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Present and Long-Term Composition of MSW Landfill Leachate: A Review

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ABSTRACT: The major potential environmental impacts related to landfill leachate are pollution of groundwater and surface waters. Landfill leachate contains pollutants that can be categorized into four groups (dissolved organic matter, inorganic macrocomponents, heavy metals, and xenobiotic organic compounds). Existing data show high leachate concentrations of all components in the early acid phase due to strong decomposition and leaching. In the long methanogenic phase a more stable leachate, with lower concentrations and a low BOD/COD-ratio, is observed. Generally, very low concentrations of heavy metals are observed. In contrast, the concentration of ammonia does not decrease, and often constitutes a major long-term pollutant in leachate. A broad range of xenobiotic organic compounds is observed in landfill leachate. The long-term behavior of landfills with respect to changes in oxidation-reduction status is discussed based on theory and model simulations. It seems that the somewhere postulated enhanced release of accumulated heavy metals would not take place within the time frames of thousands of years. This is supported by a few laboratory investigations. The existing data and model evaluations indicate that the xenobiotic organic compounds in most cases do not constitute a major long-term problem. This may suggest that ammonia will be of most concern in the long run.

KEY WORDS: waste disposal, xenobiotic organic compound, ammonia, dissolved organic matter, heavy metals, toxicological testing.

I. INTRODUCTION

The burial of municipal solid waste in landfills is the most common disposal alternative in most countries. Landfill leachate is generated by excess rainwater percolating through the waste layers in a landfill. A combination of physical, chemical, and microbial processes in the waste transfer pollutants from the waste material to the percolating water (e.g., Christensen and Kjeldsen, 1989). The biodegradability of organic content in the MSW and the compaction of the waste layers make the landfill an anaerobic environment, giving many similarities to the composition of leachates among different landfills.

Focusing on the most common type of landfill that receives a mixture of municipal, commercial, and mixed industrial waste, but excludes significant amounts

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of concentrated specific chemical waste, landfill leachate may be characterized as a water-based solution of four groups of pollutants (dissolved organic matter, inorganic macro components, heavy metals, and xenobiotic organic compounds [Christensen et al., 1994]).

The major potential environmental impacts related to landfill leachate are pollution of groundwater and surface water. The risk of groundwater the pollution is probably the most severe environmental impact from landfills because historically most landfills were built without engineered liners and leachate collection systems. More recently, regulations in many countries have required the installation of liners and leachate collection systems as well as a plan for leachate treatment. Christensen et al. (1994 and 2001) reviewed the characteristics of leachate plumes downgradient of landfills. Surface water pollution caused by leachate has also been observed, although relatively few cases have been described in the literature. The major potential effects of a leachate release to surface water are expected to be oxygen depletion in part of the surface waterbody, changes in the stream bottom fauna and flora and ammonia toxicity.

When refuse is buried in a landfill, a complex series of biological and chemical reactions occur as the refuse decomposes. Generally, it is accepted that landfills undergo at least four phases of decomposition, (1) an initial aerobic phase, (2) an anaerobic acid phase, (3) an initial methanogenic phase, and (4) a stable methanogenic phase (Christensen and Kjeldsen, 1995). Recently, an additional aerobic or humic phase of decomposition has been proposed (Christensen and Kjeldsen, 1995; Bozkurt et al., 2000). Once the refuse is very well decomposed, the rate of oxygen diffusion into the landfill may exceed the rate of microbial oxygen depletion. Thus, over time the anaerobic landfill is hypothesized to become an aerobic ecosystem. As refuse is buried in landfills over many years in a series of cells and lifts, it is quite common for different parts of the landfill to be in different phases of decomposition. As described below, there is a strong relationship between the state of refuse decomposition and its associated leachate characteristics. Thus, leachate composition can vary throughout a landfill. More detail on each phase of refuse decomposition is presented in Sections II and IV.

An understanding of leachate composition is critical for making projections on the long-term impacts of landfills. Even after a landfill stops accepting waste and a final cover is placed over the landfill, the refuse will continue to decompose. While leachate production decreases significantly with placement of the final cover, there is little data on leachate production over long periods of time. Furthermore, in assessing the long-term stability of a landfill, the possibility that the integrity of the landfill cover will decrease must be considered. Should the cover integrity deteriorate, the quantity of leachate could actually increase long after landfill closure.

In the U.S., the regulatory structure for landfills specifies a 30-year post-closure monitoring period during which the landfill is monitored. It is presumed that at the end of the 30-year period, the landfill will be stable and will no longer require intensive monitoring. However, there is little information on the long-term behavior of lined landfills and 30 years may be an insufficient long-term monitoring period. In developing assessment methods to determine whether post-closure monitoring can be terminated, it is important to understand the potential future composition of the leachate, should it be released to the environment at some time in the future.

The objective of this article is to give an overview of the composition of leachate from landfills receiving municipal solid waste. This overview is based on published reports that represent landfills that are on the order of 40 years old and younger. A second objective is to evaluate the expected long-term composition of leachate once the refuse is completely degraded. This is done by reviewing models and laboratory experiments addressing stabilization processes in landfills. Our focus is on landfills that are operated under anaerobic conditions and that receive a mixture of municipal and nonhazardous commercial and industrial waste. The composition of leachate from landfills that are maintained under aerobic conditions, as well as landfills that receive incineration residue and other inorganic wastes will be considerably different and is not addressed here.

A description of the first four phases of refuse decomposition is presented in the following section. This is followed by presentation of information on the composition of leachate based on our current knowledge. Leachate composition is considered in four categories: (1) dissolved organic matter, (2) inorganic macro components, (3) heavy metals, and (4) xenobiotic organic compounds. The final section of this article evaluates the potential for changes in leachate composition over very long (geologic) periods of time.

II. STABILIZATION PROCESSES IN LANDFILLS — AN OVERVIEW

Landfills have been controlled and monitored for about 30 years. Throughout this period an increasing understanding of the complex series of chemical and biological reactions that initiates with the burial of refuse in a landfill has been developed. Figure 1 shows the gas and leachate composition as refuse decomposes. The figure is developed from the first description of the landfill phases given by Farquhar and Rovers (1973). The first four phases shown in the figure are referred to as the aerobic phase, the anaerobic acid phase, the initial methanogenic phase, and the stable methanogenic phase. Subsequent phases of decomposition, in which the waste cell begins to turn aerobic are based on theory and are somewhat speculative because no field data are available to document the onset of aerobic conditions (Christensen and Kjeldsen, 1995). This is due to the fact that most well-monitored landfills are less than 30 years old and are still in the stable methanogenic phase.

During the initial aerobic phase, oxygen present in the void spaces of the freshly buried refuse is rapidly consumed, resulting in the production of CO_2 and

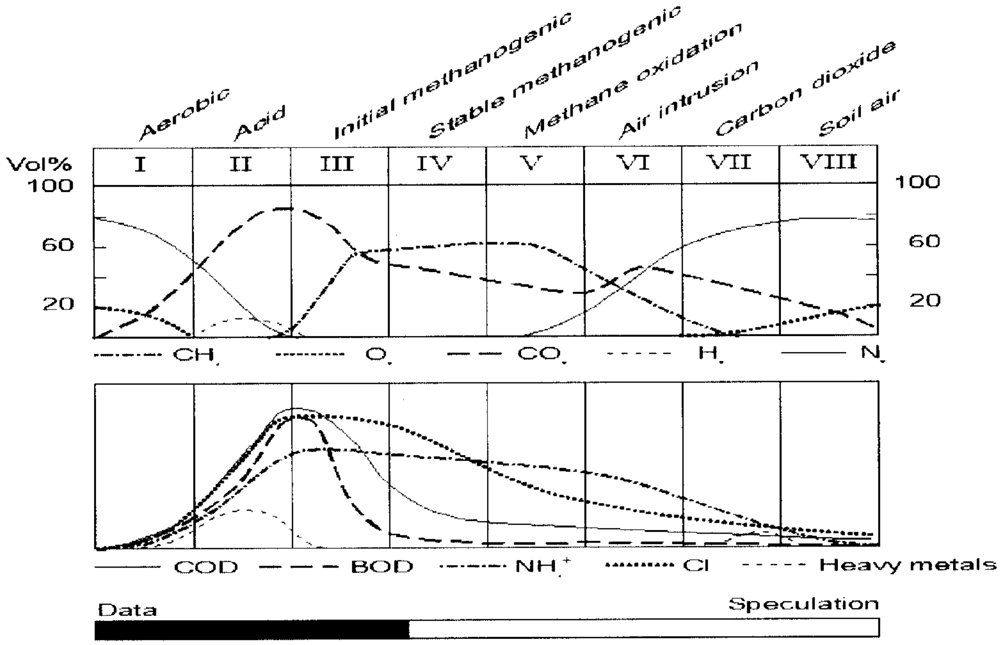

FIGURE 1. The lifetime of a landfill showing general trends in gas and leachate quality development.

maybe an increase in waste temperature. The aerobic phase in a landfill lasts only a few days because oxygen is not replenished once the waste is covered. During the aerobic phase, the waste is not typically at field capacity (Barlaz and Ham, 1993). Most leachate produced during this phase results from the release of moisture during compaction as well as short-circuiting of precipitation through the buried refuse.

As oxygen sources are depleted, the waste becomes anaerobic, which supports fermentation reactions. Cellulose and hemicellulose comprise 45 to 60% of the dry weight of MSW and are its major biodegradable constituents (Barlaz et al., 1989b). The decomposition of these compounds to methane and carbon dioxide in landfills under anaerobic conditions is well documented (Barlaz et al., 1990; Pohland and Harper, 1986; Bookter and Ham, 1982). Cellulose and hemicellulose biodegradation is carried out by three groups of bacteria: (1) the hydrolytic and fermentative bacteria that hydrolyze polymers and ferment the resulting monosaccharides to carboxylic acids and alcohols; (2) the acetogenic bacteria that convert these acids and alcohols to acetate, hydrogen, and carbon dioxide; and (3) the methanogens that convert the endproducts of the acetogenic reactions to methane and carbon dioxide (Zehnder, 1982). This process proceeds efficiently over a relatively narrow pH range around neutral. In the second phase the hydrolytic, fermentative, and acetogenic bacteria dominate, resulting in an accumulation of carboxylic acids, and a pH decrease. The highest BOD and COD concentrations in the leachate will be measured during this phase (Barlaz and Ham, 1993; Reinhart and Grosh, 1998). The BOD:COD ratio in the acid phase has been reported to be above 0.4 (Ehrig, 1988) or 0.7 (Robinson, 1995). As the pH is acidic, acid phase leachate is chemically aggressive and will increase the solubility of many compounds.

