Engineered Wetlands and Reactive Bed Filters
for Treatment of Landfill Leachate

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January 2004
PREFACE

This thesis entitled “Engineered Wetlands and Reactive Bed Filters for Treatment of Landfill Leachate” has been produced as a part of the requirements for the Licentiate degree at the Royal Institute of Technology, Stockholm.

My Master’s degree (engineering) is from the Warsaw Agricultural University (SGGW) where my interest for ecotechnology started. Thanks to scholarships from the Swedish Institute I was able to join the ecotechnological research programme that was established by Assoc. Prof. Gunno Renman in joint cooperation with my home University. During the first years I performed research together with Dr Lena Johansson Westholm until she graduated with a PhD in 1998. I also had the opportunity to work with my friends from SGGW, Dr Joanna Kwapisz and Dr Agnieszka Karczmarczyk. In 2001 I finally had the possibility to start my PhD studies.

This study was conducted in close cooperation with Telge Återvinning, the operator of the Tveta Landfill. In the very beginning of this research in which I assisted, I also had good cooperation with NCC AB. This latter contact has continued through Mr Magnus Alfredsson who organised the preparation of Polonite® for my experiments.

The thesis consists of five papers. I am the author of two of them and the other three have been written together with other people. Paper I is a literature review written by myself. The article presented as Paper II has Gunno Renman as first author. My contribution to that paper was collection of data and part of the analyses. The concept of the compact constructed wetland that is described there was developed by G. Renman. In Paper III, I am the main author and I have also collected much of the field data and performed all chemical analyses. Concerning Paper IV, I was responsible for all parts of the experiment, analyses and most of the writing. Finally, Paper V represents a “training” where all of it is my product. Unfortunately, this last paper is affected by many errors due to default proof delivery before printing.

A warm and sunny day in the compact constructed wetland. The hat is not only because of the sun, it is also to protect me from droppings produced by all sea-gulls flying over the landfill!!
ACKNOWLEDGEMENTS

This licentiate thesis was carried out at the Department of Land and Water Resources Engineering, Swedish Royal Institute of Technology (KTH). First and foremost, I would like to thank my advisor, Assoc. Prof. Gunno Renman for his continued support during my studies within his interesting and stimulating research group. Secondly, I would like to thank Prof. Dr hab. Zygmun Brogowski and Prof. Dr hab. Józef Mosiej from my home university, Warsaw Agricultural University (SGGW) for challenging me in my work. Assoc. Prof. Jon Petter Gustafsson, and Assoc. Prof. Lars Hylander are acknowledged for their valuable advice, discussions and comments on manuscripts. I am also especially grateful to Engineer Bertil Nilsson and Research Engineer Monica Löwén for discussions and technical support in the laboratory. I also wish to thank all my colleagues at Department of Land and Water Resources Engineering for the enjoying atmosphere. Thanks are due to Dr Mary McAfee for the English revision of most parts of this thesis.

This thesis was produced with limited economic resources. The kind financial support from Axel och Margaret Axson Johnson Stiftelse helped and is acknowledged. This work would not either have been possible without very good cooperation with Telge Återvinning AB.

Before I was accepted as a PhD student at KTH I travelled a lot between Poland and Sweden under the research exchange programmes headed by Prof. Renman and sponsored by the Swedish Institute.

The deepest and warmest gratitude goes to my family, which has always supported and encouraged me along my way. I especially want to send my deepest love to my Mother, who is no longer with us, for being so very important for me. I thank you for everything.

Stockholm, December 2003

Agnieszka Kietlińska
The main objectives of this study were to investigate (i) a novel wetland treatment technology and (ii) selected bed filter media for the removal of contaminants from landfill leachate. A review of the literature concerning experiences of the use of constructed wetlands (CW) for the removal of nitrogen from landfill leachate showed that at least three groups of treatment systems are in operation: sub-surface flow wetlands, hybrid systems (a combination of vertical and horizontal flow wetlands) and compact constructed wetland (CCW). Most of these types were generally effective in reducing nitrogen (N, e.g. NH$_4$-N, the dominant N species in leachate) down to effluent concentrations of about 10 mg L$^{-1}$. Unfortunately, very little evidence has been presented as regards the mechanisms responsible for the removal of N, although some data indicate denitrification. The treatment performance of a compact constructed wetland (CCW) applied at the Tveta Landfill, Södertälje, Sweden, was evaluated. Chemically purified leachate and untreated leachate were applied in periods of 7 days submergence and 7 days drainage to different sections of the CCW. The removal efficiency varied between 40 and 82%, and a mass removal rate of up to 5.1 g m$^{-2}$ d$^{-1}$ was achieved. The chemical pretreatment had a decisive role for the highest removal efficiencies obtained and it was unclear whether that treatment enhanced the efficiency because of lower toxicity and/or content of fewer competing cations. The possible combination of bed filter media and CCW as an ecotechnological treatment method for landfill leachate was investigated by bench-scale laboratory column experiments. Reactive filter media (sorbents) were selected from their known or suggested capacities for removal of heavy metals, nitrogen and phosphorus. Quartz sand or natural sand from an esker was used as a reference medium. Peat was used as an additional component in mixtures with the reactive media Polonite® (product from the bedrock opoka) and blast furnace slag (BFS). A small column study also involved zeolite. Phosphorus was efficiently removed by Polonite® and NH$_4$-N to some extent. Concerning metal removal, the best performance was again found for Polonite®, especially for Mn, Fe, Zn and Cu. The BFS showed good removal efficiency for Cu, Ni and Mo. The removal of different elements was suggested to be a combination of several factors, e.g. precipitation, ion exchange and adsorption. Prior to full-scale application of reactive filters at a landfill site, matrix selection, filter design and operational procedures must be developed.

**Keywords:** Blast furnace slag; Compact constructed wetland; Metals; Nitrogen; Polonite; Sorbents

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This thesis is based on a summary and the following five papers that are referred to throughout the text, by their Roman numerals:


Reprints are published with the kind permission of the journals concerned, and the papers are appended at the end of the thesis.
1. INTRODUCTION

This thesis deals with possible uses of constructed wetlands and reactive bed filters for the treatment of polluted water emanating from municipal landfills. The focus is on ecological engineering systems that are more environmentally friendly than many systems based on traditional techniques, as they usually require less or no input of chemicals and electric energy for the process (Mitsch and Jørgensen, 1989; Jenssen, 1996). They are also characterized as more cost-effective in the long run. These properties are beneficial for developing ecotechnological treatment processes as an interesting alternative for treatment of wastewater such as landfill leachate. However, in some cases advanced and expensive treatment systems have to be used together with ecotechnological systems to meet treatment goals.

