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Title and subtitle

Characterization and treatment of municipal landfill leachates

Abstract The efficiency of different leachate treatment methods for the removal of refractory organic compounds and ammonium-nitrogen was investigated. The methods evaluated were nitrification, denitrification, adsorption onto activated carbon, precipitation by ferric chloride or aluminum sulphate and oxidation by ozone or Fenton´s reagent. Furthermore, analyses were performed on leachates from municipal landfills of different kinds (a biocell deposit, a conventional mixed landfill containing household and industrial waste, and an ash deposit) in order to study the leachate composition in regard to various hydrophobic organic compounds as a function of the type of waste deposited.

The results suggested that, in order to achieve a satisfactory removal of both ammonium-nitrogen and organic substances, the treatment of methanogenic leachates should be performed through a process combining biological and physical or chemical stages. When the biological treatment was not combined with a physical or a chemical process a COD removal of only 20-30% was achieved, wheras the toxicity of the leachate was significantl reduced. In contrast, a combination of nitrification and either adsorption onto activated carbon or oxidation using Fenton´s reagent resulted in a COD removal of about 80%, although certain specific organic compounds, such as phthalates, were unaffected by the treatment. A combination of nitrification, precipitation by ferric chloride and adsorption onto activated carbon removed 96% of the TOC.

The analyses of leachates from municipal landfills of different types showed the leachate from the ash deposit to contain more C4-substituted phenols than the other leachates and to likewise contain alkanes,which the others did not.

Key words leachate treatment, municipal landfill, organic substances, characterization, biological treatment, physical/chemical treatment

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LIST OF ORIGINAL PAPERS

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I. Welander U. (1997) Identification by Gas Chromatography of Organic Constituents in Leachates from Sanitary Landfills. *Resource and Environmental Biotechnology* 1:283-296.

II. Welander U., Henrysson T. and Welander T. (1997) Nitrification of Landfill Leachate using Suspended-Carrier Biofilm Technology. *Water Research* 31: 2351-2355.

III. Welander U., Henrysson T. and Welander T. (1998) Biological Nitrogen Removal from Municipal Landfill Leachate in a Pilot-scale, Suspended Carrier Biofilm process (in press *Water Research*).

IV. Welander U. and Henrysson T. (1998) Degradation of Organic Compounds in a Municipal Landfill Leachate Treated in a Suspended Carrier Biofilm Process (in press *Water Environment Research*).

V. Welander U. and Henrysson T. (1998) Physical and Chemical Treatment of Nitrified Leachate from a Municipal Landfill (in press *Environmental Technology*).

NOTATIONS

AOX	adsorbable organic halogen
ATP	adenosine triphosphate
BOD	biochemical oxygen demand
Cd	cadmium
Cl^{-}	chloride
COD	chemical oxygen demand
Cr	chromium
Cu	copper
DO	dissolved oxygen
FA	fulvic acid
Fe	iron
GC/MS	gas chromatography/masspectrometry
GPC	gel permeation chromatography
HA	humic acid
Hg	mercury
HRT	hydraulic retention time
HPLC	high-performance liquid chromatography
IR	infrared spectroscopy
MBR	membrane bioreactor
Mn	manganese
MSW	municipal solid waste
$NH4^{+}$	ammonium ion
$NH4^{+}$-N	ammonium nitrogen
Ni	nickel
$NO3^{-}$	nitrate ion
PAH	polycyclic aromatic hydrocarbons
Pb	lead
Pt	platinum
SBR	sequenced batch reactor
TOC	total organic carbon
Tot-N	total nitrogen
Tot-P	total phosphorus
TOX	total organic halogen
UASB	upflow anaerobic sludge blanket
UV	ultraviolet light
VFA	volatile fatty acids
Zn	zinc

INTRODUCTION

The introduction of special sites for waste dumping in Sweden, or landfills, began during the latter part of the 19th century. Industrialization was one of the reasons for their being established (Lagerkvist 1995). Another reason was the growing knowledge of the importance of hygiene and the realization that waste should be deposited in remote areas. Previously, waste had been dumped as conveniently as possible in holes and cavities where it accumulated in and around cities. It is interesting to note that the area encompassed by "Gamla Stan" in Stockholm was tripled between the 13th century and the 18th century through an ongoing "land reclamation" which made use of waste (Hansson 1970).

As trading in merchandise increased, waste became increasingly complex, due to packages consisting of different combinations of materials being produced. The complexity of waste increased even more when the populations of cities increased and when, during the thirties, garbage chutes were introduced (Lagerkvist 1995). Around the turn of the century, landfills increased in both number and size since the proportion of the waste that could not be composted increased.

The increasing number and size of landfills led to increasing interest in environmental aspects. A consequence of the growing environmental concern has been that during the late eighties and early nineties recycling systems for different kinds of materials have been developed, above all for paper and glass. The deposition of waste in landfills, however, is part of the waste handling system that will become unavoidable in the foreseeable future, since certain materials are difficult to recycle. Furthermore, there are a great number of closed landfills in which everything from household waste to industrial waste of various kinds have been deposited. In 1994, there were some 300 active municipal landfills in Sweden alone, the number of active industrial landfills being approximately the same. In addition, there were around 4000 closed deposits (Swedish EPA 1994). The major environmental problem associated with landfills is that of the emission of methane, carbon dioxide and highly polluted leachates. The emission of methane and carbon dioxide is an environmental problem since these gases are greenhouse gases, whereas the emission of leachates is a problem due to their high content of ammonium ions and organic compounds. The ammonium ion is an acutely toxic compound. The reported 96 h LC_{50} values of NH_4^+-N for different varieties of fish lie between 0.32 and 3.10 mg/l (the lethal dose for 50% of fish at an exposure of 96 hours) (Ruffier *et al.* 1981). These values can be compared with the content of NH_4^+-N in leachates, which can be many hundreds of mg/l (based on leachate samples from 26 different Swedish landfills that were analyzed, Meijer 1980). The organic compounds can also be acutely toxic, although because of their low concentrations in many leachates this is usually not a severe problem. A more serious problem in the organic content of leachates is the risk of long-term effects due to the accumulation of various of these substances in organisms.

Today, leachates from many of the larger landfills are co-treated with municipal wastewater in municipal wastewater treatment plants. This solution may possibly be forbidden by authorities in the near future, however, due to a growing concern over refractory organic compounds (organic substances that are difficult for microorganisms to degrade) that the leachate contains disturbing the biological processes at municipal wastewater treatment plants (Linde 1995). Another reason why co-treatment of municipal wastewater and leachate may sooner or later be forbidden is that municipal wastewater treatment plants can have difficulties in meeting their requirements for nitrogen removal if leachate is co-treated with municipal sewage (Swedish EPA 1982). Problems may also arise in the end use of the sludge produced if it contains high concentrations of heavy metals (Lagerkvist 1986). In future, therefore, many leachates will probably be treated on-site in separate treatment plants using approaches optimized for their treatment, plants which are more than simply copies of municipal wastewater treatment plants.

The aim of the work presented in the present thesis was to study the characteristics and treatment of landfill leachates. Leachates from municipal deposits of different kinds, a biocell deposit, an ash deposit and a conventional landfill, were compared with regard to the content of specific hydrophobic organic compounds. In addition, different treatment methods for the removal of refractory organic compounds and nitrogen from leachates were evaluated. Special emphasis was placed on refractory organic substances since there is a risk of their being neglected in the optimization of treatment processes due to their not constituting a major environmental problem in the **short-term** perspective.

LANDFILLS

Swedish landfills built in recent years normally include a drainage layer, as well as a clay layer or engineered liner, and they generally are covered with some kind of covering layer in order to minimize the production of leachate, Figure 1 (Swedish EPA 1994). Conventional landfills contain everything from household waste to different kinds of industrial waste, since it is only recently that separation of various types of waste has started. Thus, there is a wide variety of organic compounds that can leach from landfills, the leachate composition depending, for example, on the industries that are dominant in the area. The complex composition of leachates, in combination with a flow of leachates that varies markedly over the season, makes them difficult to treat. A potential problem in the construction of the landfills described above is that the liner at the bottom of the deposit may fail to retain all the components of the leachate (e.g. various organic substances). This can result in groundwater pollution. Several cases of groundwater pollution due to leachates have been reported (Meijer 1980, Albaiges *et al.* 1986, Seman 1986, Kjeldsen 1993).

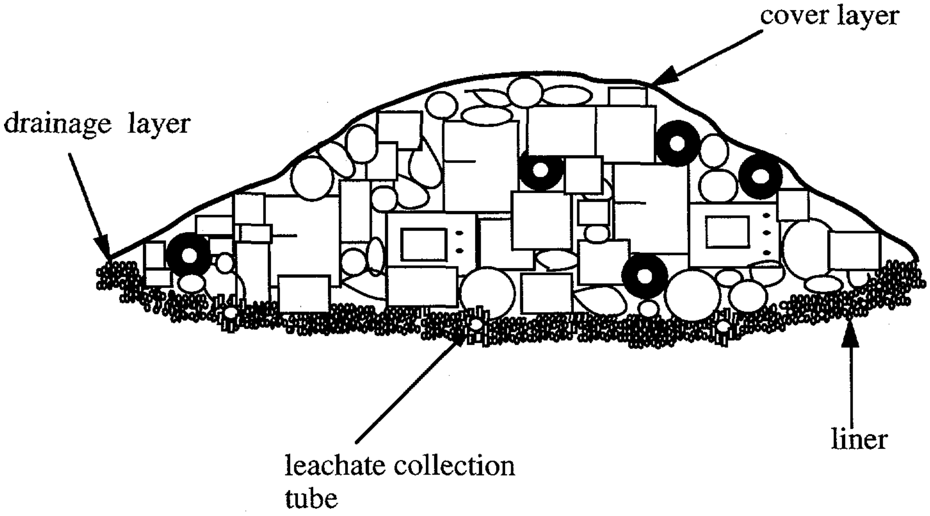

Figure 1 A schematic view of how a landfill is constructed.

