Life cycles assessments and solid waste

– Guidelines for solid waste treatment and disposal in LCA

Final Report

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Summary

This study is to a large part a synthesis of our previous studies[i,ii]. The long-term goal with the project has been to develop and demonstrate methods to handle waste treatment disposal (landfilling and incineration) in connection with LCA (Life Cycle Assessments).

The main problem when modelling landfilling of waste in LCA is the time aspects - how should long-term future emissions be interpreted in a LCA. Our approach is that the emissions are integrated over different time periods:

- a <u>surveyable period</u>, which is the period until a pseudo steady-state in the landfill processes is obtained. This period should usually be of the magnitude of one century.
- a <u>hypothetical, infinite time period</u>, which is the period from the start until the landfilled material is completely released to the environment.

In this approach the processes in the landfill must be known, but not the exact kinetics of the landfill. This makes the approach site-independent. From model studies of different types of waste and landfills we draw the conclusion that the surveyable time approach is usable for several types of waste. For several cases it is possible to define a surveyable time period on the basis of physical, chemical and microbiological processes in the landfill.

In the report emission factors for MSW landfills, biofills (biocell), incineration ashes, coal ashes, metal hydroxide sludge landfills and mine tailings are given. The emissions of metals during the surveyable time period are often rather small compared to the total emissions during the hypothetical infinite time period, from both municipal solid waste landfills and different kind of ash landfills. The emission factors are of the magnitude $<10^{-3}$ (kg emitted per kg landfilled of metal). This emphasis that the hypothetical infinite time period must not be forgotten in the LCA inventory. For metals the emission factors for the hypothetical, infinite time are in all cases =1 by definition.

Some alternatives to the surveyable time period and the hypothetical, infinite time period are discussed in the report. The differences between other ”short-term” approaches and out the surveyable time period approach seem to be relatively small. Often the uncertainties in the data are larger. Some alternative long-term time horizons are discussed, e.g. next glacial period, or when the emission reaches background level. One difficulty with these approaches is that both the processes in the landfill and the kinetics of the landfill must be known. Our approach is easier to handle since it only requires knowledge of the processes in the landfill.

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[i] Sundqvist, J.-O., Finnveden, G., Albertsson, A.-C., Karlsson, S., Berendson, J., Eriksson, E., Höglund, L.O. (1994): 'Life Cycle Assessment and Solid Waste'. AFR-Report 29; AFR, Stockholm, Sweden

[ii] Sundqvist, J.-O., Finnveden, G., Albertsson, A.-C., Karlsson, S., Berendson, J., Höglund, L.O. ,Stripple, H.(1997): 'Life Cycle Assessment and Solid Waste - stage 2'. AFR-Report 173; AFR, Stockholm, Sweden

In stage 1 of the study, we concluded that the methane emissions from municipal solid waste landfills are important. Also when landfill gas recovery systems are used, there seem to be large losses of methane to the environment. The recovered part seems to be 10 - 50 % of the theoretically available methane, but analyses of the atmosphere close to the landfill often show low methane concentration, the real losses to the environment are reduced by oxidation in the soil cover. New landfilling techniques, such as biocells or biofills, can obtain higher gas yields and also possibility to oxidise the leaking methane in the soil cover.

Landfill fires give considerable emissions to the environment of PAH, dioxins, PCB:s, etc. The landfill fires seem to be a significant pollution factor for landfilled material. Emission factors for landfill fires are proposed in this report.

The main problem when modelling incineration of waste in a LCA, is to find relevant allocation factors. We have grouped emissions into product-related and process-related. The former are often relatively easy to handle, and emission factors are often expressed as kg emitted per kg of contaminant in input. The process-related emissions are more difficult to handle. We have discussed some allocation principles, and found that there is no universal principle that always should be used. The appropriate choice of allocation method will often be dependent on the question at issue. E.g. there is a distinction between small changes (product LCA) and large changes (for example analyses of waste treatment systems). The emission factors may be expressed as average changes or incremental changes. We suggest either an allocation to the total weight or to the carbon content for the process-related emissions. We have also introduced the ”permission-controlled” or ”operation-controlled” emissions as a complement to mark that the emission actually can be controlled by operation, not only input.

[i)] Sundqvist, J.-O., Finnveden, G., Albertsson, A.-C., Karlsson, S., Berendson, J., Eriksson, E., Höglund, L.O. (1994): 'Life Cycle Assessment and Solid Waste'. AFR-Report 29; AFR, Stockholm, Sweden

[ii)] Sundqvist, J.-O., Finnveden, G., Albertsson, A.-C., Karlsson, S., Berendson, J., Höglund, L.O. , Stripple, H. (1997): 'Life Cycle Assessment and Solid Waste - stage 2'. AFR-Report 173; AFR, Stockholm, Sweden

Contents

Preface

This report is the result of the third and final stage of the project LCA and Solid Waste, financed by the Swedish Waste Research Council (AFN) at the Swedish Environmental Protection Agency. The first stage was a pre-study, where some hypothesis of how to handle waste in LCA:s was developed and tested (Sundqvist et al, 1994). The second stage involved deepened studies of some interesting topics (Sundqvist et al, 1997). An international workshop was organised during the second stage (Finnveden and Huppes, 1995). During the second stage also two papers were published in international journals (Finnveden et al, 1995; Finnveden, 1996). This third stage has been the final stage with the goal to summarise and synthesise the earlier stages.

This report has been written by myself. However, most of the results are from the earlier reports and have only been subject to editorial work. I have to give great thanks to the following colleagues who has contributed in the earlier stages:

- Göran Finnveden at the Swedish Environmental Research Institute (IVL)[3] who has been the LCA expert in the group. His knowledge about both LCA and waste have been valuable during all the work. He has also elaborated the section about metal in MSW landfills during the stage 2, and organised the Workshop Life Cycle Assessments and treatment of Solid Waste.
- Håkan Stripple at Swedish Environmental Research Institute (IVL) in Gothenburg developed the allocation model for incineration during the second stage.
- Lars Olof Höglund, Kemakta Konsult AB, was involved in modelling of landfill processes for ashes from waste incineration, coal ashes, mine tailings and metal hydroxide sludges in both stage 1 and stage 2.
- Mats Ek at the Swedish Environmental Research Institute (IVL) in Stockholm contributed to the sections about paper and paper additives in landfills during the second stage.
- Associate professor Sigbritt Karlsson, and professor Ann-Christine Albertsson at the department of Polymer Technology, at the Royal Institute of Technology in Stockholm has contributed to the sections about plastics and plastic additives in landfills during stages 1 and 2.
- Erik Eriksson and Jaak Berendson at department of electro-chemistry at the Royal Institute of Technology in Stockholm have contributed to the the sections about metals in MSW landfills.

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[3] Göran has since then moved to fms (Environmental Strategies Research Group)

Some points have been developed in this stage, e.g. there is some attempts to develop the discussions about allocation principles. I have been involved in both LCA studies (of products) and of system analyses of waste management systems (the so-called ORWARE-project). I have found that these two options require different ways to attack the allocation problem. Also the discussions about time aspects in landfilling have been developed. For example, the long-term aspects are illustrated and discussed in this report.

Stockholm 1999-10-30

Jan-Olov Sundqvist
project leader

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Notation

Some symbols and contsants which are defined and used locally are not included here.

Greek symbols

a	Degradation yield, see Figure 5.5. and Tables 5.3. and 5.4, *kg/kg*
b	Molar ratio $CH_4/(CH_4+CO_2)$ in gas, *kmol/kmol or* m^3/m^3
g	Oxidation yield of CH_4 in soil cover, see Figure 5.5. and Tables 5.3. and 5.4, *kg/kg*
e	Ratio (recovered methane)/(formed methan), see Figure 5.5. and Tables 5.3. and 5.4, *kg/kg*
f	Thermal efficiancy, *MJ/MJ*
h	Viscosity, Ns/m^2
DH	Heating value, *MJ/kg*
r_l	Air density, kg/m^3
r_p	Particle density, kg/m^3
s_y	Parameter that shows the horizontal dispersion, see Eq. A.1 in App. A
s_z	Parameter that shows the vertical dispersion, see Eq. A.1 in App. A

