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Development and Evaluation of a Filter-Bed-Based System for Full-Scale Treatment of Industrial Landfill Leachate

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Pille Kängsepp
2008

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Akademisk avhandling som för avläggande av filosofie doktorsexamen kommer att offentligen försvaras på Kemicentrum, Sölvegatan 39, Lund, Hörsal B, lördagen den 27 september 2008, kl 10.15, med vederbörligt tillstånd av matematisk-naturvetenskapliga fakulteten vid Lunds Universitet. Avhandlingen försvaras på engelska. Fakultetsopponent är Prof. Rune Bakke, Faculty of Technology, Telemark University College, Porsgrunn, Norway.

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Development and Evaluation of a Filter-Bed-Based System for Full-Scale Treatment of Industrial Landfill Leachate

Abstract

It is well known that nature offers an abundance of materials and processes that can be adapted in low-tech fashion, to fabricate systems capable of removing pollutants from wastewater. During the last decade, extensive basic and engineering research on such natural treatment systems have led to improvement of the quality of discharged wastewaters. However, the mechanisms of natural treatment and their application for leachate treatment in temperate climates are not fully understood due to a large number of interacting factors. A complex, interdisciplinary research project was therefore undertaken to develop strategies for construction and evaluation of full-scale leachate treatment systems. The main objective of this Thesis was to develop and characterise a vertical-flow filter-bed-based system for treatment of leachate from an industrial landfill containing shredder residue (SR) of end-of-life vehicles (ELV) and white goods. The strategy for development of the treatment system involved laboratory batch and column experiments, and progressed to long-term field studies in a pilot plant at the landfill site. Several locally available low-cost natural or residual waste materials and mixtures thereof were investigated for removal of different types of pollutants from leachate. A mixture of peat and carbon-containing ash was found to be most efficient for removal of a number of toxic metals and various polar and non-polar organic compounds. Effective removal of phenolic compounds and an abundance of phenol-degrading and heterotrophic bacteria observed in the filter show that it also acts as a biofilter.
Given the climatic character of Sweden, 60–70% of the annual leachate volume might be produced during a few months, resulting in possible overloading of the full-scale filter-bed-based treatment systems. To ensure stable hydraulic loading of the biofilter system, a retention pond with a storage volume of about 23,000 m3 was needed to even out annual leachate volumes up to about 35,000 m3 in the particular location. Such retention pond also equalises peak concentrations and allows pre-treatment of leachate. Sedimentation was found to be essential to decrease the initial load of pollutants (e.g. iron and suspended solids) and reduce the risk for clogging. Long-term performance of the vertical-flow biofilter was studied in the pilot and the full-scale plants. Efficient year-round performance of a filter-bed-based system can be achieved in a temperate climate zone like Sweden, if measures are taken to protect the filter from seasonal extreme of climate. The results obtained show that on-site natural treatment systems based on readily available materials with low environmental "footprint" can be used for treatment of leachates from an industrial landfill.

Key words: Landfill, shredder residue, leachate treatment, pond, vertical-flow filter-bed, peat, carbon-containing ash, organic and inorganic pollutants

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The thesis is based on the following papers, referred to in the text by their roman numerals (*e.g.* Paper I).

I. **Selection of adsorbents for treatment of leachate - batch studies of simultaneous adsorption of heavy metals.**
Sõukand, Ü., Kängsepp, P., Kakum, R., Tenno, T., Mathiasson, L. and Hogland, W.
JOURNAL OF MATERIAL CYCLES AND WASTE MANAGEMENT, 2008 (Manuscript).

II. **A batch-test-based approach towards an environmentally friendly adsorbent for leachate treatment of organic and inorganic pollutants.**
Kängsepp, P., Svensson, B.-M., Mårtensson, L. and Mathiasson, L.
JOURNAL OF ENVIRONMENTAL MANAGEMENT, 2008 (Submitted).

III. **Column studies aiming at identification of suitable filter materials for pollutant removal from landfill leachate.**
Kängsepp, P., Svensson, B.M., Mårtensson, L., Rosenquist, D., Hogland, W. and Mathiasson, L.
INTERNATIONAL JOURNAL OF ENVIRONMENTAL AND WASTE MANAGEMENT, 2008, 2 (¾). (In press).

IV. **Biofilter-based treatment of leachate from an industrial landfill containing shredder residues of end-of-life vehicles and white goods.**
Kängsepp, P. and Mathiasson, L.
WASTE MANAGEMENT, 2008 (Submitted).

V. **Performance of a full-scale biofilter with peat and ash as a medium for treating industrial landfill leachate – a three-year study of pollutant removal efficiency.**
Kängsepp, P. and Mathiasson, L.
WASTE MANAGEMENT AND RESEARCH, 2008 (Accepted).

VI. **Hydraulic performance of a full-scale peat and ash biofilter in treatment of industrial landfill leachate.**
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WASTE MANAGEMENT AND RESEARCH, 2008 (Accepted).

VII. **Bacterial community dynamics along a treatment system for leachate from a landfill of automobile shredder residues.**
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(Manuscript).

VIII. **Leachate treatment in newly built peat filters: a pilot-scale study.**
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CONTRIBUTION BY THE AUTHOR TO THE DIFFERENT PAPERS

Paper I. The author was involved in outlining the experimental strategy, performed a minor part of the experiments, was highly involved in scientific discussions and actively contributed to the writing of the manuscript.

Paper II. The author was highly involved in outlining the experimental strategy and evaluating the data, and contributed actively to scientific discussions and writing of the manuscript.

Paper III. The author was involved in outlining the experimental strategy, performed a minor part of the experiments, was highly involved in scientific discussions and wrote the major part of the paper.

Paper IV. The author was highly involved in outlining the experimental strategy, was responsible for all field work, performed all sampling and the major part of the analyses, was responsible for evaluation of all data and wrote the major part of the paper.

Paper V. The author was highly involved in outlining the experimental strategy, was responsible for all field work and sampling, performed part of the analyses, was responsible for all data evaluation and wrote the major part of the paper.

Paper VI. The author outlined the experimental strategy, performed or supervised all the experiments, was responsible for all data evaluation and wrote the major part of the paper with assistance from the co-authors.

Paper VII. The author outlined the experimental strategy for the field work, was highly involved during sampling, was involved in scientific discussions and was responsible for writing the manuscript.

Paper VIII. The author was highly involved in outlining the experimental strategy, performed and supervised a substantial part of the experiments and sampling in the field, was responsible for data evaluation, and wrote the major part of the paper with assistance from the co-authors.

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POPULÄRVETENSKAPLIG SAMMANFATTNING

Det är välkänt att naturen erbjuder en stor mängd material och processer som kan används i lågteknologiska reningsmetoder för behandling av förorenat vatten. Under det senaste årtiondet har omfattande grund- och ingenjörsvetenskaplig forskning om sådana naturnära behandlingssystem lett till förbättrade utsläpp av förorenat vatten. Mekanismerna för naturnära vattenbehandling och deras tillämpning vid behandling av lakvatten i tempererat klimat är emellertid inte helt kända på grund av ett stort antal samverkande faktorer. Tvärvetenskapliga forskningsprojekt har genomförts med syfte att utveckla strategier för konstruktion och utvärdering av fullskalig lakvattenbehandling.

Huvudsyftet med denna avhandling var att utveckla och karaktärisera ett system baserat på en filterbädd med vertikalt flöde för behandling av lakvatten från en industriell deponi, som innehåller fragmenteringsanläggningsskrot från uttjänta fordon och vitvaror. Laboratorieförsök, som innefattade kolonnförsök och skaktester, har legat till grund för utveckling av de behandlingsmetoder som sedan har vidareutvecklats genom pilotförsök och långtidsstudier i fält på en fullskaleanläggning. Flera lokalt tillgängliga och billiga naturella material eller restavfall samt blandningar av dessa undersöktes för borttagning av olika typer av föroreningar från lakvatten. En blandning av torv och kolhaltig aska visade sig vara mycket effektivt för att avlägsna ett antal giftiga metaller och olika polära och icke-polära organiska föreningar. Ett effektivt avlägsnande av fenolföreningar och en riklighet av fenolnedbrytande och heterotrofa bakterier observerades i filtret, vilket visar att den också fungerar som en biobädd. På grund av de klimatförhållanden som råder i Sverige, produceras 60-70% av den årliga lakvattenvolymen under ett par månader, vilket medför risk för tillfällig överbelastning av filteranläggningen. För att utjämna det varierande lakvattenflödet från deponin och säkerställa en stabil hydraulisk belastning av biofiltersystemet, behövs en stor damm vid den studerade anläggningen. Sådana utjämnings magasinering jämnar också ut höga koncentrationer och gör att förbehandling av lakvatten sker. Sedimentation befanns vara nödvändig för att minska den initiala belastningen av föroreningar (t.ex. järn och suspenderade ämnen) och minska risken för igensättning av filtret. Effektivitet året runt vid användning av ett filterbäddsbaserat system kan uppnås i en tempererad klimatzon som Sverige, om åtgärder vidtas för att skydda filtret från säsongsmässiga extrema väderleksvariationer. Resultaten visar att naturlig behandling på plats, som baseras på lättillgängliga material med låg miljöpåverkan "fotavtryck" kan användas för behandling av lakvatten från en industriell deponi.

ABBREVIATIONS

BOD	Biochemical Oxygen Demand
CEC	Cation Exchange Capacity
CFU	Colony-Forming Unit
COD	Chemical Oxygen Demand
DOC	Dissolved Organic Carbon
ELV	End-of-Life Vehicle
GC	Gas Chromatography
MSW	Municipal Solid Waste
MWTP	Municipal Wastewater Treatment Plant
PBDE	Polybrominated Diphenyl Ether
PCB	Polychlorinated Biphenyl
POP	Persistent Organic Pollutant
RNA	Ribonucleic Acid
DNA	Deoxyribonucleic Acid
SD	Standard Deviation
SR	Shredder Residue
SVOC	Semi-Volatile Organic Compound
TOC	Total Organic Carbon
VFSS CW	Vertical-Flow Subsurface Constructed Wetland

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1. INTRODUCTION

Anthropogenic activities increasingly contribute to loading of the environment with various pollutants. Better pollution control over point pollution sources is becoming important. There is a need for cost-effective on-site treatment systems, especially when treating contaminated waters from landfills. In 2002, about 300 municipal solid waste (MSW) landfills were in operation in Sweden [1]. Out of these, about 130 landfills have some sort of leachate treatment system, either an on-site purification plant or a leachate conditioning facility to improve leachate quality prior to discharge into municipal waste water treatment plants (MWTPs) [2]. It is apparent from the scientific literature that a tremendous number of studies have been carried out concerning MSW landfills. However, there are also other types of landfills, *e.g.* industrial landfills, in which the composition of leachate differs significantly from municipal leachate. Reports on industrial landfills are less common. Thus, in order to fill a gap and enhance specific as well as general knowledge about leachate treatment, this Thesis presents the most important findings concerning treatment of leachate from an industrial mono-landfill containing shredder residue (SR) from a recycling industry of scrap metal. At the industrial site, dismantled end-of-life vehicles (ELV) and white goods are shredded, and valuable materials are recovered for recycling. Although the recycling efficiency of ELV and white goods is gradually improving [3], the amounts of SR tend to increase due to the increasing number of ELV and white goods in the market [4]. Moreover, since the amount of different plastics used in cars has also increased, recycling has become more complex and challenging. Most of these mixed residues have to be disposed of. Leachate from this type of industrial landfill, as from any other type of landfill, has to be collected and treated. In Sweden there are no reported scientific studies concerning alternatives for treatment of leachate from landfills containing SR, and as far as the authors are aware, this also seems to be true from an international perspective. Therefore, there is a need for development of cost-effective on-site treatment systems for this type of industrial leachate.

One attractive treatment approach is to use a filter-based treatment system. This natural treatment option is considered as a low-tech and as well a low-cost alternative, when naturally abundant materials or waste materials can be used to fabricate the filters. Despite the tremendous amount of high quality research on alternative sorbents or filter materials, there are still aspects, which are not well covered from a scientific point of view. For instance,

although removal of metals by filter materials (*e.g.* peat) is extensively studied, removal of organic pollutants by the same filter materials is not frequently considered. Moreover, it is impossible to find information about simultaneous removal of polar and non-polar organic compounds together with metals, as well as common water quality parameters (*e.g.* nitrogen and dissolved organic carbon) in a filter material. Further, translations of advanced knowledge arising from laboratory research into reality *via* scaling up studies have not been made. This is probably due to lack of funds, time constrains or limited success in co-operation between universities and industries or companies, which actually require these systems. Combining results from different disciplines into one multidisciplinary approach, targeting the solution of leachate problems at specific site conditions, is not common, probably due to limited communication between scientists within different fields such as microbiology, hydrology, toxicology and chemistry. However, today, multidisciplinary co-operation between different institutions has increased, which certainly increases the possibility to address complex issues. Any natural treatment plant should be custom-developed. This means that the performance of the treatment system and its removal efficiency of pollutants should be tested on-site in each individual case. In this way, climate and local environmental conditions will be fully taken into consideration. This was the comprehensive approach taken in the work presented in this Thesis.

1.1. Objectives

The main objectives of the work presented in this Thesis were:

- To develop a filter-bed-based system for treatment of leachate from an industrial landfill containing shredder residue (SR) of end-of-life vehicles (ELV) and white goods

- To characterize the composition of an industrial landfill leachate and to investigate how well the concentrations of different types of pollutants can be simultaneously reduced in a biofilter-based system. The research has been focused on metals, polar and non-polar organic pollutants, as well as on commonly used water quality parameters

- To develop a strategy leading to the construction of a full-scale leachate treatment plant through batch and column experiments in the laboratory, as well as through field studies at a pilot plant on-site at the landfill

- To evaluate the long-term performance of a filter-bed-based full-scale treatment system for industrial landfill leachate in a temperate climate zone

1.2. Research layout

Many aspects of filter-bed treatment systems have previously been studied individually by different authors and under diverse conditions. In this study, batch, column, and three-year pilot-plant experiments, along with research on a full-scale treatment plant, were combined to develop and evaluate a filter-bed-based system for the full-scale treatment of an industrial landfill leachate. A multidisciplinary approach was an important component of the selected strategy to solve this complex task, and the work was conducted at School of Pure and Applied Natural Sciences (Kalmar University), at Department of Analytical Chemistry (Lund University) and at Centre for Aquatic Biology and Chemistry (Kristianstad University) in Sweden, and at Institute of Molecular and Cell Biology (Tartu University) in Estonia. The project was supported by the Knowledge Foundation (KK-Stiftelsen) and Stena Recycling AB. The research plan is outlined in Figure 1.

The results are presented in a series of scientific papers. In **Papers I – III**, results from the laboratory studies have been summarized. A focus in these articles was on the selection of suitable filter media for a filter-bed system among locally available, low-cost natural or residual waste materials. The batch test methodology, proposed in Paper II, was used to decide whether the selected material, a mixture of peat and carbon-containing ash, was suitable for use in leachate filters. In this judgement some environmental aspects were considered.

Papers IV – VIII include findings from field work experiments done to evaluate the performance of a filter-bed-based system for on-site leachate treatment. Papers IV and V comprise an evaluation of the efficiency of simultaneous removal of different types of pollutants in leachate in a filter-bed system, with a mixture of peat and carbon-containing ash, run for a three-year period. In Paper IV, pre-treatment options, aeration and sedimentation, were considered for reducing the load onto the biofilter. In Papers VI and VII, focus was placed on the impact of on-site environmental conditions such as precipitation and temperature variations on the hydraulic and biological performance of the full-scale filter-based system. A risk for long-term clogging of the biofilter system was also studied. In Paper VIII, the interest was directed towards the efficiency of leachate treatment during the initial period of use of a newly built peat filter.