As landfills become larger, the enormous quantities of wastes that they contain increase their potential to generate highly polluting leachates as they decompose anaerobically over many years. Among emissions generated by a landfill, the leachate is considered as the longest lasting (Kylefors et al., 2003). Leachate in an untreated form is unsuitable for direct discharge into lakes and rivers as the high metal and ammonia concentrations would have a severe impact on the ecology of the receiving water and a potential impact on human health. Persistent organic substances such as pesticides deposited in the landfill can form a long-term threat to the surrounding watercourses and later become incorporated into fish and other biota. Treatment of this highly polluting wastewater is becoming mandatory world-wide. Discharge to a municipal sewage treatment plant is often difficult and expensive since the landfill and the sewage plant are not located on the same site. However, such solutions for leachate treatment exist and efforts to produce a cleaner and sustainable sludge for recycling will be meaningless as long as leachate is mixed with municipal wastewater.

There are about 5,000 municipal landfills in Sweden, although most of these are now closed and landfilling is concentrated to about 300 active larger sites (Persson et al., 2000). These are government-approved and have different types of permits depending on e.g. volume and type of refuse handled and production of leachate. Sensitive receiving watercourses in the neighbourhood and risks for groundwater contamination may restrict the landfill activities or put high environmental demands on the operator, in accordance with current environmental legislation. However, from a European and global perspective the problems related to landfills are enormous. For example, Bulc et al. (1997) reported the existence of 60,000 illegal landfill sites and only 43 registered landfill facilities in Slovenia. In developing countries the situation is even worse, since solid waste management practices are uncontrolled and seem likely to become an expensive remediation problem in the future.

Constructed wetlands (CW) can reduce the quantity of pollutants from a variety of sources, such as primary and secondary wastewater, storm water, landfill leachate, industrial and agricultural wastewater and acid-mine drainage (Kadlec and Knight, 1996; Vymazal et al., 1998). The treatment of leachates by natural systems seems to be environmentally sustainable for
the removal of many constituents. Kadlec and Knight (1996) stress that both subsurface-flow and surface-flow wetlands are emerging ecotechnologies with the potential to treat landfill leachates. As the use of constructed wetlands to treat leachate is a relatively new ecotechnology, the data on their performance are being accumulated in different parts of the world. A state of the art review was recently published by Mulamottil et al. (1999). However, these authors concluded that more data are needed on the effectiveness of different wetland systems, such as the subsurface-flow and surface-flow wetlands, and some combinations of the system, such as peat infiltration and extended aeration. These authors were also convinced that design guidelines for treatment of landfill leachate in constructed wetlands must be developed. For better performance, it is essential that constructors of the systems understand the movement, breakdown and accumulation of pollutants in different parts of a wetland.

Characterisation of the leachate is essential since it can contain high concentrations of BOD/COD (biological and chemical oxygen demand), ammonia, metals, high or low pH and often priority pollutants of concern. Landfill leachate is generally anoxic and usually contains high concentrations of organic carbon, chloride, iron, calcium and manganese, all depending on what materials were originally placed in the landfill. Leachate quality can vary from relatively harmless to extremely hazardous waste. Landfill leachate does have some consistent characteristics but it is advised that data are collected for each site under study (Staubitz, 1989). The problem is to assess the different effects of single elements as well as their overall environmental effect. No general standard procedure has been available in Sweden until recently for the determination of toxicity and characterisation of leachates from landfills (Öman et al., 2000).

The selection of an appropriate onsite treatment technology is thus dependent on the leachate characteristics. However, both short-term and long-term fluctuations in the leachate quality and quantity must be accounted for in the CW design. In addition, the design must also consider that treatment requirements can change significantly as the landfill site matures. Historically, aerated ponds have been popular for the treatment and have proven to be successful in BOD/COD and ammonia removal (Maris and Harrison, 1984). Later, horizontal-flow reed bed systems became used to treat dilute leachates (Davies et al., 1993), polish pond treated leachate (Robinson et al., 1992) and eventually vertical-flow reed bed systems for treatment of high strength leachate were introduced (Reed et al., 1995). Recently, more complex systems have been proposed consisting of two or more treatment steps, so-called hybrid systems. Mæhlum (1998) investigated the effect of horizontal subsurface flow CWs combined with intermittent vertical flow filtration systems and alternatively extended aeration lagoons in the treatment of domestic wastewater and landfill leachate.

Many investigations have recently shown that the removal efficiency of particular contaminants can be enhanced if a filter medium of high sorption capacity is used in the CW (Mæhlum, 1998; Zhu, 1998). Another approach involving in situ CW upgrading with reactive filter media has been developed at the Swedish
Royal Institute of Technology (KTH), where separate filter wells were constructed as a step preceding the CW. In addition, other design principles have been developed for different types of wastewaters (Renman, 2003) although the results of long-term operation have not yet been evaluated. Besides the filter construction, the most important part is the medium or sorbent used. The sorbent is ‘reactive’ for one or several contaminants that have to be removed from the wastewater. The term sorbent refers not only to adsorption, but also to processes such as precipitation, ion exchange, complexation and mechanical filtration (McCay, 1996). Sorption depends heavily on conditions such as pH, concentration of pollutants, ligand concentration, competing ions and particle size. Sorbents may consist of natural materials that are available in large quantities and at a low cost, or of by-products from industrial or agricultural operations. Since they are non-expensive, these materials can be disposed of without expensive regeneration, although one must bear in mind that they can contain hazardous substances after use and have to be treated accordingly. Potential low-cost sorbents for heavy metals have been reviewed by Bailey et al. (1999). Examples of such sorbents are bark and other tannin-rich materials, chitosan and seafood processing wastes, zeolites, clays, fly ash and peat moss.