Processes in a landfill

The degradation process in a landfill passes through different phases. The first phase, which is normally short, is characterized by the aerobic degradation of organic matter. During this phase the temperature increases and only small amounts of leachate are produced (Lagerkvist 1995). When the degradation processes become more intensive, the oxygen is soon depleted, the degradation continuing anaerobically. The anaerobic degradation proceeds then through two phases, an acidogenic phase and a methanogenic phase, Figure 2. The acidogenic phase consists of two steps. The first step involves the extracellular hydrolysis of macromolecules too large to enter the microbial cells, e.g. of polysaccharides and proteins. The smaller compounds produced in this step, such as simple sugars and amino acids, are then fermented during acidogenesis to such products as acetate, propionate, buturate, ethanol, molecular hydrogen and carbon dioxide, Figure 2. The same group of bacteria, the acidogens, also produce hydrolytic extracellular enzymes (Welander 1989). The concentration of volatile fatty acids (VFA) in the leachates can become very high (thousands of mg/l) during this phase, leading to low pH. This low pH, in turn, involves a risk in regard to the mobilization of metals since the solubility of most metals increases with decreasing pH. In addition, the VFA are able to form complexes with many metal ions, increasing still further the mobility of the metals (Kylefors 1997). During acetogenesis, intermediary products of acidogenesis other than acetate and hydrogen gas are then converted to acetate and hydrogen gas by obligate, proton-reducing, acetogenic bacteria (Mah 1983). Later, during the methanogenic phase, there is a change in the microflora of the landfill to methanogens. These transform acetate, hydrogen gas and carbon dioxide, mainly to methane and carbon dioxide. The pH of the leachate increases while the redox potential decreases during the methanogenic phase. The organic matter of the leachate is dominated by refractory compounds, such as humic-like compounds, the concentration of metal ions during this phase commonly being low due to the solubility of many of these ions decreasing with increasing pH values. Furthermore, many metal ions are precipitated by the sulphide which the sulphate-reducing bacteria produce. Although metal sulphides are difficult to dissolve under anaerobic conditions, there is the risk of their dissolving when the majority of the biodegradable waste is degraded and oxygen can diffuse into the landfill. Some metal ions (e.g. lead) form complexes with humic substances, which may thus increase in concentration in the leachate (Öman 1991). In addition, it has been shown that humic substances are able to transport certain hydrophobic organic compounds such as phthalates (Gonzalez-Vila and Martin 1985). The differences between different phases in the composition of leachates from landfills can be illustrated by the study of Millot *et al.* 1987 concerning two different leachates, one from a relatively old landfill still in operation and one from a newer but closed landfill. The results of gel-permeation chromatography (GPC) showed 65% of the TOC in the leachate of the older landfill to consist of compounds of high-molecular-weight which adsorbed UV light at 260 nm and 220 nm, whereas 90-95% of the TOC of the leachate from the newer landfill consisted of volatile fatty acids. These results are in agreement with those of a study by Harmsen 1983, which showed that in the acidogenic phase over 95% of the TOC in a leachate from a landfill consisted of free volatile acids whereas in the methanogenic phase 32% of the TOC content of a leachate from a landfill consisted of compounds with a molecular weight of over 1000. The microbiological phase of a specific landfill depends on such parameters as the age of the landfill site, the moisture content in the landfill and the temperature (Ross 1990). The time needed for a landfill to stabilize, however, has been found to be reduced if the surface of it is covered (Pohland *et al.* 1979). These authors compared two different cells, one of them open, allowing precipitation to percolate through the waste, and the other a covered one to which tap water was added in amounts corresponding to the precipitation. The leachate from both cells was recycled. The results showed the concentration of fatty acids in the leachate from the covered cell to decrease more rapidly than that in the leachate from the open cell. In addition, the pH value increased more rapidly in the leachate of the covered cell than of the open cell, which was obviously a consequence of the decrease in fatty acid concentration. The difference between the two cells was explained by oxygen being more efficiently excluded from the covered than from the open cell.

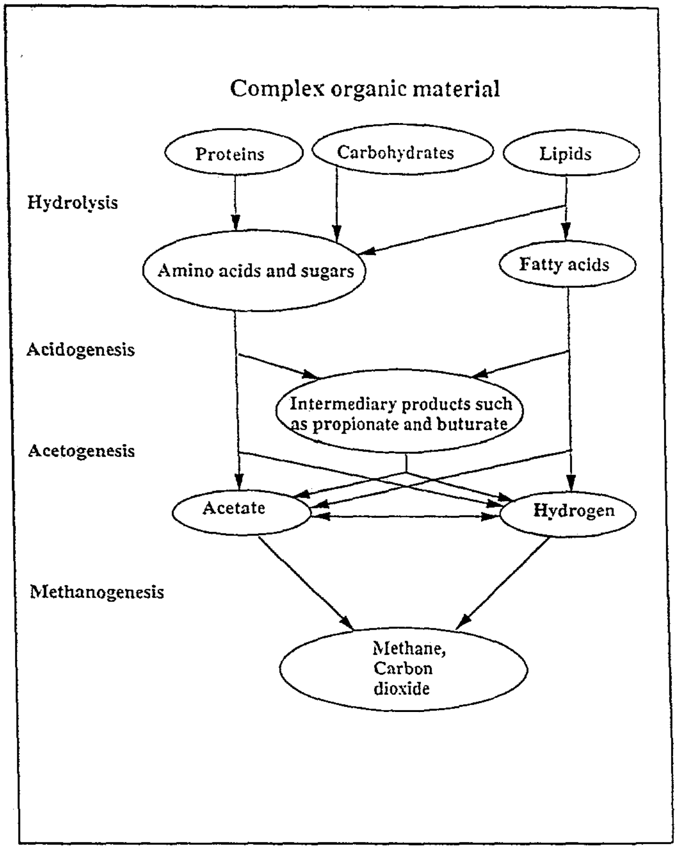

Figure 2 The anaerobic degradation sequence of complex organic material (modified from Guijer and Zehnder 1983).

Abiotic processes that influence the chemical composition of leachates include the adsorption of different compounds to the waste, the volatilization of certain substances and the water solubility of various compounds. A study performed on 12 model substances, chosen to represent substances of widely differing properties, showed that retardation in the waste was of importance for hydrophobic compounds whereas the less hydrophobic compounds were transformed or leached (Reinhart and Pohland 1991). Volatilization was of only minor importance for most of the compounds, with the exception of trichloroethene.

The generation of leachate

The generation of leachate depends on many different parameters such as the kind of waste deposited in the landfill, the biological processes there (water can be either produced or consumed, depending on the processes involved), the precipitation and the infiltration of surface- or groundwater. The amount of leachate produced is also dependent on evapotranspiration. A simplified water balance is described in equation 1, Figure 3. The equation is only valid if the landfill is built above groundwater level and if interceptive ditches are used to minimize the infiltration of surface- and groundwater (Swedish EPA 1994).

$L= P-E-R-\partial M$ (1)

L=Leachate production
P=Precipitation
E=Evapotranspiration
R=Surface runoff
∂M=The change in water content in the landfill

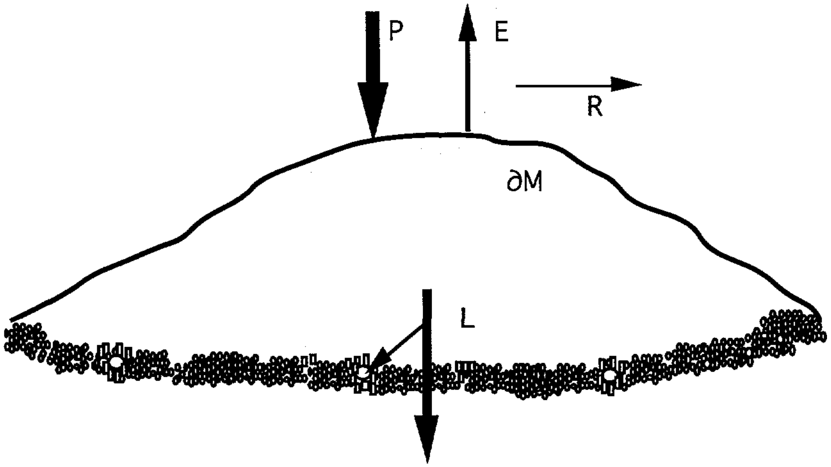

Figure 3 The water balance of a landfill.

Production of humic substances

As already mentioned, leachates coming from methanogenic landfills often contain a high concentration of humic-like substances, Figure 4. However, since the content of humic substances in the waste deposited is normally low, humic substances are usually produced in the deposit.

There are four major theories to explain the production of humic substances, Figure 5 (Stevenson 1982):

1. Reducing sugars and amino acids formed during microbial metabolism undergo non-enzymatic polymerization to form brown nitrogenous polymers, Figure 5.

2. Polyphenols are synthesized from **non-lignin** carbon sources (e.g. cellulose). The polyphenols are then oxidized to quinones and converted to humic substances, as in theory 3, Figure 5.

3. Phenolic aldehydes and acids released from lignin during microbiological attack undergo enzymatic conversion to quinones. These polymerize then to humic-like macromolecules in both the presence and absence of amino compounds, Figure 5.

4. Humic substances are derived from the incomplete utilization of lignin. The incomplete degradation includes loss of methoxy (-OCH_3) groups and oxidation of aliphatic side chains to form -COOH groups. Humic acids are produced as the initial product, whereas fulvic acids are produced as a result of further oxidation and fragmentation, Figure 5.

All four theories for the synthesis of humic substances are considered, although theories 2 and 3 are favored by most modern scientists, (Stevenson 1982) theory 4 being the classical theory (Waksman 1932). The production of humic substances in a specific soil can follow more than one of the mechanisms described above, although the relative importance of the various pathways differs from one soil to another, depending on various factors, such as the water content of the soil (Stevenson 1982).

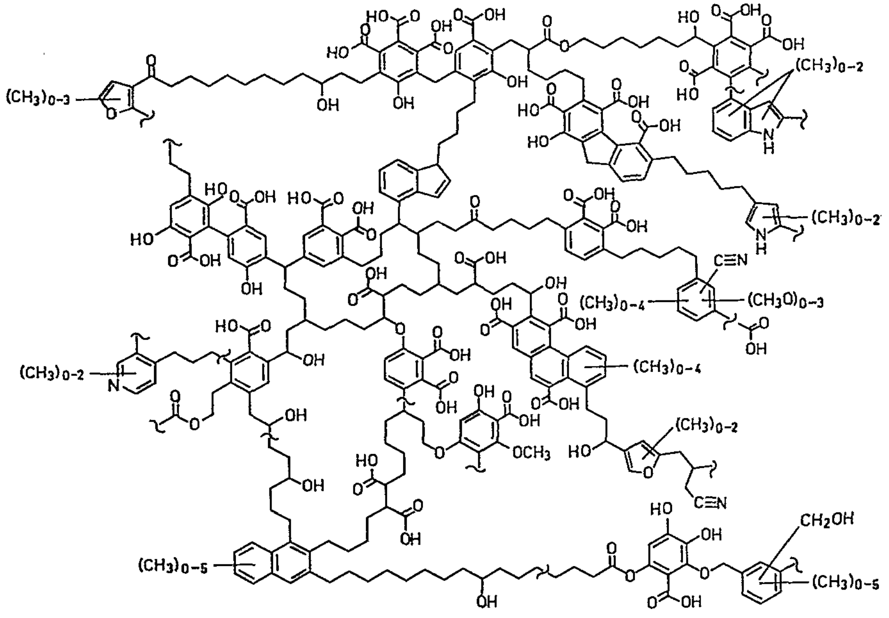

Figure 4 A schematic view of a HA structure (Schulten and Schnitzer 1993).

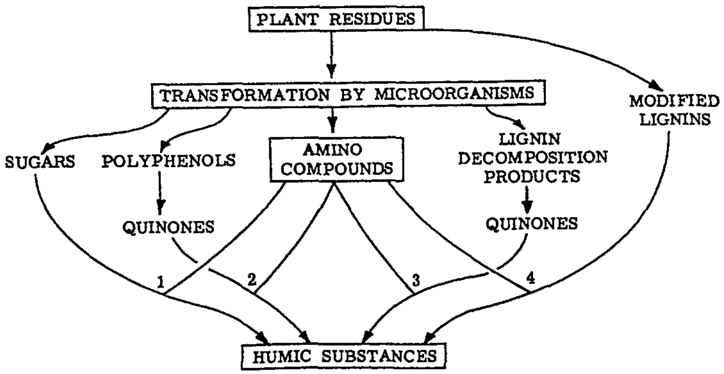

Figure 5 Mechanisms for the formation of humic substances in soil (Stevenson 1982, with the kind permission of John Wiley & Sons, New York).

LEACHATE

The chemical composition of municipal landfill leachates

Leachates vary in chemical composition due to conditions specific to the site, such as the landfill´s construction, its age, the climate at the deposit site, the kind of waste deposited and the sampling point (Jasper *et al.* 1985, Ettala *et al.* 1988, Ehrig 1989, Andreottola and Cannas 1992, Méndez *et al.* 1989).