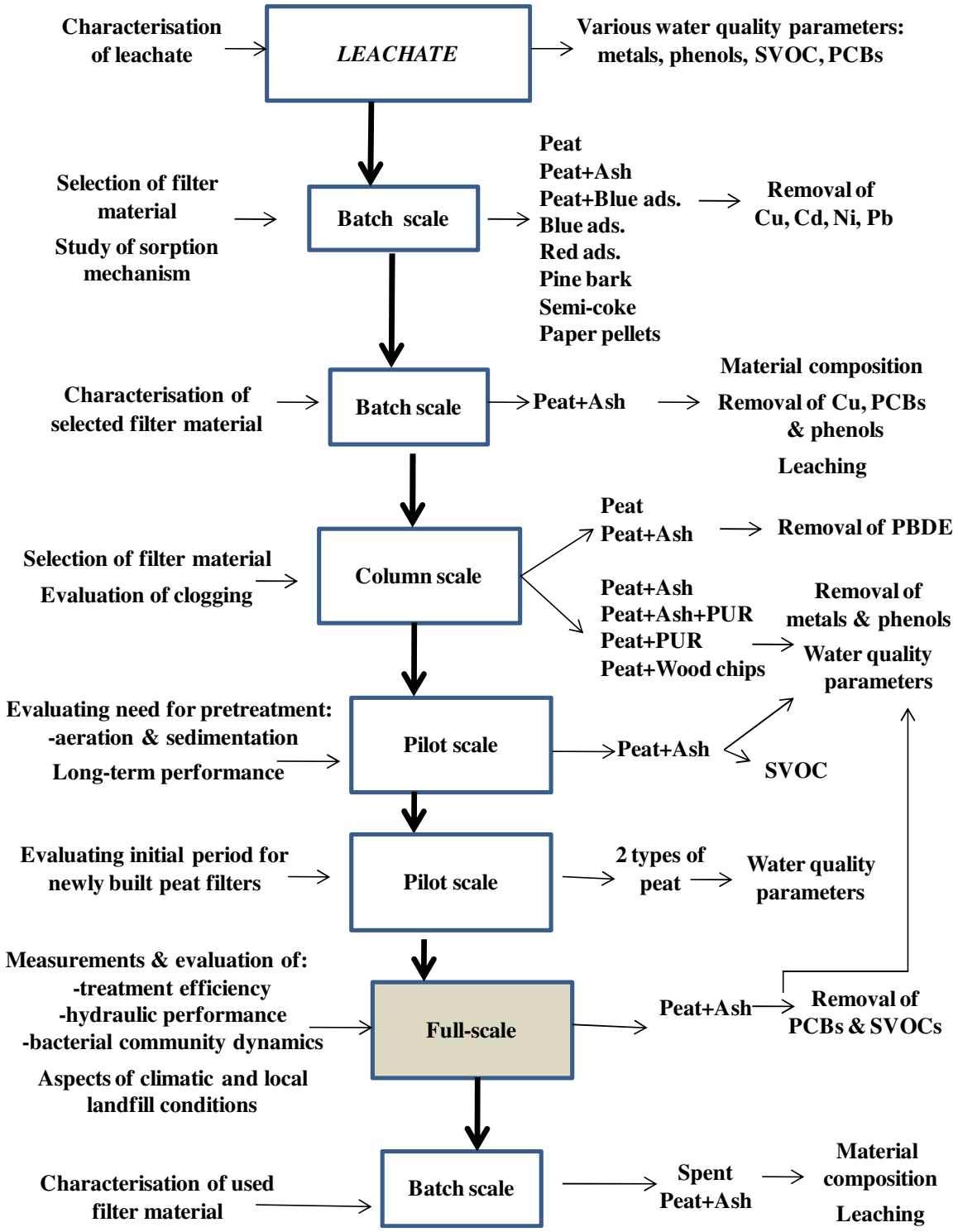

Figure 1. Research outline

2. LANDFILL LEACHATE MANAGEMENT

2.1. Leachate – an emission from landfills

One of the major environmental impacts from landfills is generation of leachate, which can pollute surface and groundwater. Leachate means any liquid percolating through the deposited waste and emitted from or contained within a landfill [5]. Among all the environmental hazards posed by landfills, the risk of groundwater pollution is probably the most severe because, historically, most landfills were built without engineered liners and leachate collection systems [6]. To reduce pollution sources, large numbers of unlined landfills have been closed down in Sweden. Nowadays, installation of bottom liners and leachate collection and treatment facilities is required. Sometimes, unlined and engineered bottom sections exist in the same landfill. To avoid leachate spills from unlined sections of a landfill, passive filter barriers are recommended in Sweden to cut off plumes [7, 8]. This option could also be an alternative for closed landfills for handling leachate during the aftercare period.

Until recent years, leachate has usually been treated in municipal wastewater treatment plants (MWTPs) together with sewage water. An advantage of co-treatment was the use of phosphorus from municipal wastewater and nitrogen compounds from leachate to achieve enhanced biological treatment for both waters. Since landfills are typically located far away from community sewage systems, construction of long pipelines or use of tanker transport becomes costly. In addition to the economical drawback, co-treatment has sometimes been shown not to be effective, since pollutants of concern in leachate are not necessarily the same as in wastewater. For instance, the removal of heavy metals in leachate requires a special type of treatment, otherwise these metals will end up in the sludge. Furthermore, leachate may be toxic for microorganisms in MWTPs, for instance by inhibiting nitrification processes [9]. Due to difficulties in optimizing the biological treatment process and utilizing excess sludge, co-treatment has become less popular. Today, landfill owners need to implement suitable site-specific leachate treatment methods rather than sending leachate directly to a MWTP.

2.2. Amount of leachate

2.2.1. Precipitation and generation of leachate in Sweden

In the European Directive on landfilling of waste, and as a further consequence of Swedish law, it is stipulated that the maximum water flow rate through a covered landfill with a bottom liner should not exceed 5 and 50 litres per m^2 and year for hazardous and non-hazardous waste landfills, respectively [10]. Estimating leachate flow is no easy task. Climatic conditions on-site, such as temperature, precipitation and evaporation, strongly influence the amount of leachate produced and thereby the design and operation of a natural leachate treatment system. Unfortunately, little attention is usually paid to the climate on-site at a landfill. Sometimes a crude rule of thumb is used to simplify the prediction of annual amounts of leachate. For instance, in the south and middle of Sweden it has been suggested that annual leachate flow from an open landfill area is 3,000 m^3/(ha year) and from a closed landfill area with vegetation is 2,000 m^3/(ha year) [11]. However, in reality, due to two reasons, the climatic conditions vary dramatically from area to area in Sweden, making the value of such an assumption questionable. First, the country is very long in the north-south direction, and second, it is located between the warm Atlantic Ocean in the west and the colder continental areas to the east [12]. Variations in altitude also contribute to the notable climatological variations [12].

The amount of precipitation fluctuates greatly over time and space in Sweden (as shown in Figure 2). Moreover, the precipitation is not always immediately available for filtration into a landfill. For instance, in the northern parts of Sweden, precipitation during the winter accumulates in a snow cover, which results in a delay of its availability [12]. In the south of Sweden, on the other hand, winter precipitation is available almost directly [12]. All these factors influence leachate production, its variation over time and the amount produced both seasonally and from year-to-year in a temperate climate. Thus, the design of a natural leachate treatment system becomes a complex hydrological matter. Understanding landfill hydrology and calculations of water budget for a landfill at the specific site is thus imperative for appropriate handling of the generated leachate.

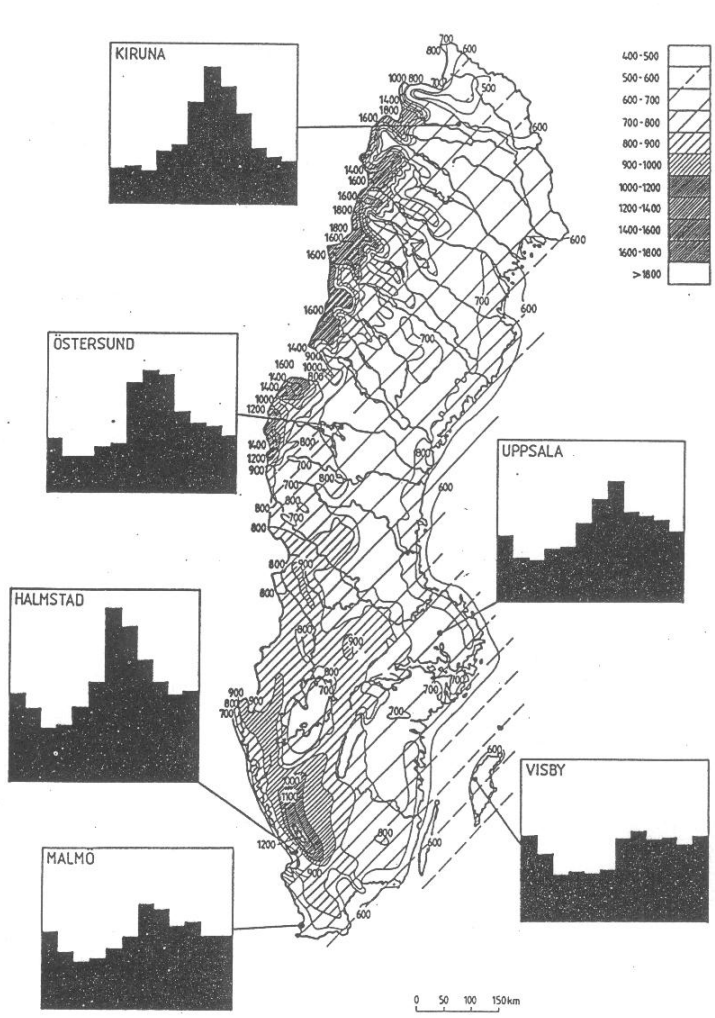

Figure 2. Mean annual precipitation in Sweden and the distribution of precipitation during the year from January to December (left to right) at the weather stations indicated. The map is based on meteorological data for the period 1931 – 1960 [12, 13].

2.2.2. Prediction of generation of leachate

Landfill hydrology is usually a key determinant factor in leachate generation, the presence and movement of water in a landfill, the leaching of pollutants therefrom and the geotechnical stability of the fill [14]. There are several methods and models available in the literature for calculation of leachate production. Some models provide an estimation of landfill leachate production through the use of simple formulas that take into account only the main factors, for example, water balance methods as have been presented by Hogland [15], RVF [16] and Hogland *et al.* [17]. Other models, such as the Hydrologic Evaluation of Landfill Performance (HELP) [18] and the Hydrologic Simulation of Solid Waste Disposal Sites (HSSWDS) [19] models are more detailed, aiming to account for the greater complexity within the system. The

HELP program uses climatic, soil and design data to generate daily estimates of water movement across, into, through and out of landfills [20]. The HELP model is the most widely used tool for the prediction of leachate emission from landfills, and for the design of leachate control and management infrastructure [14]. Calculations should be carried out on a case-by-case basis, since there are large differences in time for the availability of water in different parts of Sweden, as discussed in section 2.2.1. Long-term data series on daily leachate flows from landfills contribute to more accurate estimations of amounts of leachate that need to be treated today, and to background knowledge for simulation of future scenarios. As a common practice, detailed information about the variations of leachate concentrations during the year exists; however, knowledge about variations of leachate flow on a finer time scale is often missing.

2.3. Composition of leachate

The composition of leachate depends on a myriad of interacting factors, such as the type and depth of wastes, degree of compaction, age of fill, moisture content, rate of water application, phase of degradation, landfill design and operation, and the interaction of leachate with the environment [21]. Generally, it is accepted that over time, waste in landfills undergoes several different phases of decomposition. Depending on how the phases are defined, three to five phases have been observed at landfills and an additional one to four phases have recently been suggested [22]. Generally speaking, for example, Kjeldsen *et al.* [6] and Quasim [21] reported that at least four phases of degradation exist, including an initial aerobic phase, an anaerobic acidic phase, an initial methanogenic phase and a stable methanogenic phase. The acidogenic and the methanogenic degradation conditions are the two most important factors in determining the composition of the formed leachates [22]. The leachate composition during these degradation phases varies considerably, as described in detail by Kjeldsen *et al.* [6], Quasim and Chiang [21] and Andreas *et al.* [23]. The composition of leachate often seems to be generally similar in landfills, which happen to be in the same phase. An explanation might be that the biodegradability of organic content in the MSW and the compaction of the waste layers convert the landfill environment to anaerobic, giving many similarities to the composition of leachates among different landfills [6]. Determination of all the substances in leachate is complicated, expensive and time-consuming. Thus, organic content of leachate has been assessed utilising a sum of parameters such as biochemical oxygen demand (BOD_7), chemical oxygen demand (COD), total organic carbon (TOC), and dissolved organic carbon

(DOC). The DOC in leachate covers the degradation products of a variety of organic compounds ranging from small volatile acids to fulvic and humic-like compounds [24]. Typical surveys of leachate also include measurements of pH and conductivity, and determination of total nitrogen (N_{tot}), ammonium-nitrogen (NH_4-N), total phosphorus (P_{tot}), and heavy metals. In spite of similarities, the concentration of pollutants in leachate ("strength") at different sites varies considerably. In landfills where waste deposition started several decades ago, different degradation phases occur simultaneously within different parts of the same landfill. When a final cover is placed on a section of a landfill, leachate production from this part is significantly decreased [6]. As a result of all these factors, the amount, flow rate, composition and strength of leachate vary considerably throughout the landfill, as was also shown in the present study (Paper V). However, such variations are often difficult or impossible to observe, since the leachates from the different landfill sections are frequently discharged to the same collection system for further treatment. The average concentrations of pollutants of leachate from several landfills, including those investigated in this thesis, are shown in Table 1. Data in Table 1 show that concentrations of pollutants in the leachate from industrial landfills and MSW landfills are different.

During the last decade, the level of concern about characterisation of specific organic pollutants in landfill leachate has increased substantially. The first indication of a necessity for a more detailed characterization of leachate came from the Greenpeace organization. Greenpeace analysed concentrations of various organic compounds in leachate from 28 MSW landfills in Sweden [25]. Subsequent follow-up characterisation of leachate from MSW landfills in Sweden [26, 27] and old landfills in Denmark [28] have confirmed that a large number of different organic compounds are present in leachate from MSW landfills. The presence of organic compounds in the leachate has also been studied by other authors [29-33]. The most frequently identified groups of organic pollutants are aromatic and halogenated hydrocarbons, phenols, alkylphenols, pesticides, phthalates, aromatic sulfonates, phosphonates and polychlorinated biphenyls (PCBs). Even though the concentrations of these compounds are often low, their environmental impact can be high. Substantial progress in sample preparation methods and the rapid development of highly sensitive and selective analytical equipment have made it possible to more efficiently determine organic pollutants in complex contaminated waters at very low concentrations in everyday practice. Today, several methodologies for determination of organic pollutants, *e.g.* PCBs and flame retardants, in

landfill leachate are available [34-36]. Although the determination of specific organic compounds has become an everyday practice, only a few studies [37] have been published, which take a look at the removal of these organic pollutants in leachate by natural systems. The general opinion today is that new leachate treatment plants need to be capable of successfully treating both inorganic and organic pollutants.

Table 1. Average concentrations of impurities in leachate from the investigated industrial landfill (IL) in Sweden (standard deviations, in parentheses), and 26 other landfills in Sweden [38], landfills in Finland [39], Denmark [28] and 20 landfills in West Germany [40].

Parameter	IL, Sweden		Sweden	Finland	Denmark	Germany
pH	8.1	(0.2)	7.6	7.1	7.0	8.0
Conductivity (mS/cm)	4.7	(1.7)	0.87	0.24	0.3	na
N_{tot} (mg/l)	150	(94)	287	66	na	na
NH_4-N (mg/l)	100	(60)	260	53	110*	740
S (mg/l)	11	(3.0)	91	48	na	na
BOD (mg/l)	290	(360)	25**	na	44	180
COD (mg/l)	1270	(875)	688	na	320	3000
BOD/COD (ratio)	0.22	(0.17)	0.038	na	0.11	0.06
DOC (mg/l)	450	(240)	227	na	na	na
IC (mg/l)	380	(110)	na	na	na	na
TOC (mg/l)	470	(240)	220	na	130	na
Cl (mg/l)	630	(320)	1471	250	360	2120
SS (mg/l)	80	(55)	196	99	na	na
Cu (µg/l)	190	(210)	23	31	70	65
Al (µg/l)	160	(130)	209	820	na	na
As (µg/l)	12	(5.7)	5.5	na	16	87
Cd (µg/l)	0.70	(0.66)	0.44	5.4	6.8	5
Co (µg/l)	5.6	(0.86)	7.7	na	na	50
Cr (µg/l)	8.4	(7.3)	15.3	33	76	280
Hg (µg/l)	0.78	(0.49)	0.028	na	5.2	na
Pb (µg/l)	7.9	(8.3)	4.4	29	70	9
Sb (µg/l)	5.4	(7.6)	1.5	na	na	na
Ni (µg/l)	76	(20)	31	41	130	170
Sn (µg/l)	8.4	(7.0)	2.7	na	na	na
Zn (µg/l)	56	(31)	66	520	670	640
Li (mg/l)	1.2	(0.53)	0.17	na	na	na
Fe (mg/l)	1.5	(1.1)	6.5	42	76	15
Mn (mg/l)	0.92	(0.26)	1.15	2.5	3.5	0.7
Mg (mg/l)	81	(18)	48	na	60	250
K (mg/l)	140	(50)	411	160	140	1085
Ca (mg/l)	30	(9.5)	131	170	280	80
Na (g/l)	0.90	(0.28)	0.48	0.53	0.21	1.34

na - not analysed
*Determined as ammonia
**BOD_7

2.4. Toxicity and quality control monitored through living biomarkers

In addition to the chemical characterisation of leachate, information about toxicity is also required for evaluating the environmental impact of a landfill leachate. A large organic load from leachate will reduce the content of oxygen in surface waters [41], which could be harmful for aquatic organisms. A high concentration of ammonia is toxic to fish and zooplankton, as are high total concentrations of different heavy metals [41]. In particular, heavy metals present in the form of free cations are very mobile and can easily absorb onto and pass into the tissue of marine organisms. However, some metal species are much more toxic then others *e.g.* Cr (VI) is more toxic than Cr (III). Thus several authors have even tried to speciate metals in the leachate using chemical equilibrium in different models or programs [42], or size-charge-fractionation methodology [43].

Different toxicological tests provide indirect information about the content of pollutants that may be harmful to a class of organisms [6]. Toxicity testing has grown steadily in recent years, since becoming a useful tool in environmental risk assessment. Farré and Barceló review different methods and their usage for acute toxicity measurements [44]. Biotests can be based on micro-organisms, invertebrates, plants, fish or other bio-indicators [45]. Since the concentration of chloride ions is high in the leachate from landfills, the chloride-tolerant *Artemia salina* is often used for toxicity testing. In short, in the toxicity test, a batch of organisms is incubated at 25ºC for 24 h in a dilution series of the samples. After the incubation period the number of organisms defined as immobile (*e.g.* presumably dead or at least incapacitated) is recorded. The results are reported as an effect concentration (EC_{50}) or as a lethal concentration (LC_{50}), the concentration that affects or kills half of the test population, respectively. For instance, assessment of acute toxicity of leachate from different landfills using *Artemia salina* as a test organism has been carried out by Svensson *et al.* [46].