1.1 Aims of the study
The main objectives of this study were to investigate (i) a new wetland treatment technology and (ii) selected bed filter media for the removal of contaminants from landfill leachate. The following tasks were of particular interest in the study:

- A review of the literature concerning experiences of the use of constructed wetlands for treatment of landfill leachate with special regard to nitrogen removal (Paper I).
- Evaluation of the treatment performance of a compact constructed wetland (CCW) applied at the Tveta Landfill, Södertälje, Sweden, based on field investigations (Papers II, III).
- Investigation of selected reactive filter media with regard to their removal efficiencies of a wide spectrum of elements, with particular attention to heavy metals, nitrogen and phosphorus. Research based on column studies (Papers IV, V).
- Assessment of the possible combination of bed filter media and CCW as an ecotechnological treatment method for landfill leachate based on the results obtained in all investigations in the thesis (I-V).

2. CONSTRUCTED WETLANDS – TECHNOLOGY WITH SPECIAL REGARD TO THE CCW

Constructed wetlands are engineered systems that have been designed to utilize the natural processes involving aquatic plant species (i.e. macrophytes), soils and their associated microbial assemblages to assist in treating wastewater. The classification of CWs depends on the selection of macrophytes (See Fig. 1 in Paper I). The most developed CW is that using emergent plants such as cattail (Typha spp.) and common reed (Phragmites australis). This type of CW is often used for treatment of landfill leachate, although a few have been constructed as free-floating systems using duckweed (Lemna spp.). The latter
system has not produced convincing results in temperate climates (Vymazal et al., 1998).

By simulating the optimal treatment conditions found in natural wetlands, both FWS and HSF treatment wetlands (see Fig. 1 in Paper I for abbreviations) provide the flexibility of being applicable at almost any location. In most cases the wastewater is pre-treated by any method before it is discharged and flows horizontally or vertically by gravity through the bed substrate. Bed depth is about 0.6 m with hydraulic loading rates between 2 and 20 cm d\(^{-1}\) and a specific area for domestic wastewater treatment of between 5 and 10 m\(^2\)/p.e. (Kadlec and Knight, 1996). Constructed wetlands are suggested to remove suspended solids (SS), organic matter (BOD, COD, TOC), phosphorus (P), nitrogen (N), organic pollutants, metals and pathogens (Moshiri, 1993; Reed et al., 1995; Kadlec and Knight, 1996). The major nitrogen removal mechanisms in most constructed wetlands are ammonification and microbial nitrification/denitrification, as demonstrated by numerous studies, see reviews by Vymazal et al. (1998) and Vymazal (2001).

In this thesis most interest is focused on the vertical-flow system, since that type of CW has similarities with the CCW regarding design and operation. The concept of vertical-flow CW is based on the work by Seidel (1966). A system typically consists of two groups, or stages, of vertical-flow cells in series followed by one or more horizontal-flow polishing cells (Reed et al., 1995). Each stage of vertical-flow units consists of several individual wetland cells in parallel so that wastewater can be applied intermittently. The main advantage of this concept is the restoration of aerobic conditions during the periodic resting and drying phases. This is suggested to allow more efficient removal of BOD and ammonium nitrogen (NH\(_4\)-N) than can be obtained in the continuously saturated and generally anaerobic HSF wetland. For that reason, vertical-flow systems can be reduced in area but are designed for the same performance level as HSF wetlands.

The concept of the compact constructed wetland (CCW) was developed by Renman in 1997 (unpubl.) for a particular treatment system at the Tveta landfill, Södertälje, and later described more generally (Renman and Kietlińska, 2000; Paper II). The leachate treatment system at Tveta is shown in Figure 1. The CCW is characterized by its small area requirement and the fact that the design allows easy renewal of the bed substrate. Furthermore, this wetland is built on a unit comprising of two sections or cells with hydraulic communication via pipes. Each unit can be operated individually (see Fig. 2). Leachate is distributed into Section A by a perforated pipe for percolation through the sand/peat matrix, and further by two pipes to Section B, where no particular substrate is added. The continuous hydraulic loading stops when the water reaches a maximum depth in Sections A and B of 0.5 m and 1.25 m, respectively. The water is stored for about 6 days and subsequently drained. The ponds (A+B) then rest without leachate for about 6 days, while waiting for the next batch to be treated. Hence one treatment cycle consists of 12-14 days, including time for the filling-up and the drainage phase. The number of CCW units employed depends on the volume of wastewater that need to be treated.
The rationale for developing such a constructed wetland with alternating periods of flooding and drying is based on knowledge of how littoral zones and natural wetlands are able to remove nitrogen (Hillbricht-Ilkowska and Pieczyńska, 1993). Wijler and Delwiche (1954) first proposed that alternating periods of submergence and drying of soil might enhance N loss compared to continuously flooded conditions. They reasoned that alternating periods of aerobic and anaerobic soil conditions should facilitate the sequential coupling of nitrification and denitrification, with nitrate generated during the aerobic phase being denitrified in the anaerobic phase. However, in the particular CCW that is described here, macrophyte species are planted on the bed substrate as they are known to provide attachment sites for microbial growth, produce organic carbon for denitrification, transport oxygen to the system and perhaps improve permeability of the flow system.

Figure 2. A diagrammatic sketch of the CCW in cross-section (no scale). Water is distributed to the left section (A) where infiltration through the substrate occurs. The water slowly flows to the right section (B), via two connecting pipes. When the water depth reaches 0.5 m in the A section and 1.25 m in the B section, pumping stops. After about six days, the system is allowed to drain and dry for next six days until the next discharge period starts.

3. REACTIVE BED FILTERS – TECHNOLOGY

A reactive bed filter is designed according to the purpose of treatment, e.g. for storm water, domestic wastewater or leachate water (Renman, 2003). The construction may be a filter well or a CW where the most important component is the reactive medium or sorbent. Different types of artificial adsorbents or ion exchange materials are available as commercial products and most of them are utilized when very high treated water standards are required. However, these materials are not useful for treatment of landfill leachate because of their high cost. In the literature, numerous filter materials are described, those removing metals (Bailey et al., 1999), those removing organic compounds (e.g. O’Hannesin and Gillham, 1998), and those with removal capacities for phosphorus (P) (Johansson, 1998; Kløve and Mæhlum,
Recent studies have focused on filter materials among which emerge Blast Furnace Slags (BFS), Ex-clay filtralite, Polonite®, peat, and others (Mæhlum, 1998; Sakadevan and Bavor, 1998; Zhu, 1998; Bailey et al., 1999; Kløve and Mæhlum, 2000; Heavey, 2003; Brogowski and Renman, 2004; Renman et al., 2004).

