morello et al., 2017) and to reduce the length of aftercare in landfills (Raga and Cossu, 2014; Ritzkowski and Stegmann, 2013). In both anaerobic and aerobic methods, stabilization generates gaseous emissions and leachates, which need to be controlled and treated.

Bioprocesses are enhanced by the pre-treatment of wastes or by process conditions. Pre-treatment methods could be for example reduction of particle size or temperature treatment (Mali et al., 2012; Tampio, 2016). Process conditions such as temperature, pH, addition of water, inoculum or nutrients and recirculation of leachate are modified to enhance processes (Alkaabi et al., 2007; Mali et al., 2012). The advantages of enhanced stabilization include the improved generation and quality of produced biogas, the reduction of the environmental impact (via leachate and gas emissions, for example), and settlement, which reduces the volume of the waste (Warith, 2002).

2.4.1 Anaerobic stabilization

In the anaerobic process, no oxygen is available for the micro-organisms, thus anaerobic degradation occurs producing biogas, consisting mainly of methane and carbon dioxide. Anaerobic degradation consists of four steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Themelis and Ulloa, 2007). In hydrolysis, organic matter, which consists of carbohydrates, proteins and lipids, is degraded in sugars, amino acids and long chain fatty acids. These products are degraded in acidogenesis to volatile fatty acids and alcohols, which are degraded in acetogenesis to acetic acid and hydrogen. The final products, methane and carbon dioxide are produced in methanogenesis (Themelis and Ulloa, 2007).

The biogas is collected because methane is a strong greenhouse gas and the collected methane can be utilized in heat and electricity production or upgraded to vehicle fuel (Chai et al., 2016). On the other hand, methane utilization is limited by the low methane content of the gas (< 45%), and in that case should be flared to avoid greenhouse gas emissions (Chai et al., 2016; Haubrichs and Widmann, 2006). Methane yields and methane production rates are affected e.g. by the material composition and the moisture content of the material/process (Šan and Onay, 2001) or by the supply of inoculum, for example in the form of digestated sewage sludge to increase microbial activity as studied with fresh MSW (Mali et al., 2012).

2.4.2 Aerobic stabilization

The aerobic process occurs when oxygen is available, as micro-organisms use organic matter to produce mainly carbon dioxide. Suitable micro-organisms already exist in landfills where there is biological activity in landfills. The process requires moisture, oxygen, free pore space and degradable organics (Stentiford and de Bertoldi, 2010). Aerobic treatment can be a more rapid method for stabilization compared with anaerobic treatment, and may degrade compounds that would not be degraded in anaerobic conditions (Reinhart et al., 2002; Ritzkowski and Stegmann, 2012). Organic matter is mostly removed in gaseous form, as formed gas is mainly carbon dioxide but also containing gaseous nitrogen compounds (N_2O, NH_3, N_2) and sulfur compounds (such as H_2S) (Brandstätter et al., 2015a; 2015b). In case of insufficient oxygen amount, some methane may also be produced, thus off-gas should contain oxygen, which guarantees the sufficient aerobic conditions in the aerated material (Bilgili et al., 2006).

The availability of oxygen can be assured by aeration. In landfill bioreactors, there are various aeration concepts, for example, high- or low-pressure aeration (Ritzkowski and Stegmann, 2012). In terms of biocell structure, active or passive aeration (air venting) can be achieved through vertical aeration pipes, while off-gases are collected through parallel off-gas pipes (Ritzkowski and Stegmann, 2012). Off-gas from aerated FF must be treated. Treatment can be carried out via biological processes, e.g., an external biofilter, a methane oxidation cover or a thermal treatment (Ritzkowski and Stegmann, 2012). The aeration rate is dependent on the aerated material and the aeration structure, e.g. the number of aeration pipes and the height and shape of the aerated structure

(Ritzkowski et al., 2006). In the laboratory studies of FF, the aeration rate has been 1–2 L/kg TS/d (Brandstätter et al., 2015a; Hrad et al., 2013; Raga and Cossu, 2013).

In aerobic processes, moisture is evaporated (Reinhart et al., 2002) and addition of water is needed to prevent the material from drying out. Aeration has also effect on the leachate quality as for example ammonium nitrogen is removed more efficiently in aerobic treatment than in anaerobic treatment (Prantl et al., 2006; Raga and Cossu, 2013). On the other hand, aeration is energy consuming, thus even increased aeration would be beneficial, the aeration rate cannot be increased due to the costs of aeration. (Prantl et al., 2006; Reinhart et al., 2002). Bulking material can be mixed with material to increase porosity and thus the gas mobility within the material e.g. in oil-contaminated soil remediation resulting in reduced remediation time (Rhykerd et al., 1999). The addition of bulking material is commonly used in composting processes (Eftoda and McCartney, 2004).

2.4.3 Role of water

Biological processes always need water, thus both anaerobic and aerobic stabilization methods require water. Irrigation is necessary if initial moisture content is low, and in some cases, continuous water addition may be necessary. In the previous experiments with FF in laboratory scale studies, continuous water addition rates have been for example 0.5 L/kg TS/d (Brandstätter et al., 2015a). The continuous water addition leads to the formation of leachate. The leachate from the FF contains, for example, soluble nutrients, anions, and organic matter, much like landfill leachate (Raga and Cossu, 2013). Similarly, to landfill leachates, concentrations vary greatly due to differences in landfill composition, age of landfill, season and climate. Alternatively, water could be added non-continuously, when necessary at the beginning of the treatment to maintain sufficient moisture content, while some leachate is also formed in this case.

The collected leachate requires treatment before discharged. Treatment methods can be similar as in the treatment of landfill leachate, such as settling ponds, membrane processes and chemical oxidation (Bove et al., 2015; Wiszniowski et al., 2006). Depending if water addition continuous or not, collected leachate volumes may be high. Volume of leachate can be reduced by recirculation, which is a technique that has been used widely for fresh solid waste stabilization in laboratory scale reactors, because it can enhance stabilization by increased moisture content and reintroduced nutrients and organic compounds (Francois et al., 2007; Šan and Onay, 2001; Slezak et al., 2015; Sponza and Ağdağ, 2004; Valencia et al., 2009). However, the recirculation rate cannot be increased too much. In anaerobic conditions, too high a recirculation rate may also inhibit methane production due to the creation of acidic conditions (Sponza and Ağdağ,

2004) and the accumulation of inhibitory compounds such as ammonia. In aerobic conditions, more organic carbon was removed with the lower recirculation rate than with the higher recirculation rate (Slezak et al., 2015).

3 Research Objectives

The main objective of this thesis was to assess different biological treatment methods for the stabilization of FF from landfill mining. The hypothesis of the study is that biological stabilization can be used to improve the usability of FF from landfill mining. The main objective was divided into four sub-objectives:

1. To characterize FF mined from two landfills presenting different age and composition
2. To evaluate the potential of anaerobic and aerobic methods to stabilize FF with and without water addition or leachate recirculation
3. To evaluate the effects of the stabilization methods on FF characteristics analyzing organic matter content, biological activity and nutrient content
4. To evaluate the mass balance of the treatment of FF based on two example cases to present landfills different age and size in order to evaluate the cost structure of full scale scenario

4 Materials and Methods

An overview of objectives and experiments in this thesis is presented in Table 4. This study examined FF (< 20 mm) from two different landfills, which had differences on the age of the landfill and the composition of the landfilled waste. FF was characterized on moisture content, organic matter content, nutrient content, particle size distribution and biomethane potential (BMP). In the laboratory scale leach bed reactor (LBR) experiments, both anaerobic and aerobic methods were used to stabilize the FF. The effects of the stabilization were evaluated based on the characteristics and biological activity of the FF. Finally, based on the laboratory experiments, the theoretical evaluation of mass balances and cost structure of FF treatment in full scale of two example cases were evaluated.

Table 4. Objectives and experiments in this thesis

Objective	Experiments	Paper
Characterize FF mined from landfills	Chemical analysis, BMPs	I
Stabilize FF with anaerobic and aerobic methods	LBR experiments	II, III
Evaluate effects of the stabilization methods on FF	Chemical analysis, biological activity	II, III
Evaluate mass balance and cost structure of stabilization of FF in full scale	Theoretical calculations	IV

FF = fine fraction, BMP = biomethane potential, LBR = leach bed reactor

4.1 Sampling sites

The two studied landfills are located in Kuopio in central Finland and in Lohja in southern Finland. Both studied landfills contained mainly MSW while they were filled in different decades thus content of landfill is expected to be different. Other conditions, for example climate, are similar in both locations.

The Kuopio landfill contains MSW landfilled between 2001 and 2011. The composition of landfilled waste was affected by changes in the local waste management system, as biowaste source segregation was initiated in 2004. Since 2009, MSW has been mechanically pre-treated, and only sieved underflow (< 70 mm) has been landfilled. Regional paper, glass, hazardous waste, and metal collection systems were used during the landfill’s history. The landfill area is about 4.5 ha and a depth in the sampling site is 25–35 m. The landfill has a sealed bottom and cover structure according to the EU requirements. For this study, the landfill was sampled when the vertical gas collection system was built in July 2012.

The Lohja landfill was landfilled between 1967 and 1989. Landfill received unsorted MSW, industrial waste, construction waste and soil, but the composition, volume and placement of the waste fractions were not documented. The landfill area is about 5 ha with a depth 15 m. The landfill is closed with a top cover of 2 m of soil but has no bottom structure; the gas collection system was built in 2000. The site was sampled in June 2013.