2.5. Legal requirements

During the last decade both extensive research and practical application of wastewater treatment have led to improvement of the quality of wastewaters. There exists enough information to propose and establish clear guideline values for discharge of wastewaters to the environment. Detailed guidelines for on-site treatment of domestic wastewater in constructed wetlands are now available, including technical descriptions for design, construction and management [47, 48]. Discharge limits from WWTPs for BOD is 15 mg/l and for phosphorus

is 0.5 mg/l. If a WWTP is located on the coast, the discharge limit for nitrogen is 10 mg/l [22]. Despite the success in managing municipal waste waters, limit values for compounds in discharged landfill leachate are still missing in Sweden as well as in the EU. In conformity with the EU directive on waste, contaminated water and leachate need to be treated to an appropriate required standard prior to discharge [49]. If an assessment, based on consideration of the location of the landfill and the waste to be accepted, shows that the landfill poses no potential hazard to the environment, a competent authority may decide that this provision does not apply [49]. Thus, the Swedish policy is to not provide national limit values for leachate [22]. Local conditions at each landfill site and recipient are individually examined and requirements for discharge are provided at each landfill. This provides great flexibility, but does not eliminate the eventual need to treat this complex leachate adequately. As the composition and volume of leachate tend to vary considerably both seasonally and from year to year [12], the design of a natural leachate treatment system becomes a complex matter, especially in temperate climates. Therefore, more research is needed to provide a comprehensive background of data, before guidelines for on-site treatment of landfills in natural treatment systems can be proposed in Sweden.

2.6. On-site treatment of leachate

2.6.1. Natural treatment systems

Treatment systems, which are described as “natural”, are those that utilize processes often occurring on their own in nature for treatment of pollutants in water. In these systems vegetation, soil and microbial ensembles all play an important role. Processes which have an effect on pollutants are sedimentation, precipitation, filtration, adsorption, chelation (chemical complexation), biodegradation, microbial metabolism, uptake by plants, adsorption onto roots and soil particles, and plant or microbial die-off. A natural treatment system might include ponds or lagoons, horizontal- or vertical-flow filters, and soil-plant systems, often in various combinations [37, 50, 51]. The main advantage of using natural treatment systems instead of conventional wastewater treatment techniques is lower capital, operation and maintenance costs [52]. Furthermore, equipment which is needed in natural treatment systems is relatively simple compared to high-tech systems using techniques such as reverse osmosis, nanofiltration, or chemical treatment. Thus, there is no need for specially trained technicians to run these plants. Natural treatment systems are thus considered to be low-tech systems. The term “natural treatment system” is often used as a synonym for artificial, constructed or

engineered wetlands. Natural treatment systems are land-intensive, while conventional treatment systems are energy-intensive [53, 54].

Historically, sand filters have been widely used for wastewater treatment. Vertical-flow subsurface systems (VFSS) with macrophytes and sand, represent the latest generation of subsurface flow constructed wetlands and present the state of the art for this technology in wastewater treatment in Europe [55]. VFSS constructed wetlands have been applied as secondary (biological stage for mechanically pre-treated wastewater) or tertiary treatment (for polishing the effluent from a conventional biological treatment plant) of wastewater [56]. VFSS have been also used for treatment of, *e.g.* landfill leachate, pulp and paper wastewater, mine drainage, and wastewater from petroleum refineries and textile industries. Innumerable books give comprehensive reviews and details about the design, operating principles and treatment efficiencies of natural systems for different types of wastewater [57-59], showing their versatility.

Nowadays, when a tremendous amount of different filter materials are available, other materials alone or in combination with sand can be selected for filter beds. Filter beds without growing plants can also be used alone or as a part of existing constructed wetlands for water treatment. In this case, the filter beds are operated similarly to VFSS constructed wetlands and most processes within filtration systems are similar (except processes which are related to plants).

2.6.2. Filter-bed technology

A filter-bed treatment system is an unplanted filter. The design of filter beds, reported in the literature, varies to a large extent. A general layout of a filter bed is presented in Figure 3. The number and size of filters used depend on the volume of water that needs to be treated [60]. The filter-bed system may consist of one or more layers, and units can be placed in series or in parallel, or both.

The chemical and physical properties of the sorbent or the filter media determine its efficiency. The treatment of leachate takes place due to physico-chemical trapping processes. By judicious selection of filter materials (homogeneous or mixed), specific pollutants can be separated from the leachate. The efficiency of purification depends on the characteristics of

the leachate (composition, pH, *etc.*) and the properties of the filter material (hydrophobicity, composition, value of pH, *etc.*), as well as on details of the construction of the filter beds, the hydraulic loading rates, and whether the bed is kept aerobic or anaerobic.

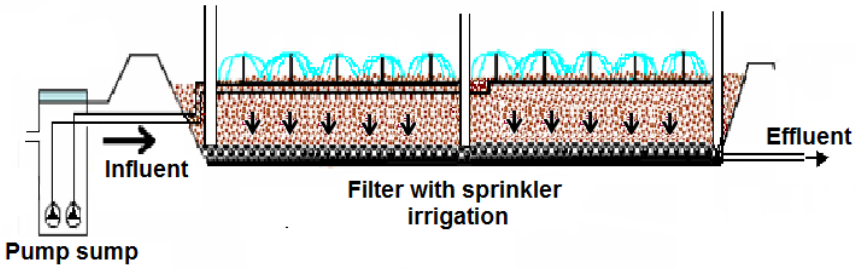

Figure 3. A general layout of a vertical-flow filter bed with sprinkler irrigation (modified from Paper VI).

Both up- [61] and downflow [62] vertical filters have been used. The filtration system might be designed to operate in batch flow mode to reduce pumping and computer control costs [63]. In systems with advanced design, an operator on-site might be able to select the operation mode depending on actual circumstances. Regulation of the water level in the filter determines whether aerobic or anaerobic conditions prevail or whether both conditions occur at different places in one filter. The aerobic or anaerobic conditions in the filter are regulated by the hydraulic loading rate and by regulating the water level in the filter. Hydraulic loading rates on filters are selected based on the hydraulic conductivity of the filter material. The characteristics and size of the filter material dictate the acceptable hydraulic load on a filter.

2.6.3. Filter-bed media

Filter materials are commonly called sorbents or adsorbents. Sorbents can provide chemical and/or biological treatment processes as a complement to physical processes [64]. Many types of adsorbents are available today. In recent years, substantial research efforts have been spent to identify and test inexpensive alternative filter materials both for leachate and wastewater treatment. To be a low-cost filter medium, the material should require little processing, should be abundant in nature, or should be a by-product or residual material from an industrial or agricultural operation [65]. Any type of sorbent alone or in combination with other sorbents, *e.g.* sand, can be selected for filter beds.

The cost of the materials and their local availability, to avoid expenses from transportation, are of great importance, since large volumes of leachate often need to be treated. At some landfill sites in Sweden, the annual amount of leachate is up to about 100,000 m^3, and thus a quite large volume of filter material is needed for its treatment. Filling a full-scale filter with an expensive and rare filter material makes the treatment cost per cubic meter very high. This is not reasonable: not only does leachate need to be treated during operation of the landfill, but also treatment must continue during an up to 30-year post-closure period after the landfill has been closed. Apart from economic factors, the suitability of a certain filter material depends on the chemical characteristics of the leachate and effluent requirements.

Peat [66-71], zeolites, bark [65], opoka [72], clinoptilolite [73], blast-furnace slag [74, 75], sawdust [76], pine bark [74, 75], carbon-containing ashes [77, 78], zeolitised coal fly ash [79] and iron oxide-coated sand [80, 81] are most commonly investigated natural and residual waste materials for treatment of metals in wastewaters or leachate [65, 68, 82]. Organic compounds have been removed by carbon-containing ashes [77, 78] and bone char activated carbon [81, 83, 84], derived from almond shells, olive stones and peach stones by heating them in CO_2 at 850ºC or by activation with chemicals.

Alternative filter materials derived from residual products from industrial or agricultural operations are, for example, carbon-containing ash from the pulp and paper industry or polyurethane waste from the automobile shredder industry, for replacing expensive activated carbon. A disadvantage of using residual products is that some media might leak, and therefore might increase the pollution load instead of reducing it due to their use in a system. However, information concerning this kind of leakage is in most cases not available.

Peat is partially decomposed organic matter, with a high water-holding capacity, a large surface area, and chemical properties making it very effective as an adsorbent. Peat is an inexpensive material, which is readily available (15% of the land area of Sweden is covered with peat [85]). Therefore, during the last decades the feasibility of using peat in filter systems has been widely investigated. Peat is a complex material, with lignite and cellulose as major constituents. As described in many studies, the polar functional groups of the lignin and humic fractions, which include alcohols, aldehydes, ketones, acids, phenolic hydroxides, and

ether groups, are involved in the formation of chemical bonds of various types with constituents of wastewater [65].

Peat has been studied as a material suitable for removal of pollutants such as NH_4^+ [77, 86], and boron [87], reducing biochemical oxygen demand (BOD) [86], COD, TOC, phenols [77], and pesticides [70]. The processes involved in removal of pollutants are adsorption, ion exchange, precipitation, and formation of layers of bacteria and fungi on the wetted surfaces of the medium [68, 70, 88]. Peat has been used for treatment of various pollutants in different types of wastewaters, *e.g.* waste waters from households, laundries [42], slaughterhouses [89] and dairy wastewaters [89], storm-water [90], sulphide mine leachate [42], and leachate from municipal [77] and industrial landfills [42]. However, peat has also been found not to be capable of removing metals to acceptably low levels for direct discharge, but was sufficient in a pre-treatment (or polishing) process at low hydraulic loadings [42, 71].

Mixing of two or more filter materials might improve removal efficiency of several types of pollutants and might improve the hydraulic properties of the whole filter. For instance, mixing of an organic fraction such as peat with filter material rich in CaO will reduce the risk for chemical clogging [91]. Renman [92] reported that adding peat to polonite improved the hydraulic conductivity of the filter. Certainly, carbon-containing ash would need to be supplemented with a more grainy material in order to avoid clogging if it is used alone, thus improving the hydraulic load of the filter material. However, Ringqvist *et al.* reported that a combination of Sphagnum peat with glauconite or clinoptilolite gave only a minor improvement in metal adsorption [42].

2.6.4. Microorganisms in filter-bed treatment systems

A large variety of microorganisms may cohabit in the landfill environment and thus they are present also in the leachate. Microorganisms use different metabolic pathways to obtain their energy and access different carbon sources to nourish their growth [93]. Since microorganism cultures are already present in raw leachate, by manipulating environmental conditions their amount can be increased. Filter material provides a place on which microorganisms can grow. If conditions are optimal for micro-organisms (*e.g.* oxygen content, nutrients), the organisms break down pollutants found in the leachate and improve water quality. Thus, if in addition to physical-chemical trapping of pollutants, simultaneous chemical and biological degradation of

the pollutants occur, the filter acts as a biofilter. The filters perform better after they have been in use for a period of time, due to the creation of a biofilm, which facilitates permanent retention of pollutants [94]. In the environment, multiple species of microorganisms can share one ecological niche (*e.g.* degradation of a pollutant); among these, one population (or a few) that best adapts to the ecosystem conditions becomes predominant, while others exist as minorities [95]. Most of these diverse microorganism populations have not been isolated and characterized in the laboratory. Nowadays, culture-independent molecular approaches (exemplified by the rRNA phylogenetic approach) have been more often applied for analysis of microbial populations existing in leachate. Molecular approaches based on 16S ribosomal DNA sequence analyses allow direct investigation of the structure and diversity of the microbial community. Moreover, individual species of microorganisms can be quantified. Although the molecular phylogenetic approach cannot directly establish that the detected microorganisms are capable of pollutant degradation, some of these organisms apparently occur in response to presence of pollution and may have a significant influence on the fate of pollutants [95].

A transformation of nitrogen by nitrification and denitrification is the most common example of a biological leachate treatment, where microorganisms are utilized to reduce pollutants in the leachate. Molecular characterisation of bacteria, which remove ammoniacal-nitrogen from landfill leachate, has been carried out in reactors with carrier to which surface nitrifying bacteria adhered [96] and also in natural leachate treatment systems [97, 98]. To analyse the population structure and dynamics of the activity of the ammonia-oxidising bacterial (AOB) community, a 16S rRNA-based approach was employed [98].

Several studies show that phenolic compounds are among the common contaminants in landfill leachate [38, 99, 100]. Landfills were shown to be a habitat of anaerobic microbial populations capable of degrading toluene, phenol and *p*-cresol, as reported by Wang and Barlaz [101]. Phenol biodegradation occurs through intermediates under both aerobic and anaerobic conditions to yield carbon dioxide in soil; however, degradation under anaerobic conditions is slower [99]. The intermediates in the biodegradation of phenol are benzoate, catechol, *cis,cis*-muconate, β-ketoadipate, succinate and acetate [99, 102-105]. According to the review of Tuah [99], phenols are metabolized by microorganisms from a variety of different genera and species. Bacteria, fungi, yeast and algae have been reported to be capable

of degrading phenol [99]. In order to gain insight into the genetic diversity of microbial populations involved in phenol degradation, the genes coding for such catabolic enzymes as phenol hydroxylase have been analyzed [106-108]. Microbial populations continuously adapt to fluctuations in the physicochemical composition of leachate, however, increasing phenol concentrations can decrease biodegradation. Phenol inhibitory levels for phenol degradation by different microorganisms are in the range 25 to 300 mg/l [99]. Both free-living and particle-attached bacteria can be found along the flow path of leachate through the treatment plant. Due to the large surface area of any filter material, the filter generally supports establishment of a bioculture (biofilm). Despite what is already learned about these biofilms, still more information has to be collected from field studies to improve the effectiveness of leachate treatment by microorganisms. Both *in situ* community analyses and pure-culture studies are indispensable for attaining a good understanding of the ecology of pollutant degradation and thus these studies should be carried out in coordination with each other [95].

2.6.5. Limitations of filter-bed technology

2.6.5.1. Adsorption capacity

The adsorptive capacity of a filter material for a particular compound is dependent on both the characteristics of the material and the compound itself. Important characteristics of a compound are its solubility, molecular structure, molecular weight and polarity. The theoretical adsorption capacity of a material for a particular contaminant is given by its adsorption isotherm, which is determined by exposing given amounts of a contaminant in a fixed volume of liquid to a given amount of adsorbent [83]. The adsorption characteristics of the material are most commonly described by the isotherms of Freundlich, Langmuir or Brunauer, Emmet and Teller (the BET isotherm) [83]. In addition to prediction of the sorption capacity of materials, investigations and explanations of the sorption mechanisms and determination of kinetics are usually also studied at laboratory batch scale. The summaries and review articles indicate that the processes and mechanisms behind removal of metals by alternative adsorbents is fairly well investigated [65, 82]. Warith [66] and Sartaj *et al.* [109] have used their results from laboratory studies for calculations of the amount of material needed for filters for full-scale applications.

2.6.5.2. Research approaches for selection of material

A comparison of results from several adsorbents tested under the same experimental conditions gives information about the best suitable one. Unfortunately, it is often difficult to

compare results from different investigations due to differences in both the used filter material and experimental set-ups. For peat, a main problem is its heterogeneity resulting in variations of the chemical and physical properties of different peat types. Another problem might be variations in pre-treatment and sampling procedures [110].

Great simplifications are often made in the study of the complex processes underlying removal of pollutants in water treatment systems based on filter techniques. Efficiencies in removal of pollutants such as metals by different filter substrates are often tested in laboratory experiments, but more seldom are the measurement extended to the field. Thus the following question often remains: Will the reduction efficiency of the pollutants be the same in a full-scale treatment plant as indicated by the results of the laboratory experiments? What are the practical problems that can be faced out in the field? Although batch tests provide useful capacity information, column studies yield more information about the dynamic performance of a filter. Loading rates, breakthrough times and the risk of clogging as well as removal efficiency of pollutants can be investigated in dynamic column experiments. For scaling-up to field design sizes, column studies are necessary to define the treatment possibilities using peat [71]. Both isotherm equations and breakthrough curves are utilized to determine the required dimensions of the filter bed [66].

Studies on breakthrough curves provide information about the volume of water containing pollutants which can pass the filter material before the filter is exhausted and reaches its removal capacity. These studies have usually been carried out using a constant initial concentration (at mg/l level) of pollutants. However, such conditions do not reflect a real situation for landfill leachate. Leachate composition and volume tend to vary considerably both seasonally and from year to year in temperate climates [12]. The value of pH in industrial landfill leachate and in many leachates in Sweden is neutral or slightly basic. In this pH range and at low initial concentrations of metals, adsorption and ion-exchange are important mechanisms for trapping. However, at the same pH range but at high initial metal concentrations, the main mechanism for trapping might be precipitation. Thus a comparison of results from landfill leachates with large differences in pollutant concentrations is probably not always reliable. If the column tests could be carried out on real leachate, that would of course be an improvement. However, the disadvantage of this approach is that with materials

with high adsorption capacities (*e.g.* peat) and relatively low metal concentrations in the leachate, it might take an impractically long time to reach the breakthrough point.

Use of a smaller column diameter or a shorter length is possible. However, wall and channelling effects are limiting in this approach, since these might lead to unrealistically low breakthrough values and clogging might stop the experiment prior to breakthrough values being obtained. Moreover, columns with small diameters (about 10 cm) on-site are more vulnerable than a pilot-plant to environmental changes. For instance, above-ground filter columns experience higher temperatures and associated data variation, which might cause performance data to be "overly optimistic" compared to below-ground filter column during the summer [63].

2.6.5.3. Temperature effects on natural treatment systems

The second major concern in cold to temperate climate zones with natural treatment systems is related to the relatively low ambient temperature and its influence on purification processes, and management of treatment systems from a hydraulic point of view. For instance, nitrification processes are strongly affected by temperature, since growth rate and activities of ammonium-oxidizing bacteria are extremely low at low temperatures [96]; however, efficient removal of ammonium from landfill leachate is often required, making it imperative to overcome this problem. Also availability of dissolved oxygen in the treatment system determines BOD removal. The solubility of oxygen in water increases as temperature decreases, but microbial activity decreases in cold water. Again, the temperature of the filter seems to be an important factor.

Leachate is most commonly distributed by sprinklers or by perforated tubes. The tubes are placed on the surface of the filter bed. The main problems with effective year-round operation of a filter-bed treatment system in cold climate, are according to Mander and Jenssen [111], hydraulic failure due to freezing; hydraulic failure due to viscosity changes of the polluted water; and inadequate purification processes due to the low temperature.