The criteria for selection of a filter material are also related to the purpose of treatment, but usually include the following:
• Availability of material
• Cost
• Physical characteristics; pH, porosity, specific surface area
• Chemical composition
• Sorption capacity

This filter technology is a treatment system where wastewater is allowed to percolate, normally by gravity, through a reactive porous medium that removes the contaminant(s) from the water. If this type of treatment is successful, the benefit is that a significant mass of the contaminant is accumulated in a finite and accessible volume of material, which allows for future collection and disposal if necessary. The latter is not the case if a natural and large wetland is used for the treatment purpose.

4. MATERIALS AND METHODS

This chapter contains a description of materials and methods applied in field and laboratory experiments according to the aims of the study.

4.1 Study site (Papers I-V)

Studies of the compact constructed wetland were performed at the place for its first implementation, the Tveta landfill, Södertälje, Sweden. Leachate was collected from the leachate pond and transported to the laboratory at KTH for experiments. The composition of leachate shows considerably variation with degradation phases of

<table>
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<td>mg HCO₃ /l</td>
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<tr>
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<td>µg/l</td>
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organic waste in the landfill. Mean concentrations and ranges of selected parameters were compiled for the period 1994-2002 for the Tveta landfill (Table 1). The Tveta landfill site has been used since the early 1960s and is operated by Telge Återvinning AB, a municipally-owned company. The landfill depot received almost 200,000 tons of waste during 2002 of which only 10% were landfilled, according to a company plan to minimise deposition of waste and to put every effort into sorting waste for reuse (Tham et al., 2003).

Leachate is collected by means of a network of pipes and wells, and treated locally according to the system shown in Fig. 1. The leachate pond has a volume of 15,000 m$^3$ and is characterized by a retention time for leachate of about 90 days. Leachate is pumped to the indoor chemical treatment, intended for metal precipitation. Treated leachate is thereafter stored in Pond 1 before distribution, or directly distributed to the compact constructed wetland (CCW). The design and operation of the system is described by Renman and Kietlińska (2000; Paper II). The CCW was developed and patented as an alternative system of landfill leachate treatment due to lower installation and maintenance costs than other existing systems.

4.2 Monitoring and sampling programme (Papers II,III)

The treatment performance of the CCW was monitored during 1997-1998 and 2002-2003. The system only operates during the warmer season (May-October). Between the years 1999-2001 the chemical treatment plant, which is the step preceding the CCW, was reconstructed. The

![Figure 1. Aerial view of the treatment system at Tveta. In reality, the leachate pond is twice as large as shown in the picture.](image-url)
chemical treatment used during the opening phase in 1997 was container-based and was removed because of technical problems. The substrate in the A-sections was replaced in 2001 with a matrix of higher hydraulic conductivity.

Sampling in the CCW during the first trial in 1997-1998 included only triplicate samples of influent and effluent water. Redox potentials and the pH of pore-water in the bottom substrate of two CCW sections were measured. The programme was extended in 2002 and covered daily recordings of climatic data (precipitation, temperature, wind speed) by a local meteorological station at the landfill site. Duplicate leachate samples were taken from the inflow to the chemical treatment and after that step, before the inflow into the CCW. Effluent samples were taken in triplicate from each section (A+B) of the four ponds after six days retention in the CCW. In the A sections, three plastic tubes were installed vertically through the soil matrix for sampling of interstitial water.

4.3 Column experiments and reactive media (Papers IV,V)

For the short-term experiment, aimed at studies of phosphorus (PO₄-P) and NH₄-N removal efficiencies by different sorbents, laboratory columns consisting of acrylic plastic cylinders with an inside diameter of 34 mm and a length of 190 mm were used. Leachate was pumped continually under saturated conditions and samples were taken once a day from the outlet of the four columns during six days. For the long-term experiment, aimed at overall studies of elements and their removal by different substrates, five columns (K0 – K4) made of PVC, each having an overall height of 60 cm and an internal diameter of 9.8 cm, were used. All columns were filled with substrate to a height of 50 cm. The top of each substrate was covered with polyester filter to prevent media scouring and clogging during leachate addition. Peat with a moisture of 77.5% was mixed with the mineral substrates in a ratio of 1:4 by volume. The column K0 consisted of sand/peat, columns K1 and K3 were filled with Polonite®/peat and columns K2 and K4 with BFS/peat. Landfill leachate was transported from the Tvetta Landfill in Södertälje to the laboratory in eight separate batches during the experimental period. The leachate was stored at a temperature of 4 °C and brought in portions of 25 L to the column test, performed at room temperature. Leachate water was distributed by a peristaltic pump through Teflon tubes to each column with a flow rate of 7 mL min⁻¹, corresponding to a hydraulic loading rate of 1.34 m d⁻¹. The system was operated intermittently for 8 h per day, and each column received approximately 300 L of leachate during the whole experiment. The experiment was run under saturated conditions.

The media used in the short-term experiment were sand, opoka, calcinated opoka (Polonite®), zeolite (Hungarian clinoptilolite), and peat. In the long-term column experiment the following substrates were used (see above): sand, Polonite®, blast furnace slag (BFS), and peat. The latter was intended to prevent clogging because of chemical reactions between sulphur and calcium. The most novel sorbents of those mentioned are opoka and Polonite®. Polonite® is a product manufactured from the cretaceous rock opoka and intended for use in wastewater treatment (Brogowski and Renman, 2004). This material is known for its high sorption capacity of
soluble phosphorus and usefulness for recycling of nutrients in agriculture. Blast furnace slag is produced in large amounts by the steel industry and most of it is reused in a variety of applications such as for road construction, liming materials in agriculture and for wastewater treatment.

