

Long-term fate of organics in waste deposits and its effect on metal release

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Abstract

The long-term chemical evolution in waste deposits and the release of toxic metals was investigated. The degradation of organic matter and hence the potential efflux of heavy metals in a long-term perspective was studied by defining some scenarios for waste deposits containing organic compounds, different longevity and functions of covers and different water and air intrusion rates. The scenarios were based on various transport processes as well as different landfill constructions. The rates of influx of oxygen into both saturated and partially saturated landfills have been estimated. Each scenario takes the form of a mathematical model. The starting point for all the studied cases is the humic phase, i.e. the phase after the methane production has stopped. Based on the different cases studied, it appeared that landfills where the waste is below the water table could have advantages over the other cases. Recognizing that this option is not accepted in most countries we, nevertheless, suggested it should be reevaluated. The main conclusion is that the degradation of humic matter and hence the release of toxic metals can be substantially decreased if potential build-up of hydraulic gradients are avoided and if the landfill is located below the water surface. A conceivable alternative construction would be to place it in a depression — either natural or artificial — and to construct it so that under normal conditions it would always be water-saturated. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Degradation; Waste material; Landfill processes; Metals; Long-term; Transport processes; Oxygen; Contaminant release; Modelling

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1. Introduction

Landfilling is the oldest and cheapest way of disposing municipal solid waste and residues contaminated with heavy metals, inasmuch as long-term effects and external costs are not considered. Landfills for industrial and municipal solid wastes contain a complex mixture of both organic and inorganic matter and are potential sources for the spreading of toxic compounds. Pollutants that are added to a landfill have various sources. Knowledge of the fate of these compounds in landfills is poor, and this makes it difficult to assess environmental risks with existing landfills, to develop plans for how future waste deposits shall be constructed and to decide what to deposit, etc. The chemical reactions taking place over decades in a reactive multicomponent heterogeneous system such as a landfill cannot be controlled or predicted accurately. Toxic compounds are therefore likely to be released, possibly centuries after deposition. There is a considerable risk that environmental problems may result from contaminated sites in future generations and this calls for a scientifically sound method of assessing the important processes and the associated risks. In order to assess the long-term pollution risks associated with deposits, knowledge is required of the processes going on in the deposit.

Degradation processes in landfills take place over a very long period of time. Until now, studies have been focused on the degradation processes occurring during the short initial aerobic phase and the anaerobic phase. The latter can be divided into two phases: a first phase called the anaerobic acid phase, and a second phase called the methane production phase. Considerable work has been done on these early phases (Farquhar and Rovers, 1973; Chian and DeWalle, 1976; Ehrig, 1989; Barlaz et al., 1992) and most of the processes during these phases can be described qualitatively and quantitatively. The subsequent phase is called 'the humic phase'. This is reached after perhaps one century and can possibly last for thousands of years. The humic phase is poorly understood and not well covered in the literature. Quantitative descriptions of processes during this

phase are missing. Prior to this phase, only a very small portion of the toxic metals deposited in a landfill will have been leached and thus the waste deposits will still have a large potential for contamination. Long-term processes that may change the leaching behaviour of the waste will therefore be of great importance. In this study, the times of interest are of the order of hundreds to thousands of years.

Studies of waste deposits in the early degradation stages (up to and including the methane production phase) indicate that the leaching rate of contaminants will decrease exponentially over time (Belevi and Baccini, 1989). This is sometimes taken as evidence that this decrease will continue and that there is a relatively small risk of long-term contamination from the leachate. However, extreme caution must be exercised in extrapolating the results from one phase to another. Changing conditions during the humic phase may lead to a substantial change in the mobility of heavy metals. One reason is that oxygen may penetrate into the landfill and generate aerobic conditions, which will fundamentally change how metals are bound in the deposit. Oxidation of organic matter will give carbon dioxide, which may decrease the pH to below neutral. These changes may considerably increase the mobility of heavy metals.

The difficulty of making statements about the behaviour of a landfill results from the multitude and complexity of processes and their interrelations. In this investigation, the purpose has been to include only those mechanisms assumed to be the most important without considering the detailed processes. Laboratory experiments and field experiments can provide much valuable information, but they are not in themselves enough to permit reliable assertions in the long-term, and the slow processes occurring in a waste deposit are difficult to study in a laboratory experiment. The assessment must thus be carried out with models that are based on fundamental principles such as mass balances, chemical equilibria between solid phases and the aqueous phase, kinetics of dissolution of phases and contaminant transport. The models must in turn obtain data from specifically designed experiments and be evaluated against observations from old landfills.

Observations of old waste landfills and other similar environments, such as peat moors (Lucisano and Bozkurt, 1998), sediments, etc. can probably contribute with valuable information about the long-term behaviour of landfills.

This paper applies the conceptual model developed by Bozkurt (1998) to different cases and focuses on the quantification of what we envisage to be the main processes governing the degradation of organic matter in landfills, and hence the emission of landfilled heavy metals, over long periods of time. The long-term behaviour of a landfill will be strongly influenced by its content of organic material and the development of the emission behaviour in landfills will mainly depend on the redox and pH conditions. The focus of this paper is on degradation processes for *humic* substances since, in a long-term perspective, i.e. after the methane gas production has become more or less negligible, the organic compounds are the most important substances that control the mobility of the metals.

In this paper, the main question we address is: At what rate will the organic matter in a landfill be exhausted and at what rate will the toxic metals be released? Another important question is: How will the design and structure of the landfill affect the leaching? These issues will be approached in the way presented in Bozkurt (1998) and can be summarised in the following way:

- Study of the influx of water and oxygen into the waste deposit.
- Study the degradation rate of organics in the waste deposit.
- Study of the release of pollutants from the landfill to the recipient.

To answer these questions, we need to look at the mechanisms that determine the chemical processes occurring inside a landfill, emphasising the processes that govern metal emissions in the long-term. The degradation of organics, and subsequently the potential efflux of heavy metals, in a long-term perspective has been studied by defining some cases for waste deposits. The different cases studied are transport processes for oxygen into a partially saturated landfill and a satu-

rated landfill because oxygen is the most important reactant. The starting point for all the studied scenarios is the phase after the methane production has stopped. The type of landfill studied is a municipal solid waste which contains organic matter. Although the quantifications are made for landfills for industrial and household waste containing much organic matter, the approach can also be used for waste that has been incinerated and contains only small amount of organic material. The assumptions and models that allow prediction of leaching rates for a period up to several thousands of years will be presented for each case.

We envisage the following evolution of the migration and release of the metals from a deposit. Water that leaves the bottom of the deposit carries with it metals in a concentration that is influenced (at equilibrium) by the sorbing materials — humics mostly, at the bottom of the deposit. The concentration may be low and decrease in time as the most mobile metal fractions are released from the waste. This is what is often observed and also is part of the assumptions of Belevi and Baccini (1989). In contrast and in addition to their conception we also look further upstream in the deposit. In the upper parts fresh water enters and gradually sweeps the metal with it. In addition, and most importantly we account for the destruction of the humic substances due to oxidation by intruding oxygen. This releases the metal from the oxidized part of the deposit, sweeping it down into the locations where there still are humics. The metal can temporarily be re-adsorbed there causing a band of more concentrated metal region to form. This moves constantly downward as more humics are oxidized. Finally when all humics along the flowpaths are oxidized and release the metal a strong increase in the effluent concentration can take place. Fig. 1 illustrates this.