A simple approach to avoid freezing is to insulate the filter with an overlayer of straw, as described in Paper VI, or with extruded polystyrene or other materials as described by Mæhlum [112], prior to the ambient temperature decrease below 0ºC. By this measure, continuous year-round performance can be maintained in temperate climate zones. Another

option is to construct a sufficiently large equalisation pond and filter areas to make it possible to irrigate the filter only when the temperature is above zero, and storing the leachate during winter.

2.6.5.4. Clogging

According to some authors, well-mineralised peat tends to become clogged [53, 113], whereas some types of peat may retain their hydraulic conductivity for many years [114]. Reported information concerning filters with peat is contradictory, probably dependent on peat origin. Several studies [72, 113, 115-118] indicate that the problem of clogging is often the limiting factor for the long-term performance of a treatment system based on the filter-bed technique. In case of clogging, the filter-bed material is changed before its sorption capacity is reached. Several causes of clogging of filter beds are reported in the literature [119]. One of the main concerns always mentioned in this context is the risk of physical blockage, caused for example by non-degradable particles washed into the filter inlet. One cause for this can be that the time allowed for biological degradation of particulate organic carbon is not sufficient due to a sudden increase of concentrations of organic matter and SS. The risk for this is realistic, since a large variation of concentration of SS and organic compounds in leachate from landfills is quite common. In addition, too high continuous loading rates or more frequently loaded leachate batches will restrict the time available for oxygen transport into the filter by diffusion and mass transport. This affects the balance between aerobic and anaerobic processes in the biofilter. As a consequence, the risk of clogging will also increase [120]. Moreover, overloading and changes in the balance between aerobic and anaerobic processes will also affect nitrogen transformation. Thus, in the event of a lack of oxygen, nitrification will be limited.

Another important reason for clogging is mineralisation of organic filter material during operation of the treatment plant. This mineralization takes time, but it leads to decreased particle size, and reduced hydraulic conductivity. Advantageous for finer material textures is that the cation exchange capacity (CEC) tends also to increase, because more negatively-charged binding sites are available [121]. One serious disadvantages of decomposition of filter material is that less water can be treated since the hydraulic load on the filters has to be decreased.

The formation of a bio-film is positive with respect to removal efficiency; however, excess biomass is not favourable [119]. It has been suggested that filter beds should be left unloaded during certain periods, to provide "rest" periods during which the amount of biomass is reduced. A decomposition of previously loaded organic compounds and suspended solids occurs during such periods, and as a result, the hydraulic conductivity of the filter most probably improves.

2.6.6. Equalisation and pre-treatment

An equalisation pond minimises and controls the fluctuation of leachate volumes from a landfill to provide optimum or designed flow conditions for the following filter-bed treatment system. It can provide continuous feeding of leachate to the filter and large concentration fluctuations can be evened out. Mixing of the contents of the pond can be done to ensure adequate equalisation of leachate. However, without mixing, an equalisation pond can also act as a sedimentation pond, where settleable solids deposit. Design of ponds generally requires a multidisciplinary input of knowledge involving biological and ecological sciences, aquatic chemistry, landscape architecture, hydrological engineering, and flow hydraulics [122]. Design of a given pond depends on its purpose, and details of size and construction can vary as described in [83, 122-124]. The size of a pond located in the front of a filter depends, for example, on the expected fluctuations in the flow rate from the landfill, on the size of the filter-bed, and on the designed hydraulic loading rate of the filter-bed treatment system. Although the existence of an equalisation pond before the treatment system is a necessity for all leachate treatment on-site [16], there are some disadvantageous in including an equalisation pond. A relatively large space is needed and additional operation and maintenance is required (*e.g.* aeration to avoid odours). This will increase the capital and maintenance costs of the system.

The option to construct very large ponds for storage of leachate during the cold months, in regions where most of the leachate is generated during this period, might not be favourable from an economic point of view for large landfills in Sweden. This is especially valid when leachate treatment comprises an irrigation of leachate onto a soil-plant system to reduce the quantity of leachate through increased evapotranspiration, and if irrigation can only take place during the plant growing season. A further drawback is that the plant growing season in Sweden may be only between 3 and 9 months [16]. For instance, in Umeå irrigation can be

take place from May to July, a much shorter time than can be used in Halmstad and Västerås (6 months) [16]. Thus, design of a natural leachate treatment system is very site-specific and the best alternatives must be searched for in each case.

Leachate can also be pre-treated in a pond, through aeration and sedimentation. The aeration mode used depends on leachate quality. Mechanical or diffused aeration units and induced surface aeration can be used [123]. However, it has been reported in the literature that several other types of aeration systems are also functional. The final selection of an aeration system depends on the type and geometry of the pond, the costs for installation and operation, and the goal of its performance [83]. An aerobic pre-treatment before the filter reduces BOD in the leachate and hence the BOD loading to filter, thus reducing the possibility of clogging of the filter surface as shown in [111]. In addition, ammonium-nitrogen is converted to nitrate by aerobic treatment.

At the same time in the pre-treatment step, metals can be oxidized and can be precipitated as hydroxides, oxides and metal-metal complexes. A decrease of iron concentration in leachate, which in many leachates occurs at high concentrations, is of great importance and can be accomplished in a pre-treatment step. Moreover, extended settling after an aeration period is required to achieve co-settling of other heavy metals with insoluble iron and calcium compounds, presumably acting as flocculating agents [125]. In this way progressive clogging of void spaces in the filter material and also of the surface of the biofilter due to formation of iron oxides is avoided. Furthermore, the removal of metals such as Cu, Pb, Cd and Ni in the filter bed will most probably be facilitated since there will be less competition with Fe for available active sites in the filter material. For instance, the concentration of Fe in leachate from a MSW landfill was reduced by more than 95% in an aerated pond, as reported by Thörneby *et al.* [37]. The reduction of Fe in aerated lagoons in Norway was 78% [50].

Treatment by sedimentation is commonly used to remove readily settleable solids in order to reduce the content of suspended solid [83]. Considering storm-water treatment by the filter it is required that sedimentation provide a low content of SS material (<10mg/l) in the water, to obtain an acceptable operational lifetime of the filter [91].

2.7. Selected natural leachate treatment systems in Sweden

In Sweden, the total amount of collected leachate is estimated to be between eight and twelve million m^3 per year [126]. Annual leachate amounts, which have passed through pre-treatment and treatment steps in natural treatment systems in some Swedish landfills are presented in Table 2. The volumes of leachate involved are quite large. Collection of leachate in an equalization pond and aeration are typical leachate treatment steps, which exist on-site in Swedish landfills [16]. Aerated ponds are used in 86 landfills in Sweden [127]. Sometimes even chemicals are added, *e.g.* phosphoric acid. The retention time of leachate in such ponds is usually between 15 and 30 days [127].

Filters with vegetation such as Salix or grass have been employed for landfill leachate treatment in some landfills in Sweden [128]. Leachate is irrigated on soil plant systems at 37 landfills out of 134 landfills, and in filters at 14 landfills [127]. In many landfills, several treatment options are combined. Thörneby *et al.* [37] have shown that a natural treatment system consisting of ponds and a soil-plant system was successfully used for leachate treatment. Efficiency of overland flow technique in pilot-scale has been studied at the landfill of Hagby Återvinningsanläggning in the community of Täby, Stockholm [129]. However, Börjesson [130] has pointed out that a disadvantage of applying leachate by overland flow is the risk of long-term salt accumulation in the soil. For example, the content of chloride is normally much higher in landfill leachate than in municipal waste water [130]. Spraying the leachate with high sulphate, sodium and chloride concentrations directly on the leaves, caused growth disturbance as shown by Ettala [39]. These problems occurred when excessive irrigation at rates exceeding 500 mm were used during the growing season and when sprinklers were used instead of irrigation by hose [39]. During the last decade, a number of on-site leachate treatment methods, their suitability for Swedish conditions, treatment efficiency and cost have been studied, evaluated and summarized (Table 3) [16].

Table 2. Annual leachate amounts for pre-treatment and treatment of leachate in natural treatment systems in some Swedish landfills

Landfill, Location (reference)		**Landfill Area (ha)**	**Waste: MSW/ HW/IW)**	**Annual leachate amount m^3/y**	**Pre-treatment in ponds (m^3)**	**Treatment**
Umeå [131]	1974	12	HW+ IW +MSW	2001-2006: 36,000 - 78,000	E: 15,000 P^a: 15,000	I
Skedala [132]	50-ies	6	MSW, IW	90,000m	P^{as}: 4,000	CW
Kavaheden, Gällivare [133]	1973	40 (12)	MSW, IW	25,000	P^a: 10,000	W
Monkebo, Trollhättan [134]	60-ies	15	IW	ca 100,000	E: 7,000 $P^{a, m, an}$: 4,000 (20 d, 10 d)	CW: 5ha
Atleverket, Örebro [134]	1978	22	MSW, IW	70,000-115,000	$P^{a, m}$: 20,000 (2-4 months)	CW:53,000 (1 yr)
Gärstad, Lindköping [134]	-	-	MSW, IW	ca 150,000	P^a: (1 yr)	CW: 2,1 ha (2-3 weeks)
Isätra, Sala [134]	1973	10	MSW, IW	40,000	P^a; SBR; E	I, SP
Måsalycket, S:t Olof [134]	1975	9-10	MSW, IW	30,000-40,000	E: 13,000; P^a: 3,000	F_{sand}, P, I_{forest}, CW
Rödjorna, Skara [134]	1971	20	MSW, IW	15,000	P^a;	Ev
Korslöt, Trosa [134]	60-ies	-	MSW, IW	10,000	P:1250	I, CW:3.1ha
Tveta, Södertälje [134]	1975	36	MSW, IW	70,000	E^a: 15,000	Chem; $CW_{chips,peat, sand}$:800 (5d); I_{forest}
Moskogen, Kalmar [37]	-	22	MSW	150,000	P^a:30,000 P:54,000 P:52,000	SP $_{Willow}$:10 ha

E - Equalization pond
$P^{a, as, m, an}$ – Pond with aeration, aeration stairs, mixing, anaerobe zone;
CW – constructed wetland with ponds (surface-water constructed wetland);
I – Irrigation area; I_{forest} irrigation to forest;
SP – soil plant system;
F_{sand} – sand filter;
Ev – evaporation;
Chem – chemical treatment;
MSW- municipal solid waste;
IW – industrial waste;
HW – hazardous waste;
W – Wetland

Table 3. A comparison of biofilter systems and other local leachate treatment alternatives at Swedish landfill sites from treatment efficiency and operation condition points of view [16]

		Treatment efficiency %							Operation conditions					
	Method	COD	BOD	N-tot	NH_4	P	Metals	Bacteria	Cost, incl. operation	Required managem.	Sensitivity, flow variations	Level of technique	Area requirem.	A need for pre-treatment
1	Aerated pond	(-)	+	(+)	(+)	(-)	+	(+)	L	L	L	L	M	No
2	Aerated pond with chem.addition	(+)	+	+	+	(-)	+	+	M-H	M-H	L	M	M	No
3	SBR-technology	(+)	++	(+)	++	(-)	+	(+)	M	M	M	M	L	No
4	SBR-technology with chemic. addition	(+)	++	++	++	(-)	+	+	H	M	M	M	L	No
5	Recirculation*	(-)	(+)	(+)	(+)	(-)	(-)	(-)	L	L	L	L	L	Yes**
6	Irrigation to soil-plant system	+	+	+	++	++	+	(+)	H	M	M	M	H+	Yes**
7	Land treatment (forest)	+	+	+	++	++	+	(+)	L	L	L	L	H+	No
8	Land treatment (peat)	+	+	+?	+?	++	++	+	L	L	L	L	H+	No
9	Constructed wetland	+	+	++	++	+	+	(+)	H	M	L	M	H+	No
10	Sandfilter	(-)	+	(+)	(+)	+	+	+	M	L	L-M	L	L	Yes
11	Natural infiltration	(+)	+	(+)	(+)	++	++	++	L	L	L	L	L-M	No
12	Chemical precipitation	+	+	(+)	(-)	++	++	(+)	H	H	H	M-H	L	No
13	Ammonium stripping	(-)	+	++	++	++	++	++	H+	H	M	H	L	Yes**
14	Ultrafiltering	++	++	+	+	+	++	+	H	H	H	H	L	Yes
15	Reverse osmosis	++	++	++	++	++	++	++	H+	H	H	H+	L	Yes
16	Activated carbon	++	++	(+)	(+)	+	+	+	H	H	H	M	L	Yes
17	Evaporation	+	++	+	+	+	+	+	H	M	H	M	L	Yes**
18	Anaerobic filter	+	++	(+)	(-)	+	++	(+)	M	M	H	M-H	L	Yes**
19	Geofilter/biofilter	+	+	(+)	(+)	+	++	+	M	M	M	M	L	Yes
20	Wastewater treatment plant	+	++	(+)	++	++	+	(+)	M	L	M	M-H	L	(No)

*** For reduction of total nitrogen is required that recirculated water is nitrified (ammonium is treansformed to nitrate before recieculation takes place)**

****For all methods which iclude irrigation of leachate e.g. throgh the nozzles pre-treatment is required to reduce Fe content and a risk for clogging**

Explanation of characters:
(-) No effect/uncertain effect
(+) Low treatment effect
+ Moderate reduction
++ Good treatment effect

Operation conditions:
L - Low
M - Moderate
H - High
H+ - Very high

3. FROM LABORATORY SCALE TO FULL SCALE – DEVELOPMENT AND EVALUATION OF A FILTER-BASED LEACHATE TREATMENT SYSTEM

3.1. The case study

Stena Recycling AB is one of the major recycling companies in Sweden, recycling scrapped cars, and other complex ferrous and non-ferrous scrap metals. After dismantling, shredding, and recovery of recyclables, some complex ferrous and non-ferrous reject materials (shredder residue, SR) still must be disposed of. The disposed waste material from the separation and flotation processes consists of cable scrap, rubber, foam, plastics and textiles. The landfill operated by Stena Recycling was opened in the 1970s. Year 2004, an area of 24,000 m^2 is in use, whereas about 146,000 m^2 has been filled and covered. The landfill is surrounded by ditches that collect runoff water. About 22,000 m^3 of leachate was produced in 2001. In order to promote a closed loop strategy and to treat the leachate close to point of origin, an on-site treatment alternative was needed. A few preliminary investigations were made to establish the best strategy for on-site leachate treatment.

First, a combination of sand filtering, biological treatment in a bioreactor, ozonation and UV-treatment, activated carbon treatment, and denitrification was studied by Cannon and Alexandersson [135] in a pilot-plant designed by Lemna Nordic AB. Their results indicated that many compounds in leachate were successfully reduced. However, more investigations were needed concerning the reduction of heavy metals (*e.g.* Cu and Ni), COD and organic substances, and on whether these pollutants could be removed at more reasonable costs. The possibility to use less costly and possibly more effective natural treatment systems was found to be of great interest for Stena Recycling AB. The work in this Thesis has been directed towards this approach.

3.2. Methodology

3.2.1. Research strategy

Generally, no standard solutions for on-site natural treatment plants exist. Each case is site-specific, and even local climatic conditions must be carefully considered. In this particular

study, a design for an optimized, low-cost filter-based treatment plant for industrial leachate was targeted. The strategy was to develop a full-scale treatment plant through experiments at different scales, including batch-, column- and pilot-scale experiments (Figure 4) that were conducted in Sweden and Estonia. The approach was expected to yield better understanding of complex systems in real applications leading to optimal performance of a full-scale plant in a temperate climate. To assess the performance of a filter-bed system, and its efficiency, an evaluation was made of physical-chemical, biological and hydraulic processes in the filter as well as in the whole treatment plant.

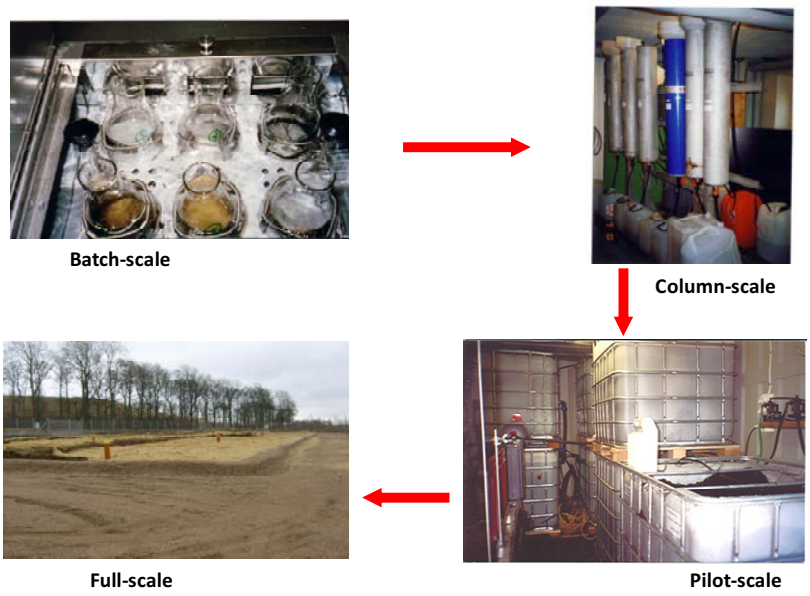

Figure 4. Overview of the successive scale-up engineering approach for the design and construction of a full-scale treatment plant for leachate from a particular industrial landfill

One of the key aspects of the research was the selection of filter material for the filters. Despite the fact that several materials have proven over and over again to be good sorbents in replicated batch and column tests, their ability to function effectively at pilot-scale or full-scale is seldom reported. In this Thesis, laboratory results have formed the basis for pilot-plant experiments at the landfill site. The design of a natural leachate treatment system for a specific landfill site is a complex matter, and knowledge obtained merely from laboratory studies, which were often carried out far away from the actual landfill site, is insufficient. A long-term on-site pilot-plant was accordingly set up to obtain more site-specific knowledge before a full-scale biofilter-based treatment plant was constructed. Such a study was important

for confirmation that under specific site conditions, the efficient treatment could be achieved by the proposed filter-bed system. Knowledge about the potential benefits from pre-treatment of leachate, particularly from an aeration and sedimentation step, as well as knowledge on climatic effects and system maintenance aspects, was sought with the pilot-plant tests. It is very usual that full-scale filters are constructed but that appropriate follow-up studies are not undertaken or published in the literature, probably due to a lack of financial resources. So far, the only study which reports about usage of peat in full-scale filters has been in Scotland [136]. Such follow-up studies, however, are of great importance if a stable and successful operation of the treatment plant is targeted. In addition, availability of reports on results and operational experiences with full-scale plants could contribute to working out general guidelines for construction of natural treatment systems for landfill leachate. In this work, the performance of a full-scale treatment plant was evaluated over a long period from several points of view.

