Modelling water intrusion impacts on pollutant transport from a municipal solid waste landfill in Sweden

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MODELLING WATER INTRUSION IMPACTS ON POLLUTANT TRANSPORT FROM A MUNICIPAL SOLID WASTE LANDFILL IN SWEDEN

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Aamir Ilyas
June 06, Lund, Sweden
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Summary

Landfills often evoke images of waste pits with obnoxious odours, unsightly vermin, and omnipresent health hazards. This negative view of landfills is associated with the past experience of unregulated land filling, which resulted in soil and water pollution. Today, the land filling is an option of last resort in the new waste management hierarchy. This policy shift may be a good omen for the future but it has not reduced the threats from older landfills, nearing their final closure. Especially, the landfills located at the sea shore or below the ground water table are a severe cause of concern. Their close proximity to sensitive coastal ecosystems and pristine ground waters places a high premium on prevention of leachate migration in the post closure period. Therefore, it is an imperative to assess the long term environmental risks of coastal landfills before their final closure. In this context, the reactive transport models, by allowing us to study the complex interplay of leachate, estuarine and ground waters, over varying times scales, can help predict the long-term impacts of these sites.

The present study attempted to model the impact of sea and ground water intrusion on leachate quality and migration, at the onset of final closure, from the Spillepeng landfill in Malmo, in southern Sweden. This objective was achieved by using a reactive transport code called PHREEQC. This program can simultaneously model solute transport and chemical reactions in 1D with an equilibrium approach. For the present study, leachate quality data were obtained from SYSAV- the regional waste management company, responsible for Spillepeng landfill’s operation and care. The data set contained the analysis results for several leachate quality parameters. The information about the barriers around the landfill, ground water quality, and soil type of the area was obtained from both published and unpublished literature.

For the transport modelling, different scenarios were considered; 1) ground water and seawater flooding the landfill at the onset of final closure period, 2) the leachate migrating from the landfill in the long run, and 3) a worst case scenario was considered by increasing the hydraulic conductivity as a result of changes in porosity of the landfill barrier. Within the aforementioned three scenarios, following processes were modelled with the help of PHREEQC.

I. A two way (1D) transport of a conservative solute (Chloride), through the containment materials.
II. Heterovalent cation exchange reactions between Na and Ca in the barrier matrix and in bottom sediments.
III. Simulation of redox buffer depletion, in bottom sediments, during degradation of DOC coupled to electron acceptors.
IV. Assessment of changes in leachate chemistry induced by seawater intrusion.

The simulation results for conservative transport showed that the seawater can breakthrough the landfill through the barriers within 100 years. The diffusion dominated transport, significantly increased this time span. Even after the 500 years the seawater could reach up to only 8 meters in the barrier. These results also highlighted the potential impact of calcium replacement from exchange sites due to sodium rich water, which causes dispersion of soil aggregates. This could lead to sealing of pore and reductions in hydraulic conductivity. The results of leachate migration, towards the groundwater, showed that leachate can pass through the bottom layer of low permeability sediments in about 30 years time. This time may
seem short, but it also entails initial dilution with ground and seawater, and subsequent changes in leachate chemistry.

The simulation of reactive processes showed that exchange of cations between Na and Ca was the dominant process in both barriers and bottom sediments. The modelling of DOC biodegradation reaction confirmed the usual sequence of redox buffer depletion, often found at the landfill sites. This process was coupled to electron acceptors such as iron (Fe$^{2+}$), Nitrate NO$_3^-$, and Sulphate SO$_4^{2-}$ etc. The iron (Fe$^{2+}$) was mobilised close to the landfill but as leachate proceeded farther-a-field, due to change in redox state of sediments, the iron (Fe$^{3+}$) was precipitated. Similarly NH$_3$ showed a decrease away from the landfill and was probably being transformed to NH$_4^+$. In contrast to sediments, the process of DOC degradation in seawater resulted in strong attenuation of pollutants like Fe$^{2+}$ and NH$_3$, through oxidation and nitrification, mainly due to high dissolved oxygen in seawater.

Keeping in view the above results it was concluded there was no significant risk of leachate migration from Spillepeng, which will continue to trap moisture and would prevent the leachate migration. Therefore, it may not be a serious long term threat to both coastal and ground waters. Several factors support this conclusion. First of all, the water intrusion was extremely slow and therefore would take decades, perhaps centuries, in order to drastically reverse the hydraulic gradient away from the landfill. Secondly, the scenarios of leachate migration are based on worst case with drastic changes in permeability of containment materials. The Na rich seawater was replacing Ca from exchange sites in the barrier. This in reality causes dispersion in soil aggregates and sealing of pores, and therefore further reduction in hydraulic conductivity can be expected. Thirdly, the concentration of pollutants, such as metals, was extremely low. The initial dilution with sea and ground water, and subsequent chemical changes will further decrease the concentrations of pollutants. Finally, even in the worst case of leachate migration, the process of natural attenuation was operating and retarded the movement of pollutants. Nevertheless, in order to reduce the uncertainty, it was suggested that the impact of extreme events such sea level rise, storms, and tidal fluctuations should be considered in future studies.
1. Introduction

There are many waste sites situated on the coasts and close to ground water (Khoury et al., 2000). The reasons for selecting sites at the sea coast or in the former mine pits, are some assumptions, not necessarily tested, regarding both their geotechnical merits and the waste decomposition process. From geotechnical perspective, a former mine pit or a low lying marshy area on a sea coast, due to negative hydraulic gradient, will allow the landfill to trap moisture, and therefore would keep the leachate from leaking out. Similarly, from stabilisation standpoint, the addition of moisture is assumed to enhance the biodegradation process which helps stabilise the waste quickly, and thus reduces the need for after-care. However, all these convincing arguments fail to mitigate the uncertainty regarding the future environmental impacts of coastal landfills. The long emission span and their close proximity to sensitive coastal ecosystem and pristine groundwater are the facts that cast doubts over the wisdom of using the coastlines for landfilling. There are several sources of uncertainty about the impacts of coastal landfills, especially, when these sites will be abandoned.

Firstly, the coastal landfill sites are often places for complex interactions among landfill leachate, estuarian and ground waters. The differences in salinity of sea, ground water will affect the partitioning behaviour of pollutants. Similarly, the high moisture levels may enhance the transport of pollutants from these sites. Secondly, the leachate produced by landfills, is a highly complex mixture of soluble, insoluble, organic, inorganic, ionic, non-ionic, and bacteriological constituents (Philips and Wells, 1974), generated as a result of equally complex physical, chemical and biological processes, occurring in the waste body over long period of time. Finally, a uniform distribution of moisture and decomposition of waste are never achieved. The studies by Bengtsen et al. (1994), and Berge et al. (2005), suggest that due to inherent heterogeneities, in the waste materials, a waste body can act as a dual porosity medium in which major portion of water actually bypasses the bulk of the waste, and moves quickly through the preferential pathways without helping the biodegradation. This essentially means that at the onset of closure period there still may be some dead spots present, in the waste, which in response to rising moisture level would start to degrade. Therefore, the emission span of pollutants may be longer than actually anticipated.

Nowadays, landfills are carefully planned and engineered with improved synthetic liner systems. Therefore provide more control over leachate production and migration. The new waste management concept places the landfill at the bottom rung of management hierarchy. With in the EU, due to rising needs of energy, there is an explicit focus on incineration of waste for energy production, and land filling of inert materials. Thus, in general, the landfilling is currently on the vane. For example, in Sweden the land filling has seen a sharp decline of almost 56% since 1994 (Naturvårdsverket, 2005). Several policy instruments such as focus on reduction/recycling, producer’s responsibility, integration into the EU policy framework, landfill taxation, and energy recovery were used in tandem to harness this decline. Considering these facts a question arises, whether the above mentioned concerns, about the coastal landfills, are over stated? The answer to this question would be negative. As these new management concepts may be a good omen for the future but they offer little or no respite to the environment from the older landfills.
nearing their final closure. Therefore landfilling of municipal solid waste still continues to be a major concern for modern societies.

The control of water pollution, at the landfills sites, poses an ultimate challenge for landfill managers. The leachate produced by landfills carries with it the pollutants present in the waste by dissolving them through a complex series of reactions (Bengtsson, et al., 1994). The problem of water quality deterioration is more complicated and often difficult to remedy, due to wider spatial and temporal scales created by leachate migration. Therefore it is quite important to understand complex interplay of various processes with site hydrogeology in order to reduce water quality impacts of coastal landfills. However, the monitoring of landfills based on frequent sampling and analysis beyond the mandatory monitoring period may not be feasible due to financial and administrative constraints.

In this context, the reactive transport models can provide a useful tool for evaluating environmental hazards (Lichtner, 1996) of the landfills. The coupled models of solute transport and chemical reactions can provide answers about the future impacts of landfills, if used in predictive manner with available data. But the predictive use of reactive transport models is still limited for several reasons. Modelling the leachate transport and its impact on ground and surface water quality is hampered as often comprehensive data about the site are not available, and whatever information is available is not tailored to the need of models. The biggest hurdle that impedes the modelling of landfill is the heterogeneous nature of the waste materials which is often difficult to incorporate into the models. Furthermore, the absence of data for validation of future predictions over longer time scales (i.e. centuries) acts as a disincentive for their use in such cases.