The onset of the initial methanogenic phase (3) occurs when measurable quantities of methane are produced. The onset of this phase is likely associated with the pH of the refuse becoming sufficiently neutralized for at least limited growth of methanogenic bacteria. During this phase the acids that accumulated in the acid phase are converted to methane and carbon dioxide by methanogenic bacteria, and the methane production rate will increase (Christensen and Kjeldsen, 1989, Barlaz et al., 1989a). Cellulose and hemicellulose decomposition also begins. COD and BOD concentrations begin to decrease and the pH increases as acids are consumed. The BOD to COD ratios will also decrease as carboxylic acids are consumed.

In the stable methanogenic phase, the methane production rate will reach its maximum, and decrease thereafter as the pool of soluble substrate (carboxylic acids) decreases. In this phase, the rate of CH_4 production is dependent on the rate of cellulose and hemicellulose hydrolysis. The pH continues to increase to steady-state pool concentrations that are on the order of a few mg/L. Some COD is present in the leachate, but it is mostly recalcitrant compounds such as humic and fulvic acids (Barlaz and Ham, 1993; Christensen et al., 1994). As discussed in the following section, the BOD:COD ratio generally will fall below 0.1 in this phase because carboxylic acids are consumed as rapidly as they are produced.

The four phases of refuse decomposition described above have been defined on the basis of both field and laboratory-scale data that have been summarized in earlier reviews (see Barlaz et al., 1990). However, environmental conditions in the landfill will have a significant impact on the rate of refuse decomposition, and subsequently the time required for decomposition to proceed to the point where methane production decreases to zero. Studies on the effect of a number of factors on refuse decomposition have been summarized (Barlaz et al., 1990; Christensen et al., 1992). The factor that has most consistently been shown to affect the rate of refuse decomposition is the moisture content, and it is generally accepted that refuse buried in arid climates decomposes more slowly than refuse buried in regions that receive greater than 50 to 100 cm of annual infiltration into the waste. Refuse decomposition can also be accelerated during the operational phase of the landfill. The most common enhancement technique is the use of leachate recycle, whereby leachate is recirculated through the refuse as opposed to it being treated and released to the environment. By recirculating leachate, the refuse moisture content is increased from its initial value, which is typically 15 to 25% (wet weight basis) to 40 to 50%. In addition, leachate recirculation results in better distribtion of nutrients, substrates, and bacteria. Other factors that can be used to accelerate decomposition include shredding and an initial aeration step in which the refuse is aerated for a period of 1 to 2 months after burial to increase the temperature and allow for the aerobic biodegradation of the initial accumulation of soluble organic carbon (Komilis et al., 1999a,b). However, field experience with shredding and aeration are considerably more limited.

Wheras the authors are not familiar with any landfills that have progressed beyond the stable methanogenic phase, in theory refuse will continue to decompose until no more degradation occurs and the landfill becomes aerobic. This process can be described as a series of four phases in which the methane production rate continues to decrease to a point at which air begins to infiltrate into the waste cell (Christensen and Kjeldsen, 1995). The four phases are discussed in detail in Section IV on long-term landfill stabilization.

III. PRESENT MSW LANDFILL LEACHATE COMPOSITION

This section presents data on the composition of landfill leachate, and further discuss the importance of the landfill phases presented in the previous section on compositional changes. Pollutants in MSW landfill leachate can be divided into four groups:

- Dissolved organic matter, quantified as Chemical Oxygen Demand (COD) or Total Organic Carbon (TOC), volatile fatty acids (that accumulate during the acid phase of the waste stabilization, Christensen and Kjeldsen, 1989) and more refractory compounds such as fulvic-like and humic-like compounds.

- Inorganic macrocomponents: calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}), potassium (K^{+}), ammonium (NH_4^{+}), iron (Fe^{2+}), manganese (Mn^{2+}), chloride (Cl^{-}), sulfate (SO_4^{2-}) and hydrogen carbonate (HCO^{3-}).

- Heavy metals: cadmium (Cd^{2+}), chromium (Cr^{3+}), copper (Cu^{2+}), lead (Pb^{2+}), nickel (Ni^{2+}) and zinc (Zn^{2+}).

- Xenobiotic organic compounds (XOCs) originating from household or industrial chemicals and present in relatively low concentrations (usually less than 1 mg/l of individual compounds). These compounds include among others a variety of aromatic hydrocarbons, phenols, chlorinated aliphatics, pesticides, and plastizers.

Other compounds may be found in leachate from landfills: for example, borate, sulfide, arsenate, selenate, barium, lithium, mercury, and cobalt. However, in general, these compounds are found in very low concentrations and are only of secondary importance. Leachate composition may also be characterized by different toxicological tests, which provide indirect information on the content of pollutants that may be harmful to a class of organisms.

A. How Representative Are Leachate Samples?

Many investigations on leachate composition have been restricted to one or a few leachate samples from each landfill. Depending on the study objectives, this may suffice if the leachate collection system averages the leachate from different sections of the landfill and the leachate is pumped out of the landfill for treatment. However, a landfill contains areas of refuse of varying ages and states of decomposition. Thus, where leachate from older methanogenic refuse is mixed with leachate from fresher refuse in the acid phase, it is not possible to relate leachate composition to processes within the waste layers. Furthermore, where leachate from refuse in the acid phase percolates through well-decomposed refuse, the leachate can be expected to reflect the composition of methanogenic leachate. This is because the high COD of the acid phase leachate will be consumed as the leachate passes through the well decomposed, and thus carbon limited refuse. In cases where leachate is released to groundwater, such as in the case of older landfills that are not lined, the spatial distribution of the leachate quality is especially important to evaluate the leaching to the underlying strata. This, however, require a large number of sampling points (Kjeldsen et al., 1998; Assmuth, 1992).

Leachate composition varies significantly among landfills depending on waste composition, waste age, and landfilling technology. Leachate sampling methods and sample handling routines may also influence the measured leachate quality. For example, colloids have a high affinity for heavy metals (Gounaris et al., 1993), thus the concentration of heavy metals measured in a leachate sample may depend strongly on the amount of colloidal matter present in the sample and the handling of the sample (see also the section on heavy metals). No standard protocols for sampling, filtration, and storage of leachate samples exist. The content of colloidal matter in a sample depends to a large extent on the sampling technique used (e.g., McCarthy and Zachara, 1989; Backhus et al., 1993). Where samples are obtained from groundwater monitoring wells, a high pumping rate will increase the colloid content of the sample significantly (Backhus et al., 1993), and the heavy metal concentration may also be increased. Therefore, leachate samples should be filtered in the field before analysis of heavy metals, especially if the sampling is done quickly. Alternatively, samples could be withdrawn under very low pumping rates and after sufficient purging of the well. Sampling and sample handling may explain part of the variation seen among landfills with respect to the heavy metal concentration in leachates. Maintaining leachate samples under anaerobic conditions until they are preserved is also important because metal solubility varies with their oxidation stage.

Leachate analysis for XOCs may be biased by loss of pollutants due to volatilization or sorption to the sampling equipment. However, recent studies have shown that the quality of leachate analysis for XOCs is seriously jeopardized only if very poor sampling protocols are used (see review by Parker, 1994).

B. General Leachate Composition

Table 1 presents ranges of general leachate parameters from various reports. The table is based mainly on data originating from newer landfills. Data from older uncontrolled landfills may exhibit lower values than the minimum values given in the table (Assmuth and Strandberg, 1993; Kjeldsen and Christophersen, 2001). In general, landfill leachates may contain very high concentrations of dissolved organic matter and inorganic macrocomponents. The concentrations of these components may typically be up to a factor 1000 to 5000 higher than concentrations found in groundwater.

Several parameters change dramatically as the landfill stabilizes. During the acid phase, the leachate may show low pH values and high concentrations of many compounds, in particular high concentrations of easily degradable organic compounds as volatile fatty acids. In the later stable methanogenic phase, the pH increases and the BOD5/COD ratio (biological oxygen demand measured over 5 days divided by chemical oxygen demand), reflecting the degradability of the organic carbon is lowered dramatically (Ehrig, 1988). The increased pH also affects many of the inorganic parameters as illustrated in Table 2, where data are presented for acid phase leachate and methanogenic leachate. For parameters not significantly affected by landfill stabilization, only average values are given.

Besides the time dependency in relation to change from acid to methanogenic phase, short-term variations in leachate quality are also expected. Seasonal variations in leachate composition have been observed in several cases. Åkesson and Nilsson (1997) observed lower leachate concentrations in the wet season in a Swedish landfill test cell. Similar observations were found by Chu et al. (1994) in a Hong Kong landfill.

C. Dissolved Organic Matter

As described previously, several bulk parameters are used to describe the content of dissolved organic matter in leachate; TOC (Total Organic Carbon), COD (Chemical Oxygen Demand), and BOD (Biological Oxygen Demand). Table 3 gives further details on observed BOD and COD values and the ratio BOD/COD in leachates from older landfills that are well into their methane phase. The table shows that the BOD/COD ratio in most cases is below 0.10 for methanogenic leachates.

One source of variability of the COD measurement that may have some effect on the values presented in Table 1 is the presence of inorganic constituents that may contribute to COD. For instance, Kylefors et al. (1999) found that Fe(II), Mn(II), and sulfide contributed up to one-third of the COD in the leachates they tested. Chloride may also contribute to COD if not accomodated for in the procedure. In addition, poor sampling methods that expose anaerobic leachate to oxygen

TABLE 1
Composition of Landfill Leachate* (Values in mg/l unless otherwise stated.)