3.2.2. Objectives of the experiments at different scales

The objectives of experiments performed at different scales during this work are presented in Table 4.

Table 4. Research objectives of the different types of experiments carried out

Scale	Objectives
Batch (Papers I and II)	First phase for the selection of filter material for the filter bed – equilibrium studies: • Determination of the maximum adsorption capacity of peat; • Evaluation of the simultaneous removal of several metals from spiked water solutions and leachate by different adsorbents; • Evaluation of simultaneous removal of metals and organic pollutants at various initial concentrations from spiked water solutions and leachate by a mixture of peat and carbon-containing ash; • Characterization of new and spent filter media (a mixture of peat and carbon-containing ash) including leaching tests and determination of composition of material.
Column (Paper III)	The second phase for the selection of filter material for the filter beds – dynamic studies: • Evaluation of simultaneous removal of metals and organic pollutants from leachate and spiked leachate by different adsorbents in a dynamic system; • Determination whether breakthrough was reached at low and varying initial concentrations of pollutants in leachate or at

	high concentrations in spiked leachate; • Evaluation of whether the selected flow rates can be held throughout the experiment without deterioration of the hydraulic performance of the filter (*e.g.* clogging).
Pilot (Papers IV and VIII)	Long-term (3-year) investigations about the treatment efficiency at on-site conditions: • Evaluation of treatment of leachate with actual influent concentrations of different pollutants; • Evaluation of simultaneous removal of different types of pollutants in a filter containing a mixture of peat and carbon-containing ash; • Pre-treatment of leachate: measuring the efficiency of extended aeration and prolonged sedimentation; • Understanding climatic effects in site-specific location (temperate climate); • Understanding system maintenance needs (*e.g.* different treatment combinations in sequence, and mitigating the risk of clogging). Research about the initial start-up period of a filter-bed treatment plant, and its efficiency in pollutant removal for different types of peat and leachates (Paper VIII).
Full (Papers V, VI, VII)	Full-scale on-site treatment of leachate from an industrial landfill: • Evaluation of simultaneous removal of different types of pollutants in a biofilter containing a mixture of peat and carbon-containing ash during a three-year period. Variable influent concentrations were studied on-site under actual conditions. Hydraulic performance of a full-scale biofilter system: • Research about climatic effects (*e.g.* precipitation, evaporation and temperature) on the generation and treatment of leachate in a biofilter system under on-site conditions; • Research about designed and actual hydraulic loading of the biofilters; • Research about changes in the hydraulic properties of the biofilter and its medium over time (*e.g.* risk for clogging, changes of infiltration capacity, grain size distribution). Evaluation of bacterial community dynamics in the leachate treatment system: • Research about abundance and spatial dynamics of heterotrophic and phenol-degrading bacteria.

The following chapters concern mainly practical aspects of the work along with a general discussion about the objectives presented in Table 4. However, more information can be found in Papers I to Paper VIII.

3.2.3. Solutions and filter materials

Leachates from an industrial landfill site in Halmstad (Sweden), a MSW landfill in Kristianstad (Sweden) and two MSW landfills in Estonia (Aardlapalu and Väätsa) were used in the studies (see Table 5). In some experiments the leachate was spiked with different types of pollutants. Artificially prepared solutions, *e.g.* distilled water or tap water spiked with different compounds, have also been used, with key components being selected based on leachate composition.

Different types of low-cost filter media where tested in this work including naturally abundant materials (in Sweden and Estonia) with no need for sophisticated processing; by-products or waste materials from industrial processes; and some artificial adsorbents, as well as mixtures of these materials:

- Peat[a], peat[d], peat[e] (from Estonia, Paper I and Paper VIII);
- semi-coke (Estonia, Paper I);
- pine bark (Sweden, Paper I);
- paper pellets (Estonia, Paper I);
- hydrophilic and hydrophobic artificial adsorbents prepared from urea-formaldehyde resin (named Blue and Red ads., respectively) (Estonia, Paper I);
- a mixture of peat[b] and carbon-containing ash (Sweden, Papers I, II, III, IV, V, VI, VII);
- a mixture of peat[a] and Blue adsorbent (Estonia, Paper I);
- a mixture of peat[b], carbon-containing ash and polyurethane (PUR) (Sweden, Paper III);
- a mixture of peat[c] and wood pellets (Sweden, Paper III);

The materials used in the different experiments are presented in Table 5. Details concerning the origin and composition of the materials are presented in Papers I, II, III and VIII.

Table 5. Solutions and filter materials used in the experiments

	Experiments						
	Batch	**Batch**	**Column**		**Pilot-scale**	**Pilot-scale**	**Full-scale**
	(Paper I)	**(Paper II)**	**(Paper III)**		**(Paper IV)**	**(Paper VIII)**	**(Papers V, VI, VII)**
Filter Materials	Peat[a] Peat[b]+Ash Peat[a]+Blue ads. Blue ads. Red ads. Pine bark Semi-coke Paper pellets	Peat[b]+Ash	Peat[b]+Ash Peat[b]+Ash+PUR Peat[b]+PUR Peat[c]+Wood chips	Peat[b] Peat[b]+Ash	Peat[b]+Ash	Peat[a] Peat[d] Peat[e]	Peat[b]+Ash
Amount of material	1 g	4 (<250*) g	0.014 m^3	0.04; 0.05 litres	0.8 m^3	1 m^3	720 m^3
Leaching by water	Yes	Yes	Yes	No	No	No	Yes
Spiked water	Metals	Metals, PCBs, phenols	No	No	No	No	No
Type of landfill leachate	Industrial	Industrial	Industrial	MSW	Industrial	MSW	Industrial
Spiked leachate	Metals	Metals, PCBs, phenols	No	PBDEs	No	No	No
Test duration	24 h	24 h	6 months	weeks	3 years	up to 22 days	3years

*leaching test

[a-c] different types of peat collected from Estonia and Sweden

Peat was selected as the main material for the filter bed, since it is a cheap and natural material, abundantly available in Sweden. The carbon-containing ash from the pulp and paper industry containing un-combusted carbon was chosen to substitute expensive activated carbon for removal of organic pollutants. By mixing peat and carbon-containing ash, a filter medium was obtained which was expected to be a good alternative for simultaneous removal of several types of contaminants in leachate. Despite the otherwise favourable properties of ashes, they are known to have fine particle size and cementing properties, causing low permeability in dynamic systems. Therefore, mixing peat with ash was intended to give good hydraulic conductivity of the medium. In addition, Mårtensson *et al.* [77] have also pointed out that use of a mixture of peat and carbon-containing ash in a filter is more advantageous than peat alone from a pollutant removal point of view. However, to obtain confirmation and appropriate comparisons, other adsorbents were included in the batch studies as well.

In batch equilibrium experiments, all filter media based on peat showed high adsorption capacity of metals compared to other tested media. The mixture of peat and carbon-containing ash was selected for further studies performed as column experiments (Paper III), instead of peat and a mixture of peat and Blue adsorbent. This decision was based on economical considerations, since carbon-containing ash is much cheaper than Blue adsorbent. Moreover, transportation of the material from Estonia for a full-scale application in Sweden would increase the capital costs of the plant. A mixture based on Blue adsorbent, could most probably be used as a polishing step. Polyurethane and wood chips (wood processing waste) were selected as an alternative to carbon-containing ash, based on good results reported in the literature [137-140]. The usage of polyurethane material was especially interesting, since it was available as a waste from the metal recycling company and could thus be obtained at no cost.

The optimal mixing ratio of different components in a filter is dependent on resulting hydrodynamic performance and also on the concentrations of pollutants which need to be removed. In Paper III, a ratio of 3:1 between peat and another component (carbon-containing ash or polyurethane) was selected due to two reasons. The aim was that both organic and inorganic compounds should be trapped in filter media. It was expected that the capacity for metal ions, preferably trapped by peat, would be the limiting capacity factor of the mixture. Thus a larger portion of peat was included in the mixture. Secondly, the mixture with a ratio of 3:1 gave good hydrodynamic performance. This does not exclude the possibility that a mixture with somewhat different proportions could perform slightly better as a filter. However, a three-year run of the full-scale treatment plant has revealed that the chosen mixture has performed satisfactorily.

3.2.4. Experimental design

A brief description of the experimental set-up is presented in this chapter. The layout of experiments is shown in Figure 5 and details can be found in referenced Papers I to VIII.

3.2.4.1. Batch and column studies

Papers I and II

Two batch-size experiments were performed (Figure 5). In the first experiment, filter media were placed into Erlenmeyer flasks (250 ml) and 100 ml of the solutions with certain

concentrations of metals were added. The Erlenmeyer flasks were shaken on a shaking table. The concentrations of pollutants in the solutions before and after shaking were determined.

In the second experiment, filter material was placed into 2-litre polypropylene bottles and 1 litre of solution was added. The bottles were agitated on a shaking table. The concentrations of pollutants in the solutions before and after shaking were determined.

Paper III

Columns were packed with filter materials and solutions were pumped onto the material. Two sizes and types of column were used in the column test. For investigation of inorganic and polar compounds, stainless steel and PVC columns with an inner diameter of 150 mm and a height of 1 m, were filled to a level of 0.8 m with the filter materials. The leachate from the industrial landfill was collected and stored in a tank (1 m^3) where aeration also took place. The leachate was then transferred from this tank to smaller containers and from these it was applied to the filter material in the columns. The leachate was pumped onto the columns (downwards flow mode) for 60 min twice a day at a flow rate of 9 mL/min (1.1 L/day), corresponding to a hydraulic loading of 61 mm per day.

For investigation of non-polar organic compounds, two glass columns with an inner diameter of 2 cm were used for experiments. One column was filled with 8.7 g of peat (P1), producing a filter length of 15.5 cm. The second column was filled with 20.1 g of a mixture of peat and ash (P1/A1; 10.05 g each of P1 and A1), producing a filter length of 18.6 cm. The relatively small inner diameter and length of the columns were selected to maximize the likelihood of forced flow-rate phenomena, because large breakthrough volumes were expected for substances such as PBDEs. The system was closed, allowing the constant feeding of spiked leachate A at a low overpressure. Two peristaltic pumps were used, operating at flow rates of 50 mL/h for P1 and 35 mL/h for P1/A1.

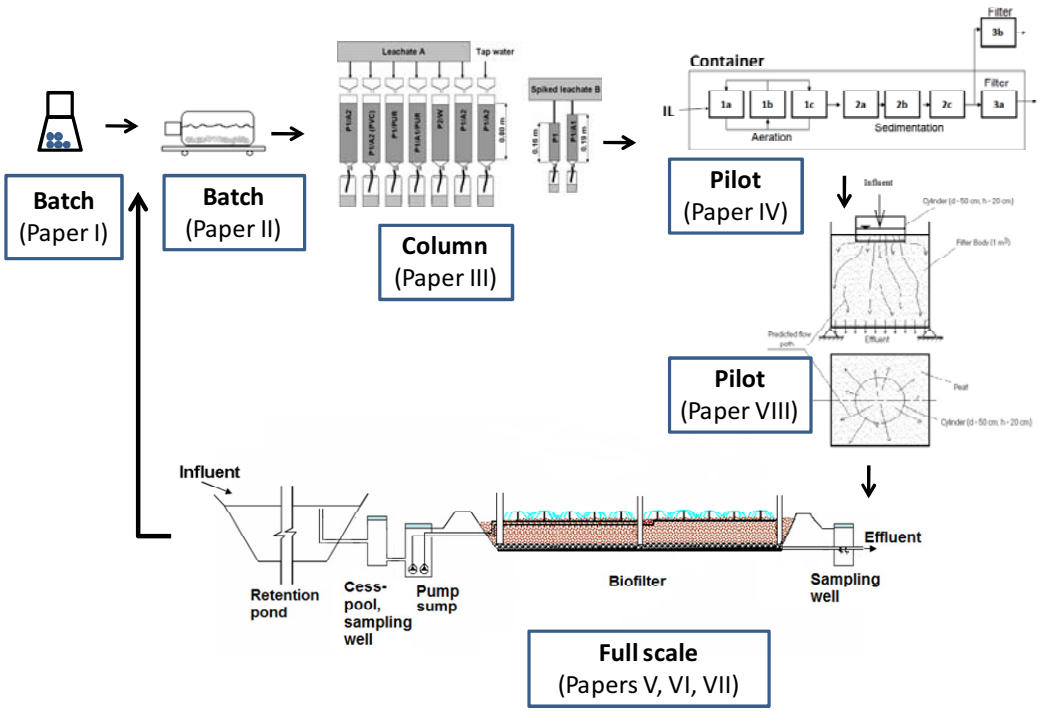

Figure 5. The layouts of experiments at different scale

3.2.4.2. Pilot-plant studies

Paper IV

The pilot plant was housed in a large metal container (4 × 2.5 × 2.5 m), positioned at the landfill site next to a pond receiving the leachate from a landfill site in Halmstad. The set-up of the pilot plant includes a series of polyethylene tanks (1 m^3 each) allowing sequential performance of three treatment steps: aeration, sedimentation and filtration through the two parallel filter beds. Leachate was intermittently loaded onto the filter by pumps regulated by timers. The flow rate to the each biofilter was 165 (standard deviation, SD, 30) L/(m^2 d) during the first month and 110 (SD 10) L/(m^2 d) thereafter.

Paper VIII

Experiments were performed in three custom-designed vertical flow peat filters with a volume of 1 m^3, rectangular shape (1 × 1 × 1 m), and a permeable floor (50 × 50 mm metal net covered with a 1 × 1 mm plastic net). Two filters received pre-treated leachate. After extended aeration during 11 days in a 3 m^3 Roth Micro-Step fine-bubble compact wastewater treatment unit the leachate was loaded on two filters for 4 h each day over a period of 4 days. The flow rates were 90.3 and 2 l/min respectively. A second type of leachate was biologically pre-treated in a compact two-phase activated sludge leachate treatment plant, which is

accomplished with an aerobic-anoxic sedimentation pond after the plant. Leachate from the outflow of the sedimentation pond was loaded onto the filter by timer-adjusted pumping during 22 days, providing 24 pumping events per day, each of which was 15 min long (2.9 m^3/day). During the experiments, the total loading of leachate on a filter was about 104 m^3.

3.2.4.3. Full-scale filters

Papers V, VI, VIII

The constructed full-scale on-site leachate treatment system consists of a pond with size of about 500 m^3 and a vertical-flow biofilter system (Papers V, VI and VII). The biofilter system consists of four parallel biofilters, each with an area 30 m × 6 m (length × width) and with a depth of the filter medium of 1 m. In Paper V, the treatment efficiency of the biofilter system during a three-year period was evaluated.

3.2.5. Sampling and analysis

Proper sampling and analytical techniques are of fundamental importance in evaluation of treatment system efficiency. Representivity and reproducibility mean that the data must represent the water or environment being sampled and that the data obtained must be reproducible by others following the same analytical protocol. The analyses were performed according to a LAQUA protocol, described elsewhere in detail [35, 45, 141]. Generally, the analytical protocol addresses parameters to be measured in leachate and gives suitable analytical procedures for carrying out these measurements. The protocol was designed based on previous studies by our research group and based on a literature screening about the composition of different leachates. The protocol makes it possible to compare results obtained in different research projects within the research group, which involve scientists from several departments and universities. Analytical research was performed by the author in the laboratories of Lund University, Kristianstad University and Kalmar University in Sweden and Tartu University and Estonian University of Life Sciences in Estonia.

The research included an extensive sampling program. The volume of the sample depended on the analysis method to be used as well as on the usual sampling requirements. In most cases, grab samples were collected manually. During one occasion in the full-scale plant, grab sampling was combined and compared with 24-hour time-integrated auto-sampling.

Prompt analysis is undoubtedly the most positive assurance against error due to sample deterioration [83]. However, due to time-consuming field work and the distance between the plant and the laboratory, determination of all field samples during the same day they were acquired was not possible. Thus, the samples were normally collected and kept in a refrigerator overnight to avoid any change in sample composition during storage and analysed the next day. In some cases, when direct analysis was not possible, the sample was deep-frozen and stored. Samples were always taken in the same way and at the same points on-site to reduce errors due to sampling.

3.2.6. Sources of errors

Heterogeneity of the filter material is a source of error in this type of study. One limitation in the presented batch and column tests was that for most sorbents, except for the mixture of peat and carbon-containing ash, no replicate experiments were done. In the column experiment, three columns containing material composed of peat and carbon-containing ash were tested. This was due to the fact that the main interest was focused on this particular mixture, while other sorbents were selected to give some comparative results. Since experiments were carried out at the same time and the experimental procedure was the same, the results should be comparable. As Lee *et al.* [63] have also pointed out, a more detailed experimental column-scale study, where three replicate filters would be used, would not be justified in terms of costs and potential scientific benefit.