But despite these shortcomings, reactive transport models can still be quite helpful, as management tools, for two reasons. One, by performing the sensitivity analysis, the critical parameters affecting the modelling outcome could be identified for future monitoring, thus making the monitoring campaign more targeted. In addition, these models can help landfill managers in understanding the important natural attenuation processes, controlling the fate of pollutants, which could be harnessed for remediation. Two, the predictions from these models can help determine the actual time needed for post closure care-current requirement is 30 years. This is especially important for development of alternative closure strategies such as opting out of the landfills early which pose lower risks, and focussing resources towards those with higher risks. Thus over all impact could be reduced environmental and financial liability for waste management companies.

In the above context, the application of existing reactive transport models to problems of leachate migration is important as this will not only help judge their suitability, pin point their shortcomings, but also highlight the future areas of action.

1.1. Spillepeng and Malmö

Malmö is the third largest city of Sweden with a population of around 270,000 inhabitants. It is situated on Sweden’s south-western coast in the province of Scania (Skåne). Since its connection with Copenhagen through Oresund Bridge, over the straights of Öresund, the city has become a centre of economic activity in the southern region (www.malmo.se). The study area of Spillepeng landfill is managed by SYSAV—a regional waste management company. This company deals with waste collection,
incineration, land filling and management, in the city of Malmö along with 14 other municipalities in the Scania region. The study area was initially an unplanned waste dump, which in early 1990s was developed into a proper landfill by reclaiming the land from Öresund or the Sound. Spillepeng was constructed as a depression below the sea level and ground water to keep the hydraulic gradient towards landfill, and thereby keeping the leachate from leaking outwards. Whether such a strategy would be successful in the long run or not, was a point of concern for this study.

1.2. Previous work and rationale

Past studies have mainly focussed on the hydrology of Spillepeng, the impact of sludge co-disposal, and the impacts of covers and the waste materials on leachate production. For example, Marques and Hogland (2003), considered the effect of Spillepeng’s construction on leachate production and its potential for pollutant emissions. Their results show that in worst cases, there is a risk of leachate migration through the 10m thick, low permeability sediment layer, and it can reach the limestone aquifer below the study site. But this study has not considered the impact of external influences, such as the effects of moisture addition on leachate chemistry and on pollutant migration in the post closure phase. Therefore, the present thesis attempted to model the impact of water intrusion on pollutant transport in the post closure phase from Spillepeng landfill.

1.3. Objectives

The major objective of the study was to

- Evaluate the impact of water intrusion on pollutant transport from the Spillepeng landfill in post closure phase with the help of the 1D reactive transport model PHREEQC.

While secondary objectives were to

1. model the intrusion of moisture from surroundings towards the landfill
2. model the leachate migration from the landfill towards the sea and the ground water
3. highlight the available attenuation mechanisms at the landfill site
4. evaluate the impacts of seawater on leachate chemistry and the impact of biodegradation on pollutant mobility
5. assess the pollution risk posed by the Spillepeng landfill to ground and seawater quality in the long run
2. Conceptual framework

This chapter aims to outline the major issues such as 1) nature and types of landfills, 2) leachate production and its quality, 3) impact of sea water on waste stability, 4) use of reactive transport models, and 5) process of natural attenuation. The aim is to understand the nature of leachate, its pollution potential, its interactions with different waters, the natural mechanism of attenuation, and explore the scope of reactive transport modelling. To achieve this end, it combines both theory and peer reviewed literature.

2.1. The science of landfills

The advent of present day engineered landfill is not more than half a century old. Today in well developed societies landfilling has evolved as a separate division of technology which combines the techniques of civil, geotechnical, chemical, environmental engineering and landscape architecture. The growing awareness about environmental problems, such as water pollution, has provided an impetus for waste recycling, waste streaming and material recovery in order to reduce the amount of waste destined for landfills. As a result, the landfilling of MSW is on the vane and more focus is on incineration and landfilling of inert ashes rather than solid wastes.

2.2. Modern landfills and their types

In contrast to conventional landfills, modern landfills are managed proactively as complex systems that can degrade the readily degradable materials. These landfills, often called bioreactors, are controlled systems in which moisture and air control are used as enhancements to create an environment capable of actively degrading the readily biodegradable organic fraction of the waste (Reinhart, 2005). The purported advantages of bioreactors include quicker waste stabilization, enhanced gas production, facilitated leachate management, volume reduction and minimized long-term liability. Table 1 gives a comparison of leachate quality which results from both land filling methods.

Table 1: Comparison of conventional and recirculation landfills

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Conventional*</th>
<th>Recirculation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iron (mg/l)</td>
<td>20–2100</td>
<td>4–1095</td>
</tr>
<tr>
<td>BOD (mg/l)</td>
<td>20–40,000</td>
<td>12–28,000</td>
</tr>
<tr>
<td>COD (mg/l)</td>
<td>500–60,000</td>
<td>20–34,560</td>
</tr>
<tr>
<td>Ammonia (mg/l)</td>
<td>30–3000</td>
<td>6–1850</td>
</tr>
<tr>
<td>Chloride (mg/l)</td>
<td>100–5000</td>
<td>9–1884</td>
</tr>
<tr>
<td>Zinc (mg/l)</td>
<td>6–370</td>
<td>0.1–66</td>
</tr>
</tbody>
</table>

Adapted from Reinhart, (2005).

Berge et al., (2005) has used operational conditions and behaviour of nitrogen as a criterion to categorise bioreactors into following four.

2.2.1. Anaerobic

In this type of landfill moisture is added on purpose to decompose waste, and to recover the gas (CH₄) from it. This moisture can come from leachate recirculation and
from ground water or seawater seepage. Excess moisture creates anaerobic conditions, in the landfills, which enhance both biodegradation rates, and methane production. Studies dealing with methane production in both laboratory and the field include Reinhart and Al-Yousfi (1996), Townsend et al. (1996), and Farquhar and Rovers (1973). From these studies it can inferred that the gas production is dependent on moisture content of the waste, which is never evenly distributed in time and space. Usually, more gas is produced in initial stages of landfilling, and less in old landfills which is mainly coming from areas previous unreachable to moisture. Anaerobic landfills have one major disadvantage which is ammonia emission. As leachate is continually recirculated, there is no removal pathway available inside the landfill (Berge et al., 2005). Other problems may come from low temperature and slow degradation rates.

2.2.2. Aerobic

The aerobic degradation of waste results in the production of carbon dioxide and water. Air is added to this kind of landfills to intentionally speed up the degradation process. Stessel and Murphy (1992), have described the concept of aerobic landfill design in detail. Others include Leikam et al. (1999), Murphy et al. (1995), Murthy et al. (2000), and Read et al. (2001). Major advantages of these systems are low metal emission, less ex-situ leachate treatment and reduced environmental liability (Read et al., 2001). Aerobic processes result in increased evaporation due to heat production inside the landfill. The temperatures sometimes can be quite high. Stessel and Murphy (1992), for example, have reported temperatures up to 66°C in such landfills. Therefore, potential advantages of aerobic system are low leachate production, and removal of nitrogen (ammonia) through oxidation pathway.

2.2.3. Hybrids

The combined or hybrid systems alternatingly use both anaerobic and aerobic conditions as landfill management strategy. For example aeration is followed by irrigation with leachate. The benefit of this approach is having best of both worlds. Because there are many advantages associated with both aerobic and anaerobic processes, therefore combining both of them is seen as maximizing the potential of bioreactors (Berge et al., 2005). This sequential aerobic-anaerobic treatment results in faster degradation of easily degradable component of the waste. The advantages include lower organic acids, and earlier onset of methanogenesis.

2.2.4. Facultative

This system relies, on ex-situ treatment of leachate, before recirculation, instead of air injection. In this way, ammonia concentrations are controlled. Therefore, practically, the environment inside the waste cells remains anaerobic because the conversion of ammonia to NH$_4$-N is achieved by external treatment. This NH$_4$-N when recirculated is converted to nitrate through de-nitrification due to anaerobic conditions in waste cell. However, only laboratory studies by Price et al., 2003 have used this method. No major field scale application of this method was available. The major disadvantages include interference in methane production due to de-nitrification, and also reliance on external treatment systems.
2.3. Leachate generation and models

The volume of leachate generated depends upon climate, internal moisture content, recirculation, the nature of the wastes and the water intrusion from the surroundings. In addition, the operation of a landfill—whether a landfill is being operated as a bioreactor or not—determines the amount of leachate generated. As an increasing number of waste sites are being operated as bioreactor sites, therefore operational conditions become important influence on leachate production. Several authors such as Blakey (1992), Bengtsson et al. (1994), Marques and Hogland (2003), and Poulson and Muldrup (2005), have used water balance models ranging from simple to computer based, to predict the generation leachate, and have elucidated key controls on it such as cover materials, waste types, waste age, and water holding capacity etc.