Parameter	Range
pH	4.5-9
Spec. Cond. (μS cm^{-1})	2500-35000
Total Solids	2000-60000
Organic Matter	
Total Organic Carbon (TOC)	30-29000
Biological Oxygen Demand (BOD_5)	20-57000
Chemical Oxygen Demand (COD)	140-152000
BOD_5/COD (ratio)	0.02-0.80
Organic nitrogen	14-2500
Inorganic macrocomponents	
Total phosphorous	0.1-23
Chloride	150-4500
Sulphate	8-7750
Hydrogenbicarbonate	610-7320
Sodium	70-7700
Potassium	50-3700
Ammonium-N	50-2200
Calcium	10-7200
Magnesium	30-15000
Iron	3-5500
Manganese	0.03-1400
Silica	4-70[a]
Heavy metals	
Arsenic	0.01-1
Cadmium	0.0001-0.4
Chromium	0.02-1.5
Cobolt	0.005-1.5
Copper	0.005-10
Lead	0.001-5
Mercury	0.00005-0.16
Nickel	0.015-13
Zinc	0.03-1000

*The ranges are based on Andreottola and Cannas (1992), Chu et al.(1994), Robinson (1995), Ehrig (1980), Ehrig (1983), Ehrig (1988), Garland and Mosher (1975), Johansen and Carlson (1976), Karstensen (1989), Krug and Ham(1997), Lu et al. (1985), Naturvårdsverket (1989), Owen and Manning (1997), and Robinson and Maris (1979).

[a] Values based on Owen and Manning (1997).

TABLE 2
Leachate Composition in Terms of Average Values and Ranges for Parameters with Differences between Acid and Methanogenic Phase (Ehrig, 1988) and Average Values for Parameters with No Observed Differences between Acid and Methanogenic Phase (Ehrig, 1983) (All values in mg/l except pH and BOD5/COD)

Parameter	Acid phase		Methanogenic phase		Average
	Average	Range	Average	Range	
pH	6.1	4.5-7.5	8	7.5-9	
Biological Oxygen Demand (BOD_5)	13000	4000-40000	180	20-550	
Chemical Oxygen Demand (COD)	22000	6000-60000	3000	500-4500	
BOD_5/COD (ratio)	0.58		0.06		
Sulfate	500	70-1750	80	10-420	
Calcium	1200	10-2500	60	20-600	
Magnesium	470	50-1150	180	40-350	
Iron	780	20-2100	15	3-280	
Manganese	25	0.3-65	0.7	0.03-45	
Ammonia-N					740
Chloride					2120
Potassium					1085
Sodium					1340
Total phosphorus					6
Cadmium					0.005
Chromium					0.28
Cobalt					0.05
Copper					0.065
Lead					0.09
Nickel					0.17
Zinc	5	0.1-120	0.6	0.03-4	

may cause Fe(II) to oxidize to Fe(III) and precipitate out of the leachate. This would result in a decreased COD relative to a study in which Fe(II) was oxidized as part of the COD analysis. This would occur when the sample was maintained under anaerobic conditions until after filtration, at which point it could be acidified to reduce iron oxidation.

Dissolved organic matter in leachate is a bulk parameter covering a variety of organic degradation products ranging from small volatile acids to refractory fulvic and humic-like compounds (Chian and DeWalle, 1977). Dissolved organic matter

TABLE 3
Observed Values of BOD, COD and BOD/COD-Ratio for Landfill Leachates Samples from Landfills in the Methanogenic Phase

BOD (mg/l)	COD (mg/l)	BOD/COD	Reference
5.7-1100	76-6997	—	Concentration ranges from 21-30 year old German landfills (Krumpelbeck and Ehrig, 1999)
290	1225	0.24	Average concentrations from 21-30 year old German landfills (Krumpelbeck and Ehrig, 1999)
44	320	0.11	Average concentrations from old, Danish landfills (Kjeldsen and Christophersen, 2001)
39	398	0.10	Composite results at Sandsfarm Landfill, (Robinson, 1995)
11	190	0.06	Composite results at Bishop Middleham Landfill, (Robinson, 1995)
38	517	0.07	Composite results at Odsal Wood Landfill, (Robinson, 1995)
1.0	53	0.02	Composite results at East Park Drive Landfill, (Robinson, 1995)
2.5	64	0.04	Composite results at Marton Mere Landfill, (Robinson, 1995)
180	3000	0.06	Average concentrations in methanogenic leachate (Ehrig, 1988)

can affect leachate composition in relation to other constituents through the complexing properties of the high-molecular-weight component of the dissolved organic matter. Unfortunately, we usually have very little information on the composition of the dissolved organic matter in landfill leachate. At the most general level, a low BOD/COD ratio suggests a leachate with low concentrations of volatile fatty acids and relatively higher amounts of humic and fulvic-like compounds. A few investigations concerning DOC in landfill leachate are available.

Harmsen (1983) analyzed an acid-phase leachate and a methanogenic-phase leachate. In the acid-phase leachate, more than 95% of the DOC content of 20,000 mg/l consisted of volatile fatty acids and only 1.3% of the DOC consisted of high-molecular-weight (MW) compounds (MW>1000). In addition, volatile amines and alcohols were detected. In the methanogenic-phase leachate; however, no volatile acids, amines, or alcohols were detected, and 32% of the DOC (2100 mg/l) consisted of higher-molecular-weight compounds (MW>1000). Also in a methanogenic-phase leachate, Artiola-Fortuny and Fuller (1982) described more than 60% of the DOC content as humic-like material. Investigating anaerobic and aerobic leachates and leachates that have passed a model aquifer, Frimmel and Weis (1991) found that only 6 to 30% of the DOC could be described as fulvic acids.

More detailed characterization of the DOC in leachate and leachate polluted groundwater is based on isolation and purification of the DOC. These procedures may potentially change the properties of the organic matter because it is necessary to purify the material to obtain results. Weis et al. (1989) compared fulvic acids from landfill leachates with those from soil and bog lake water. The fulvic acids isolated from landfill leachate had higher carbon, hydrogen, and sulfur content, lower quantities of phenolic groups, lower complexation capacities for copper and lower molecular weight. Christensen et al. (1998a) characterized 82% of the DOC in leachate-polluted groundwater sampled less than 10 m downgradient from the Vejen Landfill (DK) and found 49% fulvic acids, 8% humic acids, and 25% hydrophilic fraction. Based on molecular weight, elemental composition, and acidity, the fulvic acid fraction and the hydrophilic fraction resembled fulvic acids from other origins, whereas the humic acid had rather low molecular weight. This analysis showed that the three fractions of DOC were rather similar with respect to the features of importance to complexation of metals.

D. Inorganic Macrocomponents

The concentrations of some inorganic macrocomponents in leachate depend, as in the case of the dissolved organic matter, on the stabilization of the landfill. Table 2 shows that the cations calcium, magnesium, iron, and manganese are lower in methanogenic phase leachate due to a higher pH (enhancing sorption and precipitation) and lower dissolved organic matter content, which may form complexes with the cations. Sulfate concentrations are also lower in the methanogenic phase due to microbial reduction of sulfate to sulfide.

Table 2 also presents average concentrations for parameters with no observed difference between acid and methanogenic phase. These are the macrocomponents chloride, sodium, and potassium for which the effects of sorption, complexation, and precipitation are minor. Decreasing trends in concentration with time of these pollutants could be due to wash out by the leaching, although Ehrig (1983, 1988) did not observe any decrease in concentration for these parameters after up to 20 years of leaching.

Table 2 is based on detailed studies (Ehrig 1983, 1988) on a large number of landfills in Germany. Similar findings are presented in a study of 13 sanitary landfills in Wisconsin, USA (Krug and Ham, 1997), where equivalent concentration ranges and time dependency of the selected parameters were found.

Many investigations report concentrations of ammonia-nitrogen in the range of 500 to 2000 mg/l, and no decreasing trend in concentration with time. Ammonia is released from the waste mainly by decomposition of proteins. The only mechanism by which the ammonia concentration can decrease during refuse decomposition is leaching because there is no mechanism for its degradation under methanogenic conditions (Robinson, 1995; Burton and Watson-Craik, 1998). For this reason, several researchers have identified ammonia as the most significant component of leachate for the long term (Robinson, 1995; Krumpelbeck and Ehrig, 1999; Christensen et al., 1994; Christensen et al., 1999).

In a study of 50 German landfills, ammonia concentrations did not show a significant decrease even 30 years after landfill closure (Krumpelbeck and Ehrig, 1999). Ehrig (1988) reports that there is no significant change in ammonia concentrations from the acidic to methanogenic phase, and that the average value is 740 mg-N/L (Table 2). Ammonia concentration data from several studies is given in Table 4. These data document that ammonia concentrations will remain high even in leachate from older landfills that is otherwise low in organic content.

E. Heavy Metals

There is wide variation in the reported concentrations of heavy metals from different landfills (Table 1). However, average metal concentrations are fairly low. This has been shown by several studies in which researchers have reported metals concentrations from full-scale landfills, test cells, and laboratory studies. The ultimate conclusion of all of these studies is that heavy metals in landfill leachate at present are not at major concern (Christensen et al., 1999; Robinson, 1995; Reinhart and Grosh, 1999; Revans et al., 1999; Kjeldsen and Christophersen, 2001; Christensen et al., 1994). Table 5 gives more details of observed heavy metal concentrations. For comparison, the U.S. Drinking Water Standards are presented as well. The table shows that most heavy metal concentrations in landfill leachate are at or below the US drinking water standards.