The composition of leachates in terms of inorganic substances (such as metals and NH_4^+) is better known than their composition in terms of organic matter is. This is due to the fact that most inorganic compounds are relatively easy to analyze whereas the determination of specific organic substances is often complicated and time-consuming. Thus, the organic content of leachates is often measured through analyzing such sum-parameters as COD (chemical oxygen demand), BOD (biochemical oxygen demand) and TOC (total organic carbon). However, more is known of the composition of the organic matter in leachates from landfills in the acidogenic than in the methanogenic phase. This is due to the fact that VFA can easily be analyzed by gas chromatography (GC) or high performance liquid chromatography (HPLC), whereas humic-like substances are unsuitable for identification by ordinary GC and HPLC methods (Chian 1977, Harmsen 1983, Kjeldsen and Christensen 1984, Weis *et al.* 1989). The difficulties in determining the specific structure of the organic molecules in leachates from landfills in the methanogenic phase have led to studies being conducted to characterize these compounds by means of their molecular weight, their hydrophilicity/hydrophobicity, and the presence of specific functional groups. The methods used for estimation of molecular weight include ultra-filtration and gel-filtration chromatography. Also, HPLC analyses have been performed before and after biodegradability experiments and for estimating the degree of hydrophobicity/ hydrophilicity (Chian 1977, Chian and De Walle 1977, Harmsen 1983, Millot *et al.* 1987, Gourdon *et al.* 1989, Weis *et al.* 1989, papers IV and V). Evaluating the results of the biodegradability experiments was based on whether or not a peak disappears or decreases after treatment (Gourdon *et al.* 1989). The hydrophobicity or hydrophilicity was estimated through comparison of the retention times of the peaks of the leachate sample on a Micro pack C18 column with the retention times for phenol, p-creosol and 2,6-dimethylphenol (paper IV). Molecular weight, molecular volume and hydrophobicity (octanol-water partition constant) are important properties and are among the factors that determine whether a compound is bioaccumulable (McKim *et al.* 1985, Niimi and Oliver 1988). Infrared spectroscopy (IR) has been used to compare humic acids from municipal leachates with aquatic and soil humic acids (Artiola-Fortuny and Fuller 1982a). The results showed the humic compounds extracted from leachates to be similar to the humic substances extracted from lake ooze. A study by Gourdon *et al.* 1989 using ultra-filtration, gel-filtration chromatography and HPLC to analyze a leachate from a mixed landfill for industrial and urban waste showed that over 95% of the TOC consisted of compounds with a molecular weight of less than 500, free volatile fatty acids not being found. It was assumed that the compounds in question were industrial chemicals.

Studies have also been performed to identify specific organic compounds (Öman and Hynning 1993, papers I, IV and V). Many different classes of organic compounds have been detected in leachates from municipal landfills, among these phthalates, terpenes, monocyclic and polycyclic aromatic hydrocarbons, and phenols (Artiola-Fortuny and Fuller 1982b, Albaiges *et al.* 1986, Hallbourg *et al.* 1992, Lyngkilde and Christensen 1992, Kjeldsen 1993, Öman and Hynning 1993, papers I and IV). The presence of phthalates can probably be explained by these compounds being used as plasticizers in PVC plastics that are commonly deposited in landfills, whereas the terpenes can originate from such products as soap and cosmetics (Öman and Hynning 1993, papers I and IV). Since terpenes are also produced naturally by many plants, they can be found in garden waste (Lyngkilde and Christensen 1992). The presence of different aromatic compounds such as alkylbenzenes indicates oil products to have been deposited in the landfill (Sawhney and Kozloski 1984, Reinhard *et al.* 1984, Schultz and Kjeldsen 1986), whereas polycyclic aromatic hydrocarbons there may well originate from creosote used in the impregnation of wood (Öman and Hynning 1993). Phenols containing large substituents are used in different products as antioxidants (National Chemicals Inspectorate, Chemical Abstracts). The results just cited show that there is reason to be concerned about leachates, since many of the compounds identified in municipal landfill leachates are refractory and can have a potential for bioaccumulation.

A comparison between municipal wastewater and sanitary landfill leachates with regard to chemical composition

Analyses performed on leachates and on municipal sewage have shown that leachates usually contain higher concentrations of total-nitrogen and organic compounds than municipal wastewater. The concentrations of metal ions, however, are of the same order of magnitude in wastewaters of both kinds. Two exceptions are Fe and Mn, which are generally present in higher concentrations in leachates than in municipal wastewaters. The concentration of phosphorus is generally lower in leachates than in municipal wastewater, Table 1 (Swedish EPA 1982). The large variation in the composition of different leachates is evident in Table 2.

The high concentrations of ammonium and refractory COD in many leachates, Table 2, make the combined treatment of leachates and municipal wastewaters inappropriate in many cases, especially since the limits set for nitrogen discharge by municipal wastewater treatment plants are becoming stricter.

Table 1 Comparison between municipal landfill leachates and municipal sewage in regard to various pollutants (Swedish EPA 1982).

Pollutant	Leachate (median values of samples from 26 landfills)	Wastewater (from a housing area south of Stockholm)
Conductivity (mS/m)	340	75
pH	7.1	7
BOD (mg/l)	600	200
COD (mg/l)	800	450
NH_4^+-N (mg/l)	23	30
NO_3^--N (mg/l)	0.6	0
Tot-N (mg/l)	80	30
Tot-P (mg/l)	1.1	9
Pb (mg/l)	0.04	0.03
Cd (mg/l)	0.005	0.006
Cu (mg/l)	0.05	0.3
Cr (mg/l)	0.05	0.1
Fe (mg/l)	30	2.7
Mn (mg/l)	2.5	0.3
Ni (mg/l)	0.05	0.2
Zn (mg/l)	0.6	0.8

Table 2 The composition of leachates from 26 different Swedish landfills (Meijer 1980).

Parameter	Median value	Range
colour mg/l Pt	630	140-5600
conductivity mS/m	340	50-1400
Cl^- (mg/l)	500	50-1300
pH	7.1	5.2-8.7
BOD (mg/l)	600	90-4200
COD (mg/l)	800	180-4700
$KMnO_4$ (mg/l)	500	40-4600
NH_4^+-N (mg/l)	23	0.2-800
NO_3^--N (mg/l)	0.6	0.06-8
Tot-N (mg/l)	80	8.6-600
Tot-P (mg/l)	1.1	0.07-6.5
Zn (mg/l)	0.6	0.003-6.7
Cu (mg/l)	0.05	<0.01-0.84
Ni (mg/l)	0.05	<0.01-3.0
Cd (mg/l)	0.005	<0.0005-2.7
Cr (mg/l)	0.05	<0.01-0.21
Pb (mg/l)	0.04	<0.01-0.22
Hg (mg/l)	0.0003	<0.0001-0.0033
Fe (mg/l)	30	0.7-290
Mn (mg/l)	2.5	<0.05-40

Swedish leachates are generally not as strong as the strongest ones reported in other European countries and in the USA. This may possibly be explained by the fact that Swedish samples are often taken from leachate ditches or from lagoons in which dilution of the leachate has taken place (Swedish EPA 1992). Furthermore, many studies in which finding of the strongest leachates has been reported have been performed in pilot-scale landfills, all of the waste having been deposited at the same time (Seman 1986). COD, BOD and total-nitrogen values of up to at least 100 000 mg/l, 90 000 mg/l and 5000 mg/l, respectively, have been reported (Andreottola and Cannas 1991).

Degradation of refractory organic compounds

Organic molecules can be metabolized according to one of the following two types of metabolism (Venkatamarani *et al.* 1984):

1. A substance can serve as a source of carbon and energy.

2. A substance can be metabolized despite its not supporting growth. This type of metabolism is called co-metabolism.

Co-metabolism is a significant feature of the biodegradation of anthropogenic compounds. It may possibly be explained by a lack of substrate specificity in the enzyme systems. Examples of substances that have been shown to be metabolized through co-metabolism are DDT and certain polychloroaromatics (Venkatamarani *et al.* 1984). A special kind of metabolism that can occur when halogenated compounds are degraded is reductive dehalogenation followed by mineralization of the compound in question (Suflita *et al.* 1988).

The need for further studies of the degradation of refractory compounds under the conditions that prevail in and around most landfills is shown by the fact that the degradation of such substances as (chlorophenoxy) propionic acids may be sufficiently slow to make its transport to groundwater possible (Gintautas *et al.* 1992). Furthermore, it has been shown that during the degradation of polycyclic aromatic hydrocarbons (PAHs), slowly biodegradable compounds such as aromatic ketones, diketones and alcohols may be formed as metabolites (Gibson and Subramanian 1984, Langbehn and Steinhart 1995). The compound 3,4-dihydro-1(2H)-naphthalenone which was identified in paper IV might, for example, be a degradation product of deposited diesel fuel (Langbehn and Steinhart 1995). The kinds of metabolites that are formed in the degradation of a PAH compound are of great importance since the toxicological and carcinogenic potential of the compound in question is highly dependent on the oxidation products formed (Gibson and Subramanian 1984).

Degradation under anaerobic conditions is complex, since some substances are degraded under methanogenic rather than sulphate-reducing conditions (e.g. certain halogenated aromatic compounds such as 2,4,5-trichlorophenoxyacetic acid, 2,4-dichlorophenoxyacetic acid, various mono- or dichlorophenols and certain chlorobenzoates), whereas other compounds are degraded more quickly under sulphate-reducing than under methanogenic conditions (e.g. p-creosol) (Suflita *et al.* 1988). These authors also reported that tetrachloroethylene was not mineralized to any significant extent under either methanogenic or sulphate-reducing conditions. Metabolites such as trichloroethylene and dichloroethylene were formed, however. The degradation of the halogenated aromatic phenols and benzoates used did in most cases involve a dehalogenation process that continued until all the halogens were removed (Suflita *et al.* 1988). In the case of the chlorophenoxyacetic acids, degradation involved a cleavage of the two-carbon side chain, resulting in phenol being produced and finally mineralized. It has been shown that the degradation potential of such compounds as phthalates, lindane and certain chlorophenols differs very much between methanogenic and acidogenic conditions (Fischer and Pecher 1993, Ejlertsson and Svensson 1997). Ejlertsson and Svensson found that the transformation of the phthalates studied (dimethyl phthalate, butylbenzyl phthalate, bis(2-ethylhexyl) phthalate, phthalate P610 and diisodecyl phthalate) was much faster under methanogenic than under acidogenic conditions. It was found that the degradation stopped at the monoester level in some cases, which might explain the identification of 1,2-benzenedicarboxylicacid mono (2-ethylhexyl) ester in paper I. The concentration of the phthalates was 50 mg/l, measured as carbon in the degradation study performed by Ejlertsson and Svensson. This is many times as high as the concentrations detected in studies on normal leachates. Other studies of the degradation of phthalates under different conditions have shown them to be degradable under both anaerobic and aerobic conditions but the degradation rates to possibly be lower under anaerobic than aerobic conditions (Staples *et al.* 1997). The chlorophenols used in the study of Fischer and Pecher were pentachlorophenol, 2,3,4,6-tetrachlorophenol, 2,4,6-trichlorophenol and 2,4-dichlorophenol. Under methanogenic conditions, all the chlorophenols were partly degraded through dehalogenation processes, whereas they were relatively unaffected under acidogenic conditions. Lindane was degraded totally under both methanogenic and acidogenic conditions. The rate of degradation was much higher, however, under methanogenic than under acidogenic conditions. These results indicate it to be important that methanogenic conditions be introduced in a landfill as early as possible.

Another study of phenols in municipal landfill leachates showed the concentration of total phenol in two stabilized leachates to remain relatively constant over a period of 12 months, whereas it decreased by approximately 70% in the leachate from the newest landfill (Artiola-Fortuny and Fuller 1982 b). The leachates, contained in a 4000-litre leachate generator, were produced from municipal waste similar to the kind of waste deposited in landfills. No treatment of the leachates was performed prior to the phenol analyses (Artiola-Fortuny and Fuller 1982 b). The concentration of total phenol was about 40 µg/l and 90 µg/l for the more stabilized leachates, whereas it was approximately 1000 µg/l at the end of the 12 month period in the leachate coming from the newest landfill. These figures can be compared to the U.S. drinking water standard, set to 1 µg/l or less by the U.S. EPA (Drinking Water Standards 1972). The carcinogenic effect of phenols was taken into account there when the standard was set.