4.2 Sampling and sample processing

In both sites, samples were taken from vertical wells (0.9 m borehole) drilled with a hydraulic piling rig Casagrande B 170 (Figure 5). After the materials were drilled from landfill, the samples were stored in ambient conditions in dumpsters for 1–2 weeks. During sampling and storing, water may have evaporated or poured off the samples since the water was not collected and the dumpsters were not closed. The samples were sieved and sorted at the site manually from approximately 600 L of sub-samples collected from the dumpsters. Samples were manually sieved (Figure 5) in the Kuopio landfill to separate four particle size categories (> 100 mm, 40–100 mm, 20–40 mm and < 20 mm) and in the Lohja landfill three particle size categories the (> 100 mm, 20–100 mm and < 20 mm). Samples were weighed before and after sieving. Particles smaller than 20 mm were referred as FF. FF was stored in 10 L containers in 7 ± 2 °C.

Figure 5. Hydraulic piling rig used for sampling and manual sieving of mined material (Photos by Joni Göös, Ramboll Finland Oy)

Table 5. Sampling points, sample depths, masses and portion of fine fraction in two studied landfills.

Sampling point	Depth (m)	Sample mass (t)	< 20 mm (w-%)
Kuopio landfill			
KU1 middle	2–10	3.4	38.0
KU1 bottom	10–22	7.2	49.8
KU2 middle	2–14	8.2	50.2
KU2 bottom	14–26	9.9	38.0
KU4 middle	2–15	11.0	41.2
KU4 bottom	15–31	3.3	53.9
Lohja landfill			
LO1 top	2–5	8.8	39.8
LO1 middle	5–9	1.9	58.3
LO1 bottom	9–13	3.5	61.6
LO2	2–10	3.0	59.4
LO3	2–10	4.7	56.5
LO4 middle	2–9.3	10.7	not studied[a]
LO4 bottom	9.3–10	1.0	73.6

a = sample contained only soil and not waste materials

Samples, their sampling points and depths are presented in Table 5. In the Kuopio landfill, the samples (six in total) were taken from three sampling wells at two layers. The cutting points of the layers were chosen so that the upper layer (referred as middle layer) would present approximately the years 2006–2011 and the lower layer (bottom layer) the years 2001–2005. In the Lohja landfill, vertical samples (altogether seven samples) were taken from four wells, of which two wells were studied as single samples, one well was divided into a three-layer sample and one well into a two-layer sample. One sample was not studied further, because it contained only soil-type material without any waste materials. Depth of wells was determined so that layer between soil and waste would not be affected by sampling.

Twelve samples were further characterized. Stored samples were carefully mixed before taking representative sample for each analysis. Two samples from Kuopio and four samples from Lohja were sieved with a sieving column without drying samples. The sieves were 20, 16, 11.2, 8, 5.6, 4, 2, 1 and 0.5 mm.

4.3 Leach bed reactor experiment

LBR experiment was performed as two separate experiments for 173–180 days with four LBRs using FF from Kuopio in the first experiments (Paper II) and FF from Lohja in the second experiments (Paper III). Both anaerobic and aerobic methods were used at 35°C. In anaerobic stabilization, addition of water, recirculation of water or addition of inoculum were compared. In aerobic stabilization, aeration was done either from the bottom or from the top, and bulking material was added in one reactor. Parameters of both experiment are described in Table 6.

LBRs used in these experiments were acryl cylinders with a height of 600 mm and dimeter of 150 mm (Figure 6). LBRs had portholes in the top that were used for water or leachate addition, aeration or gas collection and in the bottom, that were used for leachate collection and aeration. On the bottom of the LBRs was a gravel layer (1–2 cm) separated from FF with mesh. All LBRs had about 2 L headspace volume at the top of the reactor. Gas was collected in gas bags. Tap water or leachate was added six times a day, one hour at a time. Leachate was collected in 1 L bottles, from which leachate was sampled weekly. In case of recirculation of leachate, the sample volume was replaced with tap water. Indoor air was used for aeration and it was moisturized before feeding to LBRs. Gas samples were taken from the tube connecting LBR and gas bag.

Table 6. Parametes of two leach bed reactor (LBR) experiments with fine fraction (FF) from Kuopio and Lohja.

LBR	FF	Water addition	Aeration	Length	Other
FF from Kuopio (Paper II)					
Control-LBR	2.9 kg (middle layer) 2.9 kg (bottom layer)	No	No	180 d	
Water-LBR	2.9 kg (middle layer) 2.9 kg (bottom layer)	Tap water, 0.023 L/h	No	180 d	
Recirculation-LBR	2.9 kg (middle layer) 2.9 kg (bottom layer)	Leachate + tap water 0.023 L/h	No	180 d	
Aeration-LBR	2.9 kg (middle layer) 2.9 kg (bottom layer)	Tap water, 0.014 L/h	2.5 L/h/kg TS From the bottom	180 d	
FF from Lohja (Paper III)					
Anaerobic-LBR	First phase: 5.9 kg Second phase: 5.5 kg	800 mL of tap water in beginning of second phase	No	173 d	
Sludge-LBR	First phase: 5.9 kg Second phase: 5.5 kg		No	173 d	800 mL of sewage sludge in beginning of second phase
Aeration-LBR	First phase: 5.9 kg Second phase: 5.5 kg	Tap water, 0.023 L/h	2.5 L/h/kg TS From the top	173 d	
Bulking-LBR	First phase: 5.9 kg Second phase: 5.5 kg	Tap water, 0.023 L/h	2.5 L/h/kg TS From the top	173 d	Bulking material, 450 g

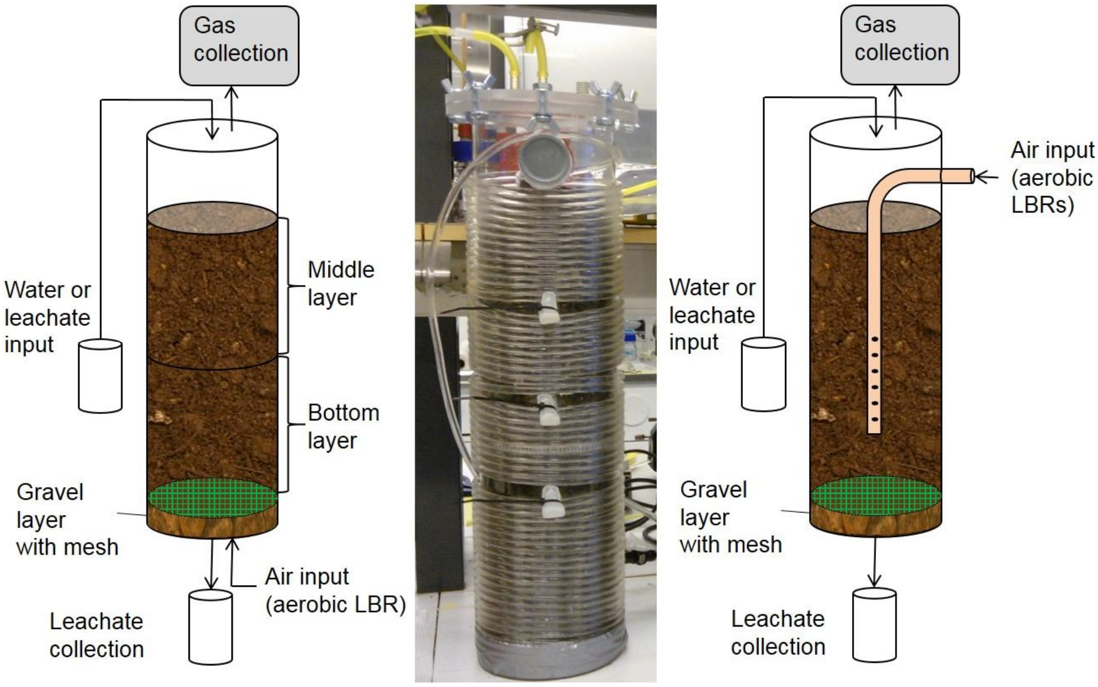

Figure 6. Schematic of leach bed reactors (LBR) used in experiments. On the left side structures for the first LBR experiment with FF from Kuopio landfill and on the right side the second LBR experiment with FF from Lohja landfill. In the middle, the photo of LBR with heating pipes.

For the FF from Kuopio landfill (Paper II), the FF in LBRs was divided in two layers, where the bottom layer contained FF mixed from bottom layer samples of three sampling points and the top layer contained FF mixed from middle layer samples. For the first 30 days of the total 180-day experiment, all four LBRs were operated similarly, in anaerobic conditions without the water addition. After 30 days, one LBR was continued as such (referred as Control-LBR) while the water addition with tap water (Water-LBR) and leachate recirculation (Recirculation-LBR) were started in two respective LBRs. In the fourth LBR (Aeration-LBR), aeration and water addition were started on day 66.

One FF sample was mixed from six samples from Lohja and used in four LBRs (Paper III). All four parallel LBRs were operated anaerobically, without water addition or leachate collection, for 101 days (phase 1). Before the beginning of the second phase, LBRs were opened, and FF mixed again and redistributed to LBRs. In phase 2, two of the LBRs were operated anaerobically and two aerobically. In one anaerobic LBR, 800 mL of tap water (Anaerobic-LBR) was added at the beginning of the second phase (day 102), while, in the other (Sludge-LBR), 800 mL of digested sewage sludge was added as inoculum to increase microbial activity at a ratio 0.02 g VS in sludge per VS in FF. In one aerobic LBR (Bulking-LBR), 450 g bulking material (wood chips) was added to increase air mobility, while the other LBR (Aeration-LBR) contained FF only. Phase 2 ended after 72 days, all four reactors were opened and material was sampled for the analysis.