It is clear from these studies that leachate production is clearly a function of these above mentioned factors. Another obvious conclusion drawn from these studies is that despite their efficiency in predicting volumes, the leachate generation models can not predict the quality of leachate. The reason commonly used hydrologic models for landfills do not take into account the biogeochemical processes. Similarly the geochemical codes predict leachate quality but not the quantity. This fragmented nature of modelling for quality and quantity may be the big disincentive for using models in comprehensive environmental risk assessment of landfills.

2.4. Leachate quality

There is a very large amount of literature available that deals with bio geochemical processes inside the landfill and their impact on the leachate quality. For example see Slack et al. (2005), Statom et al. (2004), Christensen et al. (2001), and Bozkurt et al. (2000). The reviews done by Christensen et al. (2001), and Bozkurt et al. (2000), and describe the landfill biogeochemistry and pollutant attenuation processes in detail. The review of Christensen et al. (2001), is based on both laboratory and field data. It is mainly focussed on organic contaminants, xenobiotics, inorganics and heavy metals. It was concluded that:

- DOC in leachate plumes seems to be dominated by fulvic like compounds which seem to degrade over time.
- Ammonium may constitute a significant long term pollutant in leachate plumes, because its concentrations in leachate stay high for extended time,
- Heavy metals are not a major groundwater pollution problem in landfill leachate plumes, because concentrations usually are low in the leachate, and because heavy metals are strongly attenuated by sorption and precipitation.
- Xenobiotic organic compounds in leachate are not very extensively attenuated by sorption onto aquifer material

Further more their review underscores the importance of natural attenuation which was significant in retarding the majority of the contaminants. However, much remains to be learned about natural attenuation processes in order to develop coherent risk assessment and remedial measures.

Based on their age and related biochemical process, landfill often undergoes several distinct phases (Pohland and Kim 2001). However two phases are important influence on pollutant transport. The initial acidic phase, with a high content of easily degradable organics and the final methanogenic phase with low amount of degradable organics. According to Statom et al. (2004), the later phase results in mobilisation of
metals (Mn and Fe) along with ammonia. Their study is based on 12 years of data from a landfill in Florida. The dominant quality type of leachate in post closure was as Cl-Na-HCO₃⁻, with some short term variations caused by rainfall events.

These studies highlight the fact that landfills are sites of most complex physical, chemical and biological processes. The water moving through the landfill not only forms an integral part of degradation but also washes down the products of this environmental catabolism. The landfill leachate consists of large number of pollutants such organic, inorganic and heavy metals. The dominant types of pollutants coming from landfill depend on type, management and age of landfill.

2.5. Impacts of seawater intrusion

Moisture addition is a beneficial activity as it enhances the biodegradation and helps to stabilise the waste. Despite the beneficial nature of moisture, in general, the research about seawater impacts on this process is lacking. There are two possible interactions and impacts of sea water. For MSW cells, the role of seawater in affecting the biodegradation would be important criterion for judgement. While in case of hazardous or largely inorganic waste, from incineration plants, the leaching behaviour of trace elements would important to phenomena to study. The questions that one should consider could be as following.

1. How will seawater influence the chemistry of landfill?
2. What does this seawater intrusion mean for potential migration of contaminants from the landfill in the long run?

Here, some general impacts of seawater, based on available literature, can be outlined as follows.

2.5.1. Settlement problems

Studies by El-Fadil et al. (1997), (1999), and (2000), have reported settlement, in laboratory waste columns, due to biodegradation and gas production as a result of seawater addition. El-Fadil et al. (1997), reported that the biodegradation increased the void space of the waste materials. Also, the rates of biodegradation were not uniform, this resulted in differential settlement of the waste due to over burden. However these results have not been tested in field, and do not show very significant pattern of settlement as a result of seawater intrusion. The increasing moisture, not necessarily from sea, has been reported to increase the pore pressures inside the waste body, and cause the resultant slope failures of the landfills. The pore pressure can increase greatly especially where forced entry of leachate is made for recirculation (Hendron et al., 1999). Thus enhanced biodegradation with increased void spaces and pore pressures may cause settlements and slope failures in landfills.

2.5.2. Impacts on biodegradation

The moisture along with settlement can have impacts on biodegradation and pollutant emission from landfill. Therefore its role should be further investigated in order to improve the existing models of biodegradation and leachate quality (El-Fadel et al., 1997, Khoury et al., 2000 and Fellner et al., 2003). The landfill sites in coastal areas have hydraulic connections with coastal sea or estuarine waters. This will affect the
degradation and transport of contaminant. However, the studies on impact of seawater on biodegradation and stabilisation of waste are also rare (Khoury et al., 2000, and Koga et al., 2003).

One obvious impact of seawater on leachate chemistry is pH buffering. The seawater being alkaline, has buffering capacity and is often used in industry to neutralise the bauxite residues (Menzies et al., 2004). In this sense, the seawater can provide two obvious benefits. One, being highly alkaline buffers the pH and thereby reduces the chances of metal leaching. Second, the moisture enhances the rate of biodegradation and can provide early stabilization.

However, increased moisture content creates anaerobic environment and this removes the nitrification pathway for the ammonium (Townsend et al., 1996 and Berge et al., 2005). Similarly, iron (Fe³⁺) can undergo reductive dissolution by acting as electron acceptor for organic matter oxidation. The FeOOHs are often sorption sites and their dissolution can release the metals like arsenic sorbed on them (Cummings et al, 1999, Nickson et al., 2000 and 2005). Therefore the nitrogen (NH₃) and redox sensitive metals (Fe³⁺) might be uncertain parameters in case of seawater intrusion, as due to anaerobic conditions both will be more mobile.

2.5.3. Impacts on inorganic wastes

The incinerator ashes, especially fly ashes, contain high levels of heavy metals, and salts are usually classified as hazardous waste. However the impacts of seawater on inorganic ashes are rather unclear. In one study Koga et al. (2003), have shown that due to mixing of seawater with bottom ash the flocculation occurred in finer particles. This flocculation was attributed to increasing ionic strength and pH as a result of this mixing. The minerals and alkalis in bottom ash, when dissolved, increased the pH of seawater. This rise in turn increased the flocculation. However this was studied in laboratory and bottom ash was directly dumped in to the seawater. But the floc formation, in the landfill, though not at the above mentioned scale, could result in blockage of pores and drains. Thus reduction in pore space and drain blockage could be regarded as beneficial for reduction in risk of leachate migration from the landfill.

For metal mobility, as a result of seawater, the age of bottom ash materials is very important. Bottom ashes are product of incineration and are quite reactive in nature. Polletini et al. (2005), reported that due to inherent chemical instability these materials can undergo weathering reactions when exposed to atmospheric agents. These reactions include hydrolysis, sorption/precipitation, carbonation, oxidation/reduction, and complexation etc. Studies on weathering of bottom ashes due to atmospheric agents such as air, nitrogen gas etc are quite numerous.(For example see Zevenbergen et al., 1998; Freyssinet et al.,2002; Sabbas et al.,2003;Polletini and Pomi,2004; and Lin and Chang, 2006.)

The master variables like pH and redox, and other variables such as sorption, acid neutralisation and exchange capacities would be the key to explain the changes in their chemistry as result of seawater intrusion. Apparently, high pH seawater would stop the acidification, and the alkaline pH will help reduce the mobilisation of metals. However, the extended period of submergence in seawater means low redox potentials, and this might affect the metal such as Fe³⁺, Mn etc. This mobilisation, in absence of pH reduction, is microbiologically mediated as bacteria oxidises the organic matter and utilises these metals as electron acceptor. Similarly the high amount of chloride and
colloidal materials would provide complexation avenues and increase the metal mobility, though may reduce their toxic effects. Also, the content of iron oxide minerals, such as, hematite, macknawhite both in seawater and waste is equally important. The iron oxide minerals provide sorption sites and can affect the precipitation of metals. Table 2.2 summarises the impacts of seawater on leachate chemistry.

Table 2.2. Seawater impacts on biodegradation

<table>
<thead>
<tr>
<th>Favourable</th>
<th>Unfavourable</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture addition</td>
<td>Sulphates</td>
</tr>
<tr>
<td>Nutrients</td>
<td>High salinity</td>
</tr>
<tr>
<td>pH Buffering</td>
<td>High osmotic pressure</td>
</tr>
</tbody>
</table>

(Adapted from El-Fadel et al., 1997)

2.6. Reactive transport models

The contaminants in the ground water can move by principle of mass transport. There are three basic processes (i.e. Advection, Diffusion and Dispersion) that are involved in mass transport of contaminants. Along with this, there are other processes, both of physical and chemical nature, which can cause retardation in the flow of contaminants. The models for reactive transport usually solve the advective-dispersive transport equation together, and then chemical reaction later (Appelo et al., 1997). PHREEQC by Parkhurst et al. (1995), used in this study, is based on the following equation to calculate the contaminant transport.

\[
\frac{\partial C}{\partial t} = -v \frac{\partial C}{\partial x} + D_L \frac{\partial^2 C}{\partial x^2} - \frac{\partial c}{\partial t}
\]  
\[2.1\]

The first term, on the right, in the equation is advection; the second one is dispersion, and the third represents a change in concentration with time. Where \(C\) is concentration (mol/kgw), \(t\) is time (s), \(v\) is pore velocity m/s, \(x\) is the distance (m), and \(D_L\) (m²/s) is the hydrodynamic dispersion coefficient (see 3.4). There are different numerical schemes to solve the above equation \[2.1\]. For ground water or porous medium finite elements/differences is preferred. For example in PHREEQC the transport part of A-D-E is solved with an explicit finite difference scheme.