In contrast to the model of Belevi and Baccini (1989) we try to address the chemical changes over time influencing the binding strength for the metals. We also try to account for the changes along a flowpath over time.

Although we have made the description as if only adsorption played a role there are other

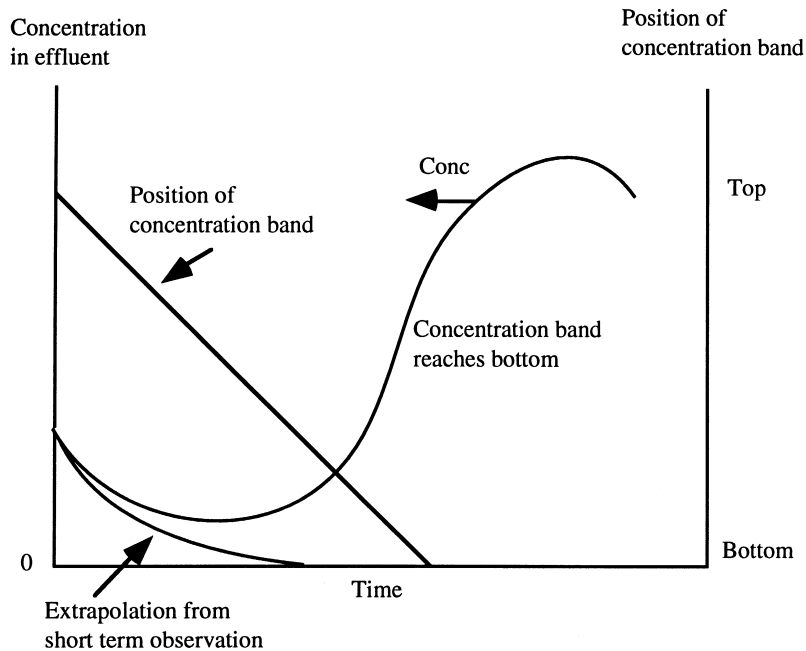


Fig. 1. Schematic illustration of a reaction front in a landfill.

mechanisms that behave similarly at the redox front notably oxidation of metals and of metals bound to sulphides.

In this paper we therefore focus on the time it will take to oxidise the humics (and other reducing substances).

Firstly, the processes occurring in a landfill are described. In Section 3, the development of mathematical models for the different cases studied are presented. These models include intrusion of oxygen by diffusion and convection into waste deposits. For the convection case, a distinction is made between natural and forced convection. The former case is due to density variations of the air while the latter case is caused by the wind. Finally, the results from the different cases will be used to identify the relevance of the different cases on the release of toxic metals from landfills.

2. Processes in landfills

After solid wastes are deposited in a landfill, physical, chemical, and biological processes modify the waste. The organic pollutants will degrade at various speeds and by various mechanisms. The

transformation of organic matter involving microbial degradation can generally be classified into either aerobic or anaerobic processes. If the biologic degradation of the waste takes place in the presence of oxygen and when the oxidation of the organics is due to reactions with oxygen, the process is said to be aerobic. When no oxygen is present the process is said to be anaerobic.

Depending on the degree of compaction of the landfill, there may be some oxygen available initially in the pores. This could then serve as an oxidising agent. However, the supply is limited and the oxygen may have already been used up after a couple of days. If there is nitrate, sulphate and ferric iron in the landfill, these can also serve as oxidising agents. During the anaerobic phase, which can be subdivided into an acidic phase and a methane production phase, these oxidants decrease and low-molecular organic matter ferments (Farquhar and Rovers, 1973; Barlaz et al., 1992). The redox potential drops and, because carboxylic acids are formed, the pH also drops (see Fig. 2). At low pH values, the mobility of some metals may increase. During the methane production phase, the intermediate degradation

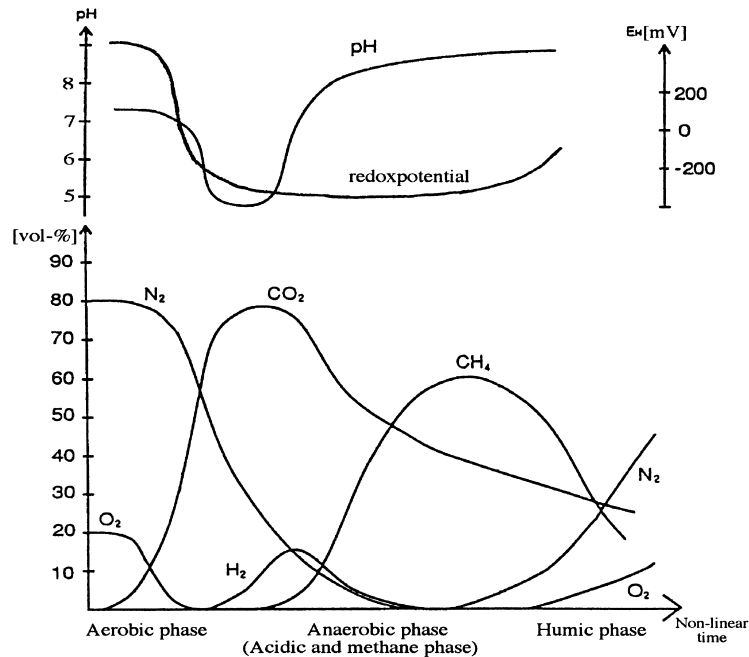


Fig. 2. Schematic illustration of the redox potential, pH (in pore waters) and gas composition during different phases in a landfill.

products — hydrogen, carbon dioxide, acetic acid, low-molecular compounds, etc. — act as substrates for methane-producing bacteria yielding a gas consisting of carbon dioxide and methane. The degradation of volatile fatty acids causes the pH to increase to an almost neutral level, buffered by the bicarbonate system. As the remaining substrate becomes more and more difficult to degrade, microbial activity slows down and the humic phase starts (Bozkurt, 1998). Because of the strong heterogeneity of the landfill, different parts of the landfill develop at different rates. There may thus be parts that enter the humic phase early and others that will not reach it for a considerable time. When all readily degradable organic matter is gone, only very stable, so called refractory organic matter remains in the waste. Parts of the organic matter are transformed to much more stable and complex compounds than the original compounds. The more stable compounds are called humic substances. Most of the remaining organic material exists in the form of humic and fulvic acids with molecular weight ranging from approximately 400 to many tens of

thousands $g\ mol^{-1}$. This material plays an important role in sorbing many of the toxic metals and retarding their release (Ciavatta et al., 1993). The complexing capacity of these substances is primarily due to their hydroxyl and carboxyl groups. The complexing capacity can be of the order of $10\text{--}20\ meq.\ g^{-1}\ C$ (Stumm and Morgan, 1996). There is, however, a competition for these sites by protons, some organic molecules and other non-toxic metals, e.g. Ca, Al and Fe. A part of the toxic metals are probably bound as sulphides. Up until the humic phase, any oxygen supplied to the landfill can primarily react with simple organic compounds. During the humic phase, the reaction rate is low and ambient air may diffuse/or flow into the landfill without being scavenged by the remaining organic (and inorganic) substances so that aerobic zones develop. If and when the conditions have turned oxidising in the landfill the humics are assumed to have disappeared or at least lost their binding properties. On the other hand, some of the common metals, e.g. iron and aluminium, can, when oxidized, form very strong 'natural' sorbents. Iron in the waste will oxidise