It has also been found that anaerobic conditions facilitate the transport of phenols to the groundwater (Sawhney and Kozloski 1984). The biodegradability of phenols under aerobic and sequential anaerobic-aerobic conditions was shown in a study of the treatment of leachates from oil shale ash to be relatively good (Kettunen and Rintala 1995). Approximately 80% of the phenols were removed. Anaerobic treatment was found to have a minimal effect on the phenol content (Kettunen and Rintala 1995). The importance of examining the degradation of organic substances under anaerobic conditions was also shown in a study by Reinhard *et al.* 1984. The study indicated that such substances as benzene, ethylbenzene, chlorobenzene, naphthalene, 1,2- and 1,4-dichlorobenzene could be persistent and mobile under methanogenic conditions. It has also been reported that under anaerobic conditions the chelating capacity of leachates (metal ions can react with organic ligands to form chelates) is not significantly reduced, even after 120 days of incubation, whereas under aerobic conditions it is reduced to 0.1-1 % of the original value after 74 days (Avnimelech and Raveh 1982).

A study by Berwanger and Barker 1988 of the aerobic and anaerobic degradation of various monoaromatic compounds found in leachates showed that benzene, toluene, ethyl benzene and o-,p-,m-xylene (BTEX) were degraded under aerobic conditions, whereas they were nearly unaffected under anaerobic conditions. Hydrogen peroxide was used as the oxygen source, leachate-impacted groundwater and waste from a landfill being used as medium and inoculum, respectively.

Lyngkilde *et al.* 1988, studied degradation of refractory compounds in diluted methanogenic leachates under aerobic and denitrifying conditions, respectively. Using externally-added compounds (22 different), they observed that the aerobic degradation of many of the compounds was faster when the leachate concentration was increased. In many cases, the lag phase was also shorter when the leachate concentration was increased. These effects were explained by microbial activity, measured as ATP (adenosine triphosphate), being higher in the experiments in which larger concentrations of leachate were used. The substances used in the study included mono- and polycyclic aromatic hydrocarbons, and phenols (e.g. benzene, nitrobenzene, dichlorobenzenes, naphthalene, phenanthrene, fluorenone, biphenyl, indene, dichlorophenols and nitrophenols). The results also showed the aerobic degradation of the aromatic hydrocarbons (50-200 μg/l) containing two or more rings to be rapid, in the range of 1 to 10 days. An interesting finding was that tetrachloromethane was degraded to chloroform under denitrifying but not under aerobic conditions. A study of the aerobic degradation of diesel oil showed that linear alkanes were degraded faster than branched ones (Geerdink *et al.* 1996). In a batch experiment it was shown, however, that branched alkanes can be efficiently degraded if the retention time is sufficiently long (Geerdink *et al.* 1996). It has been found that n-alkanes can be oxidized by various sulphate-reducing bacteria (Rueter *et al.* 1994, Coates *et al.* 1997).

The toxicity of leachates and their content of bioaccumulating compounds

The difficulty in predicting the toxicity of a leachate as a function of the type of waste deposited in the landfill is demonstrated by a study of leachates from three different types of landfills (two landfills in which primarily residential waste had been deposited, one landfill containing both residential and industrial waste and one landfill in which hazardous, municipal and industrial waste had been deposited) (Schrab *et al.* 1993). It was found that leachates from the landfills containing primarily residential waste could be as toxic as those from the landfills containing hazardous as well as municipal and industrial waste. Since organisms differ in their sensitivity, toxicity is difficult to measure, as was seen in a study using four different species of algae (Cheung *et al.* 1993) and in a study of organisms from different trophic levels (Plotkin and Ram 1984). The organisms employed in the latter study were a fish, the fathead minnow (*Pimephales promelas*), a crustacean (*Daphnia magna*), a bacterium (*Photobacterium phosphoreum*) and a green alga (*Selenastrum capriocornutum*). Sensitivity increased from the minnow to the alga. A difficulty in addition to sensitivity is that compounds giving a response in acute toxicity tests may interfere with genetic toxicity tests (Schrab *et al.* 1993).

Studies of a leachate from a conventional, mixed municipal landfill in the methanogenic phase showed a two-stage biological treatment process consisting of a nitrification and a denitrification stage to significantly reduce toxicity towards *Salmo trutta*, *Photobacterium phosphoreum* and *Ceriodaphnia dubia*, some toxicity still remaining after treatment, however. This may possibly be explained by much but not all of the toxicity being due to the high NH_4^+-N concentration in the untreated leachate (600-800 mg/l of NH_4^+-N) (paper IV).

Another important aspect of leachates is their content of fat-soluble organic compounds that can accumulate in living organisms through dissolving in cell membranes. A study by the Swedish Environmental Protection Agency 1989 showed six of the eight leachates that were studied to contain one or more compounds or groups of compounds capable of accumulating in organisms. The analyses were performed by thin-layer chromatography. The compounds were not identified but they were assumed to have an aromatic character since they adsorbed ultraviolet light (254 nm). The leachates were then degraded aerobically for 47 days, after which they were analyzed again for bioaccumulating compounds. The concentrations of the compounds in the leachates after degradation were too low to be detected by UV spectroscopy, indicating that the compounds were indeed degradable. The degradation time found in the Swedish Environmental Protection Agency study should be compared, however, with the normal hydraulic retention times of several hours found in municipal wastewater treatment plants. The importance of studying leachates as regards compounds that can be transported over membranes is also shown by experiments carried out by Schrab *et al.* 1993. Three of the four leachates examined were genetically toxic. It has been found that damage to genetic material can occur through a number of different mechanisms, the most important ones considered being chemical reactions between the toxic compound and DNA, the effects on enzymatic systems that repair and copy DNA, and the disturbance of meiotic cell division (Birgersson *et al.* 1983). The results of the Schrab *et al.* 1993 study also showed that leachates from landfills that received only residential waste posed a higher cumulative risk of cancer than the leachates from landfills in which residential and hazardous waste were co-disposed. In many cases, the carcinogenic compounds involved cause changes in the DNA in either a direct or indirect way, Birgerson *et al.* 1983. Such carcinogens are termed genotoxic. There is also another group, epigenic carcinogens, to which asbestos belongs (Birgerson 1983). These seem to do damage by having an effect on which part of the genes that are active (Birgerson *et al.* 1983).

DIFFERENT METHODS OF LEACHATE TREATMENT

A number of methods for leachate treatment have been studied, including microbiological, physical and chemical processes. The number of full-scale plants involved is still relatively small. In 1995, less than 10% of the deposits in most European countries had an on-site leachate treatment plant (Hjelmar *et al.* 1995). Some of the full-scale treatment plants in use, such as aerated lagoons, are relatively simple (Robinson 1992), while others are more complicated, involving different stages in which microbiological processes are combined with some form of physical and/or chemical treatment, such as reverse osmosis (Weber 1991). The differences in construction are at least partly governed by the characteristics of the leachate and the demands placed on treatment efficiency.

Biological methods

Aerobic processes

Two of the advantages of aerobic processes are that ammonium ions can be efficiently removed and that processes can be carried out over a wide range of temperatures (Kettunen 1997). Also, many substances are degraded at a higher rate in an aerobic than in an anaerobic environment (e.g. certain alkyl benzenes) whereas some are more or less restricted to degradation in an aerobic environment (e.g. PAHs) (Wilson *et al.* 1986, Cerniglia 1992).

Many different aerobic process designs have been studied for the treatment of leachates, for example using reactors in which the microorganisms are kept in suspension (activated sludge reactors or lagoons) or reactors in which the microorganisms grow on support media, i.e. biofilm processes (trickling filters, biorotors, suspended carrier processes or submerged aerobic filters).

Suspended growth processes

Various studies on leachate treatment in suspended growth systems, e.g. in aerated lagoons and in activated sludge processes, have shown there to be an effective removal of organic compounds from acidogenic leachates (Cossu 1981, Robinson and Grantham 1988, Robinson 1992). The COD removal was reported to be between 83% and 97% or more, and the BOD removal to be over 90%. The activated sludge process has been examined in a relatively large number of studies. Most of them, however, have been performed on acidogenic leachates, many of the studies also having been carried out at room temperature. Thus, relatively little is known regarding performance of the process when leachates containing a high concentration of refractory organic compounds in combination with a high concentration of ammonium ions are involved, as in the case of methanogenic leachates. Further studies are also needed to evaluate the process at low temperatures. A pilot-scale study performed by Knox (1985) indicated the temperature dependence of the nitrification process to be relatively strong. In addition, problems with settling of the sludge have arisen. The leachate used in the study by Knox (1985) came from a landfill in the methanogenic phase, which explains the TOC removal being relatively low, approximately 25%. One way to improve the performance of the conventional activated sludge system might be to replace the conventional settler for the sludge by an ultra-filtration unit employing a membrane bioreactor (MBR) system (Bressi and Favali 1997, Luning and Notenboom 1997). The advantages of this process as compared with conventional activated sludge treatment are that the biomass concentration in the reactor is higher, due to the process not being dependent upon the settling of the sludge, and also that the effluent is clearer than it would otherwise be after the treatment in the bioreactor. These advantages indicate that MBR can be regarded as an alternative for the treatment of methanogenic leachates. A further advantage is that, if the settler is replaced by an ultra-filtration unit, the area needed for the treatment plant is decreased (Luning and Notenboom 1997). A disadvantage of the process is the high investment and operating costs.

Attached growth processes

Aerobic biofilm processes which have been studied for the treatment of leachates or leachate-impacted groundwater include trickling filters, biorotors, submerged aerobic filters and the suspended carrier biofilm process (Knox 1985, Spengel and Dzombak 1991, Knox 1987, Pedersen and Jansen 1992, papers II, III and IV). An advantage of these processes as compared with suspended growth systems is that the risk of losing the microorganisms due to poor settling characteristics of the sludge is eliminated. It is important to effectively retain cell mass in the reactor system when removing nitrogen from leachates from methanogenic deposits, since the treatment relies on slow-growing nitrifying bacteria. Another advantage of biofilms compared with suspended growth systems is that attached growth systems are generally considered less sensitive to toxicity and to changes in the environment. Furthermore, the performance of biofilm systems is less influenced by low temperatures than the performance of suspended growth systems is. This is due to the solubility of oxygen in water increasing as temperatures decrease (Okey and Albertson 1989), which is important since the activity of the microorganisms in many biofilms is limited by the oxygen concentration available. An increase in the oxygen concentration and a lower specific activity of the microorganisms allow oxygen to penetrate more deeply into the biofilm, activating a greater number of microorganisms. The main drawbacks of attached growth systems as compared with suspended growth systems are the cost of the carrier media and the need to operate at a high dissolved oxygen (DO) concentration in order to maintain a sufficiently high nitrification rate (Ødegaard *et al.* 1994).

Anaerobic processes

An advantage of anaerobic processes is that these allow heavy metals to be removed from the leachate by precipitation as carbonates or sulphides (Callander and Barford 1983), the sulphides for precipitation being formed under anaerobic conditions by the reduction of sulphate to sulphide (Rumpf and Ferguson 1990). Further advantages of anaerobic as compared with aerobic processes are that the energy consumption for an anaerobic process is lower since no aeration is needed, and also that the production of excess sludge is lower. A disadvantage of anaerobic as compared with aerobic processes is that the high NH_4^+-N concentration is often not sufficiently reduced (Wu *et al.* 1988). Anaerobic processes for leachate treatment that have been studied include digesters, lagoons, upflow anaerobic sludge blankets (UASB) and anaerobic filters (Senior 1990, Kettunen 1997). Most studies of anaerobic processes have been performed on relatively strong leachates, having a BOD/COD ratio greater than or equal to 0.5 (Kettunen 1997).