4.4 Chemical and physical analyses

In the field trial at the Tveta landfill, influent and effluent samples were collected in 100 mL acid-washed plastic bottles. In the laboratory the samples were filtered through a 0.45 µm micropore filter (Sartorius) and kept in a freezer at -18 °C prior to analysis. The analysis of three forms of nitrogen (N) (NH$_4$-N, NO$_3$-N, NO$_2$-N) was performed using Flow Injection Analysis (FIA, Aquatec-Tecator autoanalyser). Periodic measurements of dissolved O$_2$ (DO), pH, redox potential and electric conductivity were made in the field using the following instruments: Hanna HI 8424 microcomputer pH meter (pH, redox, water temperature), Hanna HI 8733 conductivity meter, Thermo Orion model 810 DO meter.

The samples from the short-term column experiment were filtered through a 0.45 µm micropore filter (Sartorius) and immediately analysed according to Swedish Standard Procedure (SS028126). The same procedure was carried out for the long-term column experiment, although samples intended for metal analyses were preserved with a few drops of concentrated HNO$_3$ and kept in a cold-storage room at 4 °C prior to analysis. Separate samples were transferred to bottles for pH and electric conductivity determination. The analyses were performed using ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometry) for elements, and Flow Injection Analysis (Fia, Aquatec-Tecator autoanalyser) for nitrogen compounds (NH$_4$-N, NO$_2$-N, NO$_3$-N).
4.5 Evaluation of treatment performance

The mass removal rate in g m\(^{-2}\) d\(^{-1}\) was calculated as follows (Kadlec and Knight, 1996):

\[
R = \sum (Q_{\text{in}}C_{\text{in}} - Q_{\text{out}}C_{\text{out}})/ A \quad (1)
\]

Where \(\sum (Q_{\text{in}}C_{\text{in}} - Q_{\text{out}}C_{\text{out}})\) is the retention of N in the CCW, \(A\) is the wetland area, \(Q_{\text{in}}\) and \(Q_{\text{out}}\) = inflow and outflow values (m\(^3\) d\(^{-1}\)), respectively, and \(C_{\text{in}}\) and \(C_{\text{out}}\) = concentration values (mg L\(^{-1}\)), respectively. As the removal rate is expressed on a daily basis the whole treatment cycle of 12-14 days was considered, i.e. the periods of both submergence and drying.

Removal efficiency \(E\) (%) of N in the CCW was estimated as:

\[
E = 100 \times (Q_{\text{in}}C_{\text{in}} - Q_{\text{out}}C_{\text{out}})/(Q_{\text{in}}C_{\text{in}}) \quad (2)
\]

The percentage removal efficiencies of heavy metals and nitrogen by the column substrates were calculated as the difference between concentrations in influent and effluent water. Mass removals (g kg\(^{-1}\) dry substrate) of each constituent entering and leaving the columns were estimated from water quality and flow data.

5. SUMMARY OF FIVE PAPERS

The thesis consists of five papers. A summary is presented below.

5.1 Paper I

The purpose of this article was to review the experience of constructed wetlands (CW) used for landfill leachate treatment with special regard to nitrogen removal. Published data are based on treatment efficiency of different wetland systems. These are often designed as hybrid systems and also with a metal removal function. The influence of different substrates, hydraulic load, input concentrations and plant species on the ammonium nitrogen removal in various types of constructed wetlands is outlined. The literature reports that classification of constructed wetlands depends on the selection of macrophytes. The most developed CW is that using emergent plants such as cattail (\textit{Typha spp.}) and common reed (\textit{Phragmites australis}).

These treatment systems can be constructed with many different designs. In general, emergent macrophyte-based systems can be categorized into four major groups according to the flow pattern:

1) Systems with free water surface (\textit{i.e.} surface flow)
2) Systems with horizontal sub-surface flow
3) Systems with vertical sub-surface flow
4) Hybrid systems (\textit{i.e.} combination of 1, 2 and 3)

On-site “high-tech” leachate treatment systems are often avoided due to large construction and operating costs. Two questions that are crucial for the success of constructed wetlands in future landfill leachate treatment are addressed: (i) does nitrification/denitrification occur and can it be managed, and (ii) can constructed wetlands be applied for the treatment of the high ammonium content that appears in landfill leachate and do they meet high effluent standards. Many processes are involved in the nitrogen removal such as ammonification (mineralization), nitrification, volatilization and plant uptake, but most researchers argue that denitrification is the most important process. Unfortunately, very little evidence for that is presented in the literature. In one article, an acetylene block technique was used to measure
denitrification in the laboratory to simulate field conditions in a constructed wetland used for landfill leachate treatment. Results from the literature review and current research indicate that removal of ammonium nitrogen by all types of CWs could achieve values between 9% to 99%, a variation depending on different flow patterns, substrate, hydraulic load, plant species, influent concentration and climate conditions.

Many authors used hybrid systems i.e. sub-surface flow wetland and/or combination with systems with free water surface in their research. Leachate water can be also pretreated in extended lagoons before passing sub-surface horizontal flow constructed wetland. Available organic carbon is also one of the limiting factors and there are discussions as to whether carbon additives other than those found in the wetland substrates should be used. Continuous leachate application is deleterious to the treatment process. Instead, a hydroperiodical system should be used, allowing a drying period when atmospheric oxygen can diffuse into the soil. In Sweden, a compact constructed wetland of new design was recently presented. Each pair of CCWs is filled up during six days and then drained, i.e. aerated. The ponds without leachate rest for next six days, waiting for a new portion to be treated. Nitrification and denitrification can occur in such aerobic and anaerobic conditions. During the first period of full-scale operation, ammonium nitrogen was removed to 82%. Most of the constructed wetland types are generally effective in reducing ammonium-nitrogen down to effluent concentrations of about 10 mg L\(^{-1}\). However effluent limits may be as low as 2 mg L\(^{-1}\) when organisms in the receiving waters are sensitive. Guidelines for leachate treatment wetlands should be developed since the interest from the industry in this ecotechnology is growing.

### 5.2 Paper II

This paper outlines design considerations for a compact constructed wetland, primarily treating landfill leachate, and the results of a first trial. The prototype treatment was designed in 1997 as an alternative and low-cost solution at the Tveta landfill in Södertälje, south-west of Stockholm. Annual production of leachate at Tvetaverket Landfill is about 60,000-100,000 m\(^3\), depending on precipitation. Today the leachate is pumped several kilometres to a conventional municipal wastewater treatment plant. The new system tested consists of a main storage pond, chemical treatment, compact constructed wetland (16 cells connected in eight pairs ponds)(CCW), a retention pond and a polishing stage. The area available for the CCW is 2 hectares.