and precipitate as ferric oxy hydroxides which exhibit a high sorption capacity (Dzombak and Morel, 1990) for the toxic metals and could possibly take over the role of retaining substance. However, the sulphides may be oxidized and the precipitated metals may become soluble again. The pH may decrease due to sulphide oxidation and percolation of acid rain water. Metals bound to carbonates may dissolve if pH drops or if the partial pressure of carbon dioxide decreases. It is possible that the pore water can become oxic, especially in partially water-saturated landfills, and increase the solubility of some metals. More than 90% of the non-metals and more than 99.9% of the metals are still found in the residual solid at the beginning of this phase (Belevi and Baccini, 1989; Bozkurt, 1998). The oxygen supply will be a determining factor for oxidation of the metals to higher oxidation states. In a long-term perspective, an increased release of hazardous elements, e.g. heavy metals, can not be ruled out. The duration of the humic phase is expected to be very long, probably many thousands of years. It will depend on how fast and how much oxygen intrudes.

When the complex mixture of solid waste comes into contact with percolating water, such as rain water or ground water, a portion of the organic and inorganic substances (e.g. heavy metals) dissolve. The chemistry of the leachate is influenced by the substances that dissolve and react. Important mechanisms and processes are, complexation, redox reactions, precipitation/dissolution, adsorption, ion exchange, etc. The solubility of the metals is primarily determined by the pH, Eh, the presence of complexing agents (Förstner et al., 1988) such as chlorides, sulphates, carbonates, organic acids, and the solid-waste phases to and in which the metals can be bound. In reducing environments, and when the pH is almost neutral, the solubility of many of the toxic metals is low. Gradually, as the redox capacity is depleted, due to aerobic and anaerobic degradation of the organic matter, the redox state of the water will change. During the degradation process, acids and complexing agents are formed. The flowing water will carry pollutants with it out of the landfill. The concentration of substances that

leave the landfill are determined by, among other things, the sorption of individual substances into the waste, their solubility, the reaction kinetics of the waste, etc. At present little is known about what controls the leaching during the humic phase, a phase which can exist during thousands of years. It is entirely conceivable that the leaching of metals will increase substantially through, for example, complexation and uptake on colloids or dissolved humic substances.

3. Oxygen transport into a landfill

The aim of this section is to present a simple, yet robust model which can be used to simulate the long-term degradation of organic matter and hence the release of toxic metals from landfills.

3.1. Description of the model

Prior to the humic phase, only a very small portion of the toxic metals deposited in a landfill are removed by the leachate, and the larger part remains. As mentioned previously our starting point is the humic phase which is reached after perhaps one century. Its duration is expected to be very long; probably many thousands of years. When the humic phase starts, all the readily degradable organic matter has been degraded and only very stable, so called refractory organic matter remains in the waste. The remaining organic matter is mostly humic substances. The humic substances and the sulphur compounds in the waste should have a capacity sufficient to bind all toxic metals (i.e. Cd, Cr, Pb, Zn, Hg) in a deposit under ideal conditions (Bozkurt, 1998). By 'ideal conditions' it is meant that all the matter is evenly distributed and that the affinity for these metals is higher than for the other potentially competing metals (such as Ca, Fe, etc.). The calculations in this binding capacity scenario for sulphides and humic substances are based on the following complexation reactions:

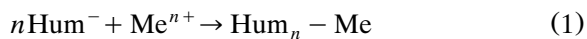


Table 1
Data used for the different scenarios

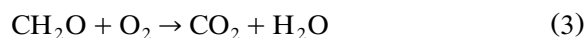
Metals and binding substances	Amount in tonnes ^a
Cd ²⁺	30
Cr ³⁺	450
Pb ²⁺	10 000
Zn ²⁺	10 000
Hg ⁺	13
Fe ²⁺	200 000
Ca ²⁺	150 000
Al ³⁺	50 000
Sum metals	420 493
Organic matter as humic substances	1 000 000
Sulphur	15 000

^aThese figures can vary in different locations in the landfill.

where Hum is a humic substance and Me is taken symbolically as a metal, and the binding capacity of humic acids for metals has been assumed to be 1 meq. g⁻¹ HS. For the different scenarios, data and information (see Table 1) from a landfill for industrial and household wastes (the Högbytorp landfill, situated 40 km north-west of Stockholm) examined previously (Aulin and Neretnieks, 1995) are used.

If no oxygen and other oxidants intrude no sulphide and no humic oxidation would occur. The system would then be stable 'indefinitely'. The apprehension is that the mobility of the metals may increase as conditions change, e.g. oxygen may penetrate into the landfill and cause

aerobic conditions (see Fig. 3). The oxidation rate in a deposit is considered to be determined by the rate of inward transport of oxygen. In the studied cases, the only reaction considered is the oxidation of organic matter. It is common practice to write the subsequent reactions of the organic matter as if it consists of a multiple of CH₂O. The overall reaction can then be written:



If the humics and the sulphides were oxidized, the metals could be released. The release of metals from the landfill will be strongly correlated to the rate by which oxygen enters the landfill. The main thrust of the modelling is thus to assess oxygen intransport rates. It may be noted that other oxidants such as nitrate, sulphate, ferric iron are assumed to have reacted at some earlier stage. The main question to address is: At what rate will the organic matter in a landfill be exhausted and at what rate will the toxic metals be released?

A number of basic scenarios are defined by making well-defined and systemized assumptions with different levels of complexity. By devising and studying different scenarios one can obtain indications of what is important and what is not. In addition, sensitivity analyses can often indicate when a process may be important and when not. This can be used as well to show where more detailed calculations and measurements must be

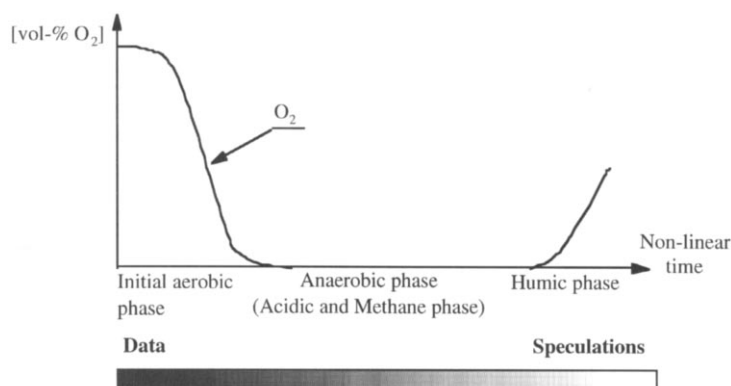


Fig. 3. Consumption and supply of oxidising agents, here represented by oxygen.

made. When it is found that a certain process will not be important under any reasonable circumstances, it need not be of further concern. The calculations are simple and are based on mass balances and simple kinetic rate equations. The different cases studied are various transport processes such as diffusion, convection and advection for oxygen into a partially saturated as well as a saturated landfill. A covered deposit where the cover is broken and covers with different resistance to oxygen transport are also explored. The different cases are studied individually and independently of each other.