Suspended growth processes

Studies of leachate treatment in different anaerobic suspended growth systems such as lagoons, digesters and UASB reactors have shown the removal of organic compounds from acidogenic leachates to be relatively efficient (Boyle and Ham 1974, Cameron and Koch 1980, Mennerich 1987, Rumpf and Ferguson 1990, Blakey *et al.* 1992, Jans *et al.* 1992, Garcia *et al.* 1996). The study of Blakey *et al.*, for example, concerning the treatment of an acidogenic leachate in batch lagoons, showed there to be a COD removal of more than 80% at a temperature of 25°C or of 10°C. The hydraulic retention time (HRT) needed was longer at 10°C than at 25°C, however.

Studies on the treatment of leachates in digesters have yielded highly varying results. A removal of 22-96% of the COD has been reported in three studies (Boyle and Ham 1974, Cameron and Koch 1980, Méndez *et al.* 1989). The lowest removals were usually obtained when leachates from old landfills were treated or, alternatively, when treatment was performed at a low temperature (10°C).

In a number of studies on leachate treatment in a UASB process, COD removals of 30-93% at temperatures of 15-35°C have been reported (Mennerich 1987, Blakey *et al.* 1992, Jans *et al.* 1992, García *et al.* 1996). The leachates used in these studies contained relatively high concentrations of biodegradable organic compounds. A study of the treatment in a USAB reactor of a high- to medium-strength leachate showed the average COD removal to be 83%, the average BOD removal being 85%. The removal of heavy metals also varied from approximately 75 to 99% for different metals. The lowest removal was for nickel and the highest removal for zinc (Young *et al.* 1988).

Attached growth processes

The COD removal in the treatment of leachate in upflow anaerobic filters has been reported to vary from 56-95% (Henry *et al.* 1987, Wu *et al.* 1988, Méndez *et al.* 1989). Both methanogenic and acidogenic leachates were studied. The advantages of biofilm processes compared with suspended growth systems mentioned in connection with aerobic treatment also apply to anaerobic treatment. The weaker temperature dependence could be an exception. The potential for better retainment of microorganisms in biofilm processes than in suspended growth systems could be of special importance in anaerobic processes, due to anaerobic microorganisms growing so slowly.

Recycling of leachate

The recycling of leachate back to the landfill is a method that reduces the volume of the leachate and decreases the concentration of organic compounds due to evaporation and anaerobic microbiological degradation in the landfill (Yuen *et al.* 1995). The method has been shown to decrease the time needed in simulated landfills for the transition from the acidogenic to the methanogenic phase (Pohland and Kang 1975). Recycling often needs to be combined with other methods, however (e.g. with treatment in an aerated lagoon), in order for a satisfactory result to be obtained, partly because of the ammonium ion concentration often remaining virtually unaffected after the recirculation of the leachate (Robinson and Maris 1985). An operational problem that has emerged when recycling leachates is that the infiltration capacity decreases with time, due to precipitates accumulating within the surface layer of the landfill (Blakey and Maris 1987). Other disadvantages are that recycling of the leachate can produce instability in the landfill and that the potential for groundwater pollution increases, due to increased hydraulic loading (Lee *et al.* 1986).

An approach that has been successfully tested on a full scale in Germany and on a laboratory and pilot scale in Sweden is to recycle leachate from a part of the landfill that is in the acidogenic phase to a part that is in the methanogenic phase (Doedens and Cord-Landwehr 1984 and 1987, Lagerkvist 1995). This approach has the advantages that many of the pollutants can be degraded by the methanogenic microflora and that some of the toxic elements are immobilized in the methanogenic deposit. Gas production can also be controlled in terms of time, space and intensity (Lagerkvist 1995). The results of the full-scale study showed that in most cases the COD removal was over 85%, whereas the BOD removal was generally over 90% (Doedens and Cord-Landwehr 1987). The recycling of leachates requires a very careful monitoring of both surfacewater and groundwater in order to ensure that recycling can be stopped immediately if the system fails (Lee *et al.* 1986).

Combined aerobic and anoxic or anaerobic processes

Because of the high concentration of ammonium ions in many leachates, a combined aerobic and anoxic treatment process could be a good method to reduce nitrogen content. In the aerobic stage ammonium ions are oxidized to nitrate ions (nitrification), whereas the nitrate ions are reduced to nitrogen gas in the anoxic stage (denitrification). The addition of an externally applied, easily degradable carbon source and a source of phosphorus are usually needed when methanogenic leachates are treated, in order for a well-functioning anoxic process to be obtained. Different process designs that have been used for the combined aerobic and anoxic treatment of wastewaters include suspended growth systems such as sequenced batch reactors (SBR), as well as pre-denitrification or post-denitrification configurations (Hosomi *et al.* 1989, Lie 1996). An approach that has been studied recently is the suspended-carrier biofilm process (Rusten *et al.* 1995, papers II, III and IV), Figure 6. An advantage of this approach compared with many other biofilm processes is the good mixing of the reactor content which is achieved, minimizing the risk of liquid´s short-circuiting and clogging the media with biomass or other solids. Good mixing also ensures an efficient mass transfer, which is very important. The main disadvantage of the process is its strong oxygen dependence, which has been shown to be approximately first-order, and not half-order as for other biofilm processes (Harremoës 1982, Hem *et al.* 1994). The suspended-carrier biofilm process has been shown to provide efficient nitrogen removal from a methanogenic leachate once the process is well established and both the dosage of carbon and the phosphorus source are optimized. The establishment of nitrification is shown in Figures 7 and 8. The degree of COD removal achieved in the nitrification process was found to be fairly low, however, approximately 20% (paper III). The process was also evaluated with regard to the removal of specific hydrophobic compounds (paper IV). It was shown that some organic substances, for example various plasticizers, were unaffected by the treatment.

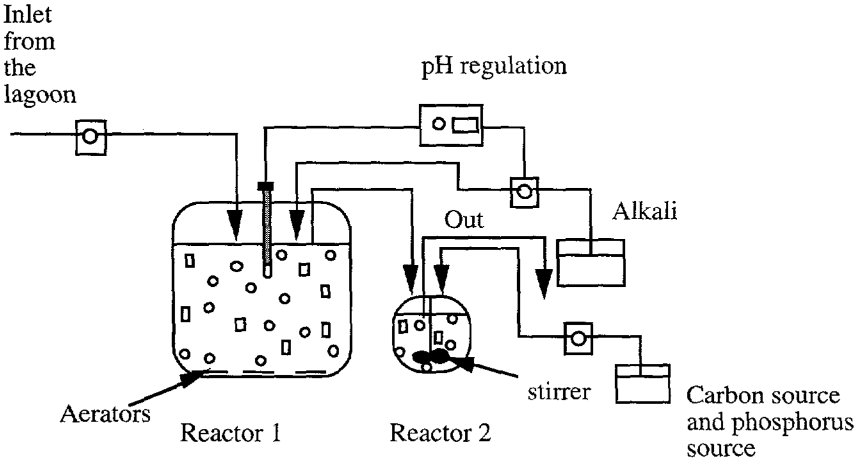

Figure 6 The suspended carrier biofilm process (paper III).

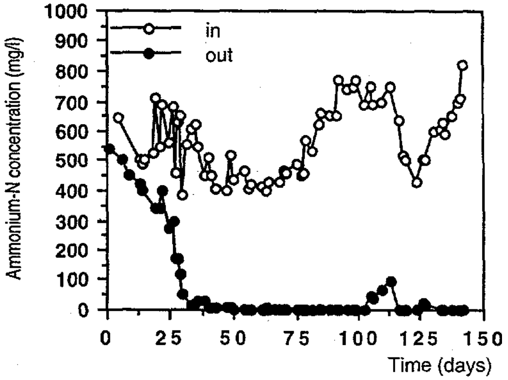

Figure 7 NH_4^+-N concentration in the inlet to and outlet from the nitrification process (paper III).

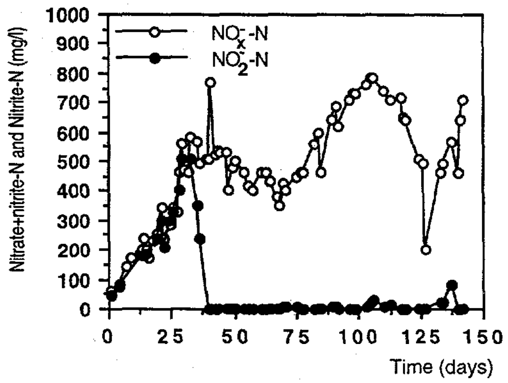

Figure 8 NO_x^--N and NO_2^--N in the outlet from the nitrification process (paper III).

The sequential anaerobic-aerobic treatment of a medium-strength municipal landfill leachate by Kettunen *et al.* 1997 resulted in a COD removal of 80-90% and a nitrification of 65->99%. The anaerobic treatment was performed on a pilot scale in a UASB reactor, whereas the aerobic treatment was performed in a laboratory-scale activated-sludge reactor. The temperature was 13-28°C in the anaerobic stage and 5-11°C in the aerobic stage. A comparison of sequential treatment with aerobic treatment alone showed removal of nitrogen in the aerobic treatment alone to mainly be due to assimilation. This poses a risk since it means that the ammonium may be released again when the sludge is used as landfilling. Another study performed on a combination of anaerobic and aerobic treatment in a biofilm system showed some chlorinated hydrocarbons to be more efficiently converted to carbon dioxide in the combined system than in the anaerobic reactor alone (Fathepure and Vogel 1991). The substances used were chloroform, trichloroethylene and hexachlorobenzene. The compounds were reductively dechlorinated in the anaerobic reactor and were then mineralized in the aerobic reactor. The overall performance of the system gave a mineralization of 61, 49 and 23%, respectively, of the three compounds, the corresponding figures after anaerobic treatment alone being 7.0, 1.0 and 16%, respectively.

Physical and chemical methods

Adsorption

Adsorption on activated carbon is a method that has been used to decrease the concentrations of hydrophobic substances difficult to remove in other ways. Aromatic compounds, for example can be removed in this manner (Rennerfelt and Ulmgren 1975). The best removal in an adsorption onto activated carbon is obtained for the fulvic acid fraction. This fraction contains substances in a molecular weight interval of 100 to 10000 (Kylefors 1997). The carbon can be used in either granular or powder form. An advantage of granular carbons is that they can be regenerated (Pinker 1997). This is a significant advantage since during the regeneration process the compounds that are adsorbed onto granular carbons are destroyed, so that there is no concentrated waste to be deposited. Adsorption onto activated carbon is a process often used as a polishing step after some other stage, so as to improve treatment efficiency. Experience during the 1990s with the treatment of leachates in this way has shown such treatment to be cost-effective (Bourke and Villers 1993). The cost per m^3 is shown in Figure 9. There is a considerable potential for post-treatment with activated carbon, since many landfills emit leachates containing high concentrations of COD and AOX, even after treatment of some kind.

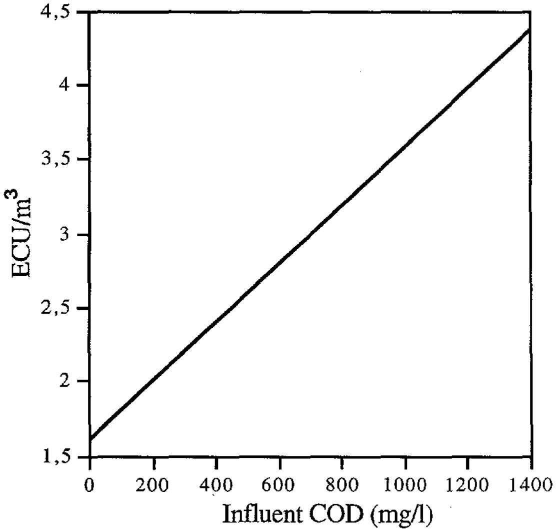

Figure 9 Operating costs per m^3 for treatment with granular activated carbon versus COD (modified from Bourker and Villers 1993).