Symbols

BOD	Biological oxygen demand, *kg*
$c(x,y,z)$	Concentration on the coordinate (x,y,z), mg/m^3
COD	Chemical oxygen demand, *kg*
E	Emission factor, *kg/kg, kg/MJ*
E', E''	Non-linear emission factor, see Eq. 3.5 – 3.6, *kg/kg, kg/MJ*
H	See also *DH*
H	Henry's law constant (phase distribution coefficient), $P\ m^3/mole$
h	Heght of plume in release point see Eq. A.1 in App. A, *m*
K_{OW}	Octanol-water distribution factor, *kg/kg, mole/mole*
pK_a	Negative logaritm of acid dissociation constant
Q	Flow of emittant, (see Eq. A.1 in App. A), *mg/s*
R	Retention factor, non-dimensional

t	Time, *years* or *h* or *s*
TOC	Total organic carbon, *kg*
U	Allocation property, see Eq. 3.3, *kg/kg*, *MJ/kg*
u	Wind speed, see Eq. A.1 in App. A, *m/s*
V	Flow gas volume, nm^3
W	Material flow, *kg/s*
W	Falling velocity, *m/s*
X	Emission flow, *kg/s*
x	x-coordinate (distance in wind direction, see Eq. A.1 in App. A)
x_C	Carbon concentration, *kg/kg*
x_H	Hydrogen concentration, *kg/kg*
x_O	Oxygen concentration, *kg/kg*
y	y-coordinate (horizontal distance from a-axis, see Eq. A.1 in App. A)
Z	z-coordinate (height from ground level, see Eq. A.1 in App. A)

Subscripts

1, 2,…n	Different materials, e.g. paper, plastic, wood, steel,…..
a, b, c, …	Different emissions, e.g. $CO_{2,}$, SO_2, CH_4, …..
b	Combustible (organic), water-free and ashfree material
C	Carbon
cal	In connection with ΔH: calorimetric heating value or upper hetaing value
eff	In connection with ΔH: effective heating value or lower heating value
H	Hydrogen
O	Oxygen
raw	Raw waste: with ash and moisture
t	Dry materialwith ash but withou moisture
Tot	Total waste (including all materials)

1. Life Cycle Assessments (LCA) and Solid Waste

1.1 Introduction

Neither waste nor Life Cycle Assessments (LCA) is something new.

Waste has been generated as long as the human being has been on Earth. Waste has been recognised as a problem, first of all a hygienic problem, since large cities became usual in the pre-industrialism era. Recycling and recovery options have been discussed at least from the late 1960:s[4]. However, during the last ten years there has been a remarkable increasing interest for the waste problem.

Earlier, the environmental problems were associated with industrial production plants as point sources of different emissions. In LCA, the environmental problem is more associated with the product from “the cradle to the grave”, thus adding together all environmental burdens, that are associated with the studied product during its whole life cycle or life time. The interest for environmental impact associated with products began to grow during the 1970:s and 1980:s. LCA was originally an abbreviation for *life cycle analysis,* but today usually *life cycle assessment* is used. The first LCA:s focused on energy consumption associated with products, but soon more and more environmental parameters were included in the LCA studies. Today LCA has a broader meaning and can be defined as a standardised process to assess the potential environmental burdens associated with a product, a process or an activity (Lindfors et al 1995; ISO 14040).

There are several connections between waste and LCA. The waste associated with the product is a part of the life cycle of the product. Every product has a waste stage, when the product is discarded or disposed. There is also waste generated during the manufacture of products, for example different types of industrial wastes.

LCA methodology is also excellent for assessing different waste management systems, especially for comparisons between different treatment and disposal systems. This ”Waste Life Cycle Assessments” has two different applications:

- Study of certain waste components in the waste. For example, there have been several studies comparing material recycling and energy recovery for paper packages and for plastic packages. In these cases the studied waste component have comprised a marginal part of the total waste stream. The methods used are equal to the methods used for product LCA. The “cradle” of the life cycle of the studied packaging waste can be defined as the moment when the used package is discarded into the waste.

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[4] However, it should not be forgotten that reuse, recycling and recovery are even older. For example, in Stockholm there were different waste programs during the first decades of the 20th century, where waste was source separated, and used for reuse, use as secondary raw material, and use in agriculture.

- Study of waste management systems, where different waste management systems for e.g. municipal solid waste are compared, e.g. incineration and landfilling. In these cases the study comprises a total waste stream. The approach to handle this kind of system analysis differs a little from the approach used for product LCA.

In this report the ”product LCA” as well as the two options of ”waste LCA” are discussed.

1.2 Scope and objectives of this study

This is the third stage of the project Life Cycle Assessments and Solid Waste, financed by the Swedish Waste Research Council. The long-term goal with the project has been to propose and test methods to handle waste disposal in connection with LCA. The work has been directed towards incineration and landfilling. In the landfilling studies municipal solid waste as well as some industrial wastes have been considered. Several reports and publications from the first two stages have been presented Sundqvist et al (1994), Finnveden and Huppes (1995), Finnveden et al (1995), Finnveden (1996), Sundqvist et al (1997). This third stage has been the final stage with the goal to summarise and synthesise the earlier stages. During this stage also some comparisons with other approaches have been done.

1.3 Waste treatment and disposal

1.3.1 Nomenclature

Waste. According to the EU Waste Framework Directive (European Community 1991), waste is defined as *any substance or object which the holder discards, or intends to discard, or have to discard*. EU has provided a waste classification system under the Framework Directive in order harmonise the view on waste. This classification system has been presented as the European Waste Catalogue (EWC)(European Commision 1994). EWC is a list of material flows and substances, which have to be handled as waste.

Wastes can be grouped into different classes, for example according to origin, composition, or disposal method. For the purpose of this report a grouping into **consumption waste** (also called post-consumer waste) and **production waste** (also called process waste) has been useful. ”Consumption wastes” are generated in households as well as in industry, business, offices, hospitals, shops, etc. and are mainly consisting of old discarded products, packages, food wastes and similar. The domestic wastes, plus a large part of the consumption wastes from industry and business, are often managed by the municipality (see explanation of ”Municipal Solid Waste” below). ”Production wastes” or process wastes are generated in industry and are often consisting of different rest products or residues from the production, e.g. slags, ashes, sludges. ”Industrial waste” has not a distinct meaning, sometimes it refers to all wastes generated by industry (consumption wastes as well as production wastes), and sometimes it refers to wastes managed by the industry.

Municipal Solid Waste (MSW). Wastes from households (domestic waste) and similar wastes from business, shops, offices, industry, etc. are in most countries disposed together. Usually the municipality has the responsibility for this waste, why it is called municipal solid waste (MSW).

1.3.2 Waste management

Waste is treated and disposed by different techniques. The most important methods are as follows.

Landfilling. The most common method to dispose the waste is landfilling. Earlier landfilling could be characterised as more or less controlled dumping. Nowadays there are several directives that rule the performance and operation of a landfill. Landfill technology has been developed and today's landfills have very few similarities with the dumps that could be found e.g. in the 1960:s.

Incineration or combustion has existed for more than 100 years. Earlier, incineration plants were performed with the main goal to decrease the volume of the waste that must be landfilled or dumped. There is some early experience from energy recovery, but it was mainly after the first oil crisis in the beginning of the 1970:s, that the interest for energy recovery began to grow. Today's incinerator plants are equipped with energy recovery facilities, which produce steam, district heating or electricity. Nowadays the incinerator plants also are equipped with effective flue gas cleaning equipment.

Composting is a treatment method based on different aerobic processes, where the organic material is micro-biologically oxidised to water, carbon dioxide plus a solid humus rich residue called compost. The compost can be used as fertiliser. Composting of organic wastes have been carried out since long. There are large scale plants that were built already in the 1920:s in Netherlands. However, composting has never had the same importance as incineration.

Anaerobic digestion is another microbial process where the organic material is converted to a biogas (methane and carbon dioxide) and a compost-like residue. Anaerobic digestion has been used for several decades in sewage treatment plants to stabilise the sewage sludge. There is a growing interest for anaerobic digestion for energy recovery and recovery of fertilisers from organic degradable waste.

Material recycling is another increasing option. Separated waste materials are used as raw material in manufacturing processes and, in most cases, substitute virgin raw materials. Metal scrap has been recycled since long, as has paper wastes. Glass recycling has increased since the middle of the 1980:s. Plastic recycling is relatively new, even if small amounts have been recycled during 10 - 20 years.

These different methods have different importance in different countries. For example, incineration is of great importance in some countries - Denmark and Switzerland incinerates more than 70 % of the MSW, while other countries only incinerate a few percentages. As an average of Western Europe about 25 - 30 % of the MSW is incinerated (OECD, 1990). In most countries the material recovery is relatively low - for several European countries the recovery rate is lower than 10 % of the MSW. In EU, as well as in North America, there is a tendency towards less landfilling and increased material recycling, energy recovery (incineration) and biological treatment (composting and anaerobic digestion).

As an example, the management of the Swedish MSW is presented in Figure 1.1. The distribution between different management systems is expected to changed considerably during a near future. The Swedish EPA has predicted that incineration shall increase by 30 %, biological treatment increase by 140 % and landfilling decrease by 50 % within ten years from 1995 (Swedish EPA, 1996).