The defined scenarios are evaluated with the aid of models. The strength of the modelling approaches lies in their simplicity. It allows us to gain insights which may be difficult to observe in the actual system due to its inherent complexity. A scenario describes an overall picture of an event and may apply to the conditions existing at a certain time or to a sequence of developments. The general picture of a landfill is schematically shown in Fig. 4. A landfill is a highly heterogeneous and anisotropic formation with a complex internal geometry that is variable in time due to biodegradation and subsequent settling. Water may flow through preferential paths while other regions may be unaffected. The studied landfill, containing a large fraction of organic matter, can be considered to be composed of a number of parallel column reactors with different degree of channelling flow, different organic matter–metal ratio, etc. In the calculations the landfill is considered as one column reactor and the variations that may exist within a waste deposit are taken into account by varying the data in the calculations. The landfill is thus modelled as a column reactor for transport in one dimension at steady state.

3.2. Main assumptions used in the model

The difficulties associated with making future assessments to embrace up to thousands of years are very obvious. At present, there is considerable lack of clarity as to which mechanisms control the chemical development and leaching in a landfill. It is also illustrated in Fig. 3 that, as we move

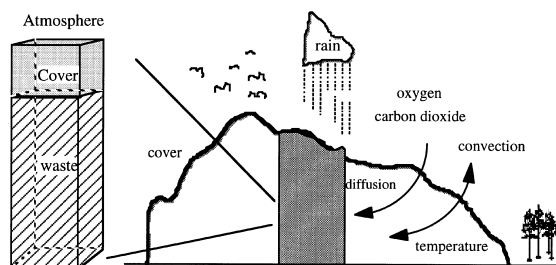


Fig. 4. A schematic figure of a landfill.

along the time axis, data become more scarce. The most difficult step is to simplify the tentative model so that it is possible, on the one hand, to extract meaningful data and, on the other hand, to describe the principal processes adequately. A number of assumptions that simplify the simulation, and highlight and demonstrate the central processes are made. These can be revisited and modified afterwards, as more data become available.

The following are the most important assumptions forming the bases for the calculations:

- The landfill is homogeneous and porous and can be modelled as a column reactor.
- The organic matter consists of humic substances.
- The chemical conditions in the landfill are determined by the degradation of the organic compounds; reactions with the inorganic matter; and the transport of various reactants (oxygen, carbon dioxide, water) and heat to and from the landfill.
- The toxic metals can sorb onto the various sorbent substances that exist or are formed in the landfill.
- The water flux through the landfill is constant over time (in the examples we use $200 \text{ l m}^{-2} \text{ year}^{-1}$), which corresponds to an infiltration-to-precipitation ratio of 0.4.
- Gases can be transported by flow and by molecular diffusion to, in and out of the landfill.

In the subsequent sections the different cases studied will be presented.

3.3. Oxygen intrusion by diffusion

The model is based on the law of mass conservation. The difference between the inflow and the outflow with flow, diffusion, injection or removal is equal to the accumulation. This fundamental principle is applied to each of the components. In Fig. 5, the reactor is illustrated for oxygen transport and reaction.

The mass conservation for an incremental control volume when reactions in every point are considered can in general be written as:

$$\left[\begin{array}{l} \text{change in mass} \\ \text{storage with time} \end{array} \right] = \left[\begin{array}{l} \text{mass inflow} \\ \text{rate} \end{array} \right] - \left[\begin{array}{l} \text{mass outflow} \\ \text{rate} \end{array} \right] + \left[\begin{array}{l} \text{mass production/} \\ \text{depletion rate} \end{array} \right] \quad (4)$$

Rearranging the different terms and expressing them in a mathematical form gives the following equation:

$$\varepsilon_p \frac{\partial c}{\partial t} + q \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} + R \quad (5)$$

This is the well known advection–diffusion equation with reaction. Here ε_p is the porosity, c is the oxygen concentration (which varies with t and x) (mol m^{-3}), t is time (s), q is water or air flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$), x is distance in the direction

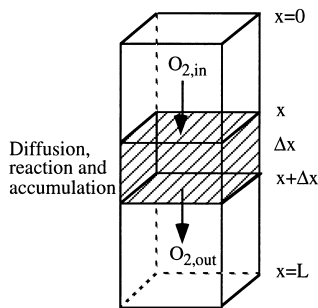


Fig. 5. Illustration of a landfill as a differential waste column reactor when oxygen is accounted for.

of transfer (m), D is the effective diffusion coefficient ($\text{m}^2 \text{s}^{-1}$), and R is taken symbolically as the mass produced/depleted per unit volume per unit time ($\text{mol m}^{-3} \text{s}^{-1}$).

The first term in Eq. (5) accounts for the accumulation of oxygen. The second term accounts for transport by convective flow. The third term accounts for molecular diffusion and the fourth for production or depletion by some process (such processes may be biological or chemical degradation or generation). The second term will be neglected at present and revisited in a later case and no accumulation of oxygen is considered. The first and second term are thus zero with these assumptions. The oxygen reaction rate is assumed to be proportional to the oxygen concentration. The oxygen consumption rate can then be written:

$$R = -k'c \quad (6)$$

where R is the oxygen depletion rate ($\text{mol m}^{-3} \text{s}^{-1}$) and k' is the first order kinetic constant (s^{-1}).

By substituting Eq. (6) into Eq. (5), the equation for the steady state case can be expressed as:

$$D \frac{d^2c}{dx^2} - k'c = 0 \quad (7)$$

To solve Eq. (7), the boundary conditions must be known. The following boundary conditions are chosen for the waste deposits:

$$\text{BC 1: } c_{\text{O}_2}(x = 0) = c_{\text{O}_2,0} \quad (8)$$

$$\text{BC 2: } \left. \frac{dc}{dx} \right|_{x=L} = 0 \quad (9)$$

The first boundary condition means a constant oxygen concentration at the upper boundary, e.g. at the ground surface. The second boundary condition means no transport over the bottom boundary. The solution of Eq. (7) for these boundary conditions is:

$$\frac{c_{\text{O}_2}(x)}{c_{\text{O}_2,0}} = \frac{e^{(x-L)\sqrt{k'/D}} + e^{-(x-L)\sqrt{k'/D}}}{e^{L\sqrt{k'/D}} + e^{-L\sqrt{k'/D}}} \quad (10)$$

where $c_{O_2}(x)$ is the oxygen concentration as a function of depth, $c_{O_2,0}$ is the oxygen concentration at the ground surface, and L is the depth of the landfill.

The model can now easily be extended by taking into account an inert soil cover. In this case the concentration at the interphase cover-landfill will be less than the concentration at the surface. Changing the first boundary condition, Eq. (8), to that in Eq. (11) allows for the influence of a soil cover with thickness L_1 and with an oxygen diffusivity D_1 :

$$BC\ 1: D_2 \frac{dc}{dx} \Big|_{x=0} = \frac{D_1}{L_1} (c(x=0) - c_{O_2,-L}) \quad (11)$$

The solution of Eq. (7) with the new BC is somewhat more lengthy and can be found in Bozkurt (1998).

A schematic picture of the model is shown in Fig. 6 and includes:

- diffusion of oxygen with no reaction in the cover, and

- oxygen consumption in the waste.

Fig. 6 also shows some oxygen profiles calculated for conditions expected to be found in a landfill with and without a cover. The curve A shows the oxygen profile when no cover is used. The curves B and C show the profiles for two different covers.