The cost of treatment with activated carbon can be compared with the estimated cost of reverse osmosis or of evaporation, reported to be 15-30 ECU/m^3 leachate (Hjelmar *et al.* 1995). (See also the paragraph concerning combined biological and physical or chemical treatment.)

Precipitation

Precipitation by chemicals such as aluminum sulphate, ferrous sulphate, ferric sulphate or lime is used in municipal wastewater treatment plants to remove phosphorus. However, the process has been shown to be capable of removing various organic substances as well (Rennerfelt and Ulmgren 1975). Precipitation is often used in combination with biological treatment (see the paragraph concerning a combined biological and physical or chemical treatment).

Reverse osmosis

Reverse osmosis is a method used in Germany, the Netherlands and Switzerland, for example, for leachate treatment in new plants. An advantage of reverse osmosis is that it is relatively insensitive to variations in the concentrations of the compounds to be removed. Also, most of the leachate compounds are well retained, with the exception of certain small molecules such as ammonia and small chlorinated organic compounds (Collivignarelli *et al.* 1993, Linde 1995). This means that a two-stage reverse osmosis process, or a combination of reverse osmosis and another method, such as biological treatment, is needed if the leachate contains high concentrations of ammonia, as is often the case. Some of the advantages of using a biological pre-treatment involving reverse osmosis are those of reduced fouling of the membrane, better permeate quality, higher flux and lower buffer capacity, the latter being relevant to pH adjustments (Linde 1995). The main disadvantage of reverse osmosis is that it yields a concentrate which needs further treatment (Seyfried and Theilen 1991). Reverse osmosis has been shown to be an efficient process for the removal of organic compounds from stabilized leachates. The leachates may have become stabilized due to the landfill being in the methanogenic phase or its having been treated in a biological process (De Walle and Chian 1977). The removal of COD from an acidogenic leachate pretreated in an anaerobic filter has been shown to be about 95%, whereas the removal from the raw leachate was about 56%. The difference is due to the fact that the reverse osmosis process removes high-molecular-weight carbohydrate humic complexes more efficiently than it removes low-molecular-weight fatty acids (De Walle and Chian 1977).

Oxidation

Experiments involving the oxidation of leachates from a stabilized landfill using Fenton´s reagent (hydrogen peroxide and Fe^{2+} ions) have shown the maximum reduction of COD achieved to be about 35% (Loizidou *et al.* 1993). The oxidation process can be carried out using different reagents, such as Fenton´s reagent, ozone or sodium hypochlorite. A disadvantage of sodium hypochlorite compared with the other reagents, however, is that toxic chlorinated organic compounds may be produced during the oxidation process (Banerjee and O`toole 1995). The oxidation process can be combined with biological treatment, since oxidation can increase the bio-availability of various refractory organic compounds (Anagiotou *et al.* 1993) (see the paragraph concerning combined biological and physical or chemical treatment).

Ammonia stripping

A study by Cheung *et al.* 1997 showed that when the pH of the leachate was increased by the addition of calcium hydroxide, 70% of the NH_4^+-N was removed when free-stripping (air flow 0 l/min) was carried out for a day. Two different leachates from landfills in the methanogenic phase were studied. The results showed that, when aeration was increased from 0 l/min to 5 l/min, removal was increased to 90%. The COD removal was 24% and 47%, respectively, for the two leachates, the phosphorus removal being 85% and 93%. The removal of COD and phosphorus was attributed to precipitation. The results showed that further treatment to remove the remaining COD and nitrogen was needed before the leachate could be discharged directly into the environment.

Evaporation processes

A new full-scale evaporation process has been built at a sanitary landfill in Finland (Ettala 1997). The approach is to let leachate evaporate on a thin plastic film. The energy consumption of the process is relatively low since only small temperature and pressure differences are utilized. Due to the low pressure difference, fans instead of compressors can be used. The capacity of the process is 130 m^3/d. The COD_{Cr} removal is more than 87%, and the NH_4^+-N removal 99.9%. The high removal of NH_4^+-N is accomplished through adjusting the pH of the leachate to 4, prior to treatment. The leachate is pretreated in a sand filter. Two disadvantages of the evaporation process are that a concentrate is created, one that needs to be taken care of, and that the low pH has to be adjusted before the treated leachate is discharged. Two other evaporation processes have been developed in the US and in Italy, respectively (Cross *et al.* 1997), both processes using landfill gas as the energy source. Most of the organic substances are stripped from the leachate and transferred to the vapor phase. Since the vapor can be malodorous, the exhaust gas may need to be further treated by thermal oxidation, for example.

Combined biological and physical or chemical processes

Due to their high content of NH_4^+-N and COD, many leachates need to be treated by means of a combined biological and physical or chemical process. The ammonium concentration can be effectively reduced by means of a nitrification and denitrification process. In methanogenic leachates, the main part of the organic matter is refractory, so that a physical or chemical method is needed to achieve an acceptable level of COD removal (Gau and Chang 1996).

Experiments involving the treatment of leachate from Atleverket (a methanogenic landfill) in which two columns filled with light expanded clay, the one column aerobic and the other anaerobic, showed the effluent from the anaerobic column to be more similar in chemical composition to the untreated leachate than to the effluent from the aerobic column, Table 3. The effluent from the aerobic column contained several PAHs that could not be found neither in the anaerobic effluent nor in the untreated leachate. These results indicate that the untreated leachate contained these substances but that they could not be identified due to the fact that they were hidden in the chromatograms by other substances that were degraded aerobically (Welander unpublished results, Table 3).

Table 3 Substances identified in untreated leachate and in leachate treated in light- expanded-clay columns.

Substance	Untreated	Aerobically treated	Anaerobically treated
Alkanes/alkenes		+	
Benzaldehyde		+	
C_3 substituted benzenes (6 isomers)		+	
C_4 substituted benzene (1 isomer)		+	
Benzophenone		+	
N-n Butylbenzenesulfonamide	+		+
Cinnamic acid m methoxy ester	+		
2,5-cyclohexadiene 1,4-dione 2,6-bis (1,1-dimethylethyl)		+	
Dibenzofurane		+	
Hexanedioic acid bis (2-ethylhexyl) ester	+		
Hexanedioic acid diester (structure unknown)	+		
3-hydroxyacetophenone	+		
Phenol	+		
2,4-Bis (1,1-dimethylethyl) phenol	+		+
2,6-Bis (1,1-dimethylethyl) phenol	+		
Chlorinated methylphenol (1 isomer)	+		
Dimethylphenols (2 isomers)	+		
2-(1,1-dimethylethyl)-4-methoxyphenol		+	
2-(1,1-dimethylethyl)-4,6-nitrophenol		+	
Methylphenol (2 isomers)	+		
Methylnitrophenol		+	
1,1-Biphenyl		+	
Naphthalene	+	+	
Methylnaphthalene (2 isomers)		+	
2-Methoxynaphthalene		+	
9H Fluorene		+	
9H Fluorene 9 one		+	
Phenanthrene		+	
Tetramethyl phenanthrene		+	
Terpene (Camphor)	+		+
Terpene (Fenchon)	+		
Terpenes, structure unknown	+	+	
Phthalates	+	+	+

A number of fatty acids were also detected in the untreated leachate and in the anaerobically treated leachate. The concentrations were very low, however.

A pilot-scale study performed on a stabilized leachate showed that the removal of COD could be greatly enhanced if the activated sludge process was combined with adsorption onto activated carbon (Copa and Meidl 1986). The removal of COD was 77% and nitrification of the NH_4^+-N 98%. Use of an activated sludge and powdered activated carbon (PACT) process which combines physical adsorption with biological treatment, makes the efficiency of the treatment higher than for either of the two component processes alone. Such a combined process can also stimulate the nitrification of wastewaters that contain inhibitory compounds, due to the toxicity being reduced by adsorption of the inhibitory compounds onto the carbon (Lankford and Eckenfelder 1990, Horan *et al.* 1997). It has been shown too that the process is able to reduce the metal contaminant content in some wastewaters (Lankford and Eckenfelder 1990). One theory put forward to explain this is that an organic compound that has formed a complex with the metal ion adsorbs onto the activated carbon. Another theory is that sulphides precipitate on the surface of the carbon due to the high sulfur content of the carbon. The metal ion may also be co-precipitated on microbiological flocs (Lankford and Eckenfelder 1990).

Experiments performed in the present study showed that 76% of the COD content of the leachate from the Kristianstad landfill (a landfill in the methanogenic phase) could be removed by a process consisting of a combination of nitrifying aerobic treatment and adsorption onto powdered, activated carbon (paper V). A study performed by Pohland and Kang in 1975 gave a COD removal of 96% when a leachate was treated in an activated sludge process followed by adsorption on activated carbon. Another study, performed on an acidogenic leachate, showed there to be a total COD removal of 99.4% after aerobic pre-treatment followed by adsorption onto activated carbon the removal after the biological treatment alone being 97.6% (Cook and Foree 1974). In the present study, a combination of aerobic treatment, precipitation with ferric chloride and adsorption onto activated carbon removed 96% of the TOC in leachate from the Kristianstad landfill (paper V).

The potential of biofilm processes to degrade refractory organic compounds has been examined in a study involving the treatment of a stabilized leachate in a fluidized bed, using activated carbon as the carrier media (Iwami *et al.* 1992). The process consisted of an anaerobic and an aerobic stage. On the average, 60% of the dissolved organic carbon (DOC) was removed whereas the total nitrogen removal was 70%. A steady-state balance performed by Iwami *et al.* 1992 showed microbiological degradation to be the main process in the removal of DOC. About 90% of the DOC removal was caused by microbiological degradation. The mechanism proposed was adsorption of the compounds onto the activated carbon, followed by degradation. These results indicated that the structure of the surface of the carrier is very important for the degradation of refractory organic compounds. A further study of the process, by Imai *et al.* 1995, showed it to be the low-molecular-weight fraction of the organic matter that was primarily adsorbed onto the activated carbon and then degraded. Treatment of the leachate in the fluidized bed process was compared with treatment of it in an activated sludge reactor. The activated sludge process gave a DOC removal of only 20%.

A treatment process consisting of three stages, namely an aerobic biological stage, oxidation by Fenton´s reagent and a final aerobic biological stage, gave a maximum COD removal of 73% (Anagiotou *et al.* 1993). This result can be compared with the COD removal attained by biological treatment alone, which was 35-47%, and the removal attained by oxidation without biological treatment, which was 22%. In the present study, experiments were performed involving the oxidation of nitrified leachate from the Kristanstad landfill with Fenton´s reagent or with ozone. The results showed Fenton´s reagent to be more efficient than ozone. The COD removal after aerobic treatment and oxidation with Fenton´s reagent was 47%, whereas it was 38% after aerobic treatment and oxidation with ozone (paper V) using an amount of reagents corresponding to approximately 40% of that theoretically needed for a total oxidation of the COD content of the leachate. The COD removal through aerobic treatment and oxidation with Fenton´s reagent was 47-80%, depending on the concentrations of chemicals used. The corresponding figures for aerobic treatment and oxidation with ozone were 29-54% (paper V). The nitrification process reduced the COD content of the untreated leachate by 20-30%. The results for oxidation with ozone agree fairly well with the results of other studies performed on leachates pretreated in an anaerobic filter or an aerated lagoon (Chian and De Walle 1976). The COD removal in this study varied between 22 and 48% depending on which leachate was studied and the amount of ozone employed. Studies on leachate treatment in a process consisting of oxidation with Fenton´s reagent in a pre-mix chamber followed by biological treatment in two aerobic SBRs showed a COD removal of 60% whereas the ammonium ion removal by nitrification was around 90% (Surmacz-Górska and Miksch 1997). The COD removal was obtained in the pre-mix chamber and in the first SBR, whereas the nitrification took place in the second reactor. The biodegradability of the organic content was shown to be low before the experiments started. Another study, which combined oxidation with ozone with recirculation of the leachate back to the lysimeters, showed the biodegradability of the leachate to increase after ozonation, the lysimeters acting as anaerobic filters (Beaman *et al.* 1997).