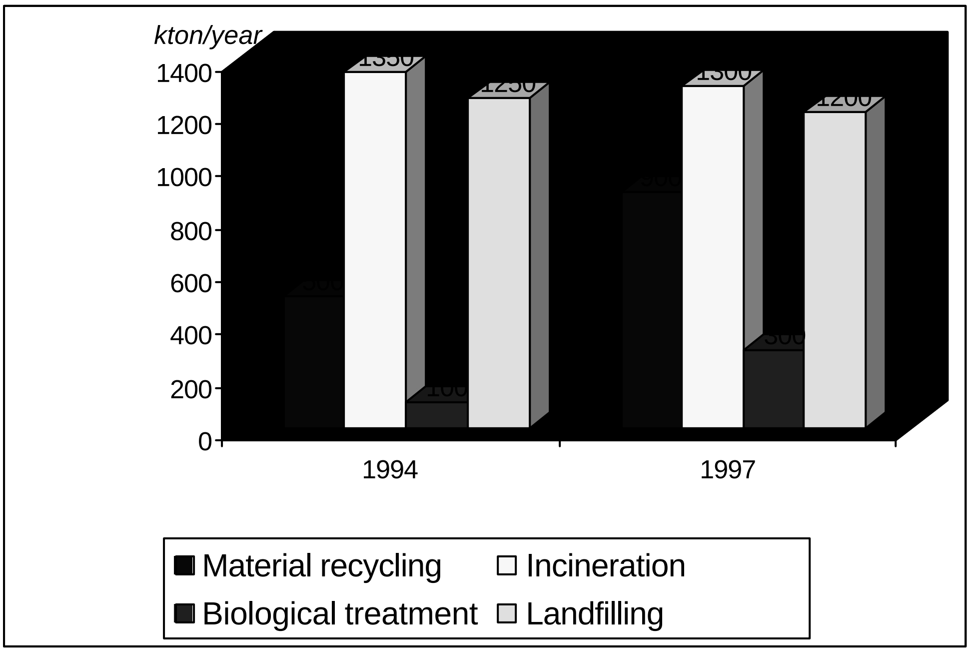

Figure 1.1. Treatment and disposal of Municipal Solid Waste in Sweden 1994 (Swedish EPA 1996) and 1997 (RVF 1998).

1.3.3 Waste management policies: ”the Waste Hierarchy” and ”Integrated Solid Waste Management”

The waste hierarchy is characterised as a hierarchical order of different options to solve the waste problem. The most common way to present the waste hierarchy seems to be (IEA Bioenergy, 1997):

1. Avoidance, reduction and minimisation
2. Reuse
3. Recycling
4. Recovery
5. Disposal

This hierarchy is equivalent to the strategy discussed by EU (European Commission, 1989; European Commission, 1996).

Characteristic for the waste hierarchy is that it presents a hierarchical order - the higher up in the hierarchy, the more preferable (from an environmental and natural resources point of view).

Integrated Solid Waste Management (ISWM) is a concept that is more and more used in waste management planning, however often in different shapes and variations. The IEA Bioenergy Agreement Task XIV has defined Integrated Solid Waste Management as *an optimised system of waste management practices designed to protect human health and the environment, based on the sound evaluation of environmental, energy, economic and socio-political considerations, and includes one or more components of the waste hierarchy* (IEA Bioenergy, 1997).

In a recent Swedish research program, ISWM was defined as *an optimised waste management system where the environmentally and economically best solution is chosen for each individual case, without regarding the waste hierarchy* (NUTEK, 1997).

A distinctive feature of these ISWM definitions is that an integrated solid waste management system should contain several of the elements of the waste hierarchy, without regarding the hierarchical order, but making a “best choice” on a multidisciplinary base for each individual case. The ISWM concept means that there is not obvious that one waste treatment method is more favourable than another, and that a waste management system should be based on several of the available treatment methods.

The methods and data presented in this report can be used for environmental assessment of different solid waste management system, when searching for the ”best choice” of system.

1.4 Life Cycle Assessments

1.4.1 General

As mentioned above, LCA (Life Cycle Assessment) is a process to assess the potential environmental burdens associated with a product, a process or an activity. Characteristic parts in a LCA are identifying and quantifying of energy flows and material flows, and evaluating the environmental impacts that are associated with these flows. The assessment should include the entire life-cycle of the studied system (the studied system can be a product, a process or an activity), including material and energy raw ware acquisition, manufacture, use and waste management.

The interest for LCA has increased dramatically since around 1990, resulting in both a development and increased harmonisation of the methodology. A ”Code of Practice” has been published (Consoli et al, 1993), as well as several guidelines (e.g. Heijungs et al 1992, Vigon et al 1993, Lindfors et al, 1995a). Also an ISO standard has recently been issued (ISO 14040). As evidenced by recommendations in these guidelines, it is now generally recognised that interventions from solid waste management and disposal should be included in a LCA. However, this is not always done today, because when trying to include the waste stages in a LCA, several difficulties will arise.

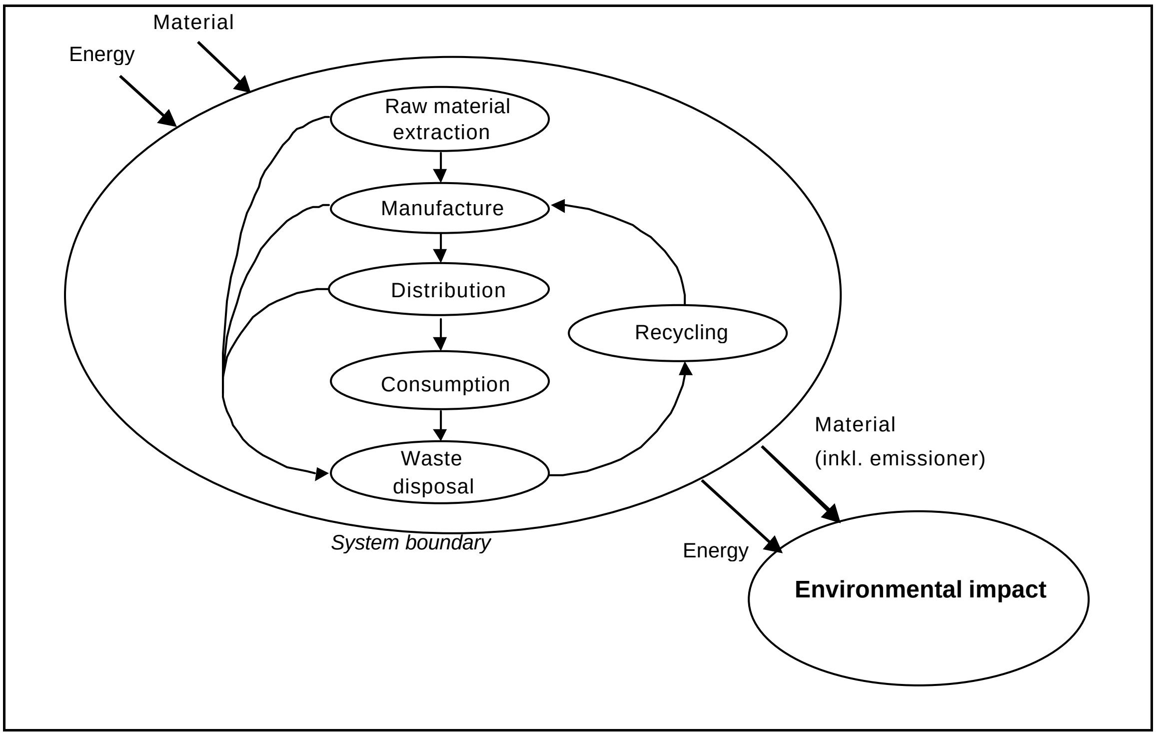

Figure 1.2. Life Cycle Assessment of products, from the cradle to the grave.