3.4. Oxygen transport by natural convection

Mass transfer due to convection involves the movement of a fluid, in this case air. There are two types of convective mass transfer depending upon the driving forces causing the fluid to flow. *Natural* or *free convection* designates the type of process wherein fluid motion results from the density difference, which may have resulted from concentration (oxygen and carbon dioxide concentration, content of water vapour) or temperature changes. When a fluid is heated or cooled (or if the composition changes), the associated density changes and the buoyancy effect produces a

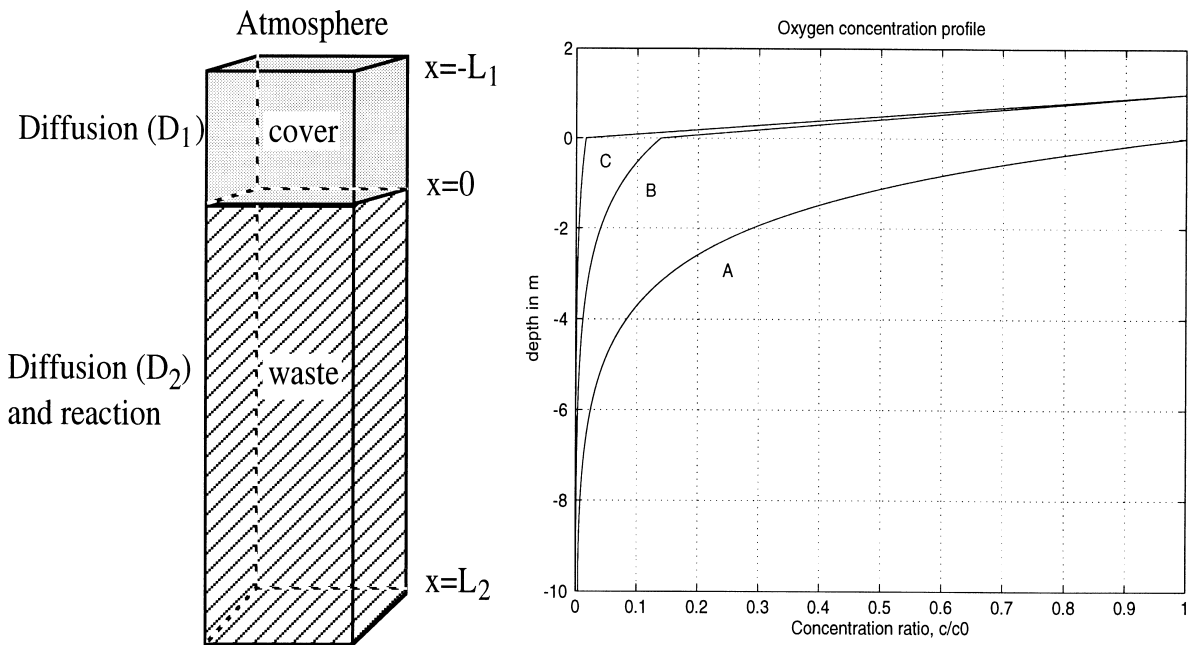


Fig. 6. (a) Concept of a waste column reactor with a cover. (b) Calculated oxygen concentration profiles in a 10-m deep landfill with a 1-m deep soil cover. A: no cover, $D_2 = 2 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$. B: $D_1 = 2 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$, $D_2 = 2 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$. C: $D_1 = 2 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$, $D_2 = 2 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$.

natural circulation in which the affected fluid moves of its own accord, the fluid which replaces it is similarly affected by the energy transfer, and the process is repeated. *Forced convection* is the name used to describe those flows in which fluid circulation is produced by an external agent such as a fan. In this section, only natural convection is studied.

The flow of a fluid at low velocities is governed by Darcy's law, the flow is being given by:

$$\vec{q} = -\left(\frac{k}{\mu}\right)(\vec{\nabla}p - \rho\vec{g}) \quad (12)$$

where q is the flux of the fluid ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$), k is the permeability of the porous medium (m^2), μ is the fluid viscosity (Ns m^{-2}), p is the hydrostatic pressure (N m^{-2}) and g is the gravitational acceleration (m s^{-2}) which is a vector in the downwards direction.

By expanding the density ρ in a double Taylor series in T and x about some reference temperature \bar{T} and some reference composition \bar{x}_i we obtain:

$$\rho = \bar{\rho} - \bar{\rho}\bar{\beta}(T - \bar{T}) - \bar{\rho}\bar{\zeta}(x_i - \bar{x}_i) \dots \quad (13)$$

where $\bar{\rho}$ is the density (kg m^{-3}) at temperature \bar{T} (K), the subscript i denotes oxygen, carbon dioxide or water. $\bar{\beta}$ is the volume expansion coefficient ($1/\text{K}$) at \bar{T} , which indicates how much the density varies with temperature. $\bar{\zeta}$ is a quantity (defined analogously to $\bar{\beta}$) that indicates how much the density varies with composition. Substitution of the first three terms in the Taylor series into Eq. (12) gives:

$$q = -\left(\frac{k}{\mu}\right)\left(\frac{\Delta p}{\Delta z} + \bar{\rho}g - \bar{\rho}\bar{\beta}g(T - \bar{T}) - \bar{\rho}\bar{\zeta}g(x_i - \bar{x}_i)\right) \quad (14)$$

If the pressure gradient in the system is assumed to be due solely to the weight of the fluid, then:

$$\frac{\Delta p}{\Delta z} = -\bar{\rho}g \quad (15)$$

and the equation of motion becomes:

$$q = \frac{k}{\mu}\left(\bar{\rho}g\bar{\beta}\Delta T + \bar{\rho}g\bar{\zeta}\Delta x_i\right) \quad (16)$$

where ΔT is a characteristic mean temperature difference defined as $(T - \bar{T})$ and Δx_i is a characteristic mean concentration difference defined as $(x_i - \bar{x}_i)$. The fluid viscosity is assumed to be independent of the temperature, and the permeability is assumed to be constant. The expression in Eq. (16) is for steady-state air flow caused by natural convection.

A sketch of the model used in evaluating free convection is shown in Fig. 7. The gas inside the landfill has a temperature T_i , a certain concentration of O_2 , CO_2 and a certain humidity, which are assumed to be constant for sake of simplicity. The air outside the landfill has a temperature T_o , a certain humidity and the normal air concentration of O_2 and CO_2 . The air density inside the landfill may be higher or lower than the density of the air outside the landfill. If the density of the air inside the landfill is lower (e.g. if the air is warmer) the pressure at the bottom of the landfill will be less than the pressure at the same level outside the landfill. This pressure difference between inside and outside air is $\Delta P = gH(\rho_o - \rho_i)$ and results in the driving force for the natural convection. This will induce air flow in the same way as in a chimney. The different mechanisms that may cause free convection are shown in Fig. 8.

3.5. Convective air flow (wind-induced)

Flow may be caused by free convection and forced convection. Here, the case where the air flow is caused by an external force will be discussed. It is assumed that the landfill is designed in the form of a mound (see Fig. 9). Wind blowing past a landfill in the form of a hill or mound will cause a pressure gradient to develop between the two opposite sides of the landfill which can cause air to flow through the landfill.