4.4 Chemical analyses

FF was characterized after the landfill mining and after stabilization. From sieved samples, VS of each size fraction was analyzed, except fractions larger than 20 and 16 mm were combined before they were analyzed. FF was also characterized after stabilization to evaluate the effect of stabilization methods. The analyses and methods used in this thesis are summarized in Table 7.

Table 7. List of analysis methods used in experiments.

Analysis	Method	Paper
TS and VS	APHA 2540 SFS 3008	I II, III
Leaching test	EN 12457-4	I
COD	SFS 5504 Soluble COD filtered with GF/A	I-III
pH	WTW ProfiLine pH 3210 with SenTix 51 electrode	I-III
CH_4 and CO_2 content	Shimadzu GC-2014 TCD gas chromatograph	I-III
Gas volume	Water replacement method	I-III
TKN	SFS-EN 13342 and SFS-EN 13654 Soluble N extracted with water at a 1:5 ratio	I-III
NH_4-N	Thermo Scientific Orion High-Performance Ammonia Electrode	II, III
P	Pretreatment: ISO 11464 and ISO 11466 Analysis: ICP-AES Soluble P extracted with water at a 1:5 ratio	I-III
Anions (Cl^-, NO_2^-. NO_3^-, SO_4^{2-})	Dionex ICS-1600 ion chromatograph	II, III
DOC and DIC	SFS-EN 1484 (Shimadzu TOC-5000)	III

4.5 Biological assays

BMP was determined for assessing methane potential of FF from all sampling points and from combined sieved samples from size fraction below 11.2 mm. Assay was performed in 1 L glass bottles with total volume of 700 mL. FF samples were added at a ratio of 0.5 g $VS_{inoculum}/VS_{sample}$, while inoculum (from Viinikanlahti sewage treatment plant, Tampere, Finland) volume was 350 mL (Lohja samples) or 500 mL (Kuopio samples). To achieve the total volume of 700 mL buffer solution (50 mL of 42 g/L $NaHCO_3$) and distilled water were added to the bottles. Bottles were flushed with nitrogen and incubated at 35 °C in a water bath. The biogas produced was collected in aluminum gas bags. The BMP assays were continued until methane production became negligible (< 5 mL CH_4/d) after 130–160 days.

Residual methane potential after stabilization experiments was measured using a batch test in duplicate 1 L glass bottles filled with a sample of 300 g at 35 °C. After 30 days, 300 mL tap water was added to each bottle to increase the moisture. At the beginning of the experiment and after the water addition, the gas phase was sparged with nitrogen gas.

The respiration activity was determined using OxiTop® (WTW) system, in accordance with Binner et al. (2012) with some modifications. 50–70 g sample of FF before and after stabilization was weighed in 1 L reaction bottle. If necessary, moisture was adjusted by adding distilled water. Soda lime pellets were added in plastic decanters, bottles were closed with OxiTop-measuring heads and bottles were kept at 20 ± 1 °C for 7 days. Soda lime pellets absorbed CO_2, thus causing negative pressure which is measured by OxiTop-measuring heads. Experiment was conducted in triplicate.

4.6 Mass balance and cost structure calculations

Anaerobic and aerobic stabilization methods were compared using full scale mass balance and cost structure calculations. Calculations were based on two example case landfills (Landfill 1 and Landfill 2), which were chosen to represent landfills different size and age. Characteristics of these landfills are presented Table 8.

Table 8. Characteristics of chosen example case landfills

Parameter	Landfill 1	Landfill 2
Age	20–40 years	10–15 years
Size	10 000 t landfilled waste	300 000 t landfilled waste
Share of FF	70%	60%
Characteristics of FF		
Moisture content	25%	30%
VS content	10% of ww	15% of ww
Methane production potential	6 L CH_4/kg VS	17 L CH_4/kg VS
Nitrogen	3 g/kg TS	5 g/kg TS
Phosphorus	1.0 g/kg TS	1.2 g/kg TS

ww = wet weight

Design parameters (Table 9) of biological stabilization methods were chosen based on the results of experiments in this thesis (paper I-III) and other literature. These parameters were used in the calculation of mass balance for two example landfills, evaluating initial and final masses in both treatments as well as volumes of irrigation water and leachate and volumes of formed landfill gas and exhaust gases.

Table 9. Design parameters for chosen treatment methods

Parameter	Anaerobic treatment	Aerobic treatment
Duration	1 year	1 year
Aeration	No	Passive aeration
Organic matter removal	2.5–5 g VS/kg TS	15–50 g VS/kg TS
Irrigation	Addition of water in the beginning, 1 m^3 per m^2 treatment area	Addition of water in the beginning, 1 m^3 per m^2 treatment area
Evaporation	5% of added water	10% of added water

The cost structure of the FF treatments used in the case studies was assessed without including the costs of the actual landfill mining, i.e., the excavation, processing, and sieving of the waste material from the landfill. The costs included the physical structures used in FF treatment and the operational costs, excluding planning costs. The costs were evaluated based on practical cases in remediation and waste sector in Finnish conditions, thus there may be differences in the costs of materials, electricity and labor when compared to other countries. The costs used in calculations were: the bottom structure, including the leachate collection system 30 €/m^2; the gas collection or aeration pipes 5 €/m^2; the treatment of the leachate 4 €/m^3, the treatment of the biogas 0.25 kWh/m^3, aeration and the treatment of the exhaust gas 0.14–0.28 €/t FF being higher in smaller landfill, electricity 35 €/MWh, machine work 5 €/t (filling and excavation), and personnel 5000 €/month (Paper IV).

5 Results and discussion

5.1 Characteristics of fine fraction

Characteristic studies on FF were made to examine the content of the FF to detect the biological stability (Table 10). The results of the study show that characteristics on landfill vary between the landfills and inside the landfill in different sampling points and depths. Moisture content is higher on the bottom of the landfill. Organic matter content and biological methane potential are lower in older, Lohja landfill than in newer, Kuopio landfill and also lower in bottom layers of landfill than in middle or top layers, because the older waste fraction have had the longer time to degrade. In addition, COD and nutrient content are lower in Lohja landfill than in Kuopio landfill.

Kuopio landfill contained 38–54% of FF and Lohja landfill 40–74%, being on the same level as in other studies, in which FF has been 40–70% of landfills content (Hogland et al., 2004; Hull et al., 2005; Quaghebeur et al., 2013; Raga and Cossu, 2013). In addition to sieve size, fraction of FF is dependent on sieving methods. Manual sieving may increase slightly amount of FF compared to mechanical sieving (Kaartinen et al., 2013) for example due to possible clogging while mechanically sieving wet material. Used sampling method in this study, drilling, may have reduced size of particles compared with some other sampling methods like bucket excavator or grab crane (Hölzle, 2017; Jani et al., 2016; Sormunen et al., 2008). Six samples (two from Kuopio and four from Lohja landfill) were sieved without drying the samples to examine particle size distribution of FF and to evaluate characteristics in different particle sizes (Figure 7). Sieving showed that 78–93% of the samples were below size fraction 11.2 mm and 51–74% below 5.6 mm (except one sample having 40% under 5.6 mm), thus large amount of landfills content has very small particle size (Paper I). In previous studies where FF has been defined as size fraction below 10 mm, FF has consisted 38–53% of landfills content (Jani et al., 2016; Quaghebeur et al., 2013; Rong et al., 2017).

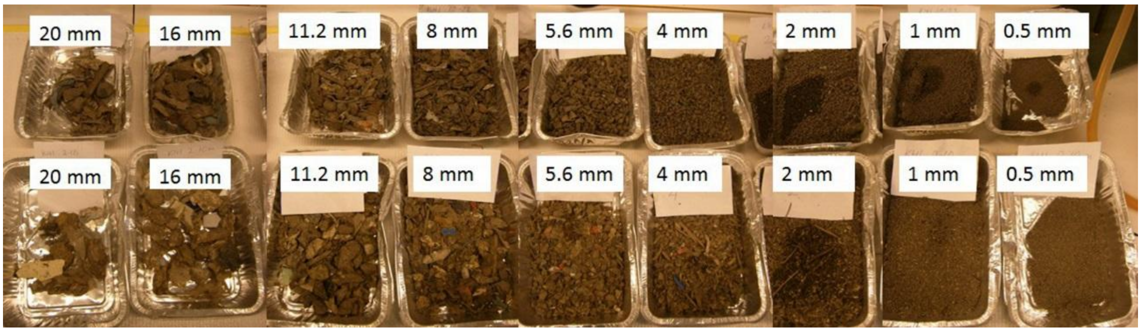

Figure 7. Different particle sizes of sieved FF samples KU1 middle layer (lower row) and KU1 bottom layer (upper row).

Moisture content varied between 20–54% being higher in bottom layers. On average, FF from Kuopio landfill had higher moisture content than FF from Lohja landfill. Organic matter content (VS/TS) in FF was 17–27% in the middle layer from Kuopio, 15–24% in the bottom layer from Kuopio and 6–24% from Lohja, while BMPs were 19–27 L CH_4/ kg TS, 0.4–10 L CH_4/kg TS and 1–10 L CH_4/kg TS respectively (Paper I). Similar methane potential has been detected in FF from Swedish landfill, 4.7 L CH_4/kg TS (Jani et al., 2016). The middle layer samples, which were the newest samples, had the highest organic matter content and BMP compared with the bottom layers and the samples from old landfill Lohja because older waste materials have had the longer time to degrade. On the contradictory, one sampling well in Lohja was divided in three layers, and the bottom layer had the highest organic matter content (24% of TS) compare with the middle (17% of TS) and the top layer (8% of TS). This is probably due to changes in landfilled waste materials, as construction material and soil in addition to MSW had been landfilled in Lohja. The same trend was detected also in BMP, where methane production was 10 L CH_4/kg TS in bottom layer, 6.7 L CH_4/kg TS in middle layer and 1.2 L CH_4/kg TS in top layer.