1.4.2 Framework for LCA

The framework outlined here is based on the ”Nordic Guidelines on Life Cycle Assessment” (Lindfors et al, 1995a) and the ISO standard (ISO 14040, 1997). According to these references a complete LCA consists of the following interrelated components:

1. Goal definition and scoping
2. Inventory analysis
3. Impact assessment
    - 3.1 Classification
    - 3.2 Characterisation
    - 3.3 Valuation
4. Interpretation

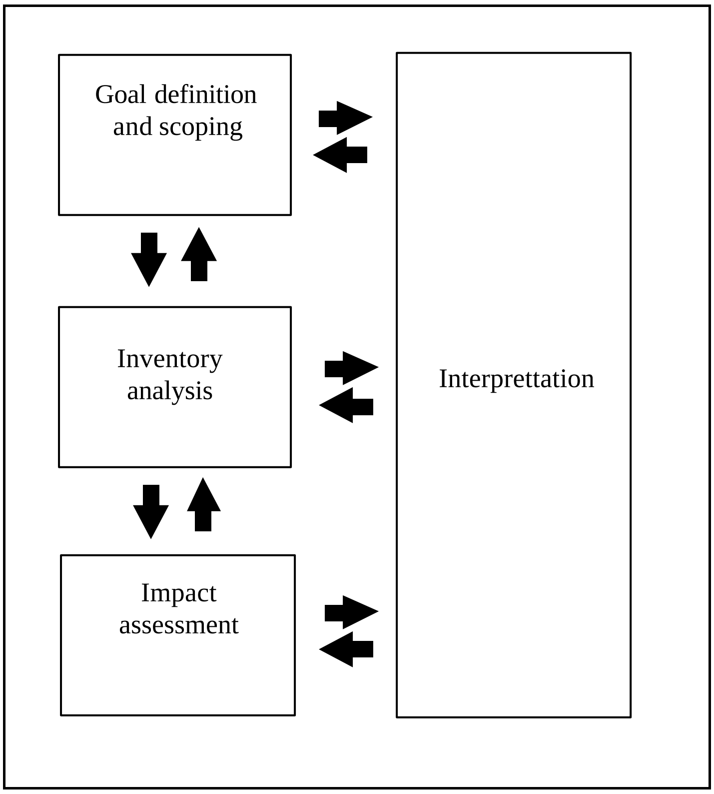

Figure 1.3. Framework for LCA according to ISO 14040

In the goal definition and scoping, the purpose and the scope of the study should be defined. This includes definition of system boundaries, data requirements, assumptions and limitations.

In the inventory analysis, the inputs and outputs of the system under study are analysed. The system is usually a product through its life-time, encompassing raw materials acquisition, production, use, and waste management. The inputs to the system are for example energy and raw materials. The outputs from the system are for example emissions from processes during raw material acquisition, manufacture, transports and waste management. The inventory analysis results in tables of inputs and outputs of the system(s) under study.

The impact assessment is a process to characterise and assess the impacts of the inputs and outputs identified in the inventory analysis. The impact assessment of an environmental LCA should consider the following major categories:

- Resource depletion
- Impacts on human health
- Ecological impacts

Each of these major categories is further divided into several impact categories, see Lindfors et al (1995 a, b).

The impact assessment is divided into three steps: classification, characterisation, and valuation. In the classification the different inputs and outputs are assigned to different impact categories. An analysis and quantification of each impact category is made in the characterisation. Valuation is the step in which the data of the different specific impact categories are weighted so that they can be compared.

The framework outlined above may indicate that an LCA can be seen as a linear process in which each component is taken in turn. However, a useful approach can be to regard an LCA as an iterative process. A procedure has been discussed in the Nordic Guidelines (Lindfors et al, 1995a) in which the first step is an initial LCA including a comprehensive, screening inventory analysis, followed by an impact assessment. A sensitivity analysis is then carried out, with the objective to identify the parts of the life-cycle that are responsible for major environmental impacts. The identified critical parts can then be further analysed in a detailed LCA if wanted.

According to the framework outlined here, emissions from treatment of solid waste should be analysed in the inventory analysis. The impacts caused by these emissions should then be analysed and assessed in the impact assessment in parallel to other emissions caused by the product during its life-time.

1.4.3 Some important terms and methods used in LCA

Functional unit

The functional unit is the basis for the calculations in a quantitative life cycle assessment. It is a product, a material or a service for which the environmental loading are quantified. In an absolute LCA the whole life-cycle of a specific product, material or service is studied, and the different parts of the life-cycle are compared with each other. In this case the functional unit should be, for example, one item of the studied product.

On the other hand, in a comparative LCA different products are compared with each other. In that case it is not always relevant to compare the different products, but more to compare the functions of the products.

The choice of appropriate functional unit is of great importance for the LCA. A relevant choice of functional unit is needed for relevant results. For waste management systems it is often preferable to work with several functional units, each one representing an essential utility that is produced from waste. For example, in a study (Finnveden, Steen and Sundqvist, 1994) where we compared energy recovery and material recycling of paper waste, we used three functional units:
- treatment of a specific quantity of paper waste
- production of a specific quantity of new paper (from waste paper or from virgin wood)
- production of a specific quantity of energy (from waste incineration or from conventional fuels such as oil or bio-fuels).

In this case the system under study had to include three different systems: the waste treatment, the energy production and the paper production.

Emission factor

In LCA:s the used data is often presented as emission factors and energy factors. Information in data bases is often expressed as emission factors. The emission factor gives the emission for a process or sub-process in the life cycle, in relation to a input parameter, for example per weight of product, per weight of a certain element in the product, or related to the energy content of the product. For example, emission of HCl from waste incineration may be expressed as kg HCl emitted per kg Cl in the input to the incinerator. Energy consumption for transport can be presented as MJ fuel (or litres of diesel oil) per kg of transported product and per km transport distance. Emission factors are further discussed in Section 3.1.

System boundaries

The system boundaries define the system that is studied. A LCA is based on the material flows and energy flows over the system boundaries. It is of absolute necessity to have well-defined system boundaries, in order to obtain unambiguous results. Different topics on system boundaries are discussed by Lindfors et al (1995b).

Allocation

A traditional problem in LCA is how to deal with processes or groups of processes with more than one input and/or output, e.g. processes or productions with co-products of economic value (multi-output processes), or waste management where several different waste components are treated in the same process with common consumption of raw material and common formation of emissions (multi-input processes). *Allocation* can, in LCA, be defined as *the act of partitioning in some proportionate shares the responsibility for environmental impacts caused by processes in a life cycle* (Lindfors et al, 1995 b).

General allocation problems in connection with LCA have been discussed by Lindfors et al (1995b). Allocation problems connected to the waste stages in LCA have been discussed in our second report (Sundqvist et al, 1997) and is further discussed in this report.

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2. Problem formulation

There are two major problems arising, when assessing the waste stages in a product LCA or when making a system analysis of a waste management system: allocation problems and time aspect problems. A third problem, as a consequence of these problems, is the interpretation of field data.

2.1 Allocation problems

Municipal Solid Waste (MSW) is a mixture of several materials and products. Several different emissions are produced when waste is treated in an incinerator plant or a landfill. Waste treatment and landfilling of Municipal Solid Waste can be characterised as **multi-input multi-output processes** (see Figure 2.1). There are several input components and each of them may cause several emissions. In a product-LCA we usually want some kind of causal relation between the studied product or material and each emission actually caused by it. Also in the case of system analyses of waste management systems, it is necessary to know the relations between changes in the emission and changes in the composition. However, available data only shows the total emissions. It is not possible to measure emissions from certain products. The causal relations must be calculated by some kind of model, to partition in some proportionate shares the responsibility for environmental impacts caused by the different inputs.

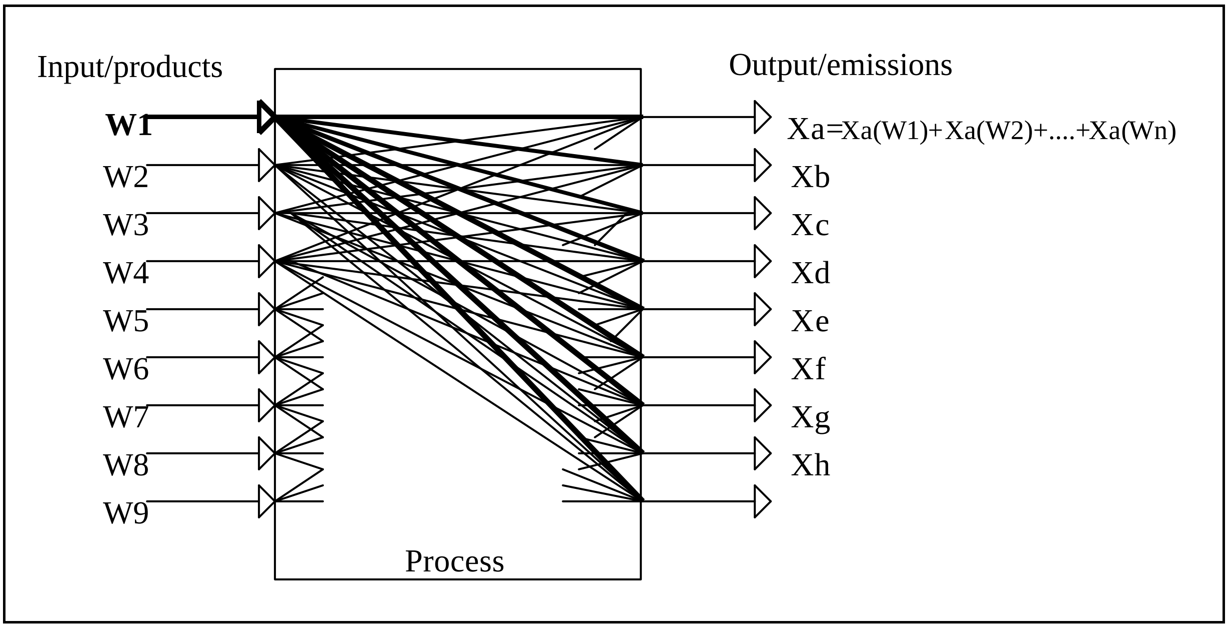

Figure 2.1. In a LCA of product W_1, we have to estimate the fraction of each emission X_i that is caused by the product W_1.