TABLE 4
Ammonia Concentrations in Landfill Leachate (All values are from older landfills in the methanogenic phase)

Ammonia-N (mg/l)	Reference
110	Average ammonia concentration from 104 old, Danish landfills (Kjeldsen and Christophersen, 2001)
233	Composite results at Sandsfarm Landfill, (Robinson, 1995)
282	Composite results at Bishop Middleham Landfill, (Robinson, 1995)
399	Composite results at Odsal Wood Landfill, (Robinson, 1995)
43	Composition results at East Park Drive Landfill, (Robinson, 1995)
30	Composition results at Marton Mere Landfill, (Robinson, 1995)
12-1571	Range of concentrations from 21-30 year old, German landfills (Krumpelbeck and Ehrig, 1999)
445	Average concentration from 21-30 year old, German landfills (Krumpelbeck and Ehrig, 1999)
740	Average concentration (Ehrig, 1988)

TABLE 5
Heavy Metal Concentrations in Leachate[a]

Reference	1	2	3	4	5	6	7	8	9	10	US Stand.[b]
Metal											
Cd	0.006	0.005	0.006	0.0002	0.0004	0.0003	0.0036	0.002 - 0.008	0.0002 - 0.018	< 0.01 - < 0.04	0.005
Ni	0.130	0.17	0.05	0.028	0.084	0.054	0.062	0.01 - 0.08	0.0036 - 0.348	< 0.01 - 0.1	---
Zn	0.67	0.6	2.2	0.2	0.36	0.085	5.31	0.003 - 0.011	0.05 - 9	< 0.01 - 0.47	5.0
Cu	0.07	0.065	0.04	0.002	0.007	0.034	0.002	---	0.004 - 0.27	< 0.02 - 0.17	1.3
Pb	0.07	0.09	0.02	< 0.005	< 0.005	0.056	0.188	0.016 - 0.067	0.005 - 0.019	< 0.04 - 0.13	0.0
Cr	0.08	0.28	0.01	0.003	0.016	0	0.002	0.033 - 0.085	0.005 - 1.62	< 0.01 - 0.05	0.1

1. Average (undiluted) leachate concentration from 106 old, Danish landfills (Kjeldsen and Christophersen, 2001).
2. Average leachate concentration in 20 German landfills in the methanogenic phase (Christensen et al., 1999).
3. Average leachate concentration in a full-scale test cell operated with leachate recirculation (Flyhammar et al., 1998).
4-7. Average leachate concentrations at four Danish landfills. Only site 4 has been closed (Jensen and Christensen, 1999).
8. Ranges of leachate concentrations in the most contaminated groundwater wells at North Bay Landfill, Canada (Christensen et al., 1999).
9. Ranges of typical leachate concentrations in a study of 21-30 year old, German landfills (Krumpelbeck and Ehrig, 1999).
10. Ranges of typical leachate concentrations at six old landfills in the UK (Robinson, 1995).
--- Data not available

a. Data listed are either the average or range for a given study in mg/l.

b. National primary drinking water regulations, USA (http://www.epa.gov/OGWDW/wot/appa.html).

1. Heavy Metal Attenuation Processes in Landfills

The reason for the low concentrations of heavy metals in methanogenic leachates is not a lack of heavy metals present in the waste. Heavy metal balances for landfills have shown that less than 0.02% of heavy metals received at landfills are leached from the landfill after 30 years (Flyhammer, 1995; Aulin and Neretnieks, 1995; Belevi and Baccini, 1989). Both sorption and precipitation are believed to be significant mechanisms for metals immobilization and the subsequent low leachate concentrations. Waste contains soils and organic matter, which, especially at the neutral to high pH values prevailing in methanogenic leachate, has a significant sorptive capacity (Bozkurt, 1999). In addition, the solubilities of many metals with both sulfides and carbonates is low, and these anions are typical in landfills. Sulfide is formed from sulfate reduction during waste decomposition in landfills, and sulfide precipitation is often cited as an explanation for low concentrations of heavy metals (Christensen et al., 2000; Christensen et al., 1994). Sulfides and carbonates are capable of forming precipitates with Cd, Ni, Zn, Cu, and Pb. While carbonates are abundant in landfill leachate, the solubilities of metal carbonates are generally higher than that of metal sulfides (Christensen et al., 2000). In general, sulfide precipitation is expected to dominate heavy metal attenuation compared with complexation agents (Reinhart and Grosh, 1999). Cr is an exception to this because it does not form an insoluble sulfide precipitate (Christensen et al., 2000). However, Cr tends to form insoluble precipitates with hydroxide (Christensen et al., 1999; Revans et al., 1999).

Investigations of the sulfur content of landfilled waste have shown that the waste does not contain sufficient sulfur to bind all the heavy metals present in the waste. The landfill sampled by Martensson et al. (1999) contained only enough sulfur to bind 5% of metals present. Occasionally, phosphates and hydroxides will also precipitate metals (Christensen et al., 1999). Hydroxide precipitates form at pHs at or above neutral, which is typically the case in methanogenic leachates (Reinhart and Grosh, 1998).

2. Heavy Metal Mobilizing Processes in Landfills

Several processes, including complexation to inorganic and organic ligands, and sorption to colloids are capable of mobilizing heavy metals by increasing the concentration in the mobile aqueous phase. Jensen and Christensen (1999) separated leachate samples from four Danish landfills into size fractions to obtain information about size distribution of colloids and associated heavy metals (Cd, Ni, Zn, Cu, Pb, Cr). A significant but highly varying fraction of the heavy metals was associated with colloidal fractions. Similarly, Gounaris et al. (1993) reported that in an American landfill a significant fraction of the Zn, Pb, and Cr were in colloidal fractions. Klein and Niessner (1998) found at a German landfill that the main fraction of the heavy metals was associated with the colloidal matter, primarily the 0.001 to 0.01 μm fraction, which is dominated by humic material. In all three investigations, comparison of the distribution of organic matter and heavy metals between the size fractions indicated that the heavy metals in the colloidal fractions were not only related to organic matter, even though the colloidal humic substances are suspected to play a major role with respect to the speciation of the heavy metals.

Different physical separation techniques have been used to divide the heavy metals in landfill leachates into different types of species. Lun and Christensen (1989) determined the distribution between different cadmium species in landfill leachate by a resin method and found that free divalent Cd^{2+} only made up a few percent of the total cadmium content. Most of the complexed fraction was characterized as labile complexes that easily could be redistributed to other dissolved species. However, a small fraction (5 to 15%) was characterized as stable soluble complexes, defined as lack of ability to exchange with a cation exchange resin. The stable complexes were considered to be organic. In leachate from three landfills containing industrial waste, Holm et al. (1995b) found by use of dialysis, ion exchange, and thermodynamic calculations, large variation with respect to the speciation of Cd. In two of the investigated leachates about 20% of the total Cd content was determined to be Cd^{2+}, while most of the Cd was identified as chloride complexes. The third leachate had a high dissolved organic carbon content (3200 mg C/l), and most of the Cd in this leachate was complexed with the organic matter. Knox and Jones (1979) showed that both low-molecular-weight compounds (<500) comparable to simple carboxylic acids and high-molecular-weight compounds (>10000) contributed significantly to cadmium complexation. Bolton and Evans (1991) performed some speciation calculations on four leachates. Their calculations showed that 38% of Cd was present in complexes with organic ligands and 32% with inorganic ligands, while Zn and Ni were present in complexes (36% and 68%, respectively) exclusively with organic ligands.

F. Xenobiotic Organic Compounds (XOCs)

Table 6 presents concentration ranges of some xenobiotic organic compounds (XOCs) found in landfill leachate. The table is based mostly on observations from landfills containing municipal solid waste. The amount of hazardous waste that has been allowed into MSW landfills has decreased significantly over the last 20 years. However, many of the landfills for which data are reported in Table 6 may contain waste from a time period when there were fewer restrictions on the disposal of hazardous waste in MSW landfills. Very broad ranges are observed, reflecting differences in waste composition, landfill technologies, and waste age.

The most frequently found XOCs are the monoaromatic hydrocarbons (benzene, toluene, ethylbenzene, and xylenes) and halogenated hydrocarbons such as tetrachloroethylene and trichloroethylene (Table 6). These pollutants are also the

TABLE 6
Xenobiotic Organic Compounds (XOCs) Observed in Landfill Leachates

Compound	Range (µg/l)	References[5]
Aromatic hydrocarbons		
Benzene	0.2-1630	a.b.d.f.h.i.k.l.m.n.o.p.q. t. x
Toluene	1-12300	a.b.d.f.h.i.k.l.m.n.o.p.q. t. x
Xylenes	0.8-3500	a.b.d.f.h.i.k.l.m.n.o.p.q. t. x
Ethylbenzene	0.22329	a.b.d.f.m.n.o.p.q. t. x. v
Trimethylbenzenes	0.3-250	b.f.o.p. t. x
n-Propylbenzene	0.3-16	T, x
t-Butylbenzene	2.1-21	T
o-Ethyltoluene	0.5-46	t
m-Ethyltoluene	0.3-21	T
p-Ethyltoluene	0.2-10	T
Naphthalene	0.1-260	c.d.f.m.n.o.p. t. x
Halogenated hydrocarbons		
Chlorobenzene	0.1-110	a.d.f.m.o. t
1.2-Dichlorobenzene	0.1-32	a.c.d.f.o. t
1.3-Dichlorobenzene	5.4-19	Y
1.4-Dichlorobenzene	0.1-16	a.c.d.f.m. t
1,2,3-Trichlorobenzene	I	Y
1,2,4-Trichlorobenzene	4.3	Y
Hexachlorobenzene	0.025-10	Z
1.1-Dichloroethane	0.6-46	a.i. t. v
1.2-Dichloroethane	<6	a.b.d
1.1.1-Trichloroethane	0.01-3810	a.b.d.f.m.o.p.q. t. x
1.1.2-Trichloroethane	2.5-16	Y
1.1.2.2.-Tetrachloroethane	1	Y
trans-1.2-Dichloroethylene	1.6-6582	a.b. v
cis-1.2-Dichloroethylene	1.4-470	a.b. t
Trichloroethylene	0.05-750	a.b.d.f.l.m.n.o.p. t. x. v
Tetrachloroethylene	0.01-250	a.b.f.i.l.m.n.o.p.q. t. x
Dichloromethane	1.0-827	a.b.d.k.m. t. v
Trichloromethane	1.0-70	a.b.d.h.i.k.o.p.q.
Carbontetrachloride	4.0-9.0	h.o.p
Phenols		
Phenol	0.6-1200	c.f.g.k.m.n. x
Ethylphenols	<300	k.l
Cresols	1-2100	c.g.i.k.l.m.n. t. x
Bisphenol A	200-240	t
3.5-Dimethylphenol	0.7-27.3	x
2.5-Dimethylphenol	0.4-4.5	X
2.4-Dimethylphenol	0.1-12.5	x
3.4-Dimethylphenol	0.03-10.4	x
2.6-Dimethylphenol	0.3-1.9	X
2-Methoxyphenol	I^{4}	x
2/3-Chlorophenol	0.03-1.6	x
4-Chlorophenol	0.2-1.3	x
4-Chloro-m-cresol	1.2-10.2	x
3.5-Di-chlorophenol	0.08-0.63	x
2.3.4.6-Tetrachlorophenol	0.079-3.0	z