In sieved samples, organic matter content was on similar level in different particle sizes, thus organic matter was found in all particle sizes. BMP of fraction below 11.2 mm was 60–100% compared to FF below 20 mm. Based on these characteristics, using smaller sieve size than 20 mm for separation of FF from mined waste materials, VS content is same while less methane is produced per VS.

Soluble COD varies slightly in between landfills, FF samples from Lohja had the lowest values 0.5–0.8 g/kg TS while higher values were measured from Kuopio landfill: 0.8–1.3 g/kg TS in bottom layers and 3.8–4.6 g/kg TS in middle layers. In Lohja, soluble COD content is not dependent on the layers being 0.63–0.79 g/kg TS in different layers, thus it can be expected that in an old landfill soluble materials have been more efficiently leached already.

FF contains less organic matter and has lower BMP when compared with landfilled waste fraction with larger particle size (Table 10). Organic matter in FF is mainly degraded organic matter, while the size fraction over 20 mm contains waste that has not been yet degraded for example biowaste, paper and cardboard. Thus the samples from larger particle sizes have higher organic matter content, as shown in Table 10 for shredded landfill waste samples and mechanically treated waste samples (Paper I, Sormunen et al., 2008). In all samples from Ämmässuo, Kujala and Kuopio landfills, waste samples have higher organic matter content and BMP in the newer waste samples compared with older waste samples (Paper I, Sormunen et al., 2008). In contrast, organic matter content and BMPs in the landfilled waste fraction are significantly lower than the fresh organic fraction of MSW, for example food waste from the UK has organic matter content 91–93% of TS and methane potential 462 ± 19 L CH_4/kg TS (Tampio, 2016).

Respiration activity, which measured biological oxygen consumption, is low in FF being 1.4–2.4 g O_2/kg TS (Papers II–III). These are lower than in other studies with FF, which have respiration index 9.8–14.4 g O_2/kg TS, while increasing in deeper layers of landfill (Raga and Cossu, 2013) and 1.7–3.2 g O_2/kg TS (Brandstätter et al., 2015a). Respiration activities of FF are significantly lower to fresh MSW fractions (80 g O_2/kg TS) (Morello et al., 2017).

In FF nitrogen content varies between 1.4–8 g N/kg TS and phosphorus content 1–1.5 g P/kg TS (Paper I). Similar nitrogen content has been detected previously in FF 0.2–9 g N/kg TS and phosphorus content 0.1–7 g P/kg TS (Hogland et al., 2004; Prechthai et al., 2008; Quaghebeur et al., 2013; Raga and Cossu, 2008; Rong et al., 2017; Zhou et al., 2015b). In fresh organic MSW, nutrient contents are higher (32 g N/kg TS and 4 g P/kg TS) and are even higher after anaerobic digestion process in digestate, which can be used as fertilizer (Tampio et al., 2015). Even FF has low nutrient content, it has been detected to increase the nutrient content of soil and promote the growth in pot experiments (Prabpai et al., 2008; Rong et al., 2017; Zhou et al., 2015b).

Table 10. Characteristics of FF from studied landfill compared to other landfilled waste fractions

	Fine fraction			**Landfill waste**		
	Kuopio – middle layer	Kuopio – bottom layer	Lohja	Shredded waste (< 20mm) from Ämmässuo landfill	Shredded waste (< 20 mm) from Kujala landfill	Mechanically treated waste (< 70 mm) from Kuopio landfill
TS (%)	55.7–67.3	46.2–56.6	59.6–81.6	50–57	65–73	20.8–47.3
VS (%)	11.7–16.9	8.8–11.2	4.9–14.3	27–35	12–35	11.9–14.7
VS/TS (%)	17.4–27.3	15.5–24.3	6.0–24	55–65	16–59	25.2–60.1
pH	6.89–7.15	6.83–7.57	7.16–7.88	7–7.6	n.a.	6.3–7.2 [a]
$COD_{soluble}$ (g/kg TS)	3.79–4.60	0.79–1.27	0.53–0.79	12–20	2.5–9.7	1.4–5.3 [a]
N_{tot} (g/kg TS)	3.5–8 [b]		1.4–5	2.4–4.6	2.4–5.2	n.a.
$N_{soluble}$ (g/kg TS)	0.041–0.398 [b]		< 0.012–0.425	n.a.	n.a.	n.a.
P_{tot} (g/kg TS)	< 1.0–1.1 [b]		< 1.0–1.5	n.a.	n.a.	n.a.
$P_{soluble}$ (g/kg TS)	< 10 [b]		< 10-12	n.a.	n.a.	n.a.
BMP (L CH_4/kg TS)	19.2–26.6	0.4–9.9	1.2–10	21–68 [c]	8–44 [c]	10–140
Respiration activity (g O_2/kg TS)	2.4 [d]	1.4 [d]	1.88 [d]	n.a.	n.a.	n.a.
Reference	Papers I-II	Papers I-II	Papers I,III	Sormunen et al., 2008	Sormunen et al., 2008	Paper I

n.a. = not available, a = fraction < 30 mm, b = sample mixed middle and bottom layers from Kuopio, c = waste shredded to fraction < 50 mm, d = analyzed from mixed sample

5.2 Stabilization of fine fraction

FF from two landfills was stabilized using anaerobic and aerobic stabilization in two experiments with FF from Kuopio and Lohja. In the first experiment with FF from Kuopio (Paper II), anaerobic treatment (Control-LBR) were compared with anaerobic treatment with water addition (Water-LBR) and leachate recirculation (Recirculation-LBR) and aerobic treatment (Aeration-LBR). In the second experiment with FF from Lohja (Paper III), moisture addition (Anaerobic-LBR) and inoculum addition (Sludge-LBR) were compared in anaerobic conditions, while aerobic treatment was examined in two LBRs with and without bulking agent (Bulking-LBR and Aeration-LBR).

Methane production was examined during the initial anaerobic phase in both experiment and then in all five anaerobic LBRs (Figure 8). Methane production yield was in Kuopio experiment 16–18 L CH_4/kg VS (Paper II) after 180 days and in Lohja experiment 9–10 L CH_4/kg VS (Paper III). Methane concentration increased during the treatment to 40–65 V-%, which is similar as in landfill gas (Chai et al., 2016). Methane production rate was slower in experiment with old FF from Lohja compared to experiment with FF from newer Kuopio landfill, probably due to higher initial biodegradable organic matter content.

The methane production was slightly reduced in LBRs when water addition and leachate recirculation were started on day 30 in experiment with FF from Kuopio compared with Control-LBR (Figure 8), however, methane was produced constantly until the end of the experiment, while in control-LBR without any water addition, methane production was slowed significantly after 100 days of experiment. Leachate recirculation did not have significant effect on the methane production compared with the water addition thus it can be expected that process was not inhibited compared to other LBRs. In experiment with FF from Lohja, methane was production continued until the end of the study. The sewage sludge addition was estimated to increase methane production up to 30% by increased microbial activity, but major part of the increased methane production is due to methane produced by sludge itself (Paper III). Furthermore, the addition of sludge affected the stability and characteristics of FF.

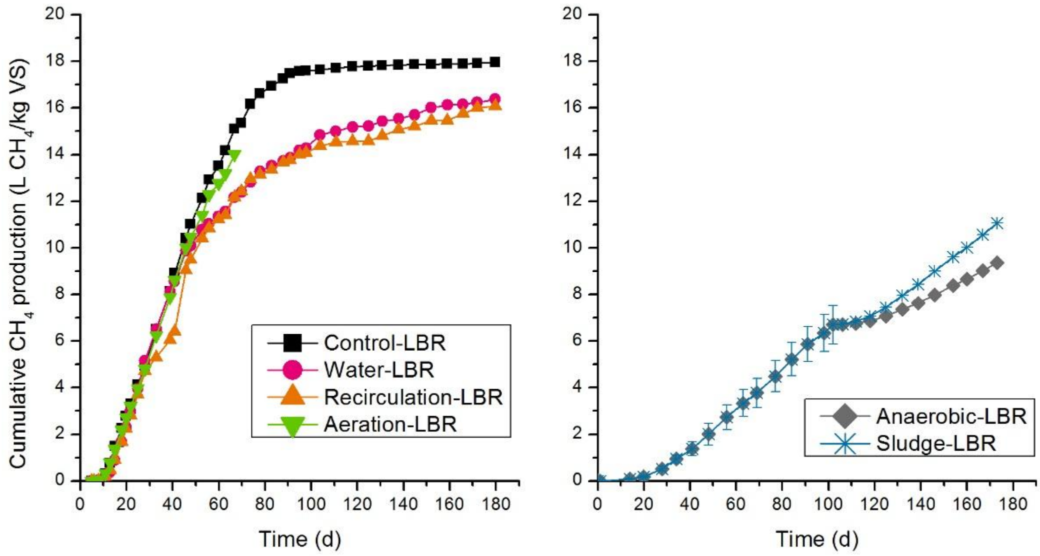

Figure 8. Cumulative methane production in LBRs during Kuopio experiment (left), where water addition and leachate recirculation was started on day 30 and Lohja experiment (right)where phase 1 was for day 1–101 and phase 2 for days 101–173 (Papers II and III).