2.6.1. Applications of reactive transport models

There is wide ranging application of reactive transport models. However, in most case these models have been applied to highlight the underlying complex processes involved in reactive transport of solutes. The use of these models for predictive application is still limited. The one single most discouraging aspect of these models is near logical impossibility that outcome of these models is true for natural conditions.

The other impediment for reactive transport is lack of data for comparison in case of long term predictions. Also most of the available models either consider biodegradation or inorganic geochemical reactions. In case of biodegradation its inclusion in the model is necessary as it exerts a major control in the landfill over alkalinity and pH of pore water solution. Similarly, microbial growth causes reduction
in hydraulic conductivity of the soil by three orders of magnitude, and affects the metal precipitation (Rittman et al., 1996 and Rowe et al., 1998).

Nonetheless, the reactive transport model can provide a useful tool for evaluating various environmental hazards (Lichtner, 1996). The problems of accuracy can be dealt with sensitivity analysis, as this would help bracket the important parameters that can be studies further. Above all, using the reactive transport models for prediction regarding leachate migration can help underline the important processes that controlling the fate of pollutants. These processes could be harnessed either for future remediation or for determining the length of post closure care. Finally, the application reactive transport models in such cases would help identify the important deficiencies in the models to be improved in future.

2.7. Attenuation processes

As pollutants in the leachate start to migrate, in the subsurface environment, they undergo many different processes which can retard or slow their movement. Although, there may be some which actually increase their mobility such as complexation. However the dominant ones are that slow their migration and they are collectively called as retardation or attenuation mechanisms. Figure 2.1, represents the hypothetical case of plume retardation in soil medium. These processes determine the spatial and temporal extent of pollutant plume migration in the subsurface environment. The major processes are dilution with ground water, dispersion, redox buffering, ion exchange, and adsorption etc.

Figure 2.1: Hypothetical case of retardation in subsurface(Yong R.N. 2001).

2.7.1. Dilution with groundwater flow

The dilution of leachate with surface or ground water may decrease the L/S ratios and thereby increase the surface areas available for sorption. Along with dilution there is phenomena of water mounds formation below the landfills due to groundwater flow conditions. Christensen et al. (2001), has reported that groundwater mounds may develop below the landfill due to low hydraulic conductivities and higher infiltration at the borders of mounding areas. This mounding can cause lateral spreading of contaminant which enhances the dilution. The dilution as a physical phenomenon is dependent on the viscosity and the density of leachate. The longitudinal dispersivity governs the concentration at the front end of leachate, while transverse dispersivity is responsible for lateral spreading of plume (Christensen et al., 2001). The development of leachate plume is a result of above factors and most plumes have length of 1000 m
and very few exceed 2000m due to dilution (Christensen et al., 2001). However this mixing and dilution is all controlled by the local hydrogeology.

2.7.2. Exchange reactions

The cation exchange capacity (CEC) is a major determinant of cation attenuation, and in aquifers with high CECs the attenuation can be significant. Several researchers have reported the role of ion exchange in attenuation of pollutants in ground waters. However the retardation by exchange depends on selectivity coefficient of two competing ions, their concentrations, exchange sites and CEC of the material itself (Appelo and Postma, 1999). For example K+ and NH4+ will be retarded significantly while Na will be the least attenuated due to ion exchange. Similarly, Ca2+ and Mg2+ will be affected by the complexation and dissolution/precipitation reactions. In case of Fe2+ and Mn the precipitation of sulphides, ion exchange oxidation and dilution are important.

2.7.3. Redox processes

The redox processes inside a landfill and development of redox front have been discussed by Crawford and Neretnieks (1999), and Bozkurt et al. (2000). They utilised PHREEQC model to model the development of redox front in a hypothetical landfill cell. By using a simplified situation with O2 transport along the infiltrating water, the time of redox buffer depletion was calculated. The results show that redox depletion is much slower in advection than in diffusion, owing to high diffusivity of oxygen. For redox development away from the landfill studies by Chappelle, et al. (1995), and Heron et al. (1998) provide the information about sensitive species, electron acceptors and H2 in ground waters. Generally, the redox potential increases with distance from landfill. The methanogenic zone is close to landfill which can deplete the dissolved electron acceptors such as O2, NO3- and SO4. The overall patterns is series of redox zones and from reduced to more oxidizing environment.

2.7.3.1. Redox buffering

Scott and Morgan (1990) have defined it as the capacity of aquifer materials to accept electrons. By virtue of this definition, the leachate because of its reduced state can be viewed as an electron donor (reduction capacity). Therefore produced electrons must be accepted by dissolved or solid aquifer- electron acceptors(Bjerg et al., 2003). Several electron acceptors can take part in this process. However which acceptor dominates the process is dependent on availability of particular element. Usually in the absence of pollutants like nitrate, the redox buffering may be dominated by Fe oxides in the aquifers. The other process that affects buffering is dilution. Heron and Christensen (1995), reported that in low dilution actually the amount and reactivity of Fe and Mn oxides controls the formation of reduced plume. In case of lime stones the plumes may move up to very long distance due to limited retardation. Similarly sulphate reduction may be insignificant due to its absence in leachate and aquifer. In glacio-fluvial sediments there is uncertainty about iron reduction as there is little evidence available to support this argument.
3. Methodology

This chapter outlines the materials and methods used for the present study, which was conducted on Spillepeng landfill situated in the city of Malmö, in south western region of Sweden. The main objective was to evaluate the risk of pollution to ground and surface water due to leachate by using a 1D reactive transport model. The data were obtained from secondary sources both published and unpublished. For site description Bevmo and Evertsson (1999), Marquis and Hogland (2003), Bengtsson et al. (1994), and SWECO (1999) (Unpublished) were consulted.

3.1. Hydro-geological character of the site

The landfill at Spillepeng was developed into a proper landfill from marshy dumping ground by SYSAV in 1976. In 1986 the company was granted with permission to reclaim land from the sea, for the land filling, and since then it has been extended three times. The new Spillepeng landfill started operations in 1990s. The landfill site is in close proximity to the city of Malmö and to the incineration plant which is seen as advantage (Bevmo and Evertsson, 1999). The present day area of Spillepeng is about 120 ha with 55 hectares on the sea floor. The average height of landfill is 32 m above the MSL with storage capacity of around 9 million m³. The Spillepeng also serves as a recreational area thanks to landscaping done there.

![The Spillepeng landfill](image)

Figure 3.1. The Spillepeng landfill (Source: Google Earth: visited 24th April 2006)

The ground water around Spillepeng exists in two aquifers. The top aquifer in glacial sediments is open to atmosphere and has a three meter depth. Immediately below this aquifer there is a thick layer of clay which extends seawards. This thick clay layer acts as an aquiclue or aquitard, due to low permeability, has created a leaky confined aquifer inside the fractured bedrock of limestone. The table 1 gives the summary of main hydro-geologic characters landfill site.
Table 3.1. Some important hydro-geological parameters of the landfill site.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Boulder Clay</th>
<th>Fill-material</th>
<th>Waste</th>
<th>Shoulders</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>$5 \times 10^{-9}$ m/s</td>
<td>$1 \times 10^{-7}$ m/s</td>
<td>$1 \times 10^{-6}$ m/s</td>
<td>$2 \times 10^{-9}$ m/s</td>
</tr>
<tr>
<td>Effective Porosity</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.16</td>
</tr>
</tbody>
</table>

The low permeability clay layer between the two aquifers seems saturated due to its leaky confined nature. The upward seepage from this layer into the landfill is collected in the bottom drains beneath the landfill cells. The regional ground water flow direction is towards the Öresund or the Sound (Figure 3.2). There is no reported incident of seawater intrusion along the south western coast of Sweden. This could be mainly due to lower dependence of urban water supply on ground water in the study area.

The Spillepeng landfill was constructed as a depression, in the ground, which can trap water from surroundings. This was aimed at minimising the chances of leakage from the landfill towards sea or groundwater during post closure period. To stop the water from moving into the landfill, during operational and monitoring phase, ground water pumps have been installed. This pumping has kept the water table 2-3 m below the sea level at the site. However after the monitoring phase is over and pumping stops, the water will start filling the landfill. This seepage of water from surroundings towards the landfill will increase the water table inside the landfill which has been estimated to rise up to 3m in the middle part of the landfill (SWECO, 1999). The soil around the landfill is composed of organic topsoil, postglacial sands and boulder clay (10m). The bed rock is part of Danian or Copenhagen lime stone system which is a dense limestone with local clay lenses, sand and flint bars.
3.1.2. Barriers/earth dams

The reclaimed area of Spillepeng is enclosed by earth dams of 50 meter thickness at the base, and 7m width at the top. A road has been constructed on the top for waste hauling. The slope of earth dams is protected, from sea water, erosion with layers of geo-textile and large stones. The barrier walls are 3 m above the sea level while 10m below it. The earth dam consists of two portions with different hydraulic conductivities. The interior mass of the earth dam has an average thickness of 23 m and it is composed of sandy loam soil retrieved from surroundings and constructions sites all over the region. This interior mass is then covered on both sides with compacted 1.5m thick shoulders of boulder clay (Figure 3.3). The volume of fill material amounts to almost 800,000 tonnes (Bevmo and Evertsson, 1999).