In batch scale tests it is difficult to obtain a suitably small sample, which is representative, even if this sample comes from a carefully mixed large supply of material. Fortunately, as shown by column test results (Paper III), the use of the same materials from different storage batches of filter material did not considerably affect the treatment efficiency of pollutants. To reduce the error when a small sample volume is collected from a large heap of material *e.g.* a full-scale filter, the principle of sub-samples was followed. For instance, to characterise the filter material after three years of use at a full-scale treatment filter, sub-samples were taken with a drill at a depth of 0 to 0.6 m from 16 sampling points, equally spread over the surface of the filter bed (180 m^2 area), and mixed to form one sample (Paper II). However, still better precision is expected when the number of samples to be analysed can be increased. This will of course also increase the costs for the investigation, very much if the analyses are complicated. This approach was used in a comparison of the bacterial population in the four

filters, when samples were taken from 16 places of the treatment plant and all analyzed individually.

3.3. Results

3.3.1. Characterisation of leachate

Since the treatment methods are dependent on the characteristics of leachate, the industrial landfill leachate was characterised. Average concentrations of different pollutants in the leachate were quite similar to those characterising leachates from MSW landfills in several countries (Table 1). However, conductivity, concentrations of DOC and TOC and concentrations of Cu and Na were somewhat higher than in the leachates from other landfills. A low BOD/COD ratio suggests that concentrations of volatile fatty acids are low. Thus, most probably the industrial landfill leachate also has relatively high concentrations of humic- and fulvic-like compounds. Detailed study of these compounds, however, was beyond the scope of this study. Landfill leachate contains a wide variety of organic compounds. In this Thesis screening of organic compounds with different chemical properties, namely *non-polar and weakly polar semi-volatile organic compounds* (SVOCs), *non-polar compounds* (with PCBs as markers) and *polar organic compounds* (with phenolic compounds as markers) was considered. Generally, a characterisation of SVOC in the leachate showed that siloxanes, ketones, phenols, alcohols, esters, alkyl phosphates and phthalates were present in the leachate, as was also common in leachate from MSW landfills [31, 37, 142]. A semi-quantitation (by comparison to the response of a known concentration of naphthalene) of the compounds found in the industrial leachate showed that the concentrations of these compounds were low. The detailed results about organic pollutants can be found in Paper V.

The spatial distribution of pollutants in leachate from several drainage pipes at the landfill revealed, that the strength of the leachate varied considerably (Paper V). This is obvious in Figure 6 in which concentrations of PCBs in the leachate from three main drains, the recipient and at other sampling points on the landfill as measured on one occasion are presented. The occurrence of PCBs in the leachate was not surprising, since already in 1998 Sakai *et al.* [143] also reported that the SR from automobile and electric appliance contains persistent organic pollutants (POPs) such as PCBs and PCDDs/DFs. The concentration of PCBs in the leachate from the industrial landfill is in the range found in other landfill leachate in Sweden [144]. As shown in Paper V, concentrations of metals in leachate from different parts of landfills varied

as well. The concentrations of Cu, Zn and Sb differed the most. It was also noticed that concentrations varied considerably during the overall three-year period (Paper V). The leachate from drains was collected to the pond, which somewhat equalises the concentrations of pollutants prior to treatment. Similar to other studies, seasonal variation in leachate composition was noticed. During rainy periods on some sampling occasions, a high flow rate of leachate was accompanied by higher concentrations of some metals (*e.g.* Cu) than normal, indicating flush out of pollutants from landfills under large flow conditions. In contrast, Åkesson and Nilsson [145] observed lower leachate concentrations in the wet season in a Swedish landfill test cell.

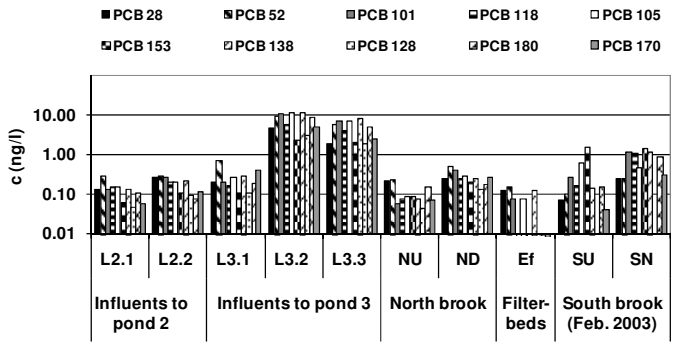

Figure 6. Concentrations of PCBs at several points of the landfill site (L3.1, L3.2 and L3.3 are from 3 drains from the landfill, which deliver leachate to the pond 3); L2.1 and L2.2 – runoff water from landfill collected to pond 2; Ef – effluent from filter bed system; SU and SN are located in a brook south of the landfill area; NU and ND – upstream and downstream of treated leachate discharge point to the recipient

3.3.2. Treatment of pollutants in filter beds at different scales

The results from all experiments at several scales indicate that a good treatment efficiency of both inorganic and organic pollutants was achieved in a filter bed with a mixture of peat and carbon-containing ash (see Table 6).

3.3.2.1. Metals and water quality parameters

As shown in Table 6, although the concentration of Cu in the influent for a mixture of peat and carbon-containing ash at different scale studies was not the same, similarly good reduction of Cu was achieved. Moreover, a good reduction of Pb, Fe, Mn, Sn and Hg was

achieved in all experiments. However, results concerning removal of metals such as Cd, Sb and Zn did not show a similar trend from the different scale studies. All the studies show that there is a leakage of As from the filter material, most probably from carbon-containing ash. In addition, the concentrations of Ca and Mg were noticeably increased in pilot-scale and full-scale filters in this study (Paper IV and V). As shown by the authors (Paper I) and other researchers [146, 147] in previous studies, Ca and Mg ions are exchangeable ions in adsorption of Cu, Pb, Cd and Ni onto peat from solutions. The result obtained in this study (Paper V) confirms that the peat used has, as does most other peat or filter media, ion-exchange properties. In most instances, these exchangeable ions are not a problem in receiving waters. For instance, sulphate is usually the exchangeable ion for activated carbon, and potassium for compost [148]. Zeolites appear to exchange sodium and some divalent cations (increasing water hardness as a result) for the ions they sorb [148].

The results show that the changes in water quality parameters, such as N_{tot}, NH_4-N and DOC in the pilot and the full-scale filters are similar to each other. A removal of TOC is slightly better in the full-scale than in the pilot-scale filters. The reduction of TOC achieved in the industrial landfill leachate by the filter composed of a mixture of peat and carbon-containing ash at full-scale is similar to the removal efficiency of 32% obtained for leachate from MSW landfill achieved with a similar filter material by Mårtensson *et al.* [77]. The removal of NH_4-N is higher than that of N_{tot}, since nitrogen is transformed from NH_4-N to NO_3-N in the system, but is not totally removed. Typical for vertical-flow filters is that conditions in the biofilter system are appropriate for microbial nitrification [149, 150], but that the denitrification is limited [150]. By using a similar mixture of peat and carbon-containing ash, the reductions of NH_4-N and N_{tot} in leachate from a MSW landfill were previously found to be 52% and 14%, respectively [77]. Although concentrations of NH_4-N and Tot-N in the industrial landfill leachate were reduced in the treatment plant, the values of Tot-N were still

Table 6. Average concentrations and removal efficiencies (R, in percentage) of different pollutants in a mixture of peat and carbon-containing ash for different scale filters

	Treatment efficiency of filter mixture of peat and ash at different scales									
	Batch I (Paper I)		**Batch II** (Paper II)		**Column** (Paper III)		**Pilot** (Paper IV)		**Full-scale** (Paper V)	
Parameters	**Conc.**	**(R%)**	**Conc.**	**(R%)**	**Conc.**	**(R%)**	**Conc.**	**(R%)**	**Conc.**	**(R%)**
N_{tot} (mg/l)	na	na	na	na	na	na	190	(23)	150	(25)
NH_4-N (mg/l)	na	na	na	na	na	na	90	(46)	100	(45)
S (mg/l)	na	na	na	na	na	na	10	(-26)[1]	11	(-4)
DOC (mg/l)	na	na	na	na	265	(50)	420	(26)	450	(28)
TOC (mg/l)	na	na	na	na	na	na	460	(21)	470	(30)
Cu (µg/l)	6680	(85)	1060	(74)	63	(72)	130	(73)	190	(72)
Al (µg/l)	na	na	na	na	67	(22)	66	(-66)	160	(-39)
As (µg/l)	na	na	na	na	14	(-66)	14	(-126)	12	(-170)
Cd (µg/l)	1960	(98)	na	na	0.2	(-45)	0.43	(39)	0.70	(60)
Co (µg/l)	na	na	na	na	2.7	(3)	3.3	(8)	5.6	(35)
Cr (µg/l)	na	na	na	na	1.5	(66)	4.8	(10)	8.4	(42)
Hg (µg/l)	na	na	na	na	0.5	(68)	0.86	(41)	0.78	(63)
Pb (µg/l)	4840	(100)	na	na	5.4	(88)	7	(78)	7.9	(55)
Sb (µg/l)	na	na	na	na	2.5	(-27)	3.2	(-26)	5.4	(29)
Ni (µg/l)	7660	(75)	na	na	37	(27)	56	(14)	76	(37)
Sn (µg/l)	na	na	na	na	1.5	(94)	6.3	(55)	8.4	(66)
Zn (µg/l)	na	na	na	na	26	(30)	37	(47)	56	(15)
Li (mg/l)	na	na	na	na	na	na	1.3	(6)	1.2	(6)
Fe (mg/l)	na	na	na	na	0.06	(75)	0.8	(74)	1.5	(55)
Mn (mg/l)	na	na	na	na	0.97	(50)	0.44	(56)	0.92	(73)
Mg (mg/l)	na	na	na	na	na	na	84	(-6)	81	(19)
K (mg/l)	na	na	na	na	na	na	175	(1)	140	(5)
Ca (mg/l)	na	na	na	na	na	na	30	(-48)	30	(-83)
Na (g/l)	na	na	na	na	na	na	1.1	(0)	0.90	(8)
Phenol (µg/l)	na	na	970	(36)	145	(94)	10	(74)	24	(89)
p-o-cresol (µg/l)	na	na	2180	(20)	93	(80)	140	(91)	690	(92)
2,4-dimethylphenol (µg/l)	na	na	890	(18)	9	(78)	16	(73)	30	(80)
4-chloro-3-methylphenol (µg/l)	na	na	1070	(20)	35	(89)	62	(60)	100	(75)
4-chlorophenol (µg/l)	na	na	1130	(36)	5	(48)	7.5	(-184)	26	(95)
Total of 29 PCBs (ng/l)	na	na	na	na	na	na	na	na	151	(70)
PCB52 (ng/l)	na	na	0.044	(99)	na	na	na	na	0.71	(62)
PCB95 (ng/l)	na	na	0.029	(97)	na	na	na	na	na	na
PCB101 (ng/l)	na	na	0.024	(91)	na	na	na	na	0.74	(45)
PCB149 (ng/l)	na	na	0.024	(86)	na	na	na	na	na	na
PCB153 (ng/l)	na	na	0.026	(82)	na	na	na	na	0.91	(52)
Di-BDE (mg/l)	na	na	na	na	na	na	na	na	na	na
Hexa-BDE (mg/l)	na	na	na	na	1	(99)	na	na	na	na
Hepta-BDE (mg/l)	na	na	na	na	1	(>99.5)	na	na	na	na
Octa-BDE (mg/l)	na	na	na	na	1	(98)	na	na	na	na
Nona-BDE (mg/l)	na	na	na	na	1	(99.5)	na	na	na	na
Nona-BDE (mg/l)	na	na	na	na	1	(99)	na	na	na	na
Deca-BDEs (µg/l)	na	na	na	na	10	(>99.5)	na	na	na	na

[1]Negative values of R indicate increase of concentration in the effluent;

– The result is not significant at 95% level;

Values of pH in influent of batch I, batch II, column, pilot and full-scale filter were 8.0, 7.7, 8.4, 8.6 and 8.1, respectively

too high compared to the legally accepted limit values (15 mg/l) for wastewater discharge to recipients in Sweden [151]. Thus, to further increase the bacterial denitrification of NO_3-N in the leachate, while at the same time decreasing the N_{tot} content, a polishing step is recommended downstream from a biofilter system. A free-surface constructed wetland with plants (similar to the one described by Bachand and Horne [152]) or a horizontal-flow filter [56, 153] can serve as effective polishing steps. Today, the effluent of the biofilter treatment system is discharged into a brook, which downstream falls to a free-surface constructed wetland, which was built for improvement of the quality of storm-water runoff from several nearby industrial areas. Thus further removal of N_{tot} most probably might occur in the constructed wetland.

3.3.2.2. Removal of organic pollutants

PCBs and PBDEs

The results from the batch test show that the removal efficiency of PCB congeners with a mixture of peat and carbon-containing ash was high from the MSW landfill leachate spiked with PCB congeners (Paper II). In addition, results from full-scale plant study (Paper V) show that the removal efficiency of ten PCB congeners was good in the biofilter, even if the concentrations in the influent were low (in the range of 0.01 to 10 ng/l) in the industrial landfill leachate. The results in Paper V also show that the average reduction of total concentration of 29 investigated PCB congeners in the full-scale biofilter was 70%.

Investigation of polybrominated diphenyl ethers (PBDEs) did not show any trace of any PBDE [36] in raw leachate from the industrial landfill. However, in leachate from a landfill located in Kristianstad, two peaks appeared at the GC retention time corresponding to BDE-153 and BDE-183 with concentrations of 3.5 ng/l and 23 ng/l, respectively [36]. Therefore, leachate from the landfill in Kristianstad was selected for investigating the removal of PBDEs as markers for non-polar organic pollutants. Due to low background concentrations, the leachate was spiked (see Paper III). Concentrations of pollutants were measured in the influent and in the effluent. Results in Paper III show that the reductions of non-polar organic compounds in the column filled with peat and ash were close to 100%. There was no tendency for breakthrough in the material, even after loading about 100 bed volumes, which is an encouraging result. This experiment also shows that although small columns with a relatively small amount of filter materials are used, long experimental time (several months or years)

might be needed to reach the breakthrough point of pollutants for adsorbents with high adsorption capacity (as was stated in chapter 2.6.5.2.).

Although concentrations of POPs, such as PCBs and PBDEs are low, removal of them from leachate is of great importance, due to their persistence, possible biomagnification and potential toxicity for receiving waters.

SVOCs

The reductions of semi-volatile organic compounds (SVOCs) were determined in pilot and full-scale biofilters and obtained results were similar. Even if the concentration of SVOCs were low, at trace level (according to a semi-quantitative comparison to the response of a known concentration of naphthalene) in the leachate, very good reduction of several compounds in the biofilter was found (Papers IV and V). However, both studies indicate that *e.g.* phthalate concentrations are not reduced in the biofilter. This might be dependent on the use of PVC-material (which contains phthalates) in the construction of both the pilot and full-scale plant. In a previous study Thörneby *et al.* [37] have shown a reduction of total phthalate concentration by about 40% in a natural treatment system.

Phenolic compounds

The results obtained from the batch experiment show that the removal of polar organic compounds occurred with only limited efficiency at neutral or slight basic pH. For instance, Viraraghavan and Alfaro [154] reported that adsorption of phenol from an initial concentration of 1 mg/l on peat and fly ash were 46% and 42% respectively, and they noted that the optimum pH range for both adsorbents was between 4.0 and 5.0. Adjustment of the pH in leachate towards lower values in the batch experiments would most probably have improved the removal efficiencies of phenols. In a full-scale treatment plant, however, pH adjustment will increase the treatment cost and would also affect other processes in the filter involved in *e.g.* metal removal. Therefore, such an approach does not seem to be the best alternative, especially when considering the results obtained with the pilot-scale and the full-scale filters. The high reduction of phenolic compounds at slightly basic pH in column-, pilot- and full-scale biofilters with a mixture of peat and carbon-containing ash was in strong contrast to values obtained in batch-scale studies.

3.3.3. Filter bed as biofilter

The high reduction of phenolic compounds in these studies (Paper IV and V) is most probably dependent on biodegradation by microbial communities. The microbial counts of phenol-degrading bacteria in the filter material acquired from the full-scale filter were abundant, in a range between 0.2×10^6 and 1×10^6 CFU/g. In addition, the number of aerobic heterotrophic bacteria in the filter materials was determined. The results show that the number of aerobic heterotrophic bacteria varied between 0.5×10^6 and 10^7 CFU/g, which is comparable with values in the range 6.3×10^6 to 1.1×10^8 CFU/g obtained in a horizontal subsurface flow filter bed loaded with wastewater [155]; and 1.1×10^7 CFU/g in a pilot test plot at a semi-coke deposit covered with a peat layer (1-2 cm) and a grass mixture [156]. These findings are expected, since unsterilized peat is home to a number of microorganisms, including bacteria, fungi and higher organisms, and even tiny plants. The number of aerobic heterotrophic bacteria in the raw peat used in the filter mixture (blank) was 1.1×10^4 CFU/g. Thus, the study shows that the abundance of microorganisms had been increased after loading of the filter by the industrial landfill leachate in the treatment system. These studies indicate that conditions for biological degradation of compounds in the biofilter are favorable and that the filter acts as a biofilter.

Straw laid on the filter for insulation before winter, becomes decomposed due to spreading of leachate by sprinklers on the top of the straw at the next warm season. The decomposition rate, or availability of organic carbon as an energy source for heterotrophic bacteria, depends on the initial litter composition, *i.e.* on the nitrogen and lignin content [157]. Plants with lower initial lignin:nitrogen ratio are usually more available for decomposition, and hence promote the growth of denitrifying bacteria. The number of aerobic heterotrophic and phenol-degrading bacteria in samples of partly decomposed straw collected from the surface of the filter system was 2.3×10^7 and 9.7×10^5 CFU/g, respectively.