In the aerobic treatment, FF in LBRs were aerated using indoor air, with the initial CO_2 concentration of 0.1–0.2 V-%, and as in anaerobic conditions organic matter was degraded into carbon dioxide, CO_2 concentration was increased to 0.5–1.7 V-%. Cumulative CO_2 production was in Lohja experiment 190–195 L CO_2/kg VS in aerobic reactors, while anaerobic reactors produced 4–5 L CO_2/kg VS during the same time (Paper II). Aeration was started in both experiment after the initial anaerobic phase (66–100 days), after which methane production was not detected in any aerobic LBR. Overall, carbon removal in gaseous form was significantly higher in aerobic conditions than in anaerobic conditions (Paper III). Previous studies have also shown that more carbon is removed in gaseous form in aerobic than in anaerobic treatment (Brandstätter et al., 2015a; Raga and Cossu, 2013).

Leachate from five LBRs contained COD, ammonium nitrogen and anions, while concentration varied during the experiments depending on the treatment. The initial concentrations of COD varied highly in different LBRs, being between 500–2500 mg/L (Figure 9). In anaerobic conditions, some peaks in COD concentrations were also detected (Paper II), possibly due to hydrolysis of organic matter and release of these materials in leachate (Slezak et al., 2015). However, in all reactors with the clean water addition, independent on anaerobic and aerobic conditions, COD concentration decreased to 100–300 mg/L at the end of the experiment. Similarly, also the concentrations of anions, dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) were reduced during the addition of clean water. Concentrations were reduced due to washout mechanism (Šan and Onay, 2001; Sponza and Ağdağ, 2004), as compounds were not recirculated back to material.

When leachate was recirculated, organic matter and anions were introduced back to LBR, thus the concentrations of COD and anions were not reduced, unless they were converted into gaseous phase or retained in the FF. COD concentration of leachate was 1000 mg/L from LBR with leachate recirculation at the end of the experiment (Paper II), and it can be expected that this is also independent on the availability of oxygen. In the similar experiment with FF from 5–15 year old landfill, COD concentrations were 1000–2000 mg/L after anaerobic treatment and 300–500 mg/L after aerobic treatment, while the initial concentrations were also higher (3900–8000 mg/L) (Raga and Cossu, 2013). In addition, the concentrations of anions were higher in LBRs with leachate recirculation than in LBR with clean water addition. Leachate recirculation may cause inhibition as inhibitive compounds are not removed from process (Francois et al., 2007; Šan and Onay, 2001; Sponza and Ağdağ, 2004), but in the present study no inhibition was detected in methane production compared with the water addition (Paper II).

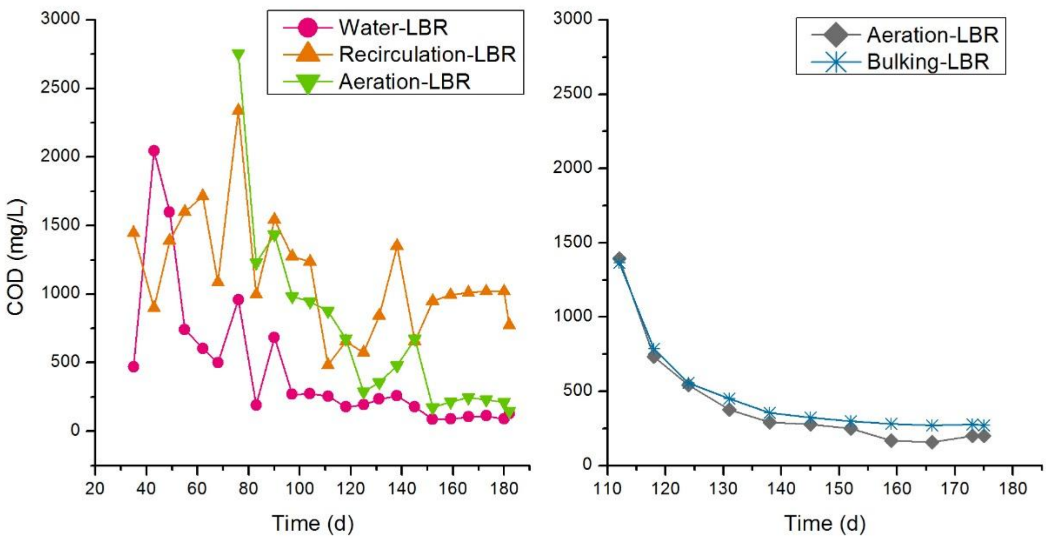

Figure 9. COD concentrations in leachates were reduced during experiments (Papers II (left) and III (right)).

Nitrogen was detected in the form of ammonium, nitrite and nitrate in leachate samples (Figure 10). In anaerobic conditions, ammonium nitrogen had similar trends as COD, as with clean water addition it was significantly reduced from initial 300-600 mg/L to 100 mg/L while concentrations were higher (300 mg/L) when leachate was recirculated (Paper II). Nitrite was not detected, while nitrate concentrations were low, less than 40 mg/L. However, aeration removed ammonium nitrogen. When aeration was started, ammonium nitrogen was decreased fast from the leachate from initial concentrations about 300 mg/L to 0 mg/L in 2 weeks (Lohja experiment, Paper III) or 4 weeks (Kuopio experiment, Paper II) (Figure 10). In Kuopio experiment nitrite and nitrate concentration in leachate are increased, while in Lohja experiment nitrite and nitrate are close to zero initially, peaking in 100–150 mg/L (nitrite) in 150–350 mg/L (nitrate). The increase in the nitrite and nitrate concentration is due to nitrification, which has been detected also previously in the aeration of FF from landfills in the laboratory scale studies (Brandstätter et al., 2015b; Raga and Cossu, 2013), until nitrite and nitrate are washed out in leachate.

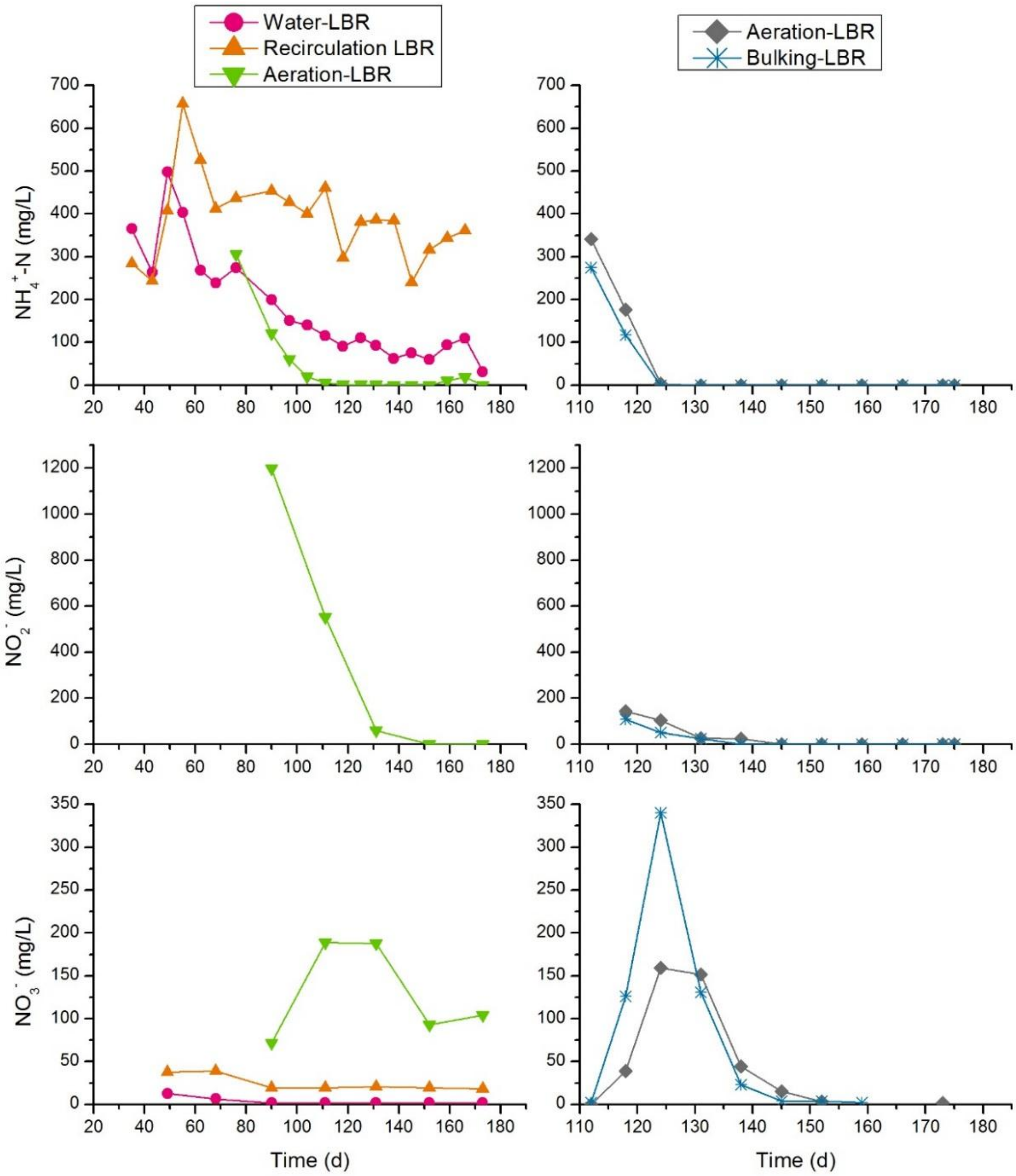

Figure 10. Ammonium nitrogen, nitrite and nitrate concentrations in leachate during LBR experiment show that ammonium nitrogen was removed fast in aerobic reactors due to nitrification (Papers II (left) and III (right)).