In order to estimate the air flow through the landfill, it is assumed that somewhere locally the approaching wind velocity is $u \text{ m s}^{-1}$. At a stag-

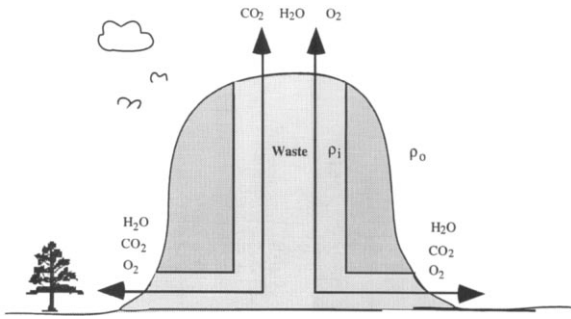


Fig. 7. Illustration of a landfill as a chimney.

nation point, the kinetic energy of the wind is transformed into pressure energy. A pressure difference between the two regions of the landfill then develops and the pressure difference, ΔP , is:

$$\Delta P = \frac{\rho u^2}{2} \tag{17}$$

where ΔP is the pressure difference (N m^{-2}), ρ is the density (kg m^{-3}), and u is the wind velocity (m s^{-1}). The pressure contribution of the elevation term, ρgh , has been neglected assuming that the air flows horizontally through the landfill. However, it is possible that not all the kinetic energy will be converted into pressure energy. The stagnation point assumption is conservative in the sense that it will overestimate the flow rate.

It can now be concluded that the air flux, q_{air} , through the cover with thickness Δz of the land-

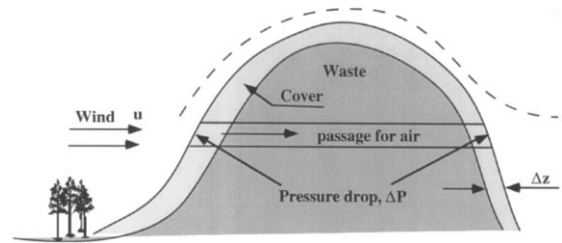


Fig. 9. Landfill created as a hill with potential build-up of hydraulic gradients.

fill can be estimated by:

$$q_{\text{air}} = - \frac{k \cdot k_{\text{rel}}}{\mu_{\text{air}}} \frac{\Delta P}{\Delta z} \tag{18}$$

where k_{rel} is the relative gas permeability which is a function of the degree of saturation (see Fig. 11), k is the permeability of the waste material (m^2) only (the intrinsic permeability), μ_{air} is the viscosity of air (Ns m^{-2}) and Δz is the cover thickness of the ingoing and outgoing sides (m). All resistance to airflow is assumed to be in the cover. The rate of oxygen transport inwards to a landfill where it reacts with the organic material will be estimated. Covers with different resistances to air flow will also be explored.

4. Results

In the preceding chapter, the description of oxygen flow into a landfill by various transport mechanisms has been presented. In the following, different cases are described and the results from the calculations are presented. The data used for the scenarios are also given.

4.1. Oxygen intrusion by diffusion into a partially saturated landfill

In our calculations, the effective diffusivity in the air-filled porous landfill is taken to be 10 times less than that in unconfined air. Preliminary calculations show that if the degradation rate constant of the organics is fast, e.g. comparable to that in a well aerated compost, the oxygen concentration will be extremely low already a few decimetres into the landfill. This is in agreement

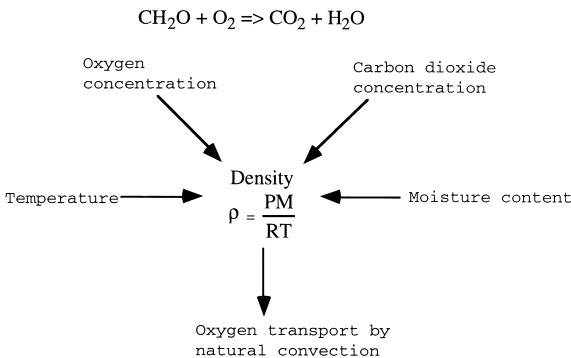


Fig. 8. Entities affecting free convection.

with observations in young landfills. In the humic phase, the reaction rate of the humic substances is expected to be very much lower. It is assumed in these calculations that it is comparable to that of peat which consists of a large amount of humic substances (Bozkurt and Neretnieks, 1998). The kinetic constant, k' , in Eq. (6) with respect to humic matter depletion is then taken to be 10 year^{-1} . This is a value that is within the range observed in peat bogs. The amount of humic substances is taken to be 100 kg m^{-3} landfill.

4.1.1. Partially saturated landfill without and with a cover

In this case, the landfill is assumed to be partially water-saturated and located over the water table. It is seen in Fig. 6 that a cover can considerably decrease the oxygen concentration and thus the influx of oxygen. Nevertheless, even with a cover that has 100 times lower oxygen diffusivity, there will be oxygen in the pores of the landfill. The low reaction rate of the humic substances will not be able to scavenge all oxygen. Other faster reacting reducing substances such as sulphide minerals will then be able to be oxidised and release the toxic metals. The influx of oxygen for the partially saturated case without a cover is $1.5 \text{ kg m}^{-2} \text{ year}^{-1}$. For a 10-m deep deposit, with this influx rate it would take more than 600 years for the humic matter to be completely exhausted. Lines B and C in Fig. 6 are for a 1-m thick cover with effective diffusivities of $D_1 = 2 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$ and $D_1 = 2 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$, respectively. The influx of oxygen for these two cases are $0.29 \text{ kg m}^{-2} \text{ year}^{-1}$ and $0.035 \text{ kg m}^{-2} \text{ year}^{-1}$, respectively, which implies that it would take 3500 and 30 000 respective years for the humic matter to be completely exhausted.

4.2. Air flow by natural convection

Natural convection is caused by the variation of the air (gas) density in the landfill as well as between the inside of the waste landfill and the surroundings. This variation may be due to temperature differences, differences in air humidity and differences in concentration (CO_2/O_2). How these entities affect the magnitude and the direc-

tion of the flow of air will be illustrated in the subsequent sections.

4.2.1. Case 1: Oxygen and carbon dioxide

It is assumed that the temperature and the humidity of the air are constant and only a change in the oxygen/carbon dioxide ratio is considered. The oxygen in the air supplied to the landfill can react with the organic compounds. During the degradation of the organic material, the oxygen is consumed and carbon dioxide is produced according to Eq. (3). In that reaction, heat is released and the air warms up. The air is assumed to consist of 79% nitrogen and 21% oxygen. If all of the oxygen is consumed and replaced by carbon dioxide in the landfill, the air density is increased by approximately 10%, since the molecular weight of carbon dioxide is greater than the molecular weight of oxygen. This increase of air density in the landfill decreases the driving force. When the oxygen is used up, the driving force is decreased and can even be reversed. This means that the air will be transported into the landfill through the *upper part* of the landfill and move downwards. The process is complicated by the fact that the heat produced will partly be used to evaporate liquid water. Water vapour will decrease the air density. This will be addressed later.

4.2.2. Case 2: Temperature

Here the only parameter to be varied is the temperature, while the other parameters are fixed. The surrounding temperature may vary considerably between day and night. A rough yet illustrative picture of the variation between day and night temperatures is shown in Fig. 10.