TABLE 6 (continued)

Alkylphenols		
Nonylphenol	6.3-7	x
Nonylphenolmonocarboxylate	0.5-3	x
Pesticides		
Ametryn	0.12	X
AMPA	3.8-4.3	X
Atrazin	0.16	X
Bentazon	0.3-4.0	X
Chloridazon	1.6	X
Chlorpropham	26	X
Dichlobenil	0.1-0.3	X
Fenpropimorf	0.1	X
Glyphosat	1.7-27	X
Hexazinon	1.3	X
Hydroxyatrazin	0.7-1.7	X
Hydroxysimazin	0.6-1.7	X
Isoproturon	1.2	x
Lindane	0.025-0.95	Z
Mecoprop[1]	0.38-150	c,e,l, u, x
MCPA	0.2-9.1	U, x
Propoxuron	2.6	X
Simazine	2.3	X
Tridimefon	2.1	X
4-CPP	15-19	X
2,4-D[2]	1.0-5	e,l
2,4,5-T	1[4]	U
2,4-DP	0.3-5.2	U, x
2,6-DCPP	0.7-1.3	X
Phthalates		
Monomethyl phthalate	1	X
Dimethyl phthalate	0.1-7.7	S
Diethyl phthalate	0.1-660	c,g,j,m, t, x
Methyl-ethyl phthalate	2-340	X
Mono-(2-ethylhexyl) phthalate	4-14	V, x
Di-(2-ethylhexyl) phthalate	0.6-235.9	s, t, u, v, x
Mono-butylphthalate	4-16	V, x
Di-n-butylphthalate	0.1-70 5.0-15	c,g,i,j,m, t
Di-isobutylphthalate	3-6	T
Mono-benzylphthalate	6-16	V, x
Butylbenzyl phthalate	0.2-8	c,g,j, v
Dioctylphthalate	1-6	T
Phthalic acid	2-14000	V, x
Aromatic sulfonates		
Naphtalene-1-sulfonate	506-616	r
Naphtalene-2-sulfonate	1143-1188	r
Naphtalene-1,5-disulfonate	<2.5-51	r
Naphtalene-1,6-disulfonate	366-397	r
Naphtalene-2,7-disulfonate	129-145	r
2-aminonaphtalene-4,8-disulfonate	73-109	r
p-toluenesulfonate	704-1084	r

TABLE 6 (continued)

Phosphonates		
Tri-n-butylphosphate	1.2-360	c.f.i.l.m
Triethylphosphate	15	f.i.l.
Miscellaneous		
Acetone	6-4400	a.i.k.o
2(3H)-Benzothiazolone	10-50	t
Camphor[3]	20.6-255.2	c.f.i.n. t. x
Cumen	0.3-7.4	
Fenchone	7.3-83	c.f.n. x
Tetrahydrofuran	9-430	a.i.k.o
Indane	0.2-20	t
Methylethylketone	110-6600	a.k. aa
Methyl-iso-butylketone	1.1-176	t. aa
Dimethoxymethane	1.1	t
MTBE	0.8-35	t
Styrene	0.5-1.6	t

[1] 2-(2-methyl-4-chlorophenoxy)propionic acid (MCPP)

[2] Dichlorophenoxyacetic acid

[3] 1,7,7-trimethyl-bicyclo(-bicyclo[2.2.1]-heptane-2-one

[4] I: Identified

[5] The references are:(a) Sable and Clark (1984), (b) Först et al. (1989), (c) Öman and Hynning (1993), (d) Harkov et al. (1985), (e) Gintautas et al. (1992), (f) Reinhard et al. (1984), (g) Albaiges et al. (1986), (h) Khare and Dondero (1977), (i) DeWalle and Chian (1981), (j) Dunlap et al. (1976), (k) Sawhney and Kozloski (1984), (l) Schultz and Kjeldsen (1986), (m) Karstensen (1989), (n) Kjeldsen (1993), (o) Barker et al. (1987a), (p) Barker et al. (1987b), (q) Krug and Ham (1997), (r) Riediker et al., (2000), (s) Bauer et al. (1998), (t) Greenpeace (1999), (u) Öman (1999), (v) Ejlertsson et al (1999), (x) Ledin et al. (2001), (y) Schrab et al., (1993), (z) Assmuth and Penttilä (1995), (aa) Plotkin and Ram (1984), (ab) Paxeus (2000).

ones found in the highest concentrations. The main reason for the relatively large number of investigations focusing on these two groups of pollutants is their well-documented negative effects in the aquatic environment. In addition, these non-polar organic compounds are relatively easy to analyze for, in spite the very complicated matrix of leachates from landfills. Finally, several of these compounds have been designated as priority pollutants by the U.S. Environmental Protection Agency.

Data on polar and ionic organic pollutants is scarcer, although the number of investigations including these more water-soluble pollutants is increasing. This can be exemplified by the group of phenols, where the older studies usually only were analyzing for phenol and the cresols, while some newer investigations also cover, for example, chloro- and nonyl-phenols (Table 6). The observed quantities of these phenols are generally in the μg/l level.

Twenty-one different pesticides were identified from a screening for a total of 101 pesticides in 10 Danish landfills (Ledin et al., 2001). The most common ones were MCPP (or Mecoprop; present in nine landfills), Bentazon (six landfills), and MCPA (three landfills). These phenoxyalkanoic acid herbicides have been identified in leachates from other landfills as well. Especially MCPP has been observed frequently (Schultz and Kjeldsen, 1986; Gintautas et al., 1992; Kjeldsen, 1993;

Öman and Hynning, 1993; Öman, 1999). However, other types of pesticides were also identified, for example, Ametryn, Atrazine, Chlorpropham, Dichlobenil, and Hexazinon (Table 6). These findings indicate that pesticides could be of importance when evaluating the impact from landfill leachates on groundwater quality.

Measurements for benzene and naphthalene sulfonates have been carried out in leachates from four Swiss landfills (Riediker et al., 2000). The results showed that benzenesulfonates (*p*-toluenesulfonate) and naphthalenesulfonates (Naphtalene-1-sulfonate, Naphtalene-2-sulfonate, Naphtalene-1,5-disulfonate, Naphtalene-1,6-disulfonate, Naphtalene-2,7-disulfonate, and 2-aminonaphtalene-2,7-disulfonate) were present in the leachates, at a concentration range from a few µg/l up to 11 mg/l (Table 6). The sulfonates include some of the surfactants used in laundry detergents and shower soaps. Another type of surfactant that could be expected to be present in landfill leachates are the alkylphenol polyethoxylates. Ledin et al. (2001) analyzed for two of these, nonylphenol mono- and di-ethoxylate, as well as two degradation products, nonylphenol mono- and di-carboxylate. They found nonylphenol monocarboxylate in leachates from two landfills, although no nonylphenol polyethoxylates were observed. The latter could be due to the relatively high detection limits for these compounds in leachates (20 to 25 µg/l).

The phthalates are also pollutants of concern. The most frequently observed phthalates are di-(2-hexylethyl) phthalate, di-ethyl-phthalate, di-*n*-butyl-phthalate, and butyl-benzyl-phthalate (Table 6). The highest concentrations are, however, observed for the degradation product phthalic acid (up to 14 mg/l) (Ejlertsson et al., 1999).

It could also be noted that MTBE (methyl-tert-butyl-ether), which is used as a gasoline additives, has been found in concentrations up to 35 µg/l in the leachates from eight Swedish landfills (Greenpeace, 1999).

More that 200 individual compounds or classes of compounds were identified in a screening for XOCs in three Swedish landfills (Paxeus, 2000). Among the compounds identified were dioxanes and dioxolans, which have not been reported previously in landfill leachates. These are synthetic cyclic ethers and are known to have very low odor threshold and high odor intensity. They originates form waste disposal from alkyd resin production and from disposed products from painting and coating (Paxeus, 2000).

AOX (Adsorbable Organic Halogen) has in some cases been used as an aggregated parameter for the content of XOCs containing halogens. Robinson (1995) reviewed concentration ranges of AOX in landfill leachates reported in the literature and performed additional analysis on 30 landfill leachate samples. The total range given by Robinson (1995) was 30 to 27,000 µg/l, but in most cases AOX results were in the range of 200 to 5000 µg/l. The usefulness of AOX measurements is limited by the lack of information on identities and concentrations on individual compounds, often with quite different health effects, and also by poor correlation between the measured AOX in the leachate, and the concentrations of identified, halogenated pollutants (Robinson, 1995). Grøn et al. (2000) saw the same problems using the related parameter TOX (Total Organic Halogens) in leachate from two Danish landfills. With commonly used screening procedures for organic pollutants, the individual halogenated compounds behind the TOX could not be found.

The results for halogenated aliphatics and the aromatics from two studies are compared in Table 7. The data presented by Krug and Ham (1997) originate from 13 municipal sanitary landfills in Wisconsin, USA, while the data from Kjeldsen and Christophersen (2001) is from 104 old Danish MSW landfills. In both studies, BTEX were the most frequently found compounds and concentrations were generally lower at the Danish landfills. Krug and Ham (1997) also reported the frequent presence of halogenated hydrocarbons at relatively high concentrations, reflecting the co-disposal of hazardous waste at some of the sites. At the old Danish sites the concentrations of the halogenated hydrocarbons were very low, probably reflecting that intense co-disposal has not been practiced on the Danish sites. Table 8 shows the results of a recent study comparing detection frequency and concentrations from old and new MSW landfills (Gibbons et al., 1999). The table shows that the concentrations of xenobiotic organic compounds at the old MSW landfills were generally higher than concentrations at the newer landfills, probably reflecting a lower acceptance of organic chemicals at the newer landfills. These data are in good accordance with an investigation of leachate concentrations over a 1-year period from 40 landfills with no co-disposal (Ecobalance, 1999).

H. Toxicity of Landfill Leachate

Risk assessment of landfill leachate is traditionally based on chemical analyses of specific compounds present in the leachate. However, risk assessment is not sufficiently developed to take into account interactions among chemicals or toxic degradation products for constituents in a complex mixture. In contrast to chemical analysis, bioassays can be used to characterize the toxicity of landfill leachate to integrate the biological effect of all its constituents. Thus, factors like bioavailability, synergistic, antagonistic, or additive effects can be assessed directly without the need for assumptions and extrapolations made from chemical analysis.