In these experiments, gaseous nitrogen was not examined but more nitrogen is removed in gaseous form under aerobic conditions than under anaerobic conditions (Brandstätter et al., 2015b). Gaseous nitrogen is mainly N_2, but also N_2O and NH_3 are emitted in aerobic conditions (Brandstätter et al., 2015b). Nitrous oxide (N_2O) is strong greenhouse gas (Ravishankara et al., 2009), thus off-gas treatment is also necessary after aerobic treatment.

5.3 Fine fraction after stabilization

Characteristics of the FF after stabilization are the most essential for the utilization of FF. A combination of different parameters, such as waste composition, leachate quality, and biological activity, can be used to evaluate the stability of waste (Ritzkowski et al., 2006). In this study, the aim was to reduce organic matter content and biological activity, thus VS, residual methane potential and respiration activity were analyzed after the LBR experiments. Nutrient content was measured to evaluate fertilizer potential of FF after stabilization.

In all LBRs, organic matter content was reduced during the treatment or was at the same level before and after the treatment (Table 11). The lowest VS contents were measured after the aerobic treatment in both Kuopio and Lohja experiments, while difference compared with VS contents after the anaerobic treatment was not significant due to the high standard deviation of average VS values. In the anaerobic treatment in Kuopio experiment, VS were lower in the middle layer of LBR than in the bottom layer, even initially VS was higher in the middle than the bottom layer. This is probably due to higher initial content of biodegradable organic matter as the middle layers had newer waste than the bottom layers. Also in the previous studies, the organic matter content of treated FF (measured as TOC) had no significant difference between anaerobic and aerobic treatment (Brandtsätter et al., 2015a; Raga and Cossu, 2013).

Residual methane potential was measured without the addition of inoculum, while water was added to increase moisture content. Residual methane potential was 0–2.9 L CH_4/kg VS (Table 11, Papers II-III). The highest residual methane concentration was in the middle layer from Control-LBR in experiment with FF from Kuopio, which had no water addition and had the lowest moisture content compared with other samples. Low moisture content apparently decreased the microbial activity during the LBR experiments (Grilli et al., 2012). Also the addition of sludge in LBR almost doubled residual methane potential, the most probably due to increased microbial activity and methane produced by inoculum itself (Paper III).

Table 11. Total solids (TS), volatile solids (VS), residual methane potential and respiration activity initially and after stabilization in both experiments with standard deviation (±).

	Kuopio experiment (Paper II)					
	Layer[a]	Initial	Control-LBR	Water-LBR	Recirculation-LBR	Aeration-LBR
TS (%)	middle	60.0 ± 5.8	70.3 ± 4.3	66.3 ± 1.8	65.5 ± 4.6	61.7 ± 2.1
	bottom	52.3 ± 5.2	60.2 ± 1.2	60.1 ± 1.6	60.4 ± 3.2	62.7 ± 3.1
VS (% of TS)	middle	22.3 ± 4.2	14.6 ± 2.7	12.8 ± 2.0	14.3 ± 1.4	13.1 ± 1.3
	bottom	20.0 ± 2.0	17.1 ± 1.7	18.5 ± 4.5	18.7 ± 1.1	13.1 ± 1.7
Residual methane potential (L CH_4/kg TS)	middle	n.a.	2.9 ± 0.5	0.9 ± 0.9	0.9 ± 0.3	0.8 ± 0.3
	bottom	n.a.	0.4 ± 0.3	0.3 ± 0.1	0.8 ± 0.4	0.0 ± 0.0
Respiration activity (mg O_2/g TS)	middle	2.4 ± 0.5	2.2 ± 0.2	2.0 ± 0.2	2.1 ± 0.1	0.9 ± 0.2
	bottom	1.4 ± 0.2	2.0 ± 0.2	2.8 ± 0.1	1.7 ± 0.3	1.2 ± 0.4
	Lohja experiment (Paper III)					
		Initial[b]	Anaerobic-LBR	Sludge-LBR	Aeration-LBR	Bulking-LBR
TS (%)		66.4 ± 7.3	59.0 ± 2.6	61.7 ± 2.1	59.4 ± 1.9	59.1 ± 1.7
VS (% of TS)		15.8 ± 5.0	18.2 ± 2.4	16.5 ± 2.1	16.6 ± 2.2	16.8 ± 2.1
Residual methane potential (L CH_4/kg TS)		n.a.	1.3 ± 0.1	2.4 ± 0.1	1.1 ± 0.1	1.1 ± 0.1
Respiration activity (mg O_2/g TS)		1.9 ± 0.4	2.3 ± 0.2	1.8 ± 0.5	1.1 ± 0.1	0.9 ± 0.3

n.a = not analyzed, a = middle and bottom layers in Kuopio experiment, b =before phase 1 of experiment

The lowest residual methane potentials were measured in aerobic LBRs. In Aeration-LBR in Kuopio experiment, no residual methane potential was detected in bottom layer, as LBR was aerated from the bottom while in middle layer residual methane potential was 0.8 L CH_4/kg VS. This shows that the availability of oxygen throughout the FF affects aerobic degradation and the aeration method affects the degradation along the reactor height, which suggests the importance of aeration systems. As in the experiment with Lohja FF, characteristics were not examined from different layers, it cannot be stated if aeration method was better than one used in experiment with Kuopio FF.

Residual respiration activity was the lowest in aerobic LBRs (0.9–1.2 mg O_2/g TS) than in anaerobic LBRs (1.7–2.8 mg O_2/g TS) (Table 11, Papers II and III). Respiration activity was on the same level or even higher than initially in FF. Respiration activity was not significantly dependent on the layer of sample in LBR. Thus, it appears that aeration is more efficient in reducing biological activity than anaerobic treatment. Similar results of respiration activity being lower after aerobic treatment than anaerobic treatment have been detected previously in the similar type laboratory scale studies with FF mined from landfill (Brandstätter et al., 2015a; Raga and Cossu, 2013).

The measured final total nutrient contents were 3.3–4.3 g/kg TS for nitrogen and 1.0–1.3 g/kg TS for phosphorus (Table 12, Papers II and III), being on the initial level. During the stabilization, soluble nitrogen was removed from FF while soluble phosphorus content was initially under the detection limit. Removal of soluble nutrients was due to washout mechanism as more nitrogen was removed from LBRs with clean water addition, and there were no significant differences in total nutrient content after anaerobic and aerobic stabilization, which was similar result to the previous study (Raga and Cossu, 2013).

Table 12. Nutrient content initially and after biological stabilization in both experiments.

	N_{tot} (g/kg TS)	$N_{soluble}$ (g/kg TS)	P_{tot} (g/kg TS)	$P_{soluble}$ (mg/kg TS)
Kuopio experiment (Paper II)				
Initial	4	0.2	1.3	< 10
Control-LBR	3.3	0.5	1.1	< 10
Water-LBR	3.4	0.1	1.3	< 10
Recirculation-LBR	3.3	0.4	1.1	< 10
Aeration-LBR	3.8	0.1	1.1	< 10
Lohja experiment (Paper III)				
Initial[a]	4.2	0.31	1.6	< 10
Anaerobic-LBR	4.2	0.18	< 1.0	< 10
Sludge-LBR	4.3	0.23	1.1	< 10
Aeration-LBR	4.2	0.04	1.3	< 10
Bulking-LBR	3.8	0.04	1.2	< 10

Standard deviation less than 10% of average results, a = before phase 1 of experiment

After stabilization, FF could be utilized for example as construction material, soil improver, cover or oxidation layer in landfill or organic fertilizer (Joseph et al., 2007; Pehme et al., 2014; Quaghebeur et al., 2013, Rong et al., 2017; Zhou et al., 2015a; Zhou et al., 2015b). Possible uses have different requirements for the material. Use of FF as fertilizer has limit values for heavy metals and organic pollutants in legislation of several countries, while the limits vary (Amlinger et al., 2004; Zhou et al., 2014). In EU, it has been proposed (European Commission, 2016) that nitrogen content of solid organic fertilizer should be 2.5% of dry matter, while if not fulfilled, material could be considered as soil improver. There are no quality requirements for biological stability, while for example suggested stabilization criteria for determination of landfill stability include biogas production below 10 L/kg TS and respiration activity below 4 mg O_2/g TS (Ritzkowski et al., 2006). If FF would be used in construction, for example as road building material, FF must fulfill the same criteria as primary building materials, structurally engineered criteria as well as environmental criteria (Wanka et al., 2016), which may require lower biological activity than use as soil improver. Disposal to landfill should be last option for the FF, but EU legislation sets limit value of organic matter content of 10% of TS for material disposed in landfill (Council decision, 2003/33/EC). According to this criteria, FF should not be disposed in landfills as organic matter content was higher than 10% of dry matter after all treatment methods (Table 11).

As FF contains also small amount, less than 10 w-% other waste materials like metals and plastics (Kaartinen et al., 2013; Quaghebeur et al., 2013), these materials could be removed before FF is utilized. While removal of impurities from FF is difficult with conventional mechanical treatment, wet separation techniques could be used (Jones et al., 2013; Wanka et al., 2016). Wet mechanical processes separate materials based on their densities, as light fraction (plastics and paper) float and dense materials (glass and minerals) accumulate at the bottom (Wanka et al., 2016). Wet separation have been widely used in ore, sand and gravel production and they can be used to separate impurities from FF, thus increasing possibilities to utilize FF (Wanka et al., 2016)

5.4 Fine fraction treatment concept

In this study, the basic idea of the FF treatment concept is to perform FF stabilization treatment separately, after landfill mining, in a heap, a biocell or a windrow (Paper IV). As during the treatment, gaseous and leachate emissions are formed, treatment structure needs to be designed to enable the collection of gases and leachate, and take into account the treatment of gases and waters. Concept of the treatment design for both anaerobic and aerobic treatment is presented in Figure 11.