The heat capacity of the landfilled material makes a landfill less sensitive to altered conditions than its surroundings. Therefore, the temperature inside the landfill is more stable and is assumed to vary only slightly with the ambient temperature. The temperature in the landfill can be higher than the outside temperature due to energy released by the chemical and biochemical reactions. However, on hot days, the outside air can be warmer! The temperature also changes with the different seasons of the year.

Due to the temperature difference between the

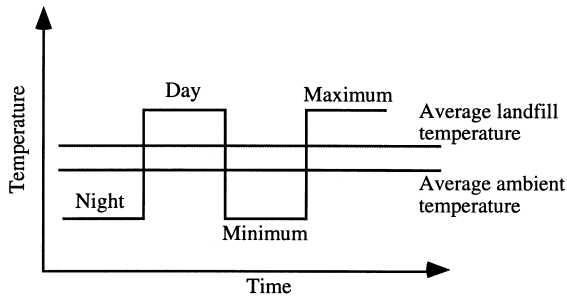


Fig. 10. A schematic chart of the day and night temperature variation.

ambient air and the interior parts of the landfill it will cause a flow of air into the landfill, induced by the buoyancy effects. The natural movement of warmer air upwards creates a 'wind' of hotter air rising through the landfill and causing natural ventilation. This will cause the air to flow into the landfill through the lower part of the deposit and out through the upper part of deposit. On the other hand, since these flows depend on the density differences due to the temperature gradients, the direction of the flow may change as the conditions change.

4.2.3. Case 3: Water vapour in the air

Another aspect to consider is the water vapour in the air. In this case, it is assumed that free water exists at the surface of the waste particles and consequently that the air in the landfill is water-saturated. For a temperature of 15°C (inside the deposit), the gas phase contains 0.0105 kg water kg^{-1} dry air. The percentage relative humidity for the ambient air, on the other hand, is taken as 20% for the same temperature (see Table 2). Then, since the relative humidity difference between the inside and the outside of the landfill is positive, the driving force is upwards, due to the lower density of the water-saturated air. The physical interpretation is that the gas flow into the waste is through the lower part of the landfill and moves upwards.

4.2.4. Summary of the different cases

Due to the complex nature of the system, several simplifications and assumptions have been necessary. The entities have, for example, been

studied independently of each other, although the free convection is simultaneously influenced by heat and mass transfer. When organic matter degrades, heat evolves. This heat will not only influence the temperature inside the landfill but also the air humidity. The rate of heat produced from the depletion of the organic matter is, however, small, and it will have no profound influence on the temperature evolution in the landfill. A summary of the different cases is given in Table 2. The air density ratio between the inside and the outside of the landfill indicates the flow direction of the air into the landfill. For air density ratio below one, the flow is through the lower part and out through the upper part of deposit. On the other hand, if the air density inside the landfill is greater than the ambient air density the flow direction will be the opposite.

For a partially saturated landfill containing waste material with a permeability of 10^{-14} m^2 and where the temperature difference between the inside and the outside of a landfill is 10°C the influx of oxygen is $3 \times 10^{-3} \text{ kg year}^{-1} \text{ m}^{-2}$. With this intrusion rate of oxygen into the deposit, the organic matter would be completely exhausted within approximately 300 000 years.

4.3. Convective air flow (wind-induced)

In the literature, many measurements of permeability made both in the laboratory and in the field are found. The permeability for materials to be used as possible cover materials can range in value over approximately 10 orders of

Table 2

Summary of the different studied cases presented as the air density ratio between the inside and the outside of the landfill

Case	ρ_i/ρ_o
Case 1: Consumption of O_2 and production of CO_2 ($\text{CH}_2\text{O} + \text{O}_2 \rightarrow \text{CO}_2 + \text{H}_2\text{O}$)	1.10
Case 2: Temperature e.g. 1 ($T_i - T_o = 10^\circ\text{C}$) e.g. 2 ($T_i - T_o = -10^\circ\text{C}$)	0.97 1.03
Case 3: Water vapour ($x_i - x_o = 8.5 \times 10^{-3} \text{ kg H}_2\text{O kg}^{-1} \text{ d.a.}$)	0.98

Note. The subscripts i and o denote inside and outside of the landfill.

Table 3
Values of permeability for various types of waste and cover materials

Material	Permeability (m ²)
Waste material	
Ground waste ^a	4×10^{-13} – 7×10^{-11}
Compost fraction ^b	8×10^{-15} – 1×10^{-12}
Ground waste ^c	1×10^{-12} – 1×10^{-11}
Cover material ^d	
Gravel	3×10^{-11} – 3×10^{-9}
Fine sand	2×10^{-14} – 3×10^{-11}
Silt, loess	1×10^{-16} – 2×10^{-12}
Till	1×10^{-19} – 2×10^{-13}
Clay	1×10^{-18} – 4.8×10^{-16}
Unweathered clay	8×10^{-20} – 2×10^{-16}

^aFranzius (1977).

^bRasmuson (1978).

^cOweis and Kehra (1990).

^dDomenico and Schwartz (1990).

magnitude, with the lowest values for unweathered clays and the highest values for gravels. Table 3 shows some representative values of permeability data for different types of waste and cover materials.

The benefit of a cover is twofold. It may act as a barrier against oxygen transport into the landfill, and reduce the amount of percolating water. Here, it is supposed that the waste is covered with a cover with properties such as silt.

The permeability for a clayey silt that could be used for cover material could be as low as 1×10^{-16} m². If silty sand were used, the permeability could be 10 000 times higher. This would mean a hydraulic conductivity for water of 10^{-9} and 10^{-5} m s⁻¹, respectively. In this case it is assumed that the cover has a much lower permeability than the waste and therefore offers all the resistance to flow, i.e.:

$$k_{\text{cover}} \ll k_{\text{waste}} \quad (19)$$

The gas permeability for a partially saturated porous media is a function of the water content. If the water content of the cover is high, its permeability will be very small and the intrusion of oxygen into the landfill will be negligible. During dry seasons, the water content of the cover may decrease and gas may flow through the cover.

The calculations are made for a dry cover with a permeability of 10^{-14} m². Calculations for a partially saturated cover are also presented.

For the air to flow, the pores must be dried out or cracks or other air-filled passages must have developed. It is known that wind does not blow constantly. During some periods of the year, it is calm while in other cases it is strong a breeze. The wind velocity and direction change with time. However, no matter from what direction the wind comes, the landfill will still be exposed to wind since it is assumed that the waste deposit is in the form of a mound. According to Krig (1997), the average wind velocity 10 m above the ground level at Stockholm, Sweden, is of the order of 5 m s⁻¹.

For a wind of 5 m s⁻¹ the influx rate of oxygen through a dry cover with a thickness of 1 m into the landfill would be 60×10^{-3} kg year⁻¹ m⁻². This amount of oxygen is able to deplete the organic matter with a rate of nearly 60 g m⁻² year⁻¹. In other words, it would take more than 15 000 years for the organic matter to be completely exhausted. The estimated value is for a dry landfill cover but, as discussed previously, the permeability depends on the moisture content in a non-linear way. As it can be seen in Fig. 11, the gas permeability decreases rapidly as the degree of saturation of the cover increases and is zero when the cover is saturated. Even when the degree of saturation is in the range of 0.8 or above, the flux of oxygen is virtually negligible.