Leachate water is collected in the storage pond via extended horizontal boreholes in the landfill. Landfill leachate is then pumped for chemical metal precipitation. After this stage, leachate water free from heavy metals is sent to the compact constructed wetland. The cattail *Typha latifolia* was chosen as the plant component in the CCW. The medium consists of peat, wood debris and sand as an organic carbon source. The main concept of the CCW is filtration through an organic substrate and alternation between oxic and anoxic conditions, promoting nitrification and denitrification processes for ammonium nitrogen removal. Leachate water enters the first cell of each pond filled with medium and planted with cattail via a distribution pipe. The landfill leachate then percolates through a 0.75-m layer medium and flows by two
connecting pipes to the next cell. Permeability investigations of the substrate showed a decrease from a value of 200 m d\(^{-1}\) to 60 m d\(^{-1}\). Due to the low soil hydraulic conductivity, it took 2 days to fill one pair of ponds. The pumping of leachate stops when the water reaches the same level in both cells. The water is then stored for 6 days followed by complete drainage. The ponds then rest without leachate for next 6 days, waiting for a new portion to be treated. Such conditions are suggested to generate aerobic and anaerobic states for nitrification and denitrification processes. Pumping and draining are operated automatically for the whole system of ponds. The system of eight pairs of ponds is designed for treatment of the annual production of leachate at the landfill.

This was the first full-scale experiment during the short time when the chemical treatment and CCW were operating together. Preliminary results indicated that chemical treatment was efficient in removal of several pollutants. The removal order was: Fe > Zn > Mn > Cd > Cu > Cl > Pb > Ni > Cr. Results indicated that removal of ammonium nitrogen by treatment of leachate in system of CCW exceeded 82%. The most surprising factor was the reduction of COD (60%) and chloride (37%) during CCW operation. Further investigations are needed to verify the processes governing nitrogen removal and to understand the COD and chloride reduction. In particular, it is very important to know whether it is possible to run this system during the cold season. Ammonium nitrogen removal in constructed wetlands is a temperature-dependent process. Leachate seeping out from the landfill has a temperature of approximately 25°C all year round. For that reason, insulation of CCW, or at least some of the cells, is proposed.

### 5.3 Paper III

On-site treatment of leachate was implemented at the Tveta Landfill, adjacent to the city of Södertälje, Sweden. The system consists of treatment steps for leachate collection in a pond, precipitation of metals with chemicals, use of constructed wetland and forest irrigation. This paper describes the constructed wetland and its effectiveness at removing ammonia in the system. Pulsed-discharge hydrology and wetland ecology formed the basis for the development of a compact constructed wetland (CCW), designed for small area requirements. Particular attention was paid to comparing the treatment effectiveness under conditions when non-purified leachate was directly discharged into two wetland sections with conditions when chemically purified leachate was discharged into two other wetland sections.

Chemically purified leachate and untreated leachate were applied to different separate sections of the CCW. A treatment cycle of about 14 days duration was used, involving a 7 day submerged phase with leachate and a 7 day period with dry conditions. The leachate entering the constructed wetlands had lower concentrations of N when it was first treated in the chemical treatment plant. A reduction of about 30% was observed after the leachate passed through the chemical treatment. The removal efficiency among the CCW sections varied between 40 and 75%. A mass removal rate of up to 5.1 g m\(^{-2}\) d\(^{-1}\) was achieved. The effect of hydroperiod upon N removal was not studied in detail in our research. However from other research it is known that the ammonium ion can be adsorbed onto organic and inorganic sediments by a cation exchange process. If the wetland substrate is exposed to oxygen,
perhaps by periodic draining, sorbed ammonium is oxidized to nitrate. The transformation of ammonium to nitrate was obvious in our experiment with untreated leachate, but was not observed in CCWs receiving pre-treated leachate. The chemical pre-treatment of leachate had a decisive role for a high N mass removal in the CCW, although it was unclear whether that treatment enhanced the efficiency because of lower toxicity and/or content of fewer competing cations. Mechanisms responsible for the NH$_4$-N removal in the CCW system have to be further investigated.

5.4 Paper IV

Leachate in an untreated form is unsuitable for direct discharge into surface watercourses as the high metal and ammonia concentrations would have a severe impact on the ecology of the receiving water. There is a strong argument for the introduction of filter systems using reactive media prepared from natural minerals or from by-products of steel production such as blast furnace slag. Such filters could be a possible solution for the removal of metals and could be used as a pre-treatment step before leachate handling for nitrogen removal in a constructed wetland. Filter materials saturated with heavy metals have to be replaced and stored in a safe way. An alternative method could be to leach out the metals with acids under controlled conditions and subsequently concentrate the solution for further work. A laboratory column study was conducted to evaluate permeable reactive filter media as a new method for removal of contaminants from landfill leachate. The screening with ICP-AES encompassed a number of 32 elements. Influent concentrations of arsenic (As), lead (Pb), cadmium (Cd), beryllium (Be), scandium (Sc) and lanthanum (La) were very low, i.e. below the detection limit of the instrument.

Filter media composed of sand/peat, blast furnace slag (BFS)/peat and Polonite®/peat were tested by loading bench-scale columns with leachate collected from a pond at Tvetaverket Landfill, Sweden. Sand and Polonite® represent natural materials, although the latter is manufactured from the bedrock opoka. BFS is a by-product from steelworks. The removal capacities of the media were assessed and the best performance was found for Polonite®, where Mn, Fe, Zn and Cu were removed to 99%, 93%, 86% and 67%, respectively. Hydroxide precipitation is suggested as the process for the high removal efficiency of metals by Polonite®, forming insoluble precipitates in the bed filter. The precipitation is primarily dependent upon two factors, namely the concentration of the metal and the pH of the water. Chemical treatment plants normally operate at a pH of approximately 9 when multiple metals are present. The superior removal capacity of the Polonite® filter is probably a combination of several factors, among which precipitation is the most important. This filter medium was also able to reduce nitrogen by 18%. The BFS showed good removal efficiency for Cu (66%), Ni (19%) and Mo (16%). Sand did not demonstrate a promising removal capacity for any of the elements studied with the exception of Cu (25%). Leaching of several elements occurred from the filter media. Most pronounced was the release of silica from Polonite® and BFS. Calcium was also leached from Polonite® but after 60 pore volumes of treated leachate, the release of Ca decreased while Si release rapidly increased. The changes in release of Si and Ca versus normalized flow for the
Polonite®/peat can be related to the weathering of the solid material and the presence of wollastonite. According to other studies, the latter material is efficient for heavy metal removal. The removal of different elements was suggested to be a combination of several factors, e.g. precipitation, ion exchange and adsorption. Prior to full-scale application of reactive filters at a landfill site, matrix selection, filter design and operational procedures must be developed.