![Cross section of barrier/earth dams](image)

The earth dams and landfill are lying on sea floor which is also mostly boulder clay with 10 m thickness over the bed rock of limestone. To prevent seepage from below the clay (2-3 meter) of glacial origin was used to compact the bottom surface of landfill.

3.2. Data sources

The data for the present study were obtained from secondary sources. The data about the landfill hydrogeology, ground water quality in surroundings and leachate quality were obtained from local companies SWECO and SYSAV- responsible for Spillepeng’s design, construction and management respectively. However, where possible the information was supplemented with published sources as well. The leachate quality data contained results of leachate monitoring period from 1991-2005. The data from two sampling points (P2 and P9) were used. These points (P2 and P9) represented the two different waste composition and conditions. The point P2 collected leachate from the MSW cells. (See figure 1).These cells have also been receiving sludge from treatment plants during operational years. The total of area of the cells was around 5ha. The point P9 collected leachate from the hazardous waste cells (Figure 1).

The data for sea water quality were retrieved from The Oresund’s Water Management Society (Öresunds Vattenvårdsförbund) reports, available on the internet (http://www.oresunds- vvf.se/). But these reports did not include the analyses for some major ions such as chloride, and also metals. Therefore, the data for sea water chemistry was modified, which may not necessarily accurately represent the quality of seawater in the Öresund straits. The data for leachate quality and groundwater showed variability with regard to sampling which was not uniformly distributed in time. The data were taken at random intervals and not all the parameters were tested.
at every interval. Some important parameters like sodium were missing. These missing parameters hampered the charge balance calculations, and also the quality analysis of the modelling results.

3.3. PHREEQC

The reactive transport code **PHREEQC** was used for modelling leachate transport and exchange flows. This was originally developed by Parkhurst et al. (1980), with the name **PHREEQE** and then later modified to version **PHREEQC** by Parkhurst in 1995. For this study, the latest version of the program **PHREEQC** (2.12.5) was used. This version was substantially modified in 1999 by David L. Parkhurst and Appelo J. Postma, and has a graphical user interface (GUI). **PHREEQC** is basically an equilibrium code with capability of modelling reactive transport through porous media in one dimension. It uses two thermodynamic data bases **phreeqc.dat** and **wateq4f.dat**. The previous version of **PHREEQC** was written in **FORTRAN** language but the current version is written in the **C** language. However, to specify the reaction rates, for kinetic modelling, the **BASIC** language code is also used.

**PHREEQC** can perform a multitude of chemical reaction calculations ranging from speciation to inverse modelling. The reactions can be performed by choosing different phases like aqueous, mineral, gas and solid-solution depending upon the nature of problem. The transport modelling is based on either advective or advective-dispersive transport in 1D. This transport in columns can incorporate stagnant zones, kinetic reactions, non ideal or binary solutions and gas phases. Another important feature of **PHREEQC** is inverse modelling which is useful to infer the initial water chemistry from final water composition.

3.4. Model building

Keeping in view the construction of Spillepeng, as a site to trap water from surrounding areas, two scenarios were considered for model building. In first scenario, the intrusion of seawater and ground water into the landfill cell was modelled at the start of post closure period. Figure 3.4, shows that it will take approx 13 years before the water reaches height of 3m inside the landfill.

Figure 3.4. Water Balance in post closure phase. (Data is from SWECO, 1999).
The flat end of the curve in the above between years 2 and 3 probably shows the storage effects as water table starts to rise in waste body after the pumping.

**Scenario 1: Intrusion of seawater and ground water**

The first scenario was seepage of seawater into the landfill through the barrier (See Figure 3.5).

![Figure 3.5. Schematic representation of water intrusion into the landfill in post closure phase](image)

The actual data about the barrier composition was not available therefore number sources were used to make inferences about the soil type, its exchange capacity and its solution chemistry. The soil for barriers was collected from surrounding areas and from construction sites all over the region. This fact helped to determine the general soil type. For physical attributes, the soil analysis results reported by Arvidsson (2001) were used. The dominant soil type was sandy loam with calcareous nature. This type of soil, according to the soil classification system developed by Food & Agriculture Organisation (FAO), of United Nations, belongs to Eutric Cambisol group. The clay content of this type was estimated 120-200gm/kg. For the soil solution chemistry, the data were obtained, and modified, from Misra and Tyler (1999).

<table>
<thead>
<tr>
<th>Soil Type*</th>
<th>Clay %</th>
<th>pH</th>
<th>Org. C %</th>
<th>Tot. N %</th>
<th>Exch. K cmol/kg</th>
<th>CEC cmol/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eutric Cambisol</td>
<td>23.3</td>
<td>6.7</td>
<td>1.07</td>
<td>0.109</td>
<td>1.86</td>
<td>12.88</td>
</tr>
</tbody>
</table>

* Based on Arvidsson (2001) and FAO’s world soil classification system.

For transport simulation, the chloride was used as a conservative solute to assess the break through time for seawater. The barrier length was discretized into 1m cells and effects of dispersion (Eqs 3.1 & 3.2) were included. Based on the information given in
Table 3.5 the pore velocity and number of shifts—the time solute would take to pass through the cells, were calculated. The processes such as mixing, exchange, sorption, etc were considered for reactive solutes, and were defined in cells. The hydraulic conductivity values were averaged for entire barrier. Similarly, averaged values of leachate quality data were used in the input files. The overall modelling approach was based on equilibrium concept. The table 3.5 gives the summary of important transport parameters.

Table 3.3. Important transport parameters used for modelling

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Scenario 1+2*</th>
<th>Worst Case</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>4.58*10^{-8}</td>
<td>6.3*10^{-6}</td>
</tr>
<tr>
<td>Pore Velocity</td>
<td>11.11 m/y</td>
<td>14.44 m/y</td>
</tr>
<tr>
<td>Effective porosity</td>
<td>0.13</td>
<td>0.16</td>
</tr>
<tr>
<td>Time step</td>
<td>3 years</td>
<td>2.5</td>
</tr>
<tr>
<td>Number of shifts</td>
<td>10</td>
<td>12</td>
</tr>
<tr>
<td>Diffusion Coeff.</td>
<td>0.3*10^{-9} m²/s</td>
<td>1*10^{-9} m²/s</td>
</tr>
<tr>
<td>Dispersion Coeff.</td>
<td>0.06m</td>
<td>0.06</td>
</tr>
</tbody>
</table>

* Scenario 2. Values are same only difference is of process which is diffusion only.

The dispersion in PHREEQC was calculated as hydrodynamic dispersion which combines both effective diffusion and longitudinal dispersion. This was calculated with help of equations given by Fetter, (2001).

\[
D_L = D^* + a_L V \quad \text{[3.1]}
\]

Here \(D_L\) is the coefficient \(\text{[m}^2/\text{s}]\) and \(D^*\) is effective diffusion coefficient which is estimated by taking account into the porosity of the medium. While \(a_L\) is a coefficient of longitudinal dispersion, \(L\) is length of flow path (m) and \(v\) is pore velocity (m/s).

\[
a_L = 0.83 (\log L)^{2.414} \quad \text{[3.2]}
\]

Scenario 2: Migration of leachate

This scenario envisions a period when a result of water intrusion from the surrounding the water table inside the landfill has risen up to reach 3 meters, well above the sea level. This rise in water table inside the landfill will reverse the hydraulic gradient, and therefore possibility of leachate migration from the landfill was considered. This was achieved by modelling the reactive transport of leachate towards sea and groundwater (figure 3.6). However, the main transport parameters for this exercise were same as previous case except the solute concentrations, and change in the direction of solute movement.
Attenuation processes

The impacts of attenuation processes such as exchange were included in the transport calculation by providing the estimated exchange capacity of porous material. The capacity of exchanger was determined by determining the CEC value for the barrier material. In order to reduce uncertainty, two different values were determined; first, by using the soil type and its given exchange capacity (FAO’s system), and second by using an empirical formula (Eq. 3.2) given by Breeuwsma et al. (1986). The values obtained from these two methods were compared and were found to be similar. The CEC value of the barrier placed it in Kaolin group. This corroborated the initial assumption of sandy loam soil with around 20% clay content and a non-swelling behaviour. Table 3.4 gives exchange capacities and surface areas of different minerals.

\[
\text{CEC} \left[ \text{meq/ (100 g)} \right] = 0.7(\%\text{clay}) + 3.5(\%\text{C}) \quad [3.3]
\]

<table>
<thead>
<tr>
<th>Material</th>
<th>CEC (meq/100g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Humus</td>
<td>200</td>
</tr>
<tr>
<td>Smectites</td>
<td>80-100</td>
</tr>
<tr>
<td>Vermiculites</td>
<td>120-200</td>
</tr>
<tr>
<td>Montmorillonite</td>
<td>70-95</td>
</tr>
<tr>
<td>Illites</td>
<td>10-40</td>
</tr>
<tr>
<td>Kaolins</td>
<td>1-15</td>
</tr>
<tr>
<td>Chlorite</td>
<td>&lt;10</td>
</tr>
</tbody>
</table>

Source: Modified from Drever (1982), and Tan, (1998).