As an illustrative example, the oxygen flux into the landfill through a cover with a degree of saturation of 50% would be nearly 6×10^{-3} kg m⁻² year⁻¹. This result is for the same wind velocity and cover thickness as in the central case.

4.4. Oxygen transport into a saturated landfill

One method of reducing the oxygen transport rate is to add a low permeability cover on top of the waste. The waste below such a cover will only be partially saturated with water. However, a cover must have a considerably lower diffusivity than the waste itself to substantially decrease the inflow of oxygen. Furthermore, in the long-term a cover will deteriorate and lose some of its pro-

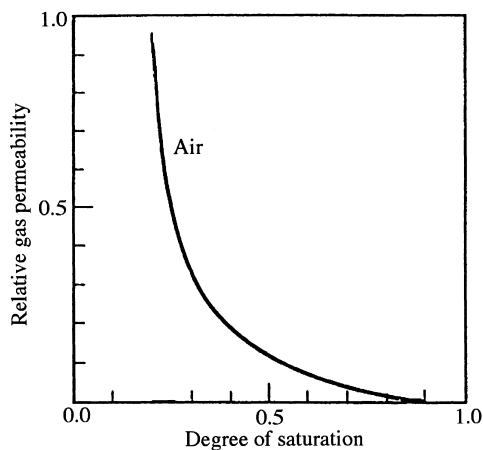


Fig. 11. Relative permeability as a function of degree of saturation.

tection capabilities. An alternative option for reducing the inflow of oxygen is to ensure that the landfill is saturated with water. In essence, wet disposal aims at maintaining the material in an unoxidised form by preventing oxygen access, and also by decreasing water transport through the wastes. The deposition of for example mine tailings under water in natural lakes has been used historically at various places around the world (Ledin and Pedersen, 1995). A general conclusion was that the reactivity of the materials was low and preliminary estimates indicated low metal flux rates.

The idea of a saturated landfill is to lower the oxygen access to the deposit. The oxygen transport rate into a saturated landfill is up to 100 000 times lower than into a partially saturated landfill. The oxygen can then enter practically only with the inflowing water (and ground water) and negligibly by diffusion or convection through gas-filled pores. The influx of oxygen with the seeping rain or groundwater is extremely low because the solubility of oxygen in water is of the order of 10 mg l^{-1} . It rains approximately $500 \text{ mm m}^{-2} \text{ year}^{-1}$ in Sweden. Assuming that only $200 \text{ mm m}^{-2} \text{ year}^{-1}$ rainwater infiltrates, then infiltrating rainwater can carry 0.002 kg of oxygen in to the landfill $\text{m}^{-2} \text{ year}^{-1}$. This amount of oxygen can oxidise $1.9 \times 10^{-3} \text{ kg}$ of organic matter. A saturated landfill would then for practical purposes be considered

to stay reducing for an extremely long time; 1 million years or more. This method may be regarded as a cover method with water acting as the cover. However, in this case, materials that are most easily degraded would probably have to be removed to avoid fermentation in the short-term.

5. Implications and discussion

5.1. How long will it take to exhaust the organic matter?

The amount of organic matter in the studied landfill is 100 kg m^{-3} landfill. Since it was assumed that the organics can be represented by CH_2O , the depletion of the material occurs according to Eq. (3). From the reaction it is deduced that water is produced as an oxidation product. The amount of produced water is $0.6 \text{ g H}_2\text{O g}^{-1}$ degraded CH_2O which is negligible compared to the rainwater infiltration. In the 'Diffusion case without a cover', the production of water corresponds to 0.45% of the infiltrated rainwater. With the oxygen intrusion rates calculated for the different scenarios, the reaction would be completed within the times presented in Table 4. It is important here to point out that these figures should be compared with each other rather than be considered individually. Heterogeneities in the landfill can be expected to strongly influence the exhaustion times of the humics along a pathway. It is not unreasonable to expect that some fast pathway could react up to 10 times faster whereas other would last for much longer times the average.

6. Conclusions

Experiments can be used as a tool to assess metal release in a relatively short perspective, over some decades. They can also indicate the immediate effects of metal releases on landfill operations such as waste mining or excavations for methane extraction in landfills. In the short-term, landfill operations that may affect the geochemistry of the deposit should be performed with caution. Control should be made not only of total concentrations of metals in the waste but

Table 4
Estimated time to deplete all organic matter for the different scenarios, in a 10-m thick landfill

Case	Time to deplete organic material (years)
Diffusion — partially saturated	
without a cover	600
with a cover (10% function)	3000
with a cover	30 000
Forced convection with a cover	> 20 000
Natural convection	300 000
Fully saturated	> 500 000

also of the concentrations in the water phase and, more important, of the acid buffering capacity of the waste after oxidation. The assessment over long times must thus be carried out by models. The experiments, in combination with simulations considering the changing geochemistry of the deposit, have been shown to be a powerful tool to gain insight into the behaviour of a waste deposit over geological times.

With a conceptual model, which is a unitary and continuous description of the overall processes in a waste deposit, as a base a number of scenarios have been studied. Landfills that are made in the form of mounds can result in the creation of high hydraulic gradients. This can lead to a lot of water flowing through the landfill. A final covering layer is often laid on the landfill to decrease the water and air intrusion. However, this means that the landfill will be only partially saturated with water. The transport of oxygen by diffusion in a partially saturated landfill can be thousands of times greater than in a water-saturated landfill. This may accelerate the oxidation of redox-buffering material. If the latter consists partly of organic matter, oxidation will also generate acids, which can form complexes with many heavy metals. The pH buffer may also be depleted by this reaction. A cover must have a considerably lower diffusivity than the landfill material itself to substantially decrease the influx of oxygen. Even so, there will be oxygen in the landfill and sulphides may be oxidised before the oxygen is scavenged by the humics.

The results of the calculations for the different scenarios show that the air flow through the cover caused by natural convection is negligible compared to the air flow caused by forced convection (wind-induced). It may also be concluded that the oxygen transport caused by forced convection may be neglected compared to the diffusional transport. However, depending on the data used, this transport process may be of the same order of magnitude as diffusion.

It was also found that the rate of inflow of oxygen into the deposit is strongly dependent on the degree of water-saturation of the deposit itself and of the covering layer. On the other hand, if the covering layer is biologically active, the oxygen could be scavenged there, until the organics in the scavenging have been exhausted.

Based on the different scenarios presented, it appears that the landfilling situation where the waste is below the water table could have advantages over the other cases. The main conclusion is that the degradation of organic matter and thus the release of toxic substances can be substantially decreased if the potential build-up of hydraulic gradients is avoided and if the landfill is located below the water surface. To substantially decrease the biological activity of a landfill, a conceivable alternative construction would be to place it in a depression — either natural or artificial — and to construct it so that under normal conditions it would always be water-saturated. The location should be an area of natural discharge which means that it receives ground water in contrast to a recharge area which releases water to the ground water flow. However, in this case, materials that are most easily degraded would probably have to be removed to avoid fermentation in the short-term.

We have found that there are many uncertainties and that we have been forced to make several assumptions that can not at present be confirmed. This points to several areas where further research is needed. Some of the main areas are: intrusion rate of oxygen, microbially mediated degradation rate of organics, in organic rich as well as organic poor waste, long-term impact of heterogeneities.

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