5.5 Paper V

This paper describes a short-term laboratory experiment using landfill leachate. Particular attention is devoted to the use of filter media to test their efficiency in removing phosphorus (PO$_4$-P) and ammonium nitrogen (NH$_4$-N) from landfill leachate. Twenty litres of leachate were brought from the Tveta landfill near Stockholm. The average concentration of PO$_4$-P was 233 µg L$^{-1}$ and of NH$_4$-N was 139 mg L$^{-1}$.

The leachate water was pumped continually through Teflon tubing connected to a multi-channel peristaltic pump to four columns. The columns automatically received about 5 litres of leachate at the same rate. The filter substrates chosen for the experiment consisted of natural opoka, calcinated opoka (Op-Polonite®), zeolite (clinoptilolite), peat and sand. The first column (I) contained natural opoka with a particle size of 0-1 mm. The second one (II) was filled up with sand (particles < 2 mm). In the third column (III), the calcinated opoka (Op) and zeolite (Ze) were mixed in volume proportions 1:1. The grain size was 0-2 mm and 2mm, respectively. The peat and Op (IV) were mixed together in the same proportions as above. All columns were applied with landfill leachate under saturated conditions. Samples were taken once a day from the outlet of the columns during 6 days. The paper reports high PO$_4$-P removal efficiency for columns filled with Op/Ze and Peat/Op during the entire experimental period. The results showed that more than 90% of PO$_4$-P was removed by columns III and IV. Natural opoka and sand filter exceeded about 42% and 39% removal efficiency, respectively. The results showed that Op/Ze medium (column III) had the highest affinity to ammonium-nitrogen: over 30% of NH$_4$-N was removed, the main mechanism of removal probably being the ion exchange process. Column IV, filled with Peat/Op, achieved only 11% removal of ammonium nitrogen.

Furthermore, it was found that the mixture of calcinated opoka and zeolite (column III) had a high capacity for PO$_4$-P removal. In spite of the low phosphorus concentration in the landfill leachate, the removal efficiency reached over 90%. The results for removal of NH$_4$-N by filtration through the selected reactive media were not promising. However, the reactive filter technology can be a promising solution as a part of combined systems to meet high discharge standards.

6. DISCUSSION AND CONCLUDING REMARKS

This chapter contains a general and critical assessment of the results obtained in the attached papers and proposals for further investigations. The results themselves, with relevant figures and tables, are discussed in detail in the papers and are not repeated here.
6.1 Critical remarks on the work

The reliability of removal efficiencies obtained in the field-scale experiment as well as in the column experiments can be questioned. The CCW at the Tveta landfill has not been in operation for many years and the system may be biologically unstable. Mæhlum (1998) made these two statements in his thesis about the maturation processes in CWs: 1) CWs require at least two years of operation to mature the biological degradation processes; 2) Pond systems and intermittent filters require less time than CWs (i.e. horizontal sub-surface flow systems). Concerning the CCW, one must bear in mind that this system, in contrast to other CWs, involves frequent restoration of the bed substrate in order to control and keep the N removal efficiency at a high level. It is still unclear how often the bed substrate has to be exchanged. What is known is that the bed material can be easily replaced at a low cost because of the CCW design.

The accuracy of removal efficiency data remains uncertain due to several other factors. As regards the influence of climatic factors, I tried to make corrections for dilution caused by rainfall and for evapotranspiration during investigations of the CCW performance (Paper III) by a simple method of water gauge observations. These parameters could have been carefully recorded by other methods. Chloride concentration, for instance, is commonly used as an indicator of dilution (Heavey, 2003). The negative chloride ions appearing in the leachate should not attach to the cation exchange sites within the peat and hence any difference in concentration should be taken as rainfall dilution. The water balance could have been calculated from general data on potential evapotranspiration, although this method of estimation would be of low accuracy for such a small area as the CCW. Installation of flow meters (influent and effluent), continual measurement probes connected to data loggers for water level changes, and estimates of transpiration from the vegetation, would have been the best solution for interpreting the impact of climate on the results. Finally, another weakness in the study of the CCW was the manner of water sampling. Optimal conditions for obtaining representative samples would be to have an automatic sampler installed in the outlet well to which the CCW-treated leachate was drained.

The column experiments (Papers IV,V) were not run until breakthrough for many of the elements studied, i.e. until influent and effluent concentrations were equal ($C_e/C_i =100\%$). Hence the calculations of mass and percentage removal efficiencies by different media have to be treated with caution, as they merely provide an indication of the true capacities. The chemical and physical conditions in columns change constantly with time, which makes the evaluation of sorption kinetics and the finite removal capacity difficult. Columns work like ‘uncontrolled’ reactors, especially when landfill leachate with its varying quality is used for the experiment.

Replication of the experimental part of this study would be possible. The CCW design is open for other field trials and can be replicated on condition that a similar quality of landfill leachate and chemical pre-treatment are used. The kind of peat used in the bed substrate may have a role and must be considered. The filter media tested here are in commercial use, although both physical and chemical characteristics within the
same particle size fractions can limit any replicate study to some extent.