The solid structures of the treatment process, meaning bottom and sides of a cell or a heap, should be materials that do not allow the penetration of water or gases, for example asphalt, concrete, compacted clay or plastic liner (Benson et al., 2007; Oonk et al., 2013). Leachate collection would be built on top of the bottom structure, horizontal pipes inside the drainage material like gravel (Benson et al., 2007; Oonk et al., 2013). The top structure on top of FF and irrigation and gas collection structures is made to cover a heap or a cell and prevent the irrigation of FF due to precipitation (Oonk et al., 2013). Height of the structure could be between 4–8 meters (Oonk et al., 2013).

Irrigation needs to perform evenly, in which horizontal pipes inside the structure have been found to be effective (Benson et al., 2007). Irrigation is necessary at least in the beginning, and in case water washing is needed, continuous water addition and leachate recirculation should be considered. Water for irrigation can be collected rainwater or surface water. In addition, leachate can be recirculated (Paper II) and it would reduce collected leachate volume that needs to be treated. Inhibition needs to be taken into account when planning leachate recirculation, while in laboratory study no inhibition was detected in comparison with clean water addition (Paper II).

Anaerobic treatment needs vertical gas collection pipes. Gas collection can be active or passive, and passive system is typically used when gas volumes are small like in case of FF from landfills (9–18 L CH_4/kg VS). Pipes convey biogas to the permeable layer, from which gas is collected (Oonk et al., 2013; Willumsen and Barlaz, 2010). As biogas contains methane, it can be treated for energy use or flared if utilization as energy is not profitable to minimize greenhouse gas emissions (Chai et al., 2016; Haubrichs and Widmann, 2006).

Aerobic treatment requires vertical pipes, where the half of the pipes is used for aeration (passive air venting or active aeration) and the half to collect off-gases for treatment (Ritzkowski and Stegmann, 2012). Active aeration is energy consuming increasing costs (Reinhart et al., 2002), thus passive system, meaning natural convection, may be suitable for treatment of FF. In addition, more water is evaporated during active aeration compared to passive aeration, thus passive aeration requires less irrigation (Kasinski et al., 2016). Passive aeration can be as efficient as active aeration, while proper ventilation system is needed to provide enough oxygen (Barrington et al., 2003; Kasinski and Wojnowska-Baryla, 2014; Ritzkowski and Stegmann, 2012). Addition of bulking material may be necessary to increase porosity of material and mobility of gases and water. Bulking materials are commonly used in composting processes and treatment of contaminated soils (Eftoda and McCartney, 2004; Rhykerd et al., 1999). Sufficient amount of bulking material should be further studied, as addition of bulking material did not increase stability in laboratory scale study (Paper III), also different types of bulking materials have different qualities depending for example on moisture content (Barrington et al., 2003). Off-gases contain for example nitrous oxide and ammonia (Brandstätter et al., 2015b) and methane in case of insufficient aeration, thus treatment of off-gases is necessary using for example biofilter or thermal treatment.

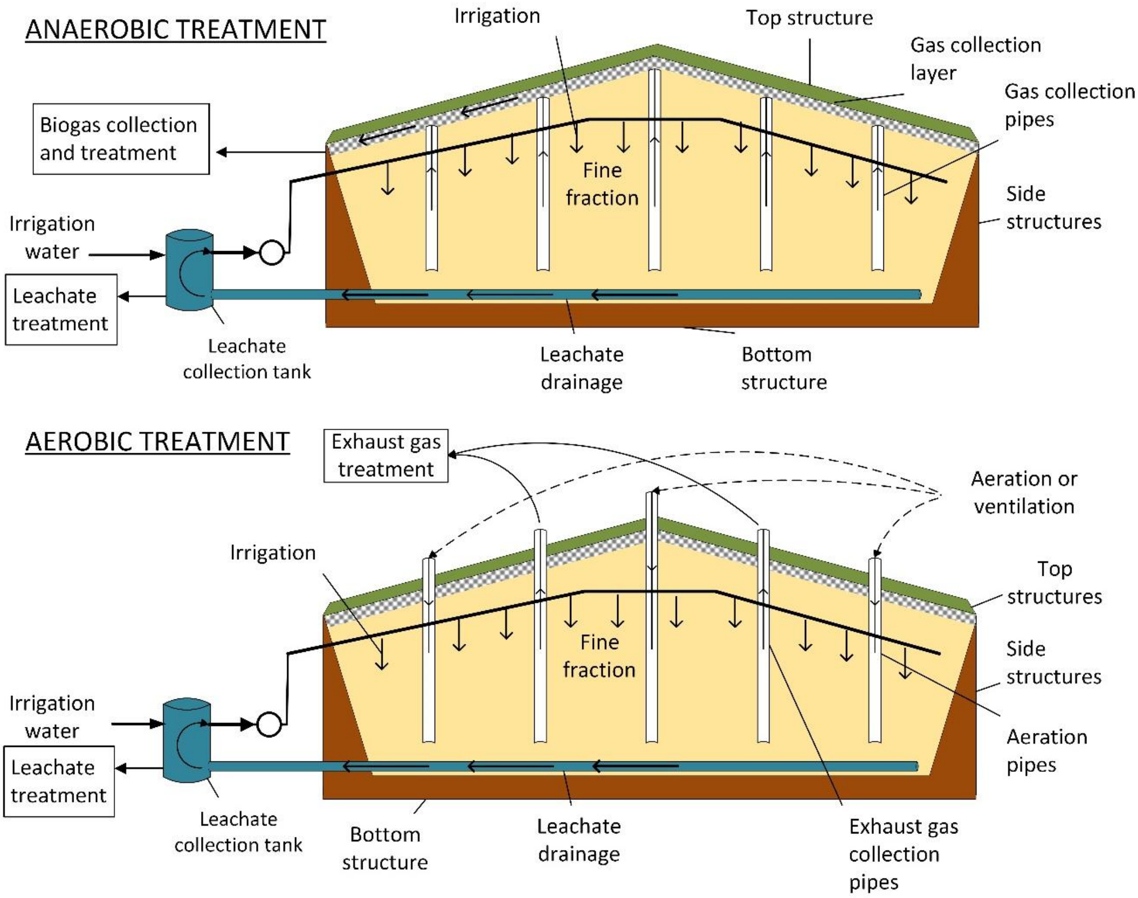

Figure 11. Concept of anaerobic and aerobic treatment system for landfilled fine fraction (not to scale), inspired by Oonk et al. (2013) and Mertoglu et al. (2006) (Paper IV).

5.4.1 Mass balance of example cases

The mass balance of two example cases was based on design parameters presented in Table 9. The inputs and outputs are presented as the masses of the FF, TS, VS and nutrients in tons in Table 13. Organic matter is reduced in both anaerobic and aerobic treatments, but it is reduced more rapidly in the aerobic treatment, which has been shown to be more efficient in reducing organic matter content and biological activity in laboratory experiments (Brandstätter et al., 2015a; Papers II and III). Volumes of needed irrigation water and formed leachate are evaluated based on the laboratory experiments, but may differ in full scale application due to challenges in scaling up. Even distribution of water in FF may be more difficult in full scale as water channels and dry pocket can be formed more than in laboratory scale.

Table 13. The mass balance of two example landfills used for FF treatment under anaerobic and aerobic conditions based on 1 year treatment time (Paper IV)

	Landfill 1		**Landfill 2**	
	Anaerobic treatment	Aerobic treatment	Anaerobic treatment	Aerobic treatment
Inputs				
FF				
Total mass	7 000 t	7 000 t	180 000 t	180 000 t
TS	5 250 t	5 250 t	126 000 t	126 000 t
VS	700 t	700 t	27 000 t	27 000 t
Nitrogen	16 t	16 t	630 t	630 t
Phosphorus	5.5 t	5.5 t	152 t	152 t
Water	1940 m^3	1940 m^3	50 000 m^3	50 000 m^3
Outputs				
FF				
Total mass	7 000 t	6 900 t	180 000 t	173 700 t
TS	5 200 t	5 170 t	125 400 t	120 000 t
VS	680 t	620 t	26 400 t	20 700 t
Nitrogen	15 t	13 t	580 t	520 t
Phosphorus	5 t	5 t	150 t	150 t
Leachate	1 840 m^3	1 750 m^3	47 000 m^3	45 000 m^3
Methane	4 900 m^3		459 000 m^3	

As methane production potential is evaluated to be 6 L CH_4/kg VS in old landfill and 17 L CH_4/kg VS in new landfill, the total methane volume is 4 900 m^3 from the landfill 1 and 459 000 m^3 from landfill 2. In LBR experiments, methane concentrations were 40–65 % during the experiments (Papers II-III), thus total biogas volumes may be up to 12 000 m^3 in landfill 1 and 1,1 Mm^3 in landfill 2. However, it can be expected that methane production rate and concentration decrease to the end of the treatment period, especially in case of newer FF, as in the laboratory scale study with FF from Kuopio (Paper II). However, due to the climate impact of methane, even low biogas volumes need to be collected and treated.

Irrigation water is generated into leachate, retained in FF or evaporated. If expected initial moisture content to be near the water holding capacity of FF, the most of irrigation water forms leachate. It is evaluated that more water is retained in anaerobic treatment than in aerobic and that 5% of water is evaporated in anaerobic treatment and 10% in aerobic treatment, but these are dependent on the climate. Active aeration would increase evaporation and thus need for irrigation (Read et al., 2001). Soluble nutrients are removed in leachate, reducing the nutrient content of FF.