Analyses of the aqueous samples from the pilot plant and along a flow path in a full-scale treatment system have confirmed that phenol-degrading bacteria were present in aqueous samples at both plants (Papers IV and VII). A comparison between the abundance of heteretrophic bacteria in the solid filter samples and in the leachate samples at full-scale plant show that the number of bacteria was 1-2 orders of magnitude higher in the solid samples than in the leachate in the ponds. This indicates that conditions for bacterial growth are more

favourable in the filter system than in the leachate. This is consistent with results obtained by others, *e.g.* Haglund *et al.* [158], who showed that the abundance and activity of free-living bacterial communities in streams is lower than in the biofilm and sediment. Several other studies have also shown that particle-attached bacteria are often more abundant and more functionally active than their free-living counterparts at the same location [159, 160]. Additional findings on abundance and spatial dynamics of heterotrophic and phenol-degrading bacteria as well as on the functional and metabolic diversity of the bacterial community along a collection and treatment system for leachate from the industrial landfill are presented in Paper VII.

3.3.4. Leaching

Since residue products such as waste materials were used in the filter mixture, the leaching of different pollutants was assessed by several tests at different scales. This investigation included a batch test with mixtures, which were unused and which were used during the 3-year period of full-scale plant operation. Moreover, a column test, in which tap water was loaded on the material, and also a full-scale study where tap water was re-circulated through one of the filters, was carried out. The results are presented in Table 7.

In batch tests reported in Paper II, compositions of filter materials were also determined. Comparisons between total concentrations of different species in solid filter material (data in Paper II) and in the eluate from the leaching test (according to SS-EN 12457-4 [161]) were carried out. The results show that even if the composition of the filter medium contains certain pollutants as “background value of pollutants”, values in the effluent are very small. This confirms that a very small part of the background components was leached out and that the unused mixture of peat and carbon-containing ash in this respect is a good material for a filter-based treatment plant. The leaching tests with tap water applied to a mixture of peat and carbon-containing ash generally did not show any significant leakage of pollutants from the filter material in batch, column or full-scale studies (Papers II, III and V). One exception was As, which most probably leached from the carbon-containing ash component in the filter. In Paper II, it was also shown that the leakage test using water envisioned by the EU gave one order of magnitude lower pollutant concentrations in the effluent than when using real landfill leachate for leaching of solid material. Thus the development of a synthetic leachate solution

for this type of testing seems to be a good idea, since it should give more realistic results expected in real leachate treatment systems.

Table 7. Leaching of pollutants from a mixture of peat and carbon-containing ash (average concentrations (Conc.) and standard deviations (SD))

	Batch (unused mixture) (Paper II)		**Batch (used mixture)** (Paper II)		**Column** (Paper III)		**Full-scale*** (Paper V)	
Parameters	**Conc.**	**(SD)**	**Conc.**	**(SD)**	**Conc.**	**(SD)**	**Conc.**	**(SD)**
pH	7.6	0.3	8.3	0.3	6.8	(0.1)	na	na
Conductivity	0.03	0.003	0.02	0.002	0.88	(0.1)	na	na
DOC (mg/l)	53	6	97	35	21	(1)	23	(11)
SS (mg/l)	na	na	na	na	na	na	36	(10)
Cu (µg/l)	34	(24)	193	(163)	4.4	(4.2)	1.5	(0.5)
Al (µg/l)	29	(16)	1576	(1887)	11	(3.7)	3.5	(2.9)
As (µg/l)	12	(1.0)	5.4	(3.5)	13	(2.3)	8.8	(5.9)
Cd (µg/l)	0.10	(0.03)	0.38	(0.25)	0.02	(0.02)	0.12	(0.06)
Co (µg/l)	1.1	(0.21)	1.9	(1.2)	0.52	(0.32)	0.7	(0.4)
Cr (µg/l)	3.0	(0.38)	6.1	(3.6)	0.1	(0.3)	1.4	(0.3)
Hg (µg/l)	0.08	(0.032)	0.78	(0.72)	0.15	(0.13)	0.04	(0.02)
Pb (µg/l)	0.16	(0.06)	6.3	(6.8)	0.29	(0.45)	1.6	(0.8)
Sb (µg/l)	1.2	(0.25)	4.2	(1.5)	0.82	(0.43)	0.3	(0.2)
Ni (µg/l)	4.0	(0.20)	23	(13)	3.0	(3.5)	2.2	(1.0)
Sn (µg/l)	0.04	(0.01)	3.6	(3.6)	0.02	(0.02)	0.02	(0.01)
Zn (µg/l)	84	(7.1)	101	(15)	21	(7.9)	85	(44)
Fe (µg/l)	37	(32)	1039	1080	24	(32)	36	(36)
Mn (µg/l)	46	(42)	55	(18)	45	(72)	350	(210)
Mg (mg/l)	48	(9.9)	33	(7.0)	na	na	na	na
K (mg/l)	5.8	(2.0)	63	(4.4)	na	na	na	na
Ca (mg/l)	542	(112)	34	(12)	na	na	na	na
Na (mg/l)	187	(49)	267	(25)	na	na	na	na

*Recirculation of tap water

3.3.5. Toxicity

In the studied industrial landfill leachate, the concentration of Cu was specified to investigate the fraction of the total Cu concentration that is in the form three cations. In the biofilter treatment plant, the total copper concentration was 259 µg/l in the influent and 64 µg/l in the effluent of the biofilter, but the concentrations of freely dissolved copper (Cu^{2+}) were only

16.3 µg/l and 8.7 µg/l respectively [162, 163]. This investigation shows that a small part of the Cu is in the freely dissolved form in the considered industrial leachate.

Moreover, the acute toxicity of untreated leachate from the industrial landfill on *Artemia salina* was determined (Paper V). The acute toxicity test showed that untreated industrial landfill leachate was not toxic to *Artemia salina.* This is most likely due to the comparatively low concentration of ammonia in the studied leachate, since previous studies [46] indicate that a high concentration of ammonia rather than metals or phenols in landfill leachates has been found to be responsible for the major part of the acute toxicity of *Arthemia salina*. Standard leaching tests and toxicological analyses of shredder residues generated in the metal shredding industry have also earlier shown low toxicity and weak mutagenity [164]. However, it is also of great importance to verify that the filter medium itself does not contribute to any acute toxicity. Previous studies of the same filter medium as used in this Thesis, have shown that the toxicity of a MSW leachate even decreased after passage through the filter medium [46].

3.3.6. Experiences from performing different scale studies

3.3.6.1. Batch equilibrium experiments

In studies reported in the literature, peat is usually sieved and only a certain particle fraction is then used for batch or column tests. For instance, in the study presented by Ringqvist *et al.* [42], peat particles smaller than 45 µm were removed. Since sieving of the filter media to be used in the full-scale filter was not planned, this step was also avoided in the laboratory batch equilibrium studies. Leachate was used in batch equilibrium tests, which is not so common. Leachate was spiked with high and low pollutant concentration (Papers I and II) for assessment of pollutant removal efficiency. An important phase of the batch equilibrium experiments was investigation of whether the removal efficiency of pollutants was the same in spiked deionised water as in spiked leachate solutions. In most batch equilibrium tests reported in the literature, different metals have been dissolved in distilled water, not in leachate or tap water. The latter approach is beneficial for simplification and better understanding of the complicated processes behind competition of selected metals. However, results obtained with distilled water as solvent do not reflect results that would be obtained in real system because of the complexity of leachate "matrix". As shown in Paper II, batch tests carried out with distilled water gave better removal efficiency than authentic leachate, leading

to an overestimation of filter efficiency if distilled water matrix is used. Obviously, contaminated waters contain pollutants that affect the removal of targeted compounds.

3.3.6.2. Column experiments

Column-scale investigations (Paper III) demonstrated that the material comprising the column wall was important. Leachate may corrode the column walls and cause leakage of pollutants from the column construction material. A three-week experiment, with a stainless steel column filled with industrial leachate, did not show any significant increase in the concentrations of Ni, Cu, or Fe in the effluent. However, the prolonged feeding of leachate into the similar stainless steel columns with filter material resulted in markedly increased concentrations of Ni, Cu and Fe in the effluent. Furthermore, a visual inspection of the stainless steel columns after six months showed signs of severe corrosion. In a similar way phthalates were leached from columns made of polyvinylchloride (PVC). Thus, for investigation of metals, PVC columns, or even better, polypropylene columns, should be used; for investigation of organic compounds, stainless steel tubes may be sufficient, and polypropylene may be an alternative.

In column experiments presented in Paper III, the effluent flow rate was measured during the whole experimental period. There was no noticeable tendency towards a reduction in flow rate of the effluent during a six month period and thus there was no indication of clogging. To reduce the risk of clogging, pre-treatment of the leachate such as sedimentation is in any case certainly recommended to reduce the concentration of particles in water distributed to the filter medium. Accordingly, investigation of the long-term performance of a filter-system is more properly performed in a pilot plant set-up, where pre-treatment steps can be included.

3.3.6.3. Pilot-scale experiments

There are achievements in effective on-site leachate treatment, but pilot-plant studies for leachate treatment based on natural systems are still rarely reported in the literature. Pilot-scale studies concerning leachate have been carried out *e.g.* by Luna *et al.* [79] and Heavey [86], but when concerning filter-based treatment systems such reports are very rare in Scandinavia. One recent example is the investigation described in Paper IV. In this pilot-plant study it was shown that in spite of large variations in concentrations of pollutants in the influent, stable removal of pollutants during the time-scale of the experiment was achieved.

Pre-treatment steps such as aeration and especially sedimentation, are considered to be essential to decrease the load of pollutants on the biofilter. For instance, the reduction of iron (47%) and suspended solid (72%) in the pre-treatment step reduce the risk for clogging of the biofilters and thus should guarantee a stable hydraulic performance and hence extend the lifetime of the filter. Investigation about the effect of aeration showed that consumption of oxygen was considerably higher in the industrial landfill leachate than in the leachate from MSW landfills. However, this aspect needs further investigation. The results of the on-site pilot study suggest that a biofilter can work for at least a three-year period without problems with clogging. However, to avoid problems due to temperate climate the filter bed should be insulated before winter to prevent freezing.

For an operator of a treatment plant, a key question always remains: how much time does it take to activate a filter-bed system? In order to study whether filter-bed treatment systems based on peat can work from the first day or whether they need a certain initial start-up period, a pilot-scale experiment was made (Paper VIII). Three filters with different types of peat were selected to treat leachate from two MSW landfills (one from the methanogenic phase and one from the acidic phase) in Estonia. The choice of peat was based on local availability. The results confirmed that all three types of peat, were suitable for the removal of N-tot and NH_4-N from leachate during the start-up period. The well-mineralised fen peat (*Phragmites* and *Carex*) with natural undisturbed structures demonstrated significantly better purification of COD, N-tot, and NH_4-N (concentration reduction in percentage) of old landfill leachate than milled and poorly mineralised *Sphagnum* peat. Fluffy well-mineralised *Sphagnum* peat was good for treatment of leachate from the acidic phase giving a high P-tot reduction. Even if a considerable amount of COD, N-tot, and NH_4-N in mg/(m^2×d) were removed in newly built peat filters, concentration values in the effluent were still above the Estonian limit values for wastewater discharge to the environment, except for P-tot. However, at least the results indicate that fen peat filters as well as semi-natural fen areas close to landfills could be used for secondary or tertiary treatment of landfill leachate, and the peat filters were operational from the first days.

It is likely that the purification mechanism in peat varies over time. In the beginning, the physico-chemical removal of pollutants dominates, since it takes time for development of a fixed bio-film around the peat particles, as was also shown in Paper VII. The physico-

chemical processes in a good peat filter are sufficient to achieve adequate trapping of most of the offending compounds already at the very beginning. But to efficiently remove biodegradable pollutants with high mobility such as phenols, and in order to realize transformation of NH_4-N to NO_3-N, time should be given for the establishment of microorganisms to obtain purification through biological processes.

3.3.7. Advantages and limitations of different scale studies

The development and evaluation of a full-scale filter-bed-based system proceeded through several steps, each of which had benefits and limits (Table 8).

Table 8. Advantages and limitations of experiments on different physical scale

Scale	Advantages	Limitations
Batch	• Simple design of the system; • Easy to perform; • Economic; • Quick; • Facilitates theoretical and more detailed studies (*e.g.* concerning mechanisms); • Replicates.	• Carried out indoors, which does not reflect real climatic conditions; • Experimental set-up is too simplified; • Different Liquid/Solid ratio than in reality; • Results highly dependent on experimental set-up (*e.g.* mixing rate).
Column	• Dynamic system; • Real Liquid/Solid ratios; • Controlled inflow conditions; • Controlled hydraulic loadings; • Careful extrapolating of results is possible;	• Design, set-up, and maintenance more difficult, time consuming and expensive; • Channelling and wall effects; • Often carried out indoors, which gives unrealistic environmental conditions; • Limited surface area of the column: diffusion of atmospheric oxygen is limited;
Pilot	• Both indoors and on-site under real climatic conditions; • Inflow concentrations vary in time, which reflects actual conditions; • Gives more reliable results in scale-up since magnification is less than in batch and column set-ups;	• Difficult to maintain if far away from the research laboratory or office), therefore subject to operational failures; • Difficult to run at low temperature, high rainfall *etc.;* • Higher capital cost; • More complicated design: • Construction more difficult: • More intensive sampling needed; • Time consuming.

Full	• On-site and real treatment system;
	• Actual climatic conditions;
	• Possible to study a complex system, where interaction occurs between different processes and procedures.
		• Difficult to maintain;
		• Difficult change the design if required for research;
		• Occurring processes are complex - "black box";
		• Affected by climatic conditions (low temperature, rainfall *etc.*), which add variables and complicates interpretation of results;
		• More difficult to control, since it is influenced by the situation outside the treatment plant:
		  o Hydraulics;
		  o Treatment efficiency;
		• More intensive sampling needed.

4. OPERATION AND MAINTENANCE OF FULL-SCALE FILTER-BED-BASED TREATMENT SYSTEMS

A full-scale on-site treatment plant was commenced to treat leachate from the industrial mono-landfill containing SR of ELV and white goods. The treatment plant consists of a 500 m^3 equalisation pond and vertical-flow biofilter system filled with a mixture of peat and carbon-containing ash. The biofilter system was constructed at the end of 2002 by Laqua Treatment AB (Sweden). In this chapter the function of the on-site full-scale treatment plant will be followed up, its hydraulic year-round performance studied and its suitability for weathering adverse climatic conditions will be evaluated.

4.1. Design and construction

The biofilter system consists of four parallel biofilters, each with an area of 30 m × 6 m (length × width) and with a depth of the filter medium of 1 m (Papers V, VI and VIII). The size of the filters in the system were in accordance with recommendation by McLellan and Rock [71]. They have suggested that the width of the peat filter should be 5 to 10 m and the length should be 10 to 30 m with multiple filters being constructed, if necessary, to stay within the loading guidelines. While a depth of 30 cm gives adequate removal, 100 cm is recommended both as a safety factor for flow surges and to increase filter longevity [71]. This selection of size gives a large degree of construction efficiency and makes filter replacement easier to perform. The existence of several parallel filter beds in the treatment system facilitates exchange of exhausted material, which can be done at different times without leaving landfill leachate untreated.

4.2. Seasonal operation

A simple water balance for the industrial landfill including precipitation, potential evapotranspiration and leachate generation at the landfill site has been presented bt Kängsepp and Dahlblom [165]. Results showed that there are quite large changes in temperatures and precipitation over a year in Halmstad. It was also seen that the leachate volume has increased during the period from 1995 to 2001 [165].

The biofilter system was constructed at the end of 2002 and leachate treatment started at the end of December 2002. In temperate climatic zones, water freezes and ice covers the filter

beds if they are not insulated. Although the filters still operate, their efficiency of removing some pollutants decreased, especially for particle-bound pollutants such as PCBs. Thus, the filters were covered by a layer of straw before arrival of the cold season. If possible, filter beds should be launched in the spring, rather than in the autumn. Careful preparation against the cold season is recommended. In this particular case, sprinkler irrigation was combined with drip irrigation *via* perforated pipes located underneath the insulating straw layer, which appeared to yield sufficient thermal insulation for the biofilters during cold winters. Sprinkler irrigation is more attractive in warm climates since it has an additional benefit, *e.g.* aerating the leachate encouraging evaporation. Further, aerobic degradation of organic compounds by micro-biological processes might occur, and such processes are generally enhanced by higher temperature. Drip irrigation systems are easy to insulate, and therefore are more common in temperate to cold climate. For continuous operation throughout the year both irrigation modes were installed and studied in the full-scale biofilter system. As far as the authors are aware, this is the first full-scale biofilter system functioning in cold to temperate climate, in which both distribution systems can be utilized in a single plant and in which the operator can manipulate the leachate distribution mode depending on climatic conditions. It has been found possible to run the plant continuously even with ambient air temperatures below zero for several months. The outcome of temperature measurements in the filter was that this was sufficient to avoid freezing of the filter surface. The lowest average temperature of 4.6ºC (SD 1) in the biofilters was registered during the winter (January 2006), and the highest temperature of 23ºC (SD 1) was registered in late summer (August 2004) (Papers VI and VII).

The distribution of water (Paper VI) in the cases of these two types of irrigation is, however, different. This fact, along with the shape of the filter beds and their hydraulic loading, must be considered. One challenge with irrigation of leachate on filters is to use the whole available surface of the filter for irrigation. The degree of uniformity obtainable with a set sprinkler system depends largely on the water-distribution pattern and the spacing of the sprinklers [166]. In an irrigation-uniformity study on leachate applied onto a full-scale filter (Paper VI), it was found that the corners of the filter received very little leachate, indicating that these parts of filter were not used efficiently. This should be taken into consideration when calculating the size and hydraulic loads for a new full-sale treatment plant. It was also found that a major portion of the leachate have effectively been applied to one longitudinal half of the biofilter, possibly due to wind directed from the north with an average speed of 0.2 m/s

during the study. The measurements show that the distribution of leachate upon a filter surface using sprinkler irrigation is strongly dependent on wind speed and direction, as also pointed out by Keller and Bliesner [166]. They pointed out that wind distorts the application pattern and that the higher the wind velocity, the greater this distortion [166].