2.2 The time aspects

The emissions may be considered “instantaneous” in most stages of the life time of a product. The emissions from e.g. transports are generated during the transport - and the total transport time is often in the magnitude of a day to a week. The time for manufacture of a product is also rather short. Also most waste treatment methods can be considered instantaneous – the residence time for waste in a combustion plant is about one half hour. However, the waste that is put into the landfill today will generate emissions for several thousands of years in the future, see Figure 2.2. Thus, the time aspects need special considerations for landfilling. The time aspects cause problems both when interpreting field data, and when comparing future emission with instantaneous emissions. To make the landfill emissions compatible to the emissions from the other stages of the LCA we need a method to make a “capitalised value” of the future emissions. Since we can not measure the future emission, we have to develop models that describe the emissions in different time horizons.

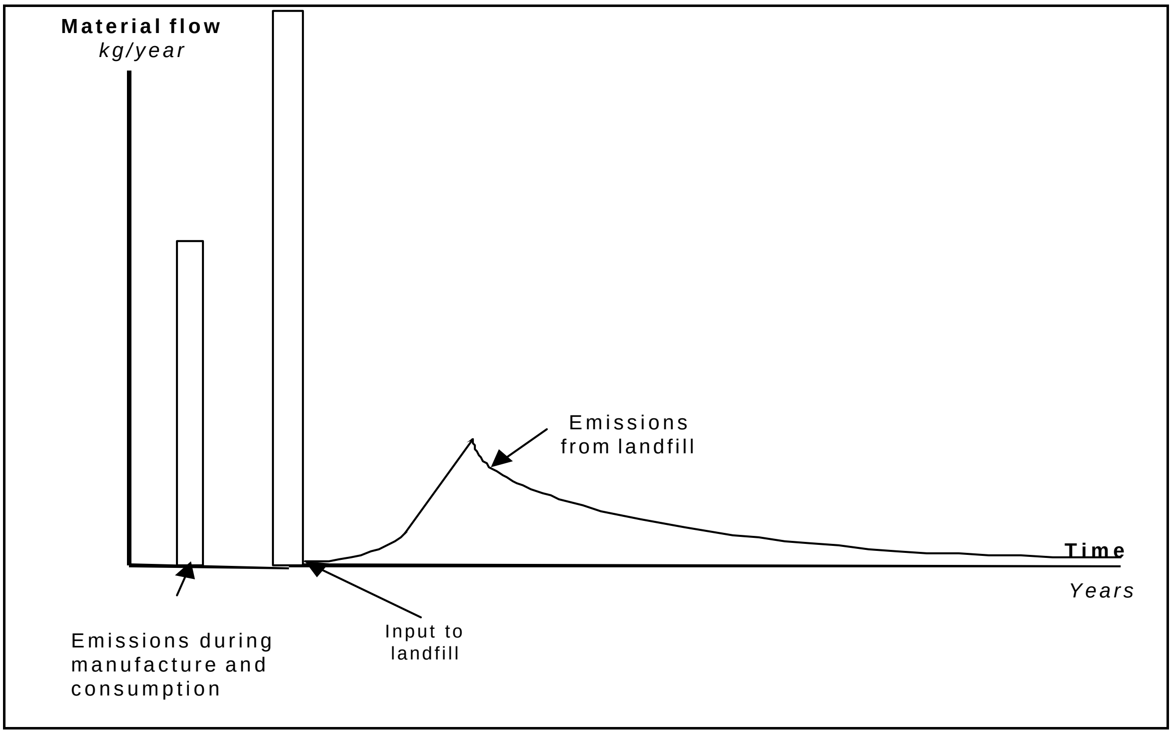

Figure 2.2. Time aspects of input and output in a landfill

2.3 Interpretation of field data

A consequence of these problems is the difficulty to interpret emission data from waste treatment plants and waste landfills. Available emission data from waste treatment plants only show the total emission from the plant, without relation to specific products in the input or to the waste composition. Available emission data from landfills only show the emissions during a short period in the landfill's history, and give no information of the long-term emissions. When handling the waste stage in connection with a

LCA, the emissions from the waste stage must be compatible to the emissions from other stages during the life time.

Another difficulty, when interpreting field data or process data, is the variability of the composition of municipal solid waste. In practice, it is impossible to establish the exact composition of the waste and at the same time making quantitative analyses of emissions. The complexity of the waste makes it difficult on the whole to analyse the composition. Often emissions and other output are measured frequently, while analysis of the input composition is neglected.

3. General approaches

3.1 Allocation

3.1.1 Emission factor

As mentioned above, a traditional problem in LCA is how to deal with processes or groups of processes with more than one input and/or output. The difficulties lies in how the emissions shall be shared between different input parameters. Waste treatment processes are examples of such processes.

Allocation can, in LCA, be defined as *the act of partitioning in some proportionate shares the responsibility for environmental impacts caused by processes in a life cycle* (Lindfors et al, 1995 b).

In LCA practice, when handling multi-input processes, there have been several allocation methods suggested (Lindfors, 1995 a, b):

- by weight
- by volume
- by price or cost
- by causal physical-chemical parameters

Discussions about allocation in connection with waste treatment are given by Clift and Azapagic (1995), Huppes and Frischknecht (1995) and other contributors in the workshop LCA and Treatment of Solid Waste (Finnveden and Huppes, 1995).

In the practical LCA work, emission data is often presented as emission factors. The emission factor can be expressed in different ways depending on the chosen allocation principle. The emission factor is usually defined as the quotient between the emission flow and some input flow expressed in e.g. mass, energy, monetary units, volume units, etc., depending on the chosen allocation principle. The use of emission factors is described in the following.

Let the total waste input to a waste treatment plant be called W_{tot} (expressed e.g. in kg/h), see Figure 2.1. The total input consists of different individual components called W_1, W_2 to W_n (*kg/h*), such as:

$$W_{tot} = W_1 + W_2 + ... + W_n \qquad \textit{(Equation 3.1)}$$

W_1 may be the quantity of newsprint paper in the waste, W_2 may be the PE-plastic, W_3 may be the food wastes, etc.

Let X_a be the flow of a specific emission (e.g. *kg/h*), e.g. CO_2 from an incinerator plant. Each of the materials 1,2,3,...n is supposed to give a potential contribution to X_a:

$$X_{a,tot} = X_{a,1} + X_{a,2} + ... + X_{a,n} \qquad \textit{(Equation 3.2)}$$

where $X_{a,n}$ represents the emission contribution from the material n.

Let U_a be a certain intensive property characterising the input material a. U can be e.g. concentration, energy content, and price depending on the chosen allocation principle. The emission factor can then be expressed by the relationships shown in Equations 3.3.a and 3.3.b:

$$E_a = \frac{X_{a,tot}}{W_{tot} \cdot U_{a,tot}} \qquad \textit{(Equations 3.3a)}$$

$$E_a = \frac{X_{a,j}}{W_j \cdot U_{a,j}} \quad \text{or} \quad X_{a,j} = E_a \cdot W_j \cdot U_{a,j} \qquad \textit{(Equation3.3.b)}$$

When studying a certain process or plant, the total flow of emission a ($X_{a,tot}$), the total input (W_{tot}), and the chosen allocation property or characteristic of the total input $U_{a,tot}$, $U_{a,tot}$, must be known. The emission factor for emission a, E_a, is then calculated by the first equality in Equation 3.3. When making a LCA of the material j, the searched emission $X_{a,j}$ is calculated from the second equality in the Equation, when the allocation property or characterisation of the studied material, $U_{a,j}$, is known (W_j is the amount of the studied material).