Other oxidation processes for the treatment of leachates that have been studied are electrochemical oxidation and a combination of electrolysis and electro-coagulation (Cossu *et al.* 1997, Tsai *et al.* 1997). The result of the electrochemical oxidation study showed both organic compounds and ammonium ions to be oxidized in treatment. The leachate used was treated biologically prior to oxidation, making it stabilized. The COD removal was 75%, the ammonium ion removal when the optimum experimental conditions prevailed being nearly 100%. The electrical power consumption was 60 kWh/ kg COD. The experiments also showed that the density of the current needed to achieve a COD removal of 75% could be decreased to a fourth if the leachate was pretreated by flocculation with an aluminum salt. The combined electrolysis and electro-coagulation process have been found to give a COD removal of 30-50% (Tsai *et al.* 1997). Two different leachates were used in the study, one from a landfill more than 15 years old and one from a landfill less than 15 years old.

Experiments performed using a combination of aerobic treatment and precipitation of leachate from the Kristianstad landfill showed the maximum COD removal attained to be 68% when ferric chloride was employed as the precipitation reagent and 54% when the reagent was aluminum sulphate (paper V). It was mainly high-molecular-weight compounds that were removed by precipitation with ferric chloride. The removal achieved in the present study was higher than that achieved in studies in which stronger leachates were employed (Ho *et al.* 1974), but they are in fair agreement with the results obtained in a study by Amokrane *et al.* 1997. The maximum COD removal obtained in that study was 55% using ferric chloride and 42% using aluminum sulphate. The leachate used was stabilized.

Soil application systems

Approximately 50 of the Swedish active municipal landfills have some kind of local leachate treatment (Linde 1995, Swedish EPA 1993). Most of them have some kind of irrigation system (e.g. energy forest). Since it is difficult to take representative samples from irrigation systems, the treatment efficiency for the removal of organic compounds that can accumulate in organisms has been poorly evaluated. The importance of such an evaluation is shown by the fact that studies performed in Denmark and the US on the degradation of organic compounds in soil have shown some substances to be relatively refractory under anaerobic conditions (Reinhard 1984, Kjeldsen 1986, Kjeldsen *et al.* 1990). The strategy for the treatment of leachates in a natural system such as used in the UK during the early 70s has been subject to increasing criticism for the results of treatment varying strongly with the strata (Walker 1993).

THE COST OF ACTIVE LEACHATE TREATMENT

A comparison of the costs of three different leachate treatments was carried out by Doedens and Theilen in 1992. The three alternative treatment were as follows:

1. A combination of biological treatment, adsorption onto activated carbon and precipitation-coagulation.

2. A two-stage, reverse-osmosis treatment, with evaporation of the concentrate.

3. A combination of biological treatment with a two-stage reverse-osmosis treatment in which the concentrate is evaporated.

The cost of the first and second treatments was estimated to be 25-30 US $ /m^3, whereas for the third alternative it was estimated to be 30-35 US $/$m^3$(Doedens and Theilen 1992). No sludge incineration was included in the costs of the first alternative, no hazardous waste disposal being included in the cost calculations for the second and third alternatives. Another estimation of the costs for leachate treatment showed biological treatment to be the cheapest method, with a cost of 5-10 DM/m^3 leachate, whereas evaporation in combination with combustion was the most expensive, costing more than 100 DM/m^3 leachate (Linde 1995), Table 4. The costs of treating the leachate coming from a landfill serving a town of about 100.000 citizens can be estimated by multiplying the cost per m^3 with the leachate volume produced per year, which in Sweden is normally between 70.000 and 100.000 m^3 .

Table 4. An estimate of the costs of various leachate treatment methods (Linde 1995).

Treatment	Cost (DM/m^3)
Biological treatment	5-10
Chemical oxidation	35-40
One-stage reverse osmosis	10-15
Two-stage reverse osmosis	15-20
Evaporation and stripping	60-65
Evaporation and combustion	>100
Two-stage reverse osmosis evaporation and stripping	35-40
Two-stage, reverse osmosis Evaporation and stripping Transport 200 km Concentrate combustion Transport 200 km Deposit of residue	65-70

Some Swedish calculations of the costs of different treatment alternatives for a leachate amount of 40.000 m^3 leachate/year show that the running costs for an SBR reactor should be around 8 SEK/m^3, whereas the investment costs has been estimated to 2-2.5 million SEK. The corresponding costs of ammonia stripping in a tower is 19 SEK/m^3 leachate and 3 million SEK, respectively. The costs of precipitation with lime were calculated to be 12 SEK/m^3 and 1-1.5 million SEK, respectively (RVF 1996).

The high costs show the importance of an optimization of the leachate treatment. One way of obtaining a treatment that is adjusted to the leachate in question is to separate the waste so that only waste of similar properties is deposited in a specific area (Kylefors 1997). The leachates from different areas are then collected and treated separately (Kylefors 1997).

WASTE HANDLING/ LEACHATE TREATMENT IN OTHER COUNTRIES

European Union (EU) countries

A waste handling practice in many countries has been to enclose waste as much as possible and minimize the volume of the leachate. This trend is changing, however, in the UK, for example, towards forced waste stabilization through leachate recirculation (the biological reactor approach). This is because under the conditions of leachate minimization the waste in many landfills becomes too dry for optimal degradation to take place, a matter illustrated by the fact that newspapers can remain intact for decades after deposition (Young *et al.* 1988). The idea of forced stabilization through recirculation of the leachate is that each generation should be able to take care of its own waste. Landfills are supposed to be stabilized after 30-50 years so that no further active leachate treatment is needed (De Rome and Gronow 1995). Another advantage is the possibility that gas production can be intensified if recirculation is performed correctly (De Rome and Gronow 1995). In January 1990, a project was started in the UK to evaluate the composition of leachates from different types of landfill sites. Eleven different categories of landfills were chosen (Robinson and Gronow 1993). Typical parameters included in the project were COD, BOD, pH, Cl^-, NH_4^+-N and certain metal ions. In a limited number of samples, certain specific organic compounds were also analyzed in order to develop recommendations for further analytical studies (Robinson and Gronow 1993). One of the aims of the project was to develop technical guidance in leachate treatment.

In Denmark the principle is the same as in the UK (that each generation should take care of its own waste). This means that, if waste is to be deposited, its leaching characteristics must be well known. The strategy in Denmark is that landfills should be located near the sea in order to ensure that the potential for dilution of the leachate is sufficiently large if the active environmental protection systems should be changed to inactive systems. The strategy was also for the deposition of combustible waste in landfills to be forbidden after 1997 (Johannessen *et al.* 1995). There are, some exceptions, however, in cases in which the incineration of organic wastes is not financially or environmentally feasible, namely in areas of low population density where landfills are to be constructed as "biological reactors" (Johannessen *et al.* 1993).

In Germany the strategy is to minimize in as far as possible the organic content of the waste that is deposited. Waste to be deposited in ordinary landfills must meet certain prescribed requirements, therefore. Waste exceeding these values are allowed to be deposited in monofills or in monosections of ordinary landfills (Stief 1993). The separation of organic matter from ordinary deposits is aimed at decreasing the organic load of leachates. The generation of leachate is to be minimized, so that the mobilization of pollutants is avoided and the costs of possible leachate treatment are kept low.

In July 1992 a new concept was introduced into French law that of "final waste". Starting in the year 2002, only final waste is to be accepted for deposition. Final waste is defined as waste that cannot be treated further under the current technico-economical conditions (Kopp *et al.* 1995). New deposits are to be constructed to replace the old municipal landfills. The owner of the landfill has to pay into a security fund so the financing of monitoring, of restoration of the site and of remediation in the case of accident is assured. This new policy means a decrease in the organic content of the deposited waste, since biodegradable organic waste is treated by composting or anaerobic digestion, whereas paper, board and plastics are recycled or incinerated (Kopp *et al.* 1995). Due to a lack of knowledge concerning emissions from these new landfills, many questions are yet to be answered in connection with the needs for leachate and gas treatment.

Various aspects of the EU directives

A directive from the European Union states that in the future the amount of organic waste deposited as landfill is to be gradually decreased (Petersen 1997), so that in the year 2002 a maximum of 75% of the amount of organic waste deposited in 1993 is to be deposited. The amount should then be decreased still further so that in 2005 a maximum of 50% of the amount deposited in 1993 is to be deposited, and in 2010 25% of that same figure (Draft Proposal Commission of the European Communities 1997). Two aims of this directive are to reduce the "global greenhouse effect" and to alleviate stability problems in landfills. A further result will be that the area needed for landfilling will decrease. Another regulation in the directives which is new is that the joint disposal of hazardous and non-hazardous waste is forbidden, which means that the control of landfills must be improved.

Waste handling in the USA

In the US, interest in the "bioreactor approach" is growing, despite the fact that the regulations are geared to a "dry" approach to landfilling (Teplitzky 1995). The groundwater monitoring program there involves three steps: detection monitoring, assessment monitoring and corrective action. Detection monitoring includes analyses of a set of indicator parameters. If a constituent is found to be at a higher concentration than is common in the environment, the owner is forced to continue assessment and monitoring, which involves analyses of a broad list of compounds. The aim of assessment monitoring is to evaluate the nature and extent of the release. If a compound is found at a concentration exceeding the groundwater protection standard, the owner must start corrective measures. The groundwater monitoring program must be adhered to by the owner of the landfill unless it can be shown that there is no potential for hazardous constituents migrating from the landfill to the upper aquifer during the active life of the landfill and the post-closure care period. From 1993 to 1994, the number of active landfills in the US decreased by 20%. In 1994 there were reported to be 3558 landfills (Steuteville 1995).

Waste handling in the Moscow Area of Russia

In the Moscow area, nearly all waste is deposited in landfills since the implementation of such methods as recycling and reuse have declined recently. Incineration is not utilized there (Lifshits *et al.* 1997). Landfilling is undertaken under varying conditions. The greatest problem in the area is the increase in the illegal dumping of waste (Lifshits *et al.* 1997). Among the underlying reasons for the problems there are the economic situation, organizational problems and lack of awareness on the part of the public and of politicians (Lifshits *et al.* 1997). However, it has been proposed that the same quality demand as used for EU should be accepted as the Intended Quality of the Moscow Area. In the proposal for Moscow, the time limit for the implementation of this demand is set for the year 2010, however, as compared with the year 2000 for the EU. A new waste management strategy has been established in order to enhance both the prevention and the reuse of waste, and to achieve environmentally sound landfilling. Deposition is to remain the dominant method of disposal in the Moscow area in the near future.

Waste handling in South Africa

Waste disposal in South Africa is based on a set of minimum requirements, introduced mainly for new landfills. These requirements are based on a landfill classification system in which general waste, e.g. domestic waste and commercial and industrial dry waste, is deposited in one class of landfills and hazardous waste in another (Ball *et al.* 1995). Hazardous waste is separated further into different categories depending on how hazardous the waste is, on the size of the landfill and on the risk of leachate production. This risk depends on precipitation, the water content of the deposited waste and other site-specific conditions. There are requirements for leachate treatment if a site generates significant amounts of leachates. The requirements are based on to which class the landfill belongs. Interest in the South African model indicates that it may also be introduced in various other developing countries.