6.2 Nitrogen transformations in CWs
A successful constructed wetland for treatment of N must provide suitable conditions for both nitrification and denitrification to occur. The efficiency of N removal is also influenced by the following factors: the N composition of the landfill leachate, total N loading rate, hydraulic loading rate, redox potential, temperature and the macrophytic vegetation (Kadlec and Knight, 1996). In this thesis, the use of pulse-discharge hydrology i.e. hydro-periods is proposed for obtaining desirable nitrogen transformations. A limited amount of research has been undertaken on the effects of the hydroperiod for wastewater treatment in CWs. The effect of hydroperiod on sediment redox potentials has been studied by Busnardo et al. (1992). According to their results from an experimental mesocosms wetland, the sediment redox potentials were significantly higher after the sediments drained than just before drainage. A few measurements in the CCW at Tveta also indicated such changes in the redox potential (Paper III). A principal piece of research in this area is reported by Kruzic and Schroeder (1990) who studied ammonia nitrogen removal on overland flow treatment systems. They concluded that: 1) Cation exchange was probably responsible for N being removed one day and then being released in the form of nitrate on a subsequent day; 2) nitrification is a significant process; and; 3) denitrification is responsible for the removal of some of the nitrate nitrogen produced as a result of nitrification. These results were recently confirmed by Tyrrel et al. (2002) although they did not draw any conclusions on the influence of submergence and drying for N removal, despite it being a major factor in their experimental system.

The literature review (Paper I) revealed no clear answers to the questions that are crucial for the success of CWs in future landfill leachate treatment. Nitrification and denitrification do occur according to many authors, although strong evidence from scientifically produced measurements is lacking. Management of CW system parameters has been manipulated to achieve higher N removal efficiencies, mostly by means of pretreatment facilities with aeration. According to literature published to date, it seems that CWs can be used for the treatment of high ammonia content in landfill leachate and meet high effluent standards. However, one can observe that many different systems are in use and with varying performance. Constructed wetlands may be described as engineered if they involve design modifications, process additions, replaceable bed substrates and advanced system operations. The CCW described in this thesis represents the engineered wetland and the results of its operation shown here (Papers II, III) must be considered as promising.

The transformation of ammonium to nitrate was obvious in our experiment with untreated leachate in two of the CCW ponds. Further transformation was not observed, instead a release of nitrate was found. When comparing these ponds with those receiving chemically pretreated leachate, it appears that ion exchange of ammonium on peat cation exchange sites is not a long-term sink for ammonia removal (cf. Vymazal et al., 1998). Rather, sorption of ammonium is assumed to be rapidly reversible. The untreated leachate may be responsible for many inhibiting effects
for ion exchange and other N transformations. An investigation has to be carried out to clarify why the pretreated leachate showed better N removal performance in the CCW than the untreated (Paper III).

Constructed wetlands must be vegetated. The CCW at Tveta has sparse or no vegetation and it can be questioned whether this is a constructed wetland or an infiltration system with ponds. When constructing the system, interest was more on the bed substrate than on the vegetation (Paper II). For the continued operation of the system it is believed that some of the sections could stay devoid of vegetation, making possibilities for comparisons with the role of plants. It is known that plant uptake can be an important N removal pathway in wetlands if plant harvest is regular, as in the case of *Glyceria maxima* studied by Sundblad and Wittgren (1989). However, again the suggested toxicity and the high chloride content of the leachate can inhibit both the establishment, nutrient uptake and growth of the plant species.

### 6.3 Reactive filter media for leachate treatment

Different types of filter media or sorbents have proved to be efficient for the removal of phosphorus and nitrogen in constructed wetlands or in specially designed traps as described earlier in this thesis (see Reactive bed filters – technology). It was shown in Papers IV and V that the sorbents used were capable of removing several contaminants from the landfill leachate. The role of Polonite® as a medium for multi-element removal was pronounced, although sorption of organic compounds was not studied. However, the property of sorbing many elements can be considered to be both positive and negative. The positive value is that only one layer of filter medium is needed for a proposed filter plant, instead of several layers or filter steps where different pollutants can be removed. The negative value is that elements which are considered a resource, *i.e.* nitrogen, phosphorus and potassium, should be recycled in agriculture.

When replacing the reactive material from a sorption facility, one has to handle a material in which contaminants have accumulated (Renman, 2003). If the material can be regenerated, an even more concentrated product can be obtained. However, it is not believed that this product will be such an interesting substance for re-use. Instead, it will be considered as waste and must be sent to a specialised plant for destruction. Landfill leachate is a mixture of contaminants but also a resource. As a liquid containing contaminants, it is a problem and has to be treated. However, the content of high strength ammonia nitrogen in leachate could be used for production of multi-nutrient fertilizer. Recovery is possible by precipitation of magnesium ammonium phosphate (struvite) with chemicals (Li *et al*., 1999; Li and Zhao, 2003), or by using the reactive medium Polonite® (G. Renman, pers. com.).
6.4 Leachate treatment system using reactive bed filters and compact constructed wetlands

This thesis raises the question of whether reactive bed filters and the CCW can be used together as a treatment system for landfill leachate. The answer is that a pilot-scale treatment plant has to be built and tested and a schematic layout of such system is shown in Figure 4. This thesis showed a preliminary function and removal performance of a full-scale CCW as individual units, while the reactive bed filter remains untested. Further research on processes governing contaminant removal by selected sorbents must be carried out the laboratory, as well as at pilot- and full-scale treatment plants.

7. FURTHER INVESTIGATIONS

On-site treatment of landfill leachate based on ecotechnological principles has to be further developed in order to meet the needs arising at many landfills worldwide. Research is needed to further understand and improve the compact constructed wetland system and reactive bed filters and should include in particular:

1. Enhanced data collection as a basis for mathematical modelling, in order to predict the removal efficiencies and lifetime of CCW and reactive filter media;

2. Measurement of nitrification/denitrification processes in the CCW in relation to external added and internal wetland carbon sources, and to length of the hydroperiod;

3. Evaluation of the disposal possibilities for saturated filter media and for bed substrate used in the CCW;

4. Improvement of nitrogen removal by filling the B-sections of the CCW with substrate (organic carbon source) to the same depth as the A-sections and distributing leachate by irrigation to both of them;

5. Investigation of the possibility of nutrient recovery from landfill leachate by struvite formation;
6. Investigation of reactive filter units, specially adapted for heavy metal removal preceding the CCW;

7. Improvement of the metal sorption capacity of the best mineral reactive filter media in column experiments by manipulating factors such as particle size, cycles of drainage and submergence and organic additives.

8. REFERENCES