We have found that causal allocations are preferable when studying the waste in LCA. Causal allocation means, in this context, that the emission factors will be based on causal properties of the studied product, mainly weight, chemical composition or physical-chemical properties, for example energy content. For different allocation methods, the allocation property or characteristic U_a in Equation 3.3. correspond to:

- *Weight allocation*: U_a corresponds to a constant = 1. This means that every material in the input gives the same emission per kg. In the case of CO_2 this would mean that 1 kg of steel will be assigned the same quantity of emission as 1 kg of paper or 1 kg of coal. If the formed CO_2 is 1,1 kg per kg of waste, then the assigned emission of CO_2 from 1 kg steel will be 1,1 kg, from 1 kg of paper will be 1,1 kg, and from 1 kg of coal will be 1,1 kg.
- *Allocation according to contents of a specific element or substance*: U_a corresponds to the concentration (kg/kg) of an element or substance. In the case of CO_2, and using the carbon content as allocation parameter, this would mean that the amount of emitted CO_2 is proportional to the amount of the studied material and its content of carbon. If 1 kg of waste with a C content of 0,3 kg/kg gives 1,1 kg emission of CO_2, then 1 kg of paper with e.g. 0,4 kg/kg C will give 1,467 kg emission of CO_2, 1 kg PE (polyethylene) with 0,8 kg/kg C will give 2,93 kg CO_2, and 1 kg of carbon-free iron will give 0 kg CO_2.
- *Allocation according to a material property, e.g. energy content*: U_a corresponds to the e.g. the heating value (MJ/kg) of the material a. In the case of CO_2, and using the lower heating value as allocation parameter, this would mean that the amount of emitted CO_2 is proportional to the amount of the studied material and its heating value. If 1 kg of municipal solid waste with a heating value of 10,9 MJ/kg gives a CO_2 emission of 1,1 kg, 1 kg of paper waste with a heating value of 14 MJ/kg will

give a CO_2 emission of 1,41 kg, 1 kg of PE plastic with a heating value of 46 MJ/kg will give a CO_2 emission of 4,64 kg.

3.1.2 Product-related, process-related emissions and permission-controlled emissions

In causal allocation, the major problem is to find a relevant causal relationship between the emission and the material studied. In stage 1 of this project, we divided emissions into *product-related* and *process-related* to understand the formation of different emissions (Sundqvist et al, 1994; Sundqvist et al, 1997). Actually we used the notations ”product-derived” and ”process-derived”, but as consequence of some of the difficulties associated with these notations (see below in Sections 3.1.2 and 3.1.3), the notation ”derived” has been changed to ”related” to indicate a weaker dependence. In the Dutch approach for landfilling the notations ”product-specific” and ”process-specific” were used (see Appendix in Sundqvist et al, 1997).

The product-related emissions are related to the chemical composition of the studied material, while process-related emissions are formed by the process and are difficult to relate to a specific element or compound in the waste. In Figure 3.1 the principles of product-related and process-related emissions are shown. Some examples are as follows.

Heavy metals and non-degradable compounds may be regarded as product-related. They occur in the input material. They will not be formed or destroyed in the process, and the total output will be equal to the input. In an incineration plant they will be emitted through the flue gases, be trapped in the flue gas cleaning system, be trapped in the fly-ash separation system, or be discharged in the bottom ash or slag. These product-related emissions are rather easy to handle in the LCA, since they are often related to the ultimate analysis.

The process-related emissions are results of the process. In the combustion process PAH, CO, dioxins, etc. are formed from all combustible materials. The formation of these emissions is more a result of the process conditions (especially the combustion temperature and oxygen excess), than of the composition of the waste or fuel. For these emissions it is difficult to find relevant causal relations to the composition or to physical-chemical properties.

The process-related emissions will usually cause trouble since they have no obvious relation to the input. Often the process-related emissions are handled as pseudo-product-related, e.g. they are considered to be bound to an element or a property of the material.

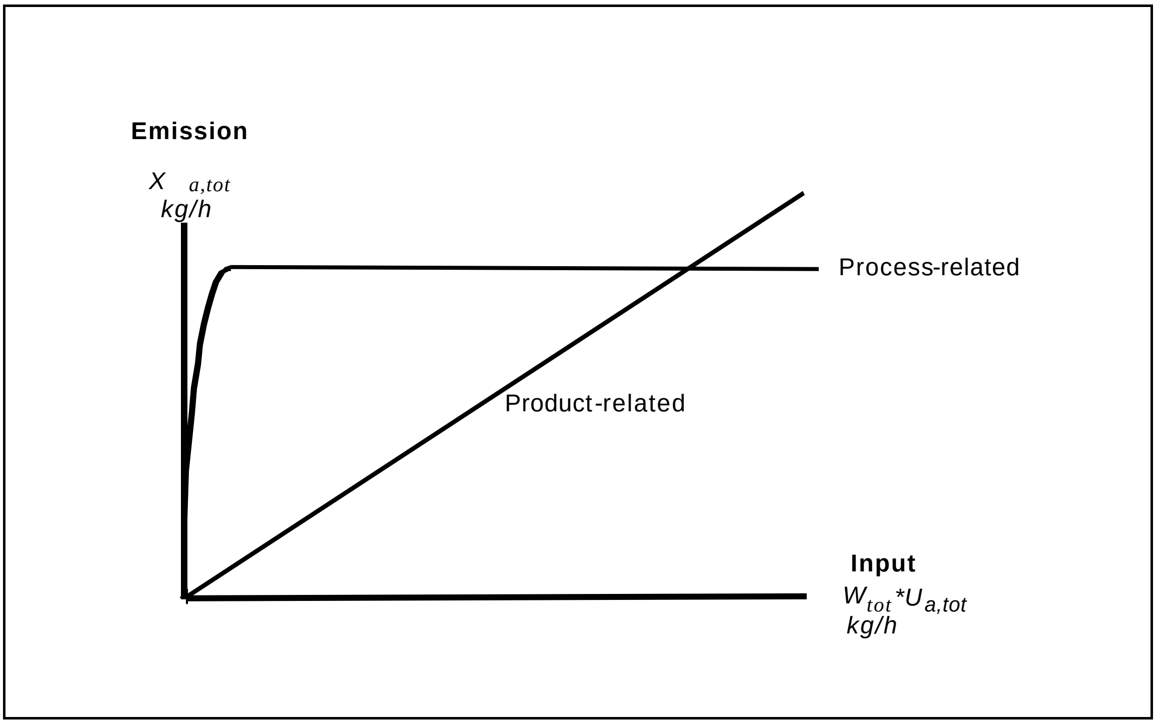

Figure 3.1. Illustration of product-related and process-related emissions

Some emissions may be both product-related and process-related. NO_X from incineration is derived from both

- nitrogen bound in organic and inorganic waste materials (product-related)
- thermal NO_X from the nitrogen - oxygen equilibrium (process-related)

There is also prompt NO_X, which is formed by hydrocarbon radicals reacting with air nitrogen in the flame.

Most emissions are governed by several parameters. Actually both the input and the process control the formation of emittants. The operator of the treatment facility can usually control the process, and also the emissions from the facility. E.g. an increased concentration of chlorine in the waste can be balanced by an increased water flow in the wet scrubber and thus keeping the HCl emission at the same level (kg/h, or mg/nm^3). The waste treatment facilities (in most countries) have to keep the emissions below certain permitted limits. The operating conditions are then often governed by the permitted emissions. Therefore we have suggested *operation-controlled emissions*, or *permission-controlled* as a complement to the product-related and process related emissions. Both product-related and process-related emissions can be controlled by operation (and by permission) at the same time.

The permission-controlled emissions are often relatively easy to handle in waste incineration. Mostly the permitted emission is expressed in mg/nm^3 dry flue gas at CO_2 10 vol-%. This is actually the same as relating the emission to the carbon content of the waste. 1 nm^3 dry flue gas at 10 % CO_2 contains 0,05355 kg elementary carbon. 1 mg/nm^3 thus is equal to 18,675 mg per kg C_{tot}. Using the permission-controlled method is thus equivalent to use an allocation to the carbon content. However, in some countries, e.g. Holland, the flue gas emissions standards are expressed per m^3 dry flue gas at 11 vol-% O_2 - this is not equivalent (but approximately close to) 10% CO_2.

3.1.3 Discussion of allocation and modelling of emissions

According to the definition in Section 1.4.3 allocation is *the act of partitioning in some proportionate shares the responsibility for the environmental impact caused by processes in the life cycle*. It must be observed that emission factors are not generally valid for calculation of the real emissions, e.g. estimation of exactly how the emissions would change if the composition of the waste changes – possibly it can be used for estimating potential emissions. Allocation in LCA is only the question of how the environmental burden shall be shared between different products. This difference between allocation model (model for sharing the environmental burden) and model for calculation of emissions can be discussed in at least two different angles of approach:

- Small changes and large changes.
- Linearity and non-linearity relationship between input and output.

Small changes and large changes

In the case of small changes, the operation and process condistions can be assumed to be constant. The studied component changes the total waste composition only marginally (a few %), and the waste treatment process does not change due to the changed waste composition. Examples of small changes are LCA of products or LCA of specific waste components (e.g. assessing different waste disposal options for paper packages).