The exchange reaction was assumed between Na and Ca, where Na replaces the Ca from exchange sites in the barrier. This assumption was based on the high sodium content of seawater and also from the studies of salinization in aquifers affected by seawater intrusion. The half reaction (Eq. 3.4) is given by Parkhurst and Appelo (1999), and PHREEQC models it as ion association reaction according to Gains-

\[
\text{Table 3.4 Cation Exchange capacities of clay minerals and other materials (Meq/100g)}
\]
Thomas convention. The thermodynamic constants are included in both data bases \textit{phreeqc.dat} and \textit{wateq4f.dat} of the program.

\[
\text{Na}^+ + \text{X}^- \leftrightarrow 4\text{NaX} \quad K_{\text{NaX}} \quad [3.4]
\]

Where X\(^-\) is the exchanger defined as meq/100g and \(K_{\text{NaX}}\) is the thermodynamic constant.

\textbf{Worst case scenario}

Finally, the worst case scenarios were modelled, by assuming the increase in hydraulic conductivity values due to frost action, settlement, mineral dissolution and erosion etc. However the impact of storm surges and sea level rise were not included as model did not allow it. For worst case, the hydraulic conductivity values were increased in two increments, such as 30 and 50\% while keeping the other parameters same as previous case.

In order to assess the impact of leachate on seawater and ground water, the transport of leachate along with redox depletion due to organic matter oxidation in tandem with electron acceptors was modelled. This was achieved by modelling the degradation of CH\(_2\)O in incremental reaction steps. The electron acceptors such as O\(_2\), Fe\(^{2+}\), SO\(_4\), and NO\(_3\) were included in the model. However amount of reactant CH\(_2\)O was taken hypothetically as leachate quality data did not provide any information about this. In addition, the movement of leachate through the barrier, under worse conditions, and its impact on seawater quality was modelled.
4. Results and discussions

This chapter would describe and discuss the results of, 1) assessment of seawater intrusion, 2) important reactions of solute in the barrier, 3) worst case scenarios 4) depletion of redox buffer, and 5) evaluation of natural attenuation process.

4.1. General character of leachate

The table 4.1 presents the leachate quality data for a waste cell containing mostly MSW which has been closed since 1994. The leachate samples were collected from 1994 to 2004 and analysed for the parameters given below. The values given in the table(4.1) are simple averages of test results and from the data it can be inferred that only one sample was taken at each random interval.

Table 4.1. Leachate quality data (mg/l) from a (MSW) waste cell.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Average</th>
<th>Min</th>
<th>Max</th>
<th>S.D.</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.30</td>
<td>6.40</td>
<td>8.50</td>
<td>0.34</td>
</tr>
<tr>
<td>EC</td>
<td>1500</td>
<td>261</td>
<td>4020</td>
<td>576.34</td>
</tr>
<tr>
<td>SS</td>
<td>120</td>
<td>10</td>
<td>1600</td>
<td>258.73</td>
</tr>
<tr>
<td>Cl</td>
<td>3700</td>
<td>1500</td>
<td>32900</td>
<td>4738.30</td>
</tr>
<tr>
<td>BOD_7</td>
<td>40</td>
<td>17</td>
<td>8200</td>
<td>1529.83</td>
</tr>
<tr>
<td>COD_Cr</td>
<td>740</td>
<td>360</td>
<td>11000</td>
<td>2178.53</td>
</tr>
<tr>
<td>TOC</td>
<td>200</td>
<td>72</td>
<td>6200</td>
<td>1180.82</td>
</tr>
</tbody>
</table>

As shown in the results, the leachate had higher values of salts, biological oxygen demand, and ammonia. It seems that cells are progressing towards the methanogenic phase. The values of leachate quality indicator in the above table correspond to the ranges reported by Christensen et al. (2001). The metal concentrations were generally in low range and only exception was iron which seemed moderately high. The data values from hazardous waste cell showed a similar trend in case of metals and nutrients. However other parameters such as chloride, conductivity, and BOD were quite high in this case. This could be due the fact that the cell was still operational.
The low metal content could be explained by presence of the bottom ash from incineration plants, and also the neutral pH which favours adsorption. Similarly slaked lime, used for removal of gases, was deposited here which has known quality of scavenging metals, and therefore may have played its role.

4.2. Assessment of water intrusion

As mentioned in the methodology that ground water in study area was controlled by pumping, and is currently kept 2 meter below the sea level. At the onset of final closure, pumping will stop and ground water will rise up to a height of 3m inside the landfill. Therefore, due to the hydraulic gradient towards the landfill, first the intrusion of seawater into the landfill cell was modelled.

Model conditions

- Barrier width: 37 meters (average) discretized into 1m cells
- Boundary conditions: Flux−Flux (Neumann Boundary)
- Time step calculated: 3 years
- Time Intervals: 30, 60, 100 years
- Flow Conditions 1: Forward (Advective-Dispersive)
- Flow conditions 2: Diffusion only (For longer than 100 years intervals)
- Solute: Chloride for conservative transport
- For Exchange: Na vs. Ca

Assumptions

1. The barrier/earth dam around the landfill has a uniform hydraulic conductivity
2. The barrier soil resembles the dominant soil type of the region in physical and chemical character.
3. The barrier is saturated and pore water chemistry represents calcareous soil
4. At longer time intervals diffusion dominated transport would occur as the differences in hydraulic gradient would not be significant.

The longitudinal dispersion along the flow path was calculated, after Fetter (2001), which was quite low in this case (.06 m). The pore water in the barrier was assumed to be in equilibrium with calcite. The simulation results showed that chloride movement through the barrier seemed to be concentration limited. The results, in figure 4.1, show that the solute front will move up to 12 meters inside the barrier over a period of 30 years. It is clear that even after 100 years chloride will reach only about 37th cell or 37 meters in the barrier. Therefore, the breakthrough may be later than 100 years. This could be due to the low hydraulic conductivity of the barrier materials and retardation provided by them. The steepness of curves suggests that flow is advective in nature. In barriers of clay for conservative solute like chloride the advection, and dispersion are more important (Mathur and Jayawardena, 2005). However, in the above case there is very low dispersion (0.06m) which makes it quite realistic to assume that the flow is mainly advective. The low dispersion could be due to finer grid size (1m) and the assumption of homogeneity. Although, the dispersion or macro dispersion is clearly related to heterogeneities of aquifer materials. Schaffer and Kinzelbach (1996) have reported that dispersion is a function of hydraulic conductivity distribution which increases with increasing heterogeneity. However in this case given the engineered nature of barrier, the assumption of homogeneity seems realistic.
Figure 4.1: Chloride transport through the barrier over 100 year period

4.2. Long term movement of seawater

In order to highlight the process that will operate in the long run, the movement of seawater was simulated by diffusion dominated flow. Here only flow conditions were changed to **Diffusion Only** in the program while rest of the parameters were kept same as before. The figure below gives the results for four time intervals 200, 250, 500 and 1000 years.

Figure 4.2: Long term evolution chloride in diffusion dominated movement

The comparison of above figure 4.2 and figure 4.1 highlights the contrast between the two processes affecting the chloride movement. In flow dominated by Advective – Dispersive transport, the break through was fast as compared to diffusion dominated scenario in which breakthrough may not occur even after 1000 years. The reason for modelling diffusion dominated flow at long time scale is the assumption that at longer time scale the effects of hydraulic gradient would be lower and differences in solute concentrations of leachate and surrounding moisture would drive the transport. Guacher et al. (2004) have reported that at longer time scales diffusion is dominant process in contrast to advection dominated flow in the initial phase. The above figures 4.1 and 4.2 suggest that due to low permeability and low diffusion coefficients, the solute movement will be extremely slow. Considering the fact that the seawater
intrusion often causes precipitation mineral (i.e. calcite), it can be assumed that barriers porosity will decrease over time.

4.3. Solute transformations in barrier matrix

The impact of different processes due to sweater intrusion is an important aspect to consider. This will help in understanding the natural capacity of barrier material for retardation. The figure 4.3 is a representation of these processes in the barrier. The dominant processes, considering the nature of material, seems to be exchange, adsorption (sorption), precipitation and mixing etc.

![Conceptual representation of attenuation processes in the Barrier.](image)

**4.3.1. Exchange reactions.**

In order to simulate the exchange reactions, the composition of exchanger ‘X’ was defined in the model. This was based on value of CEC for the dominant soil type in the area. The pore water in the barrier was assumed to be in equilibrium with calcite, and similarly seawater was assumed to be at equilibrium with CO₂. The extremely slow transport of conservative solute (Cl) suggests that reactions would occur at faster rate than water movement. This makes the use of equilibrium approximation for modelling plausible. The dominant exchange process was assumed to occur between Na and Ca, due to sea Na rich seawater.