Until about 1980, there were only a few published studies on the toxicity of landfill leachate (Cameron and Koch, 1980), but since then the toxicity of landfill leachate has been assessed by several authors using a number of different organisms (Table 9). Single species or test batteries consisting of several different species or organisms from different trophic levels have been applied to landfill leachate to detect both specific and general toxicity. Fish (usually fathead minnow or rainbow trout), crustaceans (daphnids), and luminescent bacteria (commercially available as Microtox test kits) are among the most frequently used methods. Considerable differences in the sensitivities of different test organisms have been observed in most studies (for example, Plotkin and Ram, 1984; Clément et al.,

TABLE 7
Leachate Concentrations for Halogenated Aliphatic Hydrocarbons and Aromatic Hydrocarbons Found in Two Reviews of Regional Landfill Leachate Data

	Krug and Ham (1997)			Kjeldsen and Christophersen (2001)		
Compound	Percentage of sites detected	Average (μg/l)	Range (μg/l)	Percentage of wells detected	Average (μg/l)	Range (μg/l)
Halogenated Aliphatics						
Tetrachlormethane	n.m.	-	-	50	0.23	<0.1-0.28
Chloroform	40	7.1	7.1	50	0.39	<0.1-1.5
Dichlorodifluoromethane	40	219	2-1030	n.m.	-	-
Tetrachloroethane	n.m.	-	-	0	-	<0.1
1,1,2-Trichloroethane	40	1487	0.5-7130	n.m.	-	-
1,2-Dichloroethane	10	612	212-1030	n.m.	-	-
1,1-Dichloroethane	90	210	6-4120	n.m.	-	-
Chloroethane	40	112	9-410	n.m.	-	-
Tetrachloroethylene	40	19	0.8-44	57	3.2	<0.1-5
Trichloroethylene	n.m.	-	-	70	5.6	<0.1-24
1,2-t-Dichloroethylene	10	5.6	5.6	n.m.	-	-
Aromatics						
Benzene	70	123	2.2-1630	55	22	<1-400
Toluene	100	417	2.8-12300	49	38	<1-900
Ethylbenzene	100	110	4.8-1280	60	30	<1-100
Xylenes	70	461	10-3010	73	210	<1-2400
Naphthalene	n.m.	-	-	70	34	<1-280

n.m.: not measured.

TABLE 8

Leachate Concentrations for Halogenated Aliphatic Hydrocarbons and Aromatic Hydrocarbons Found in a New Review of American Landfill Leachate Data (Gibbons et al.,1999) (The landfills have been categorized in old and new landfills.)

Compound	Old MSWLF		New MSWLF	
	Percentage of locations detected	Average (μg/l)	Percentage of locations detected	Average (μg/l)
Halogenated Aliphatics				
Chloroform	3	n.g.	4	n.g.
Tetrachloroethane	0	-	0	-
1,1,1-Trichloroethane	2	5	23	178
1,1-Dichloroethane	17	400	41	116
Chloroethane	8	n.g.	8	n.g.
Tetrachloroethylene	5	n.g.	4	n.g.
Trichloroethylene	13	51	71	5.6
1,2-t-Dichloroethylene	33	492	32	104
Vinylchloride	10	107	15	51
Aromatics				
Benzene	49	65	13	7
Toluene	69	583	89	406
Ethylbenzene	57	198	39	60
Xylenes	20	461	n.m.	210

n.m.: not measured.
n.g.: not given
a Old MSWLF are landfills first receiving waste before 1985
b New MSWLF are landfills first receiving waste after 1985

TABLE 9

Biological Species (Number of Species) Used in Toxicity Tests for Assessment of Toxicity of MSW Landfill Leachate

No. of MSW landfills	F	C	D	A	B	G/M	Other	Study
1	1							McBride et al. 1979
7	2	1						Cameron and Koch 1980, Atwater et al. 1983
1	1	1		1	1			Plotkin and Ram 1984
1	1							Wong 1989
8*						1		Omura et al. 1992
19	1	1			1			Kross and Cherryholmes 1993
4					1	3		Schrab et al. 1993
2				4				Cheung et al. 1993
2			1		1		2	Devare and Bahadir 1994
1	2	1						Ernst et al. 1994
1					1			Nohava et al. 1995
9			1					Clément and Merlin 1995
35		1						Assmuth and Penttilae 1995
2						4		Helma et al. 1996
8		3	1	1	1		2	Clément et al. 1996
1						1		Beg and Al-Muzaini, 1998
1						3		Cabrera and Rodrigues, 1999
10				1	1	1		Ledin et al. 2001

F: Fish, C: Crustacean, D: Duckweed, A: Algae, B: Bacteria, G/M: Genotoxicity/mutagenicity, Other: Protozoans, Rotifers, Plants.
*=Leachate collected from mixing tank covering 8 landfills.

1996). As with the case for other complex environmental matrices, batteries of bioassays with organisms from different trophic levels increase the potential of detecting toxicity in a leachate sample (Kross and Cherryholmes, 1993; Keddy et al., 1995).

The toxicity of leachate from more than 98 different landfills has been reported in the studies listed in Table 9. In general, high toxicity was observed in leachate from landfills receiving MSW (Cameron and Koch, 1980; Atwater et al., 1983; Plotkin and Ram, 1984; Schrab et al., 1993). Clément et al. (1996) found that the most toxic leachates were those originating from landfills with co-disposed hazardous industrial waste, but other studies found that leachates from landfills with no known co-disposal were as toxic as leachates from mixed landfills or hazardous waste landfills (Schrab et al., 1993). In a lysimeter experiment with municipal waste Cameron and Koch (1980) found that 5 years of aging of the waste resulted in an 80-fold decrease in toxicity toward rainbow trout.

Ernst et al. (1994) concluded that ammonia was the primary cause of acute toxicity of municipal landfill leachate, whereas the chronic effects of the range of XOCs identified in the leachate could not be determined. Based on toxicity testing of 27 landfill leachates, Clément et al. (1997) concluded that ammonia and alkalinity were the most probable factors contributing to the observed toxicity. In the bioassay studies by Cheung et al. (1993) and Clément and Merlin (1995), it was also concluded that ammonia was the main cause of the toxicity measured in the biotests. As illustrated in Table 10, other studies have indicated that factors like pH, conductivity, and the concentrations of chloride, copper, or zinc may also be of major importance to aquatic toxicity assessed by aquatic bioassays (Cameron and Koch, 1980; Atwater et al., 1983; Kross and Cherryholmes, 1993; Assmuth and Penttilae, 1995; Clément and Merlin, 1995).

The chronic effects of landfill leachates have not received much attention, but of the different long-term effects mutagenicity/genotoxicity has been studied to some detail. Omura et al. (1992) covered leachates collected from eight MSW landfills. It was found that the leachates were mutagenic after preconcentration, and the authors suggested that organic compounds in the leachate caused the mutagenic activity. Based on multiple genotoxicity tests of leachate from MSW landfills, Schrab et al. (1993) found that three of four tested samples exerted genetic toxicity. This was also found by Helma et al. (1996) in a study of different aqueous samples, including landfill leachates. These experiments showed that landfill leachates had higher genotoxic potency than effluents from pulp production and wastewater. Moreover, risk calculations indicated that MSW leachates may present as great a cancer risk as those from co-disposal and industrial solid waste landfills (Schrab et al., 1993; Brown and Donnelly, 1988).

A number of studies reported that analytical measurements of XOCs did not correlate well with the toxicity observed in bioassays (Plotkin and Ram, 1984; Kross and Cherryholmes, 1993; Lambolez et al., 1994). It is, however, a general observation that landfill leachates may contain a large variety of XOCs that are

TABLE 10
Relationships between Toxicity of Different Landfill Leachates and Physico-Chemical Parameters (Estimated by stepwise multiple linear regression.)

Organism	Relation	R^2	Study
Fish	$\log LC_{50,\,96h} =$	0.94	Cameron and Koch, 1980
(Salmo gairdneri)	$1.427-0.386[NH_3]-101400[H^+]-0.000539[Tan]-4.074[Cu]$		
Duckweed	$EC_{50}=-23.3\log[Alk]-9.0\log[NH_4^+]+78.9$	0.96	Clément et al., 1997
(Lemna minor)	$EC_{50}=-21.6\log[Alk]-15.5\log[Cond]$	0.89	Clément and Merlin, 1995
Crustacean	$LC_{50}=27.73[Zn]^{-1.05}$	0.97	Altwater et al., 1983
(Daphnia pulex)	$\log LC_{50}=0.969-0.00884[Zn]-0.00152[Tan]+3.804[NH_3]$	0.83	Clément and Merlin, 1995
Crustacean	$EC_{50}=2489\,[NH_3]^{-0.62}[Alk]^{-0.386}$	0.96	Clément et al., 1997
(Daphnia magna)	$EC_{50,\,48h}=102-0.00507(LC_{50,24h})^2-0.396(\log[Cl^-])^2$	0.73	Assmuth and Penttilae, 1995

Tan: Tannin; Alk: Alkalinity; Cond.: Conductivity

acutely and chronically toxic (e.g. Brown and Donnelly, 1988; Schrab et al., 1993; Ernst et al., 1994). Despite all of the aforementioned studies, the importance of XOCs as a contributor to leachate toxicity remains largely unknown. This maybe due to the fact that toxicity caused by the sample matrix (e.g., ammonia, alkalinity, and salts) masks the toxic effect of XOCs (Baun et al., 1999). A way to overcome this problem is to use fractionation and preconcentration procedures such as the ones used in mutagenicity testing (e.g., Schrab et al., 1993; Helma et al., 1996). This approach was used by Ledin et al. (2001) in a study of the toxicity of leachate from 10 Danish MSW landfills. In general, the organic fraction of all landfill leachates were toxic toward algae and bacteria, whereas genotoxic responses were found in only a few of the samples in the concentrations used (Ledin et al., 2001).

I. Summary

The review on present leachate concentrations shows relative high concentrations of many parametres, especially COD, ammonia, and in some cases salts and xenobiotic organic compounds. There is still relatively little information regarding XOCs. The concentrations of heavy metals are generally low, even in cases where considerable amounts of heavy metals have been disposed of at the landfill. In the next section potential changes in leachate composition over periods of time beyond the currently available data are considered.