Large changes are followed by a significantly change of the waste composition, which changes the operation conditions.

The difference between small changes and large changes is illustrated in Figure 3.2.

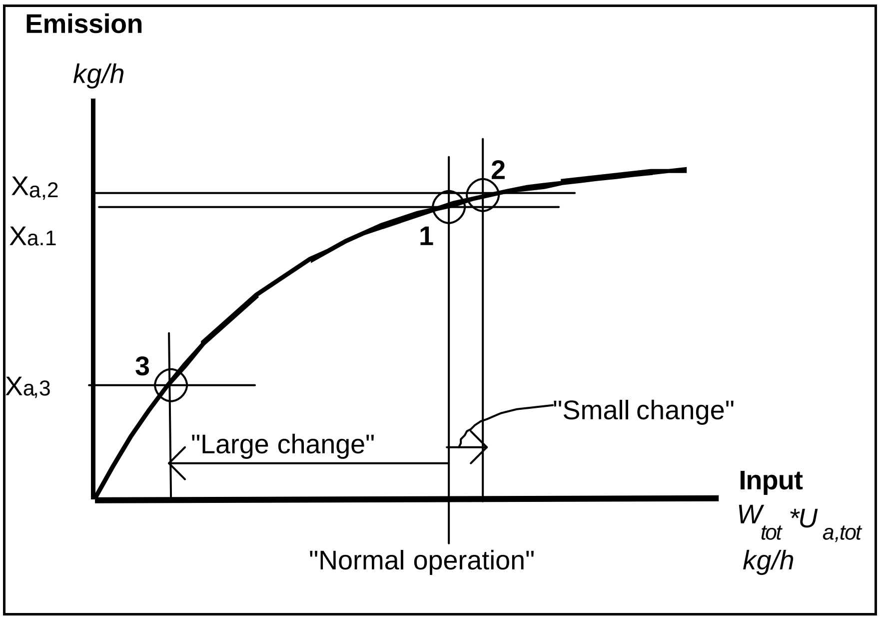

Figure 3.2. Illustration of small changes and large changes.

Emission factors, for use in LCA, are often based on small changes. They are derived from normal emissions at normal operation. They should not generally be used when assessing large changes. Instead other sets of emission factors are needen

Linearity and non-linearity

Let us assume that the emission factor for HCl in waste combustion is 0,06 kg/kg (kg HCl emitted per kg chlorine in input). This means that every chlorine-containing material or product will emit 6 % of its chlorine content as HCl, thus assuming a linear relationship between input and output, see curve 1) in Figure 3.3. However, this linearity is a simplification. In a real plant there may be at least two effects that causes non-linear effects:

a. Even at constant operation condition the relation between input and output can be non-linear, due to for example solubility equilibrium effects. Theoretically the curve can be as curve 2) in Figure 3.3.

b. The emission can be controlled by changes of operation conditions, see curve 3) in Figure 3.3. Thus the emission is depending on both the input and the operation:

$X_{a,tot}$ = *Function(input, operation)*

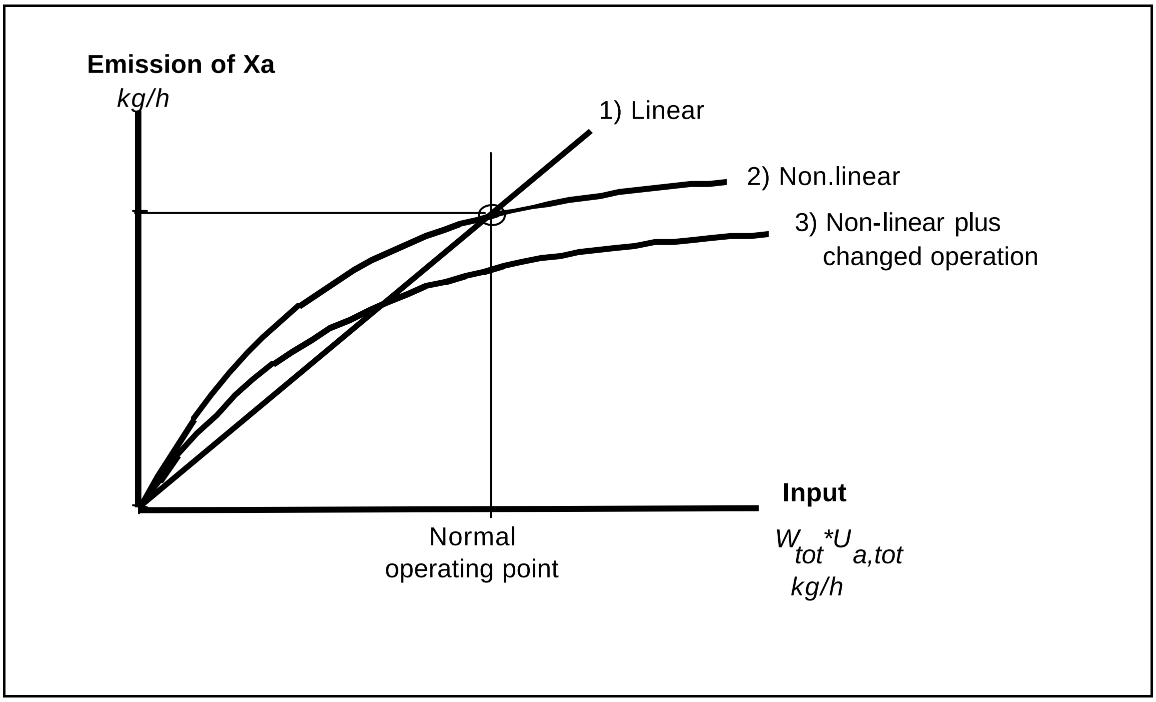

Figure 3.3. Illustration of linear and non-linear relations between emissions and input to waste treatment plant.

The linear emission factor approach is relevant for small changes, e.g. LCA of products, where a specific emission has to be allocated to a specific product. However, this approach is not useful to estimate emissions from a specific treatment facility (large changes).

<u>*Discussion on emission factors for product related emissions*</u>

The above discussion about small and large changes, respectively linear and non-linear output-input relations, shows that the question ”emissions from products” can be studied in several perspectives (the examples given below is based on product-related emission):

1) <u>Allocating the environmental burden from a waste treatment plant between the components in the input</u>. This is the typical question for the waste stage in LCA of products, and when different waste treatment alternatives are studied for individual products. Normally the studied product represents a marginal part of the waste stream input. For this option the linear emission factor approach is applied. This approach may also be called the *average change* approach, see Figure 3.4. The following equation defines the emission factor E_a, and show how the emission $X_{a,j}$ associated with the product j can be calculated:

$$E_a = \frac{X_{a,tot}}{W_{tot} \cdot U_{a,tot}}$$

Equation 3.4

$$X_{a,j} = E_a \cdot W_{tot} \cdot U_{a,j}$$

2) Estimating emissions from a waste treatment plant if the composition of waste is marginally changed (small changes). Let us consider a case where the question at issue is to assess how the emissions are changed if a certain product disappears from the waste stream (or if a new product will occur in the waste stream). This question will require a special approach. It is still ”small changes” that are studied, but the non-linearity must be considered. For solving this problem, the non-linear relation between the input and output must be known. Thus the function $X_a = f(W_{tot} \cdot c_{a,tot})$ must be known, at least at the normal operation point A1. This approach may also be called the *incremental change* approach, see Figure 3.4. The following equations define the incremental emission factor E'_a and show how the incremental emission $X'_{a,j}$ is calculated:

$$E'_{a,A1} = \left[\frac{dX_{a,tot}}{d(W_{tot} \cdot U_{a,tot})} \right]_{A1} \qquad \textit{Equation 3.5}$$

$$X'_{a,j} = E'_{a,A1} \cdot W_{tot} \cdot U_{a,j}$$

3) Estimating emissions from a waste treatment plant when the waste composition is changed considerably (*large changes*), see Figure 3.4. For solving this problem, the non-linear relation between the input and output must be known. Thus the function $X_a = f(W_{tot} \cdot c_{a,tot})$ must be known. An emission factor E''_a can be expressed by the equation:

$$E''_{a,A1} = \frac{(X_{a,tot})_1 - (X_{a,tot})_2}{(W_{tot} \cdot U_{a,j})_1 - (W_{tot} \cdot U_{a,j})_2} \qquad \textit{Equation 3.6}$$

As an alternative, the function $X_a = f(W_{tot} \cdot c_{a,tot})$ can be used to calculate the emission for point A2.