4.3. Hydraulic loading

The biofilter system was dimensioned for a total capacity of 35,000 m^3 per year (with a daily hydraulic load of about 130 $l/(m^2 \times d)$. The results from the studies presented in Paper VI indicate a considerable time delay between precipitation events and increased leachate flow rates from the landfill. The daily loading rates on the biofilter, however, varied to a large extent and the actual daily load deviated considerably from the designed one. These loading rates cannot be automatically adjusted according to the weather data, since there was a considerable time delay between precipitation events and increases in leachate flow rates (Paper VI). In order to equalise the flow in temperate climate zones, where 60-70% of the annual leachate volume might be produced during a few months, a large retention pond should be constructed. The size of the existing pond (500 m^3) in the site, located before the entrance to the biofilter, appeared to be too small. During the three-year research period, the biofilters were two to seven times overloaded (up to 700 m^3/d) for about 10% of the total time (mainly in late autumn and winter). When the loading frequency is high on an intermittently loaded vertical-flow-bed, the time period when atmospheric air is drawn into the pore spaces of the substrate (during the drying period) will be much reduced. During the loading period, air is mostly forced out of the substrate, and thus, anaerobic-aerobic conditions will be prevailing. Diffusive oxygen transport to the substrate is enhanced during the drying periods, as the diffusion of oxygen is approximately 10,000 times faster in air than in water [167]. To ensure that the leachate flow-rate from the pond to the biofilter system is in accordance with the designed daily load and that it will not be affected by variations of leachate flow from the landfill, a relatively large equilibrium pond is needed. This question will be discussed below in section 4.4.

The research results in Paper VI also showed that during the investigated three-year period the biofilters were not loaded at all for about 40% of the time, mainly in the summer. Proper sizing of the pond will not only equalise uneven flow events, but will also guarantee continuous loading during the summer period, especially if leachate flow from the landfill is

very low. The dry period provides a rest period for the filters. During such a rest period, a decomposition of previously loaded organic compounds and suspended solids occurs and the hydraulic conductivity of the filter is most probably improved. Züst and Schönborn [119] achieved regeneration of hydraulic permeability due to degradation of the surplus of biomass in the filter, by exploiting the four-month-long interruption in loading. The retention pond can also serve to accommodate leachate during the regeneration period of the filters, when the filter material should be kept dry (to degrade an excess of biomass in it). During the study period reported in the Thesis, unplanned interruptions in the irrigation occurred both in the pilot plant (*e.g.* from November 2002 to August 2003) and the full-scale plant (from June to August during all three years). However, since measurements of infiltration capacity before and after these periods were not controlled, it is difficult to ascertain whether or not these interruptions contributed to the successful long-term performance of the biofilter systems. However, despite considerable variations in hydraulic and climatic conditions, the described natural treatment system (with too small equalisation pond and biofilters comprised of a mixture of peat and carbon-containing ash) has turned out to be sufficiently robust to give good removal efficiency of a wide variety of pollutants for a period of at least three years.

4.4. Scaling of the retention pond

An existing pond (500 m^3) that received and equalised leachate flows from three drains from different parts of the landfill before the biofilter system, turned out to be too small to provide a year-round equalised load of leachate to the biofilters. The required storage volume for such a retention pond was calculated based on actual daily hydraulic loads on the biofilters measured for a time period of three years (see Paper VI). During the considered three-year period, the maximum hydraulic storage volume needed was 23,000 m^3 (year 2005), with a total annual leachate volume of about 32,000 m^3. This storage volume is not astonishingly high, when one compares with the annual amount of leachate and volumes of existing ponds at other landfills in Sweden (Table 2). However, a limitation in the calculation approach used in the present study is the short period of data collection (three years). Longer time series of data, concerning daily generation of leachate, would provide for a more accurate estimation of the size of the pond required. Once again, the importance of having weather stations in each landfill, as proposed by the EU Landfill Directive, becomes obvious.

The biofilters tolerate periodic brief stress situations with overloading without deterioration in treatment efficiency. However, long-term overloading will most probably affect the balance between aerobic and anaerobic processes in the biofilter. Thus, nitrification might be limited and the biological degradation of particulate organic carbon might decrease, thereby increasing the risk of clogging of the biofilters.

4.5. Depletion and clogging of filter beds

Over time, chemical and physical changes take place in the filter medium. The filter material depletes if it reaches its adsorption capacity or becomes clogged. Measurements on the full-scale biofilter showed that the infiltration capacity of the biofilter was not reduced during the first two years (see Paper VI). Generally, infiltration capacity values for a mixture of peat and carbon-containing ash (between 0.4×10^{-4} and 7.0×10^{-4}) were similar to values for biofilters composed only of peat (between 1.1×10^{-5} and 2.0×10^{-3} m/s) obtained by Castonguay *et al.* [168].

In spring 2007 (after four years of operation), pools of leachate were formed on the surface of biofilter 2, which had received the largest volume of leachate. Although the pollutant removal efficiency still was not significantly changed (Paper V), this appeared to be an indication that clogging of the surface layer might have occurred. The reduced infiltration was probably due mainly to decreased permeation through the insulating straw layer, which was not removed after winter. Decay of the lower layers of the straw had formed a thick mat. Earthworms, which were introduced in 2004 to the biofilter to make the whole biofilter more porous, have probably not been present in sufficiently large numbers to entirely avoid reduced permeation through the biofilter surface. To avoid or slow down the clogging process an introduction of additional earthworms to the biofilter every year might be a solution. Removing the old straw layer did not solve the problem of clogging. Hence, clogging was not only dependent on compaction of the partly decomposed straw layer and retainment of solids at the surface of the filter. The mechanical skimming off and replacement of the surface layer, which is the usual remedy in case of slow sand filters [169], is hence not an option here.

Another possibility for clogging could be the decomposition of organic filter material with time, and further investigation of the grain size distribution of filter material was needed. Analysis of filter material samples taken at different depths of the filter media showed that the

grain size distributions at different levels in filter 2 were somewhat similar (Figure 7). Thus, an exchange of the whole filter 2 or a significant reduction of hydraulic daily load was the only alternatives in this case. After four and half years, the filters with peat and carbon-containing ash have been changed because of reduced hydraulic conductivity, although pollutant removal efficiency was still not affected (Paper V).

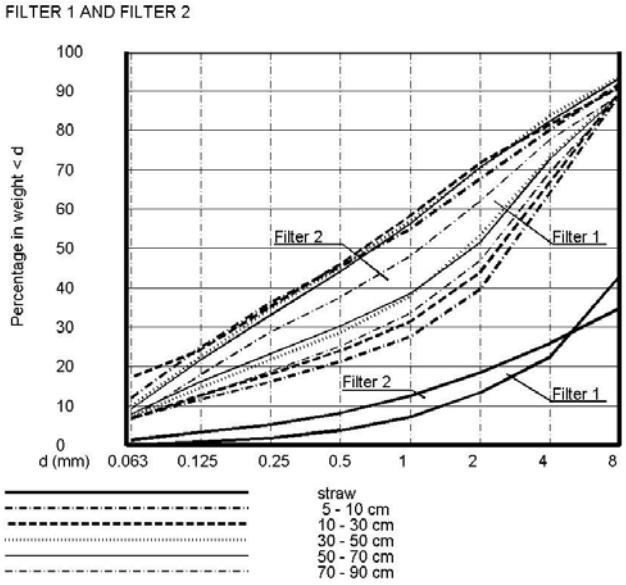

Figure 7. The grain size distribution of filter media from different depths of two biofilters (Filter 1 has been in operation the shortest time and had received less leachate than Filter 2, which has been in operation the longest time)

The grain size distribution in the biofilter medium comprised of peat and ash has changed during four years of usage in the biofilter system. Clogging might have been caused by degradation of the peat, in combination with accumulation of SS from the percolating leachate. As found in this study, about 3.0 tons of particles from the leachate had been trapped in the biofilters during three years of operation. This finding obviously motivates a need for a large sedimentation pond.

4.6. Handling of spent filter material

When employing this leachate treatment technology, several types of contaminants are accumulated in the filter media. This brings up questions about the handling of spent filter material. First of all, the content of hazardous compounds in the filters must be determined. Very little is known about the composition of filter materials, which have been used in full-

scale plants, and will be replaced. Therefore, characterisation of filter material after three years of usage for treatment of industrial landfill leachate was carried out. Heavy metals were determined to be adsorbed throughout the profile of the vertical-flow filter. It was not possible to observe any difference between accumulations of metal in various layers, *e.g.* between top and bottom layer (data not shown in this Thesis). Thus removal of a few cm of surface layer from the filter does not make a difference from the metal disposal point of view. Information about the composition of the spent mixture is presented in Table 9. In the study in Paper II, a comparison between composition of a new and three-year used filter material showed that the concentrations of Ca and Al had decreased while concentrations of Cu, Mn and Cd had increased in the used material during its three-year usage in the full-scale biofilter system. The metal concentrations in three-year-old filter media were similar to values usually found in peat in the Swedish environment, which was acquired from different parts of Sweden and represents about 13,000 ha studied area [85]. Concentrations of Cd and Hg in the filter material were however somewhat higher than in peat in the Swedish environment, but could still be considered to be low. For comparison, in addition, guideline values for contaminated soil [170] are given in Table 9. The boundary between "Slightly serious" and "Moderately serious" is used as the guideline value for sensitive land use. The nitrogen content was higher in the used mixture than in the unused one, probably due to loading of nitrogen-rich leachate. It is important to analyse the composition of a material batch intended for use in a new filter. This is especially true when residual waste materials are reused, since relatively large inhomogenities then can be expected. The results obtained in this study have shown that even if the chemical composition of peat and ash from different batches are slightly different, a good treatment efficiency of the leachate can be obtained (Paper III). For used filter material additional characterisation is needed to facilitate decision-making about further handling.

Possible handling methods include:

a. Disposal in landfills for municipal or hazardous waste
b. Containment by various materials, followed by disposal
c. Incineration
d. Regeneration

Table 9. Average concentration values for a used mixture of peat and carbon-containing ash and unused mixture (relative standard deviations, RSD, in %, are shown where n=2-4) (Paper V), concentrations of different components in peat from Swedish environment [85] and guidelines values for contaminated soil [170]. Units for elements are mg/kg

Parameter / Component	Unused mixture Average	RSD (%)	Used mixture (n=1)	Peat	Slightly serious	Moderately serious
$pH_{(KCl)}$	6.5*		7.3	na	na	na
Loss of ignition (%)	46*		63	na	na	na
C	330 000*		400 000	na	na	na
N	11 000*		16 000	na	na	na
P	na		na	na	na	na
S	4 100*		2 000	na	na	na
Br	17*		100	na	na	na
Cl	280*		1 300	na	na	na
Al	33 000	1	20 000	na	na	na
As	8.2*		4.6	4.3	<15	15-45
Ca	26 000*		14 000	na	na	na
Cd	0.34	24	0.84	0.23	<0.4	0.4-1.2
Co	21	25	8.6	34	<30	30-90
Cr	46	15	19	120	<120[a]	120-360[a]
Cu	47	15	300	228	<100	100-300
Fe	14 000	13	13 000	na	na	na
Hg	0.54	84	0.28	55	<1	1-3
K	4 300*		4 500	na	na	na
Mg	4 800*		5 900	na	na	na
Mn	210	9	700	na	na	na
Na	4 000*		5 500	na	na	na
Ni	46	14	41	102	<35	35-105
Pb	29	10	31	64	<80	80-240
Sb	0.04*		0.28	na	na	na
Sn	0.44*		5.1	na	na	na
Zn	60	6	130	227	<350	350-1050

*n=1

na – not analysed

[a] Applies only if Cr (VI) is absent

Placing of filter materials in secure landfills is the most common method of disposal. A disadvantage of placing used material into a landfill is that some metals might eventually be released. McLellan and Rock [71] found that desorption of metals was a substantial problem. Applying deionized water to the peat columns after they received leachate for 76 days showed that approximately 50% of the adsorbed metals leached back into solution [71]. Infiltration of water to the peat would need to be minimized in order to control the desorption of metals from exhausted peat intended for ultimate disposal [71]. Results presented in Paper II demonstrate that the metal concentrations in a used mixture of peat and carbon-containing ash are similar to values usually found in peat in the Swedish environment [85], except for Cd and Hg for which the values were somewhat higher. Results of leaching tests on the used mixture (Paper II) showed that the concentration values of different components in eluent are generally low (µg/kg level), compared to the proposed limit values included in waste acceptance criteria [171]. However, the DOC value is too high compared to waste acceptance criteria [171]. McLellan and Rock [71] suggest that the used peat could be encased in plastic and then landfilled. This would greatly reduce the amount of water reaching the peat, reducing and in the best case almost eliminating the impact of DOC. Anyhow, in any deposition option the material should be embedded in a cement matrix in order to avoid future leakage into leachate. Such strategies considerably reduce the mobility of contaminants. Direct deposition of spent filter material in municipal landfills is becoming an unaccepted option even if its content of hazardous material is not large, because water-rich and organic-rich wastes are to be diverted away from landfills, as proposed by the EU Landfill Directive.

Desorption of the metals from the filter matrix is an alternative solution. In this case, acidification of peat with 1 M HCl releases the adsorbed metals and the peat can be used again, according to Dissanayake and Weerasooriya [172]. This method entails an expense for hydrochloric acid. As reported by Viraraghavan and Rao [67], a cheaper method is by desorption of the peat in deionised water. Their study showed that 50 to 60% of the cadmium and chromium were desorbed from peat. However, in a large scale filter this approach most probably will also be too costly.

Several authors, including McLellan and Rock [71], have suggested that the peat from filters could be incinerated. One option is co-incineration with municipal or other waste. The disadvantage of this treatment option is that this kind of activity will lead to the increased

production of ash that has a high content of heavy metals. Moreover, there is a danger that the metals could be released to the atmosphere, and that the residual fly ash would require secure landfilling [71]. This is typically not a problem in municipal incinerators with appropriate flue gas cleaning systems.

As mentioned before, the concentrations of different metals in the three-year-used material are similar to those in natural peat, which directly or as pellets is already used for energy production *e.g.* in Sweden. As reported in Paper II, the effective heating value of the used filter mixture was 16.4 MJ/kg (in dry weight). This is similar to values for biofuels for incineration, which are usually between 17.2 – 21.5 MJ/kg, *e.g.* 21.5 for peat, 19.2 for bark chips and 17.4 for straw [173]. Given the present knowledge on solid waste handling, this alternative (incineration) seems to be the best one of the alternatives for disposal discussed above.

5. CONCLUSIONS

Results of studies presented in this Thesis show that

- On-site natural leachate treatment systems can also be used for treatment of leachate from an industrial landfill containing shredder residues (SR) of end-of-life vehicles (ELV) and white goods;
- Different type of compounds (*e.g.* ammonium-nitrogen, DOC, TOC, metals and polar and non-polar organic pollutants) in the industrial landfill leachate can be simultaneously reduced in a biofilter filled with a mixture of peat and carbon-containing ash;
- The performance of a filter-bed-based system is satisfactory even in a temperate climate zone, provided measures, such as filter bed insulation and possibility to use different irrigation systems during summer and winter, are taken to optimise performance year-round; However, to provide the designed annual irrigation rate on the biofilter, in region with large seasonal variations in precipitation, a relatively large pond is needed;
- Both laboratory tests and *in situ* experiments are indispensable for attaining understanding of and assessing the suitability and effectiveness of a methodology for the treatment of leachate from a specific landfill site. For best results, these studies should be carried out in coordination with each other;
- The strategy for development of a treatment system outlined in this Thesis, which proceeds through laboratory batch and column experiments, and ends up in field studies in a pilot plant at the landfill site, gives the necessary knowledge for construction of a well functioning full-scale leachate treatment plant.

6. FUTURE PERSPECTIVES

The study presented herein shows that the filter-bed system has undoubtedly become a valuable technology for treatment of leachate on-site. However, some further questions still remain and more research and experimental investigations are needed concerning the following questions:

- What causes very high oxygen consumption in the industrial landfill leachate and how do we achieve effective aeration of this type of leachate?
- Does dissolved organic carbon (DOC) in leachate from industrial landfill containing SR of ELV and white goods consist of refractory fulvic and humic-like compounds, similar to the situation with MSW landfill leachate?
- How should one take care of used filter material: in particular is the option of incineration really viable from an environmental point of view?

- What are the results of modern evaluations of the costs for different natural treatment systems for landfills in general, however, including study presented here?

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ERRATA LIST

In the Thesis:
On the page corresponding to „The thesis based on the following papers….", paper IV: An author Mårtensson, L. should be added in the end of the author list.

On page 27, in Table 2:

1) The header of 5th column „Pre-treatment in ponds (m^3)" should be changed to „Pre-treatment in ponds: size in m^3 (retention time);
2) The header of 6th column „Treatment" should be changed to „Treatment: size in ha or m^3 (retention time)";
3) Landfill in Umeå: Waste: „HW+IW+MSW" should be changed to „HW, IW, MSW";
4) Annual amount of leachate from landfill in Skedala is 90,000 m^3;
5) SBR – Sequencing Batch Reactors; $CW_{chips, peat, sand}$ – constructed wetland with wood chips, peat and sand filters.

On page 43, in Table 6:

1) In the column „Batch II (Paper II)", the removal efficiency (R%) of phenol should be changed to (-36).
2) In the column „Batch II (Paper II)", the concentrations of PCB52, PCB95, PCB101, PCB149 and PCB153 should be changed to 44, 29, 24, 24 and 26 ng/l, respectively.

On page 48, in Table 7:
Conductivity in eluent of unused mixture (batch) should be changed to 3 mS/cm (SD 0.3) and in used mixture (batch) should be changed to 2 mS/cm (SD 0.23).

On page 61, in Table 9:
The concentration of Hg in peat from Swedish environment [85] should be changed to 0.055 mg/kg.