The figure 4.4 confirmed the replacement of calcium (Ca) by sodium (Na). Usually, sodium (Na) is the most easily replaced cation, and soils with sodicity problems (excess sodium) are treated with gypsum to remove Na from the exchange complex. However, in case of seawater intrusion due to higher concentrations of Na in the solution, Ca is forced out of the exchange sites. This can be compared to the softening process in drinking water treatment, where an exchanger is regenerated by pumping brines (rich in Na) into the exchange columns. The figure 4.4 and 4.6 show the progression of exchange in the barrier with increasing time intervals. From the figures it is obvious that exchange reactions are occurring only in parts of barrier affected by seawater.
Flow direction → Distance (m)

Figure 4.4. Exchange reactions after 30 years

Figure 4.5. Exchange reactions after 60 years

Figure 4.6. Exchange reactions after 100 years
Impact of sodium rich water on barrier stability

The exchange of calcium with sodium cause problems in soil structure, and this problem has been demonstrated in irrigated lands (Pupisky and Shainberg, 1979), in sea water intrusion (Goldenberg, 1985), and in migration of Na⁺ rich leachate plume at Norman landfill (King et al, 1999). All these studies have demonstrated that an increase in clay dispersion, in the subsurface, resulted in low hydraulic conductivity. The decrease in hydraulic conductivity of barrier would be desirable for reducing seawater intrusion through the barriers. But on the other hand this dispersion causes loss of soil structure and increases soil erodibility (Pupisky and Shainberg, 1979; Lebron et al. 1994). However it is not possible to test the erosion resulting from sodium. Therefore with slow movement of water and a neutral pH due to mixing, it can be assumed that dispersion will further reduce the hydraulic conductivity.

4.3.2. The effects of seawater on pore water chemistry

One important aspect of seawater intrusion is mixing with pore water and resultant changes in the chemistry. This was studied by simulating mixing of two fluids in the model. Here, mixing ratios presented a problem as exact volume of mixing fluids was not known. However in case of pore water it was calculated by assuming the whole barrier was saturated, duly correcting for porosity. The sea water was assumed to contributing up to 10 %. This is in line with the slow pace of intrusion into the barrier. Both sea water and leachate were in alkaline pH range. The sea water might buffer the leachate in case its pH was in acidic range. Similarly it can limit the alkalinity in case leachate has pH higher than 8 or sea water itself. In this scenario consideration should be given to the equilibrium of Ca²⁺ and Mg hydroxide with CO₂ from the air which gives slightly alkaline yet well buffered reaction of seawater. Therefore, NaCl as the main compound does not contribute to pH change because it is a salt of equally strong acid (HCl) with base (NaOH) of the same strength.

The pH evolution as a result of mixing and transport is shown in the figure 4.7. The pH increases in the pore water as result of seawater entry. The rise in pH clearly seems to be related to the extent of seawater intrusion, as pH has risen in cells affected by seawater. While the cells beyond the 16th cell are still in same pH range as before the seawater intrusion. The sudden drop of pH between 14 and 17 meters is difficult to explain, as sea water penetration beyond this point is not occurring. Such variations are more often because of numerical method used by the model, and may not represent any real change. The finite differences scheme, as used in this case, sometimes results in problems of over shooting and numerical diffusion (Fried, 1975). The reason could be that the time step and spatial dimensions were not adjusted. The mass of water that was assumed to pass the cell had a concentration that could not be absorbed in this representative volume. There are no definite solutions for these problems. Only the trial and error approach, in adjusting the time step and dispersion coefficient, along with grid size, is available to rectify this problem. This approach, given the large number of parameters, was not very helpful.
Figure 4.7. pH evolution in the barrier as result of mixing and transport

The impact of mixing on minerals can be seen in the figures 4.8 and 4.9. The change in mineral saturation indices before and after the mixing in pore water is evident. The calcite at the start of mixing is in equilibrium with pore water. However, after the mixing it becomes deficient which could be due precipitation of calcite. It is difficult to comment on the rate at which calcite precipitated. However precipitation of calcite seems plausible as the process of exchange has increased the concentration of Ca\textsuperscript{2+} in solution. The increase of Ca\textsuperscript{2+} as result of exchange process causes calcite to precipitate (Appelo and Willemsen, 1987).

The deficiency of other minerals such as dolomite, anhydrite and halite in the aftermath of exchange seems to be related to Ca\textsuperscript{2+}. The reason is shear amount of Ca\textsuperscript{2+} displaced from the exchanger which may even push the Mg from the exchange complex. This displacement might can cause super saturation of solution with respect of Ca and Mg. Therefore the precipitation of other Ca and Mg containing minerals (Dolomite) seems quite logical outcome. For minerals like Halite-a sodium containing mineral, an increase in under saturation is not unexpected as sodium is being exchanged for calcium, therefore leaving the solution. The ultimate results would be precipitation of calcite and a concomitant decrease in porosity. This phenomenon has been demonstrated by Appelo and Willemsen(1987), in case of seawater intrusion in coastal aquifers.
4.4. Assessment of leachate migration

The results in figure 4.10 show chloride movement in the barrier over 30, 60 and 100 year period. It would take leachate almost hundred years to reach the sea water. The results of leachate transport are no different from the previous simulation as there was very little difference in parameters used in this simulation. Same was true for cation exchange occurring inside the barrier (Figure 4.11) except that Ca is replacing sodium from exchange complex. The replacement of sodium with calcium in case of leachate migration was simulated because sodium was missing from the results. Therefore it was not possible to say if this exchange mechanism was dominant or real.

Figure 4.10. Chloride movement through the barrier

Figure 4.11: Cation exchange in barrier soil

The changes in chemistry were deduced by using the 30 year mineral saturation indices values. The figure (4.12) provides the clues about possible retardation of
leachate outside the barrier matrix. Iron oxide minerals are important in the context of attenuation as they provide the majority of the sorption sites for metals owing to their large surface area. From the figure 4.12 it is clear that hematite, and to some extent hydroxyapatite are in saturation range in initial length of barrier. The saturation of Hematite was clearly limited to the extent of leachate penetration in the barrier beyond which it is still in under saturation. Here hematite is important because of its high concentration and for scavenging of metals such as Cd, As, Cu etc, present in the leachate.

For example, Jeon et al. (2003), have reported significant metal sorption ranging from 4% in case of Zn and 20% in case Fe+Co, on to hematite, in both single and binary systems. This sorption followed this sequence Fe>Zn>Co>Ni>Cd. Usually, the initial sorption of metals is very fast (ca. >1 min) which is followed by a slow uptake (approx. 1–5 days) probably due to slow conversion from outer to inner-sphere complexes (Jeon et al, 2003). Therefore, it can be assumed that hematite and other minerals (figure 4.12) will act as scavengers for metals, especially once the leachate comes into contact with oxic environment of sea /sediments.

![Figure 4.12. Saturation indices of iron oxide minerals](image)

4.4.1. Long term evolution of the leachate plume

In this case, there were convergence problems and it was not possible to simulate for longer time scales even with changing the flow and boundary conditions. The parameters, like Iron, Nitrogen and Phosphorus failed to converge which resulted in failure of mathematical method. Both data bases were tried, both the iteration limit and convergence tolerance were substantial increased. But despite all these manipulation the problem persisted. This shows that the difference between values given to the model, and the ones computed by the model was significant which resulted problem of convergence. Therefore here results for shorter time scale are employed to make inferences about the leachate migration. The figure 4.13 shows the evolution of pH during all the three time intervals. It is evident that alkaline leachate is buffering the pH of the pore water which was weekly acidic. This control on pH has benefits such as low metal mobility and barrier stability as highly alkaline pH can be erosive.

Figure 4.14 shows the metal transport through the barrier. From the landfill leachate the main concern is leaching or transport of metal to receiving waters. It is obvious
that metal would be transported without going through any significant transformation in the barrier matrix. The reason for metal mobility through the barriers is lack of oxygen and stable conditions of pH and temperature. Some metals such as iron (Fe\(^{2+}\)) may participate in exchange processes as suggested by figures 4.15 and may get adsorbed on the surface. However it is difficult to assess the amount and period of time for this exchange will occur.

Figure 4.13. pH evolution in barrier pore water

Figure 4.14. Metal transport through the barrier

Figure 4.15. Exchange of sodium with iron
This exchange between Na and Fe$^{2+}$ seems to be acting as counter process, in contrast to dominant pattern of Na sorption on exchange complex. However it is not possible to determine whether it is a competitive process and significantly affects the sorption of Na onto exchange complex or not.

Like the previous cases, the possibility of long term transport is considered through the diffusion as a dominating transport mechanism. Figures 4.16 and 4.17 give the results of chloride and iron movement through the barrier towards the sea. Both of them are retarded and preclude any possibility of breaking out to seawater or becoming long term source of pollution.