5.4.2 Cost structure of example cases

The cost structure of two example cases for anaerobic and aerobic treatment is presented in Table 14. Feasible costs for the treatment of FF are important as previous studies have shown that in case FF is redisposed, disposal costs are the large fraction of the total costs of landfill mining (Wolfsberger et al., 2016).

The costs of physical structures are directly proportional on the size of the planned treatment heap. If the height of a treatment heap is 4 m, required treatment area is 1950 m^2 for landfill 1 and 50 000 m^2 for landfill 2. For the larger landfill, the overall time of landfill mining is evaluated to be several years, thus the treatment of FF could be performed for example in three cycles, and thus physical structures can be reused in different cycles. This would reduce required treatment area from 50 000 m^2 to 17 000 m^2. Thus the total costs of structures would be 87 200 € for landfill 1 and 743 000 € for landfill 2. Increasing the height of the heap to 8 m, which has also been used in treatment of MSW (Oonk et al., 2013), would reduce treatment area and costs in half.

Table 14. Cost structure and evaluated total costs of FF treatment in euros (€), when treatment area is 1950 m^2 for landfill 1 and 17 000 m^2 in three cycles for landfill 2 (Paper IV).

	Landfill 1		**Landfill 2**	
	Anaerobic treatment	Aerobic treatment	Anaerobic treatment	Aerobic treatment
Structures				
Bottom structure	58 000	58 000	500 000	500 000
Gas collection pipes	9 700	-	83 000	-
Aeration pipes	-	9 700	-	83 000
Cover structure	19 500	19 500	160 000	160 000
Treatment				
Treatment of leachate	7 400	7 000	187 000	180 000
Treatment of biogas	100	-	4 000	-
Aeration and treatment of exhaust gas	-	2 000	-	25 000
Operation				
Personnel	120 000	120 000	720 000	720 000
Machine work	70 000	70 000	1 800 000	1 800 000
Total costs (€)	285 000	286 200	3 454 000	3 468 000
Total costs (€/t FF)	40.7	40.9	19.2	19.3

\- = no costs for the treatment

For operational costs, 1-year treatment time was used for both anaerobic and aerobic treatment. Leachate volume is significant for total costs. In this example, FF is irrigated initially with clean water and leachate volumes are 1700–1900 m^3 from landfill 1 and

45 000–47 000 m^3 from landfill 2. If FF would be irrigated continuously at rate 0.2 L/kg TS/d which has been used in the laboratory scale studies and recirculating leachate as much as possible, total leachate volumes would increase to 35 000 m^3 in landfill 1 and to 500 000 m^3 in landfill 2 multiplying leachate treatment costs. However, in some cases washing the FF might be necessary in addition to anaerobic and aerobic treatment.

In these example cases, total costs would be 40 €/t FF for landfill 1 and 20 €/t FF for landfill 2. With passive aeration and without continuous irrigation, there is no significant difference between anaerobic and aerobic treatment methods. As aerobic treatment has been found more efficient treatment methods (Papers II-III), treatment time could be reduced. Reducing treatment time to 0.5 year instead of 1 year, costs would be reduced to 75%.

In this calculation, costs of mining operation like excavation and processing of waste material and site-specific costs like transportation of materials and price of treatment space are not taken into account in this calculation. Costs of excavation and processing vary in different landfills depending on size of landfill and processing technologies (Van Passel et al., 2013). Transportation costs can be limited by performing the treatment near landfill mining site, but transportation of FF to existing treatment location reduces need for investment in new structures and treatment facilities. Mobile machines and plants can be reused at other sites thus reducing capital investment per mined landfill (Frändegård et al., 2015).

The presented cost structure lacks the benefits obtained from treatment. Produced biogas may be utilized as energy source, reducing costs. After treatment, the value of FF is increased compared with untreated FF and it could replace some other material like natural soil in construction. In addition, as the redisposal of FF is avoided, original landfill space increases its value in other purpose. Value of the land vary between 3 and 300 €/m^2 depending on the location and use (Van der Zee et al., 2004; Van Passel et al., 2013). When landfill is mined and FF is not redisposed in landfill, aftercare costs of landfilling are avoided, including costs of maintenance of landfill structures and monitoring of emissions and receiving systems while aftercare period is at least 30 years according to EU landfill directive or even longer if necessary (Laner et al., 2012).

6 Conclusions and recommendations for future research

This thesis studied FF mined from MSW landfills and the aim was to characterize FF, evaluate the potential to use biological methods to stabilize FF and effects of stabilization methods on characteristics of FF and also evaluate mass balance and cost structure of treatment of FF in full scale. FF is major fraction, 40–70% of landfills' content, thus it has significant effect on the execution and economics of landfill mining. FF is the smallest particle size fraction of landfill and resembles soil type material, containing less than 10% of waste materials. Characteristics of FF vary in different landfills depending on the composition landfilled waste, age and location of a landfill. In this thesis, FF from two MSW landfills was examined. The first landfill in Kuopio was new, landfilled 2001–2011 and FF was 38–54% of landfills content. The second landfill in Lohja was old, landfilled 1967–1989 and had higher content of FF (40–74%) than the first landfill.

The studied FF contained low amount of organic matter (VS 6–27% of TS), while organic matter content was the highest in the samples from new landfill, as in old landfill organic matter content has been mainly degraded. The nutrient content varied in both landfills, for example total nitrogen was 3.5–8 g/kg TS in Kuopio landfill and 1.4–5 g/kg TS in Lohja landfill. The highest BMP values were measured in the newest, 1–5 year old, middle samples from Kuopio (19–27 L CH_4/kg TS), while similar BMP was both in the bottom layer, 6–10 year old samples of Kuopio landfill (0.4–10 L CH_4/kg TS) and in 24–46 year old samples from Lohja landfill (1–10 L CH_4/kg TS). Respiration activities were also low in both landfills (1.4–2.4 g O_2/kg TS).

Biological stabilization of FF was studied using anaerobic and aerobic methods in laboratory scale reactors for 173–180 days. Methane production was found to range from 9 to 18 m^3 CH_4/t VS being lower in FF from older Lohja landfill than in FF from newer

Kuopio landfill. Aerobic treatment removed organic matter efficiently as carbon dioxide and carbon removal in gaseous form was calculated to be higher during aerobic treatment (15 g C/kg TS) than anaerobic treatment (0.5–0.8 g C/kg TS). Carbon removal in leachate was not dependent on the availability of oxygen, as adding clean water removed organic matter similarly in anaerobic and aerobic conditions, while leachate recirculation introduced organic matter back to FF. In addition, soluble compounds were washed out from LBRs during clean water addition. Composition of leachate was similar in anaerobic and aerobic treatment, except ammonium nitrogen, which is converted into nitrate and nitrite fast in aerobic treatment

Biological stabilization was expected to increase stability of FF and thus improve usability of FF from landfill mining. Both anaerobic and aerobic treatment turned out to be suitable for reducing organic matter content and biological activity. Aerobic treatment was found to be more efficient resulting in lower respiration activity and residual methane potential of treated FF than after anaerobic treatment. Soluble nutrient content was removed more from FF with clean water addition than from FF without water washing or with leachate recirculation. Moisture addition was necessary for biological activity during the treatment as the highest residual methane potential was measured from dried FF. The addition of sludge as inoculum increased methane production but had negative impact on treated FF for example increasing residual methane potential.

Based on the laboratory scale experiments, the treatment concept for the biological stabilization of FF was designed. When the treatment concept is planned for the treatment of FF after landfill mining, the process can be better optimized for FF. Anaerobic treatment would include biogas collection and treatment, irrigation and leachate collection. Aerobic treatment would include aeration, off-gas collection and treatment, irrigation and leachate collection. If possible, leachate can be used in irrigation, thus reducing the need for clean water, especially if continuous irrigation is needed to remove soluble compounds from FF. Physical structures at the bottom, sides and the top of a heap or a cell need to enable gas and leachate collection, while preventing uncontrolled emissions.

Treatment costs are dependent on the volume of FF, treatment area, treatment time and chosen treatment technology. In large landfill mining projects, which last several years, it is beneficial to perform FF treatment in several cycles, thus physical structures can be scaled for one cycle and reused. Treatment time could be reduced especially in aerobic treatment, which was found more efficient in the laboratory scale studies, and reducing treatment time in half would reduce costs by 25%. Active aeration would increase costs compared with passive aeration, but most likely would also reduce treatment time. If continuous irrigation is needed, the volume of leachate for treatment is high despite leachate recirculation. Investment costs could be minimized by utilizing existing treatment facilities and planning future use for built structures. Overall, the treatment of FF is beneficial as the value of FF is increased, the value of previous landfill space is increased and the aftercare costs of disposed FF are avoided.

In the future, studies on the biological stabilization of FF should include pilot scale experiments to evaluate the removal of organic matter and biological activity in large scale. In pilot scale studies, it would be interesting to study needed irrigation rate and how much leachate can be collected. In aerobic treatment, aeration rate and even distribution of aeration air should be examined. In addition, removal of other compounds, such as heavy metals or hazardous compounds should be analyzed, as these compounds have significant effects of potential reuse of materials. The treatment of FF has potential to increase the value of FF for various utilization possibilities, for example as soil improver and in construction, while at the same time landfill space is relieved for other purposes, landfill aftercare and emission during that are avoided and the need for natural soil could be reduced.

In addition to the evaluation of biological stabilization, future studies should examine the sieving and separation process of mined waste materials in order to separate FF efficiently from other fractions. The costs and environmental impacts of biological stabilization of mined FF could be compared e.g. with the in situ stabilization of landfill. The utilization possibilities of FF should be further studied as FF has potential for example as soil improver for example mixed with other organic fractions.

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