IV. LONG-TERM MSW LANDFILL STABILIZATION PROCESSES AND LEACHATE COMPOSITION

The decomposition of refuse and its relationship between the state of decomposition to leachate composition has been described in the previous sections. This description is appropriate for landfills in which decomposition is enhanced such that decomposition may occur over a period of as little as 5 to 10 years, as well as for landfills where there is sufficient moisture infiltration to promote decomposition over periods of 20 to 50 years. At landfills in arid regions, where the waste is dry and there is little infiltration, the landfill may remain in the acid phase or early methane production phase for decades or longer. To evaluate the long-term properties of landfills, it is important to look beyond the time when gas production is significant and to evaluate how the landfill ecosystem can be expected to behave over centuries and even thousands of years, once the bulk of the degradable components of the refuse have been degraded.

The objective of this section is to look into the future at the end of the refuse decomposition cycle, recognizing that the time to reach the end of the decomposition cycle will vary based on environmental conditions, primarily moisture, within the landfill. An understanding of the likely changes in leachate composition over long periods of time is required to assess the safety of landfills for the environment in the future. Interestingly, the time frames in this section go well beyond the 30-year post-closure monitoring period prescribed for landfills in the U.S.

A. Long-Term Landfill Phases

The four phases following the stable methanogenic phase are shown in Figure 1. The phases are based on speculation because we are not familiar with any landfills that have progressed beyond the stable methanogenic phase. Figure 1 is generally valid for a single waste cell, that is, a mass of waste having the same age. However, the last four phases on the figure are valid for a whole landfill because any age differences of the waste layers will be small compared with the total time to decompose beyond the methanogenic phase. In the following description we also focus on the gas composition within the landfill. The evolution of the landfill environment, and by that any potential changes in leachate composition will be driven by the supply of oxygen to the waste layers. The intrusion of oxygen to the landfill is dependent on the remaining degradable organic carbon, the gas generation rate, the cover properties, and the topography of the landfill. Phases V and VI may be seen as intermediate phases between the stable methanogenic phase and the carbon dioxide phase, where the landfill becomes aerobic.

1. Phase V. Methane Oxidation

Through the stable methanogenic phase, the gas production rate is continuously decreasing. For landfills with an active gas extraction and utilization system, air might be pulled into the landfill due to overpumping in the later phases when gas generation rates are lower. For landfills without active gas extraction, a small elevated gas pressure drives gas emission, and air intrusion will only occur due to barometric pressure changes, which might push air into the upper waste layers. If the landfill is equipped with a tight cover and open gas vents (assuming that active gas extraction or gas flaring has ceased), then air intrusion may be very local and probably of minor importance. For more permeable covers, the oxygen intruding into a landfill at high barometric pressure or due to diffusion will promote oxidation of the methane still produced in the landfill. The methane oxidation will mostly take place in the cover soil or in the waste mass surrounding the gas vent screens, that is, an insignificant part of the landfilled waste will be intruded by air.

2. Phase VI. Air Intrusion

As methane production continuously decreases, air will intrude through the cover and into the actual waste mass. At some time, almost all of the methane produced should be oxidized in the waste mass and the cover. This means that the average methane content within the landfill will continuously decrease, and carbon dioxide concentrations can be expected to increase through its production in the methane oxidation reaction. In addition, the nitrogen content will increase due to air intrusion, while oxygen will nearly be undetectable due to its rapid consumption. The overpressure produced by the generated methane will approach zero.

3. Phase VII Carbon Dioxide

Methane production will be insignificant in Phase VII, and the oxygen intruding the landfill will be consumed for oxidation of any residual methane, organic materials and reduced inorganic species (sulfur, nitrogen, and iron-containing species) buried in the landfill. Under aerobic conditions, additional refuse oxidation will occur, as some of the lignocellulosic substrate is more degradable under aerobic conditions relative to anaerobic conditions. The gas composition will be comprised largely of carbon dioxide, oxygen, and nitrogen. The resulting CO_2 production has been reported to result in a pH decrease in both refuse (Revans et al., 1999) and harbor sediments (Calmano et al., 1993). Another factor that exerts downward pressure on the pH of the refuse mass is the oxidation of reduced sulfur, nitrogen, and iron as each of these oxidation reactions releases protons (Calmano et al., 1993). It has been suggested that as the pH decreases, metal carbonate precipitates will dissolve and the carbonate release will buffer the pH, although this has not been demonstrated experimentally (Calmano et al., 1993). This phase has also been called the humic phase (Bozkurt et al., 1999).

The most important factors governing the exchange of air between the landfill and the atmosphere are diffusion, wind-induced exchange, natural convection (density differences), and barometric pumping (Bozkurt et al., 1999). Concentration differences between the landfill and the atmosphere will drive a *diffusional transport* of atmospheric components in and landfill gas components out of the landfill. One very important parameter is the diffusivity of the cover and the waste layers, which again is mainly governed by the water content. The presence of an engineered top cover aiming at preventing infiltration to the waste layers often includes a clay layer with a very low gas diffusivity at normal water content. In waste layers saturated with water, gas diffusion is not active and the diffusion through water is 10,000 times lower than in air. Diffusion will also be reduced significantly by the presence of plastic flexible membrane liners.

For above grade landfills, winds blowing past the landfill will create a pressure gradient between the two opposite sides of the landfill. This will cause air to flow through the landfill, the magnitude governed by the wind speed and the permeability of the cover/waste layers. However, Bozkurt et al. (1999) suggested that this process is of minor importance in most cases.

Natural convection is governed by differences in gas densities between the atmosphere and the interior of the landfill. This variation may be due to temperature differences, differences in air humidity and differences in concentration (CO_2/O_2), the interior gas being both lighter or heavier than the atmospheric air (depending on time over the day as well as over season) (Bozkurt et al., 1999).

If the landfill has a cover with a lower permeability than the waste layers, and if the cover has imperfections such as cracks, or open gas vents, or maintenance shafts, any *barometric pressure changes* will pump gas in or out of such openings. The flow direction will depend on the direction of the pressure change. Christensen et al. (2000) have successfully used passive venting as a remedial technology for removing volatile pollutants from sandy unsaturated layers covered by clay and showed that barometric changes led to a significant exchange of air through the installed screened wells.

Bozkurt et al. (1999) modeled oxygen intrusion considering the above stated processes (except barometric pumping) assuming that all oxygen is used for the oxidation of organic matter, and that all organic matter is available for oxidation. They used the model for different landfill scenarios (with/without cover, water saturated/unsaturated) and showed that in most cases the diffusional oxygen transport dominates the other transport processes. They estimated the time required to deplete all organic matter and found values ranging from a few centuries (for an unsaturated, uncovered landfill) to more than half a million years for a landfill fully saturated with water (i.e., waste placed under the groundwater table).

B. Long-Term Leachate Composition

In this section, changes in leachate composition through phases V–VII are evaluated. This evaluation is based on theory and limited laboratory data as field data from landfills in phase V-VII are not available.

1. Dissolved Organic Carbon and Inorganic Macrocomponents

The concentration of dissolved organic carbon and inorganic macrocomponents in landfills containing well-decomposed refuse was summarized in Section III.C and III.D, and these concentrations were shown to be relatively high, certainly higher than what can be allowed for direct release to the environment. Data from specific landfills generally show a slow decreasing trend with time (Kjeldsen and Christophersen, 2001). BOD/COD ratios below 0.1 can be expected and the absolute concentrations of BOD and COD are likely to decrease slowly through Phase V–VII until a BOD of near zero and a COD that consist of recalcitrant humic matter is all that remains. Ammonia-nitrogen will be converted to nitrate as a landfill becomes aerobic.

Belevi and Baccini (1989, 1992) developed a model to predict the long-term leachate concentration from landfills based on the extrapolation of leachate fluxes from the early phases of decomposition coupled with laboratory leaching experiments. Their objective was to evaluate when a landfill leachate would meet Swiss water quality standards without treatment. Elements modeled include C, N, P, Cl, Fe, Cu, Zn, Pb, and Cd. They present a first-order decay model that requires both a remobilization rate and knowledge of the fraction of a given element in MSW that can be mobilized. This fraction was determined in laboratory leaching experiments conducted with multiple distilled water extractions. The total volume of water used for the extractions was intended to simulate 2000 years of infiltration at a hypothetical landfill. The proposed methodology is largely empirical and does not take into account (1) the effect of biological transformations and stabilization on element release, (2) potential changes in the redox potential of a landfill, (3) the potential for dissolved organic matter to complex certain elements and increase their solubility, or (4) immobilization (sorption, precipitation) reactions.

2. Heavy Metals

As described in Section III.E, the concentration of heavy metals is low under the long, stable methanogenic phase, and the metal release is not considered problematic through this phase. However, as the refuse mass turns aerobic, a number of biological and chemical reactions will occur (see Section IV.A), and these reactions may well effect metal mobilization. As previously described, a decrease in pH is expected through the carbon dioxide phase, and it is well known that metal dissolution is enhanced at low pH. During anaerobic refuse decomposition, sulfates in refuse are reduced to sulfides that form insoluble precipitates with most metals (Christensen et al., 2000). However, as the refuse mass turns aerobic, these sulfide precipitates can be expected to oxidize to metal sulfates. The sulfate compounds are considerably more soluble than the corresponding metal sulfides that may increase metal mobilization. For example, pK_{so} for PbS and $PbSO_4$ is 27.6 and 7.73, respectively.

In addition to pH, metal solubility will be influenced by the oxidation-reduction potential (E_h). As the E_h of the refuse mass increases, the solubilities of some metals may increase. For example, Charlatchka and Cambier (2000) showed that the concentrations of Pd, Zn, Mn, and Fe increased with increasing E_h in agricultural soils polluted with metals. While there are certainly analogies between soils and decomposing refuse, the concentrations of organic carbon and the diversity of biological and chemical reactions are likely broader in landfills.

Other factors that will influence metals solubility include the cation exchange capacity of the refuse and how it changes during refuse oxidation and the presence of more oxidized functional groups on the solid humic matter as well as humic matter in leachate (Martensson et al., 1999). For example, carboxylic acids are known to act as chelators, and there could be an increase in carboxylic functional groups on humic compounds because the refuse is oxidized. Finally, iron hydroxides and oxyhydrates are formed during refuse oxidation. These compounds are reactive and may adsorb other metals (Calmano et al., 1993 Bozkurt et al., 2000).