Figure 4.16. Diffusive Chloride transport through the barrier

Figure 4.17. Iron transport through the barrier

4.5. Worst case scenarios

For worst conditions, it was assumed that the barrier has not prevented the seawater seepage into the landfill cell. This situation is based on the fact that barrier may produce cracks due to mechanical actors such as settlement, and freezing/thawing etc. The cracking will raise porosity and seawater intrusion or leachate migration may occur at faster rates than imagined. For simulation of worst case the hydraulic conductivity in this case was raised incrementally (30% and 50%). The Figure 4.18 shows the effect of increase in hydraulic conductivity due to cracks or preferential flow paths. With 30% increase in hydraulic conductivity the sea water under same conditions of transport, can penetrate to longer distance (23 m). However, at the 50% there was not much difference between two curves (25 m). This gives rate of 0.61m/year in contrast to normal conditions where rate is 0.37m/year. It is evident that if
time is doubled then seawater will reach the landfill at the same hydraulic conductivity around 40-50 years.

4.5.1. Interactions of leachate and sea water

In worst case, along with the transport of conservative solutes, the migration of reactive solutes was also considered. This was done in order to determine the changes in chemistry of leachate and seawater, and also to assess the extent of attenuation. The seawater and leachate quality data was used to simulate batch reaction and mixing between the two liquids in order to understand the changes in their chemistry. Following assumptions were made.

Assumptions
- Seawater is oxidized
- Density difference were ignored as leachate too have high TDS values
- Calcite and CO2 were assumed as final equilibrium phases.
- DOC will go through oxidation coupled to electron acceptors

The degradation of organic matter present in leachate was simulated by representing it as CH$_2$O in the model. The model used transport and exchange along with a reaction in which CH$_2$O reacts with seawater in incremental steps. The concentration of the CH$_2$O was given hypothetical and in series of steps. The solution present in last cell was used (number 37) which helped incorporate the changes that occurred during the transport through the barrier.

The figure 4.19 shows the results of this simulation and it is clear that close to landfill due to reduced environment the elements are in solution and as the front progresses they get oxidised. Therefore the concentration of iron and ammonia will reduce in case such a leakage occurs. Also the concentrations are very low which makes pollution incident on bigger scale a remote possibility.
Figure 4.19. Mixing of leachate with seawater

Similar pattern is repeated in case of seawater going into landfill (Figure 4.20). However the effects of seawater may not be spread uniformly in the landfill. Also the landfill unlike seawater represents a porous medium. Therefore, retardation processes such as mineral precipitation, dispersion and exchange etc would have strong effects.

Figure 4.20. Mixing of seawater and leachate in the landfill cell

Finally, the figure 4.21 represents pH which in both cases of mixing was quite low and apparently seems due to degradation of DOC which is results in production of CO$_2$. The buffering effect of minerals is visualised by the curve rising but remaining in the acidic range. This effect is greater in case of leachate as it is often quite rich in calcite.
4.5.2. Impacts on ground water

In this case the migration of landfill leachate towards the ground water was modelled. In order to reach ground water leachate will have to pass through confining layer first. This layer was assumed to be connected with the ground water bodies and was therefore oxic. It was assumed that DOC in leachate would go through oxidation coupled to electron acceptors, and therefore would deplete the redox buffer in the bottom sediments. Figure 4.22 shows the results of redox buffer depletion close to landfill. In this case one peculiar difference was that the pH did not affect as strongly as in case of seawater. This stable pH could be due to buffering effect of calcite mineral present in the soil, which is calcareous. The results confirm that if in worst case scenario, the leakage of leachate occurs, the oxic and calcite rich bottom sediments would attenuate the migration of contaminants to seawater. However it was not possible to quantify the attenuation because of limitations of model which only simulated it in batch reaction mode. The combination of this with transport mode was not possible, which could have provided the spatial extent of this attenuation mechanism.

![Figure 4.22. Depletion of redox buffer in bottom sediments](image)

4.6. Sensitivity analysis

A simple sensitivity analysis was performed to check the uncertainty in the model outcome. For this purpose parameters (i.e. Hydraulic conductivity, CEC etc) were varied in the same model for the period of 30 years. The hydraulic conductivity was varied in
the model in same fashion as in worst case. The effect of hydraulic conductivity change was not as pronounced as expected (see figure 4.18). But the model was more sensitive to changes in cation ion exchange capacity (CEC) of natural containment materials i.e. barrier soil and bottom sediments. From the simulations results a ratio of Ca to Na was computed and then plotted against the CEC values. Appelo and Wellemsen (1987), used ratio of Ca/Mg in order to draw conclusions about the ability of model to represent mixing and exchange.

The result in Figure 4.23 show a large variation in response to change in cation exchanged capacity. The first value on the x-axis was empirically computed value by using the formula given by Breeuwsma et al. (1986). While the rest of the values on x-axis were taken from CEC range (1-14 meq/l) of Kaolinite by assuming the clay is a non swelling typing. By changing the CEC values with a constant interval the ratio of cation in solutions to the one sorbed increases over several order of magnitude. This shows sensitivity of model to CEC values. It highlights the underlying uncertainty in the estimation of CEC values especially at lower values up to 4 meq/100 gm. The sensitivity of exchange to CEC values demands that attention should be paid this parameter as it can have important consequence for retardation pollutants. Although, it may not affect the breakthrough of conservative solute (Chloride) but in case of reactive components (salts, metals) it effects their movement through the barrier and sediments. Therefore the correct estimation CEC will help guide determination of exact nature containment materials at the landfill sites.

![Figure 4.23. Changes in Ca/Na ratio with respect to CEC of the medium](image)

Figure 4.23. Changes in Ca/Na ratio with respect to CEC of the medium
5. Conclusions and recommendations

The present study modelled the intrusion of moisture from the surroundings into the Spillepeng landfill in the post closure phase with the geochemical code PHREEQC. The study tried to highlight the interactions between leachate, ground water and seawater. Based on the results and discussion the following conclusion can be drawn:

1. The seawater can breakthrough into the landfill in 100 year’s time, in case of advective-dispersive transport.

2. The breakthrough time was significantly higher (>500 years) in case of diffusion dominated flow.

3. The exchange of Ca with Na was the dominant ion exchange process in containment materials.

4. The replacement of Ca from exchange sites by Na rich seawater will cause dispersion and therefore reduction in hydraulic conductivity of containment materials.

5. This exchange process will cause precipitation of calcite due to increase Ca concentrations. Therefore a further reduction in porosity of barriers can be expected.

6. The leachate can break through the bottom sediments in 50 years, and can reach ground water in lime stone aquifer. However this can happen only in worst cases and needs a drastic reversal in hydraulic gradient away from the landfill. Even if this change occurs, the initial dilution with ground water and seawater will decrease contaminant concentrations to a harmless level.

7. The leachate was rich in iron oxide minerals such as hematite and it can be expected that in the oxidised environment of seawater and sediments, these mineral will precipitate and will provide sorption sites for the metals (i.e. As, Cd, Cu etc) present in the leachate.

8. The simulation of DOC degradation coupled to electron acceptors showed strong attenuation of pollutants in the bottom sediments and seawater.

9. The simulation results also highlighted that attenuation process would be stronger in the seawater than in the sediments owing to high content of dissolved oxygen in the seawater.

10. The concentrations of pollutants such as metals were generally low except in case of iron and ammonia which were moderately high.

11. The sensitivity analysis highlighted the uncertainty regarding the estimation cation exchange values for barrier materials.
12. The PHREEQC model despite the paucity of information provided the estimates of water intrusion, leachate migration, exchange, and DOC degradation with reasonable accuracy. Therefore can be successfully applied as a pollution simulation and monitoring tool in landfill studies.

13. Based on the results it can be concluded that threat leachate migration from Spillepeng was not significant. Therefore it may not be serious threat to coastal and ground water quality, in the long run.

5.1. Limitations

The limitations of study mainly stem from the uncertainties regarding the data which were derived, and modified, from secondary sources. It was difficult to verify it especially in long run scenarios as there is not data available for centuries. Similarly, this paucity of data and time constraints also meant that there was no way of validating modelling observations by experimental or field work. Although the equilibrium modelling approach was appropriate for most processes due to extremely slow movement of water. However reactions like mineral precipitation and dissolution, and biodegradation could also have been modelled with kinetic approach. This would have made comparison of two approaches possible and therefore would have the uncertainty.

There is uncertainty regarding parameter such as CEC values used to model the exchange reactions. This stems from lack of information about the soil materials used for containment. Also the biodegradation reaction was modelled in hypothetical fashion as data about the composition of organic matter, stoichiometry, and reaction rates were not available. Never the less, the results have highlighted the some important parameters and issues which could be targeted in future work.

5.2. Recommendations

Based on the conclusions, the following examinations are suggested:

- Biodegradation is important for both metal mobility (electron acceptors) and the stability of waste. The results gave some clues but there were uncertainties regarding stoichiometry, composition and reaction rates. Therefore further modelling of biodegradation processes is suggested.

- The kinetic modelling of mineral precipitation and dissolution along with biodegradation is suggested. The results should be compared with equilibrium approach. This will help reduce the uncertainty regarding modelling results.

- The study estimated the water intrusion under normal conditions and did not include extreme events such as sea level rise and coastal flooding. This can have bearing on stability of barriers and also on pollutant transport. Therefore, these aspects need to be integrated in future studies on the Spillepeng landfill.
6. Literature cited


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