FUTURE LEACHATE TREATMENT

As already indicated, waste handling in the European Union is to a large extent focused on the idea that each generation should take care of its own waste. Thus, after 30-50 years, landfills are supposed to have reached a point of degradation such that active treatment systems for leachates can be replaced by inactive systems. There are, some research results which suggest, however, that this time perspective is far too short (Hjelmar *et al.* 1995). Gromadecki 1995 reported that the main problems of leachates from two landfills that were being operated according to the new waste management measures introduced in Germany were centered around the content of the nitrogen compounds COD and BOD. Thus, the same parameters need be taken into account when leachates from this new type of landfill as when leachates from older landfills are treated, despite the waste deposited in the new landfills containing much less organic matter. A relatively large part of the nitrogen, however, was found to be discharged as nitrate ions, which is in contrast to most older landfills, in which the nitrogen is almost exclusively discharged either as ammonium ions or in an organically bound form. The results of Gromadecki´s study indicate the time needed to establish the methanogenic phase to be shorter in the new type of landfills. The landfills included in the study were in the methanogenic phase after three years of operation, which can be compared with the results of a study by Kruse *et al.* 1994 showing the average time for establishment of the methanogenic phase to be six years. The differences in leachate composition and the time needed for the development of methanogenic conditions between older landfills and landfills for which the new regulations have been implemented are illustrated in Figure 10. The data for the oxidation phase of traditional landfills are mainly based on laboratory experiments (Mennerich 1984). The time required for the after-care period was estimated as decreasing from approximately 500 years to 250 years when the landfill management practices have been developed and implemented (Gromadecki 1995). The long time needed for after-care, even for deposits that accord with the new regulations, shows there to be an urgent need for further research on leachate treatment in active systems. The results also indicate that it is not possible to create landfills which during their active phase need no other treatment than by an inactive system such as irrigation, for example. Leachates from the new type of landfills must be analyzed for specific organic compounds so that treatment can be optimized. However, it is probable that a combination of microbiological and physical or chemical treatment will be needed for leachates both from the newer landfills and from existing ones. This conclusion is based on its having been shown that the BOD/COD ratios in the leachates from the new landfills were low (Gromadecki 1995). Thus, an acceptable level of COD removal is difficult to attain in a process based on microbiology alone, at the same time as an acceptable level of nitrogen removal at a reasonable cost is difficult to attain with most physical or chemical methods alone. There is an obvious need for more research on new landfills in which the amount of organic waste has been decreased considerably compared with older deposits.

The need for continued research on leachate treatment is also shown by the rapidly increasing number of landfills equipped with an on-site treatment plant (Hjelmar *et al.* 1995), as well as by the fact that the oldest on-site leachate treatment plants have been in operation for only about 20 years and that most treatment plants are less than 5 years old (Hjelmar *et al.* 1995).

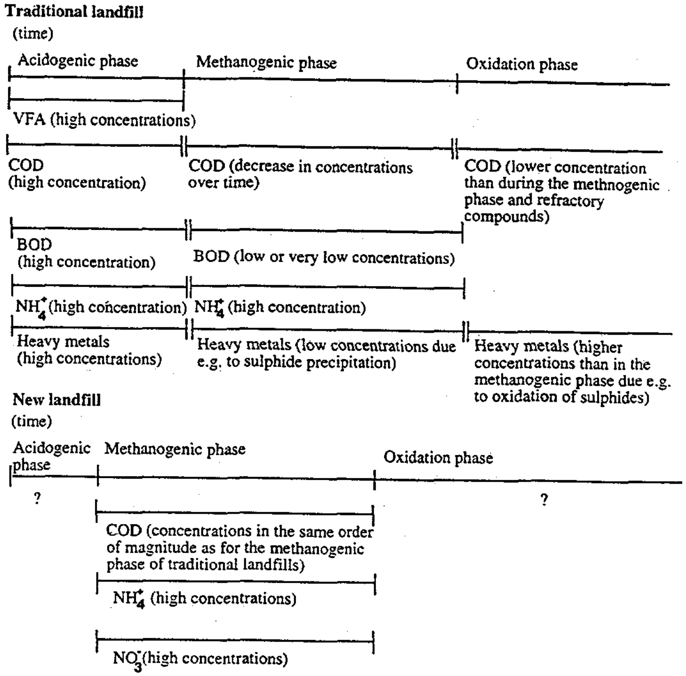

Figure 10. The differences in composition and stabilization time for leachates from traditional landfills and landfills operated in accordance with the new regulations.

CONCLUSIONS

The treatment processes for leachates from methanogenic landfills should consist of a combination of microbiological and physical/chemical stages so that the content there of both nitrogen and of refractory organic compounds can be effectively decreased. The nitrogen content can be reduced satisfactorily by a nitrification and denitrification process according to the suspended carrier biofilm technology, for instance. Since this process is relatively insensitive to low temperatures, it works efficiently, even during the cold season. In addition, since the process is flexible it is relatively simple to change the sequence of the stages when the chemical composition of the leachate changes with time. Since the removal of COD is low, however, treatment should be improved in some way. One way of attaining an increase in COD removal is to combine the biological process with some physical or chemical treatment, Figure 11. The optimal order of the stages depends on which kind of physical or chemical treatment method is used. An oxidation stage, for instance, can be carried out prior to the anoxic stage, since treatment may render some refractory organic compounds more available to the microorganisms, so that they can use them as a carbon source for the denitrification process, Figure 12. The aerobic stage following the anoxic treatment, as shown in the figures, is needed in order to remove any excess of the carbon source.

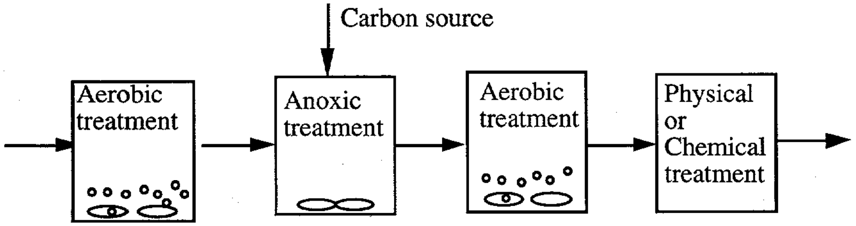

Figure 11 A strategy for the treatment of a methanogenic leachate.

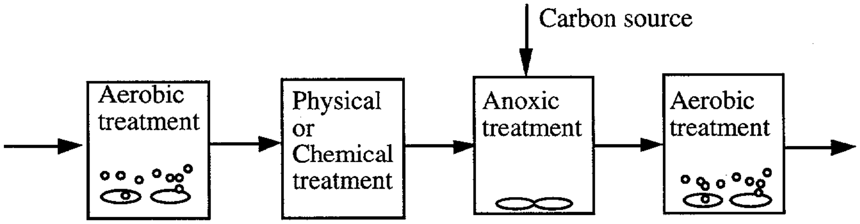

Figure 12 An alternative strategy for the treatment of a methanogenic leachate.

The results of the pilot study showed the suspended carrier biofilm process to be effective in nitrogen removal under realistic conditions, even when there is a fairly large variation both in temperature and in leachate composition. Evaluation of the process also showed it could reduce the toxicity of the leachate considerably but that some organic compounds were unaffected by treatment, such as various plasticizers and polycyclic aromatic compounds. One reason that these compounds were unaffected was probably their low concentration in the untreated leachate.

The study of different chemical and physical processes showed adsorption onto activated carbon to be the most efficient method of removing low-molecular-weight compounds. This indicates the importance of further evaluating the process, using other leachates. The results of the study also showed it to be possible to remove TOC from methanogenic leachates effectively through the combining of several processes, (such as nitrification, precipitation and adsorption onto activated carbon).

GC/MS analyses of untreated leachate, nitrified leachate and nitrified leachate treated with Fenton´s reagent or by activated carbon showed it to be very important to evaluate a treatment process thoroughly, using the specific leachate for which the process is intended. Neither oxidation by Fenton´s reagent nor adsorption onto activated carbon removed such plasticizers as phthaltes. This shows it to not be sufficient to evaluate a process for the removal of COD, BOD and TOC alone, since the composition of leachates from different landfills varies considerably with regard to specific organic compounds.

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POPULÄRVETENSKAPLIG SAMMANFATTNING

Ansamlingen av avfall i soptippar skapar miljöproblem då mikrobiell nedbrytning av lättnedbrytbart avfall ger upphov till växthusgaser (metangas och koldioxid). Dessutom bildas förorenade lakvatten på grund av att regnvatten tränger igenom deponin. Lakvattnet innehåller ofta höga halter av ammoniumjoner samt organiska ämnen. Ammoniumjonen är en giftig förening som ger akuta effekter på miljön medan många av de organiska ämnena kan ansamlas i olika organismer och ge negativa effekter på lång sikt. Problemen med många av de organiska föreningarna beror på att de är svåra för mikroorganismer att bryta ner. Ett skäl till detta är att dessa ämnen i många fall inte är naturligt förekommande, utan substanser som har tillverkats av människan. Detta innebär att mikroorganismerna inte har haft tillräckligt lång tid på sig för att anpassa sig till de organiska föreningarna. De höga koncentrationerna av ammoniumjoner och organiska ämnen medför att lakvattnet måste renas innan det släpps ut i naturen. I dag sker lakvattenreningen i många fall tillsammans med reningen av kommunalt avloppsvatten vid de kommunala reningsverken. En kombinerad rening är dock ingen optimal lösning då den kemiska sammansättningen hos kommunalt avloppsvatten och lakvatten skiljer sig markant. Dessa skillnader gör att det finns risk för att de svårnedbrytbara organiska ämnena i lakvattnet stör de biologiska reningsprocesserna vid det kommunala reningsverket och att reningsverket kan få problem med att uppnå de hårda krav som ställs på det renade vattnet med avseende på kväve. Dessutom finns det en risk att tillförseln av lakvatten till de kommunala reningsverken kan ge höga halter av tungmetaller i överskottsslammet, vilket då inte kan användas som gödselmedel på åkermark. Dessa faktorer gör att den kombinerade reningen av kommunalt avloppsvatten och lakvatten kommer att successivt avvecklas och att flera reningsanläggningar för behandling av lakvatten kommer att byggas i anslutning till soptipparna.

Reningsprocesser för lakvatten måste anpassas till speciella krav då den kemiska sammansättningen hos lakvattnet förändras med tiden och lakvattenflödet dessutom varierar kraftigt med årstiden. Den komplexa sammansättningen hos lakvattnet gör att behandlingen i många fall bör ske i en kombinerad process där mikrobiologiska och fysikaliska och/eller kemiska reningsmetoder kan samverka. Den mikrobiella reningen är i de flesta fall effektiv för avlägsnande av ammoniumjoner medan den fysikaliska/kemiska reningen behövs för att reningen skall bli effektiv med avseende på svårnedbrytbara organiska ämnen.

Mitt arbete omfattar en utvärdering av olika reningsmetoders effekt på både de svårnedbrytbara organiska ämnena och kvävet. Speciell vikt har lagts på de organiska föreningarna då det finns en risk att dessa glöms bort vid optimeringen av reningsprocesser på grund av att de inte ger tydliga synbara effekter på **kort** sikt. I mitt arbete har också ingått att analysera lakvatten från olika typer av kommunala deponier (en biocell där avfall med högt organiskt innehåll har deponerats, en askdeponi samt en "ordinär" blandad deponi för hushålls- och industriavfall) med avseende på specifika organiska föreningar.

Förhoppningarna är att resultaten som redovisas i min avhandling skall kunna vara till nytta då nya reningsanläggningar för lakvattenbehandling skall planeras och byggas.