In summary, there are multiple processes that can affect metals mobilization during refuse oxidation. Bozkurt et al. (2000) developed a model to predict long-term emissions of metals from landfills. Their model considered the oxidation of all refuse organic matter, humic substances that bind metals, and metal sulfide precipitates as well as pH buffering associated with calcite dissolution. They considered oxygen diffusion into landfills with a range of landfill geometry's and degrees of saturation and predicted that heavy metals mobilization will not occur for thousands of years. Belevi and Baccini (1989) suggested by measuring the alkalinity of landfill samples and assuming that 50% of the organic matter content of the waste is degradable and produces protons that landfills contain sufficient buffer to maintain alkaline conditions for more than 2000 years. Consequently, they did not expect remobilization of heavy metals due to lower pH for many centuries.

There has been a limited amount of experimental work to measure metals mobilization under an aerobic phase in well-decomposed refuse. Martensson et al. (1999) identified a number of factors that could effect metal's mobility in well-decomposed refuse that is undergoing a transition from anaerobic to aerobic conditions. Working with samples of 20-year-old refuse excavated from a landfill, they showed that the concentrations of Zn, Cd, Cr, S roughly doubled when the refuse was decomposed under aerobic conditions relative to anaerobic conditions in reactors. They suggested that aeration of the decomposed refuse resulted in the production of chelating agents that enhance metals mobility. This suggestion was supported by an increased metal binding capacity of aerated leachate. In contrast to the results of Martensson et al. (1999), Revans et al. (1999) conducted work in experimental columns that were filled with well-decomposed refuse to which Cd, Zn, and Cr had been added. Columns were aerated to simulate an aerobic phase after anaerobic decomposition. The concentrations of Cd, Zn, and Cr in leachate did not increase after aeration began and a mass balance showed that less than 1% of the added Cd, Cr, and Zn were leached after passing three bed volumes of leachate through the columns. Interestingly, the pH decreased approximately 0.8 pH units when the columns were turned aerobic. Altmann and Bourg (1997) carried out laboratory experiments with aqueous systems containing solid phases (aquifer solids from a landfill leachate plume). They showed that Cd that was previously trapped as a sulfide precipitate was released to the aqueous phase when conditions changed from anaerobic to aerobic. The released Cd was removed subsequently from solution by adsorption on iron oxyhydroxide phases or by precipitation as a carbonate mineral. Revans et al. (1999) suggested that the same processes were responsible for the low metal release in their columns.

Flyhammer et al. (1997) studied the fractionation pattern of heavy metals present in the fines and paper fractions of MSW based on the assumption that metals present in plastics, metals, rubber, and leather were not leachable under landfill conditions. They showed an increase in the fraction of Cu, Zn, Pb, Cr, Ni, and Cd present as carbonates and sulfides in anoxic refuse relative to aerobic refuse. This observation confirms the widely held expectation that metals are largely immobilized by precipitation under anaerobic conditions.

Finally, it must be recognized that the form of the added metal will likely impact its behavior in a landfill. Flyhammer et al. (1998) suggested that approximately 30% of the metals in MSW were available in reactive solid form based on a sequential extraction procedure. Using a similar set of extractions, Prudent et al. (1996) reported on the concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in 14 components of MSW. Prudent's work showed that much of the total metal content was present in forms that were not likely to be reactive in landfills. For example, plastics were the major contributors of Cd and scrap metal and rubber were major contributors of Zn. Interestingly, fines were high for each metal analyzed although the components of MSW that contributed to the fines fraction are difficult to define.

In summary, there are a number of factors that influence metals mobility because refuse goes from an anoxic to an aerobic state, including pH, E_h, functional groups on humic matter, and the sorptive capacity of the refuse mass. Given the complexity of the refuse system, it is not surprising that the limited amount of experimental work that has been conducted has resulted in both increased metals mobilization (Martensson et al., 1999) and no observed metals mobilization (Revens et al., 1999; Flyhammer et al., 1997). The experimental work to data is limited and the number of parameters that affect metals solubility makes models complex and uncertain. It is also unrealistic to expect that there will be field data forthcoming given the long period of time that is theoretically required for a landfill to turn aerobic. Thus, laboratory simulations that include the aerobic oxidation of organic matter that remains after anaerobic decomposition are required. These laboratory simulations should probably be performed over a long time frame because the aerobic oxidation may be a slow process.

3. Xenobiotic Organic Compounds (XOCs)

To evaluate long-term leachate concentrations and the time required to reach final storage quality, a multiphase approach is needed, including several processes: volatilization to gas escaping the landfill, diffusional loss through top covers, leaching and degradation. Kjeldsen and Christensen (2001) developed a simple box model, MOCLA, to evaluate the long-term fate of XOCs. The model is based on equilibrium assumptions concerning phase distribution mechanisms, and a first-order degradation process. Model runs using MOCLA showed that most XOCs will be released (for most volatile organic compounds through the gas produced) within a few decades. For more strongly sorbing compounds, such as naphthalenes the release will take several decades. Sensitivity analysis showed that degradation might play a very significant role in the long-term fate of XOCs. However, data in the literature on degradation of organic chemicals in landfills are still very rudimentary and, unless specific degradation rates are retrieved for a specific case, default values on degradation cannot be suggested (confer detailed literature review in Kjeldsen and Christensen, 2001).

Several factors may lead to significant extended life times of XOCs in landfills. The presence of special waste types having very high sorptive capacities such as activated carbon (Kjeldsen and Grundtvig, 1995) or insulation foams releasing CFCs (Kjeldsen and Jensen, 2001) may extend the life time considerably. Also, slow desorption from the organic waste may govern the release of XOCs, a fact observed in several studies with soils (Luthy et al., 1997). Recently, Sanin et al. (2000) reported on the behavior of toluene, acetone, and 1,2-dichoroethane in decomposing refuse. All three compounds were shown to biodegrade. Moisture stimulated toluene but not acetone biodegradation, and it was speculated that higher moisture increased the solubility and subsequent bioavailability of the hydrophobic toluene. In addition, some toluene was shown to be associated with the humic fraction (humic acid, fulvic acid, and humin) of the refuse after decomposition. This association may influence bioavailability and may also provide an alternative stable endpoint for some aromatic compounds.

In many older landfills and in some sanitary landfills, XOCs have been disposed of in intact iron containers such as drums. There is a considerable chance that such containers in some cases might have kept their integrity through the operational phase of the landfill. Thus, any release from such containers is relying on progressive corrosion eventually combined with the physical load of overlying waste layers. It is well known that iron can corrode under anaerobic conditions, especially during the acid phase when low pH will lead to substantial corrosion (Scully, 1990). However, in many cases only slightly acidic pH is reached due to the presence of adequate buffer in the waste (Christensen et al., 1992). Anaerobic corrosion of iron follows the reaction (Reardon, 1995):

$$Fe(s) + 2H_2O(l) \rightarrow Fe^{2+} + 2OH^- + H_2(g) \rightarrow Fe(OH)_2(s) + H_2(g)$$

As shown, the reactions produces hydroxide-ions leading locally to increased pH, precipitating calcium, magnesium, and iron present in the leachate on the iron surfaces. This phenomenon is known as passivation (Scully, 1990), because it prevents further anaerobic iron corrosion. Iron corrosion in the stable methanogenic phase therefore is expected to be relatively slow.

As previously discussed, acidity may be produced in the landfill phases following the methanogenic phase due to oxidation of organic matter and sulfides. This may locally dissolve the passivating layer on the iron surface and expose it to aerobic corrosion. Aerobic corrosion is generally faster, especially if pitting corrosion takes place literally forming open holes in the iron surface (Scully, 1990).

Sudden releases of organic chemicals in the carbon dioxide phase of a landfill due to corrosion of containers may lead to enhanced sorption and degradation of the pollutants. However, studies have shown that the dehalogenation of chlorinated compounds, which in many cases has been observed in landfills, is especially active at low redox potentials (Leahy and Shreve, 2000; Kromann et al., 1998), but inactive under oxidizing conditions, suggesting that a delayed release of chlorinated compounds in the carbon dioxide phase may leave the landfill unattenuated. Of course, this is a worst case scenario based on a container with chlorinated solvents that does not release the solvents until a landfill is largely aerobic. Such containers may also collapse shortly after refuse burial due to compaction of freshly buried refuse.

V. CONCLUSIONS

The release of leachate to the environment is one of the major environmental impacts related to disposal of waste. Disposed waste in landfills undergoes a series of phases where the waste is decomposed. During the decomposition leachate is generated by excess rainwater infiltrating the waste. The leachate contains four groups of pollutants: dissolved organic matter, inorganic macrocomponents, heavy metals, and xenobiotic organic compounds.

Existing data show that the composition of leachate is highly dependent on the degradation stage of the waste. In the acid phase, concentrations are generally higher due to enhanced formation of dissolved organic matter and release of ammonia. In the methanogenic phase, the content of dissolved organic matter significantly decreases and the composition of the organic matter changes indicated by BOD:COD ratios below 0.10. The ammonia concentration seems not to follow the same decreasing trend and may constitute one of the major long-term pollutants in landfill leachate. The content of heavy metals in the leachates is generally very low as a result of attenuating processes (sorption and precipitation) that take place within the disposed waste.

Leachate contains a broad variety of xenobiotic organic compounds. A very broad concentration range for each pollutant is observed in most cases. The most frequently observed compounds are aromatic hydrocarbons and chlorinated aliphatic compounds. Some later studies have also reported more polar compounds, which have not been observed previously with older analytical methods used.

A landfill will in a long time frame undergo several phases in which oxygen from the atmosphere will penetrate deeper and deeper into the waste. The oxidation of residual organic matter not decomposed anaerobically, and oxidation of sulfur, nitrogen, and iron-containing compounds may lead to lower pH and higher redox potentials, which may result in enhanced long-term releases of heavy metals. However, model calculations and a few laboratory experiments suggest that the enhanced release might not occur within a timeframe of several thousands of years. The xenobiotic organic compounds are affected by volatilization and degradation as well as leaching and are in most cases not believed to be a long-term problem.

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