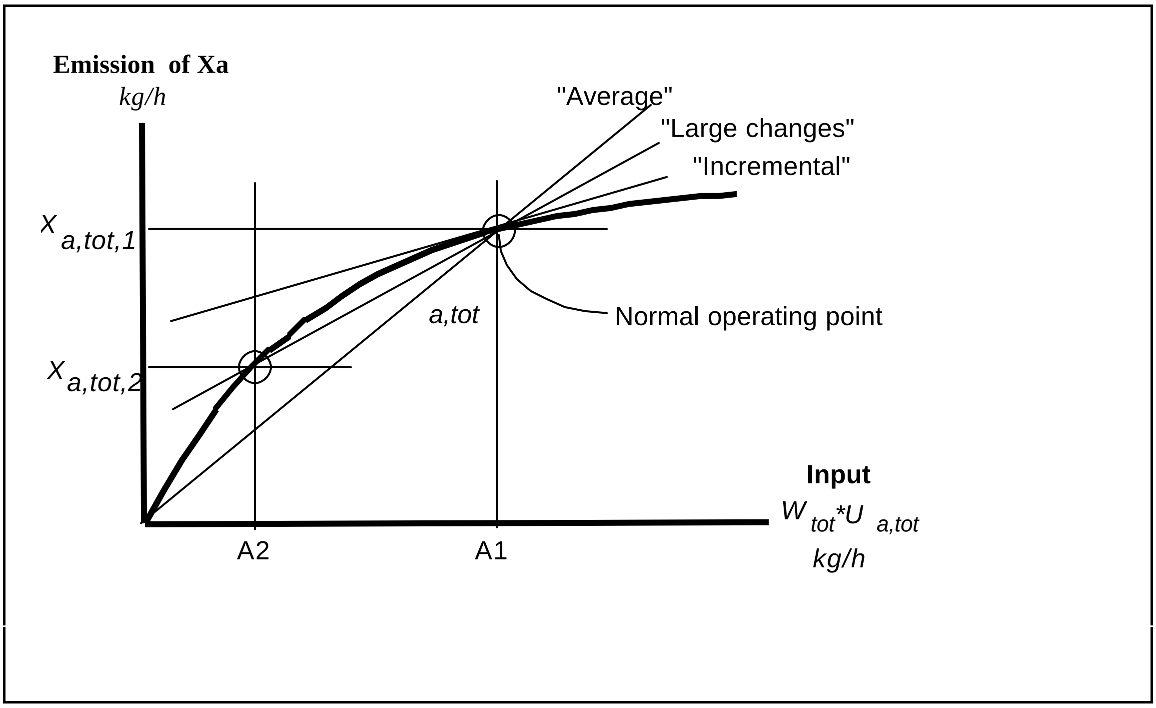

Figure 3.4. Illustration of average changes, incremental changes and large changes.

Discussion of process-related emissions

The process-related emissions are more difficult to handle. By definition these are formed more by the process conditions than by the composition of the input. An example of input-output graph of process-derived emissions is shown in Figure 3.5. The different operating conditions may be characterised by different air excess. When the composition of the input is changed from A1 to A2 (*kg/h*), assuming ”small changes”, the process conditions will not be changed, and the outlet of X_a will be unchanged (operating point will be changed from point 1 to point 2 in the Figure). This corresponds to an emission factor $E_a = 0$. However, if the process conditions (air excess, temperature, etc.) are changed by any other reason, from process conditions C1 to process conditions C2, and without any change in the waste input, the emission of X_a will be changed to point 3 in the diagram. This change corresponds to an emission factor $E_a = 8$. In that case we do not have any dependence between the composition and the output.

There are several possibilities to handle process-related emissions in LCA. In the case of small changes (e.g. LCA of products) the following methods can be used:

1) Simple weight allocation (total weight). It is assumed that every product in the waste participates in the process. Then it is reasonable to assume that the emission caused by the waste should be shared according to the weight of each product.

2) Pseudo-product-related allocation. It is assumed that some physical-chemical property or some element is controlling the emission. However, this is an approximation. In incineration the process-related emissions can be allocated to the combustible material, the total carbon content or the energy content. However, this

kind of assumptions will lead to a lot of objections because they are only approximations.

3) Permission controlled emissions. For several emissions there will be legislative limits of the level of certain emissions. The emissions have to be below these limit values. These limits are often expressed as permitted amount of emittant per nm^3 of dry flue gas at CO_2 10 %. As pointed out in Section 3.1.2, this is actually the same as relating the emission to the carbon content of the waste. 1 mg per nm^3 dry flue gas at 10 % CO_2 corresponds to 1 mg emission per 0,05355 kg elementary combustible carbon in the input. Using the permission-controlled method is thus equivalent to use an allocation to the carbon content.

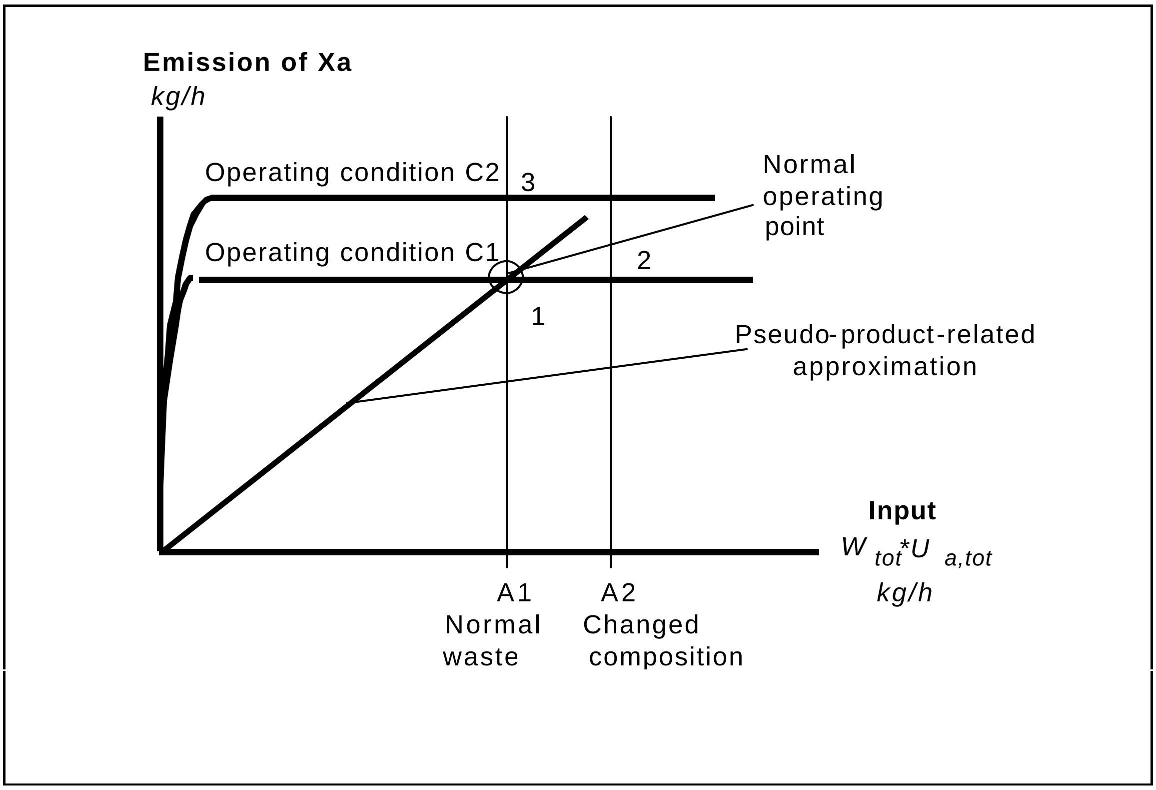

Figure 3.5. Process-related emissions

The nomenclature "product-related" emission and "process-related" emission was introduced to explain the difficulties with finding methods for relevant causalities between emissions and input. However, when deepening the discussions about allocation principles, it seems that these notations may cause some confusion. Probably, this confusion is arisen because no emission is product-related or process-related to 100 %. All emissions are probably more or less both product-related and process-related (and also at the same time operation-controlled, at least within some limits). Product-related and process-related is based on assumption that combustion, or any other studied treatment method, is regarded as one process. Product-related and process-related is also based on the assumption that the formation of the emittants, and/or the distribution of different discharge pathways (emission, flyash, sludge, slag, or similar) is governed by physical-chemical properties only. However, in reality an incinerator plant has several processes in series. For example, let us assume that the incinerator may be divided into two processes: combustion (oxidation) and flow gas cleaning, see Figure 3.6. Let us also assume that the output from each step may be either product-related (possible to express as a certain fraction of the input) or process-related (independent of the input but depending on the process conditions). The output from step 1 can be divided into product-related outputs and process-related outputs. Each of these output flows from step 1 will be an input to step 2, and will in turn give either a product-related or a process-related output from step 2. Thus, when referring to the input to step 1, the emissions from step 2 will be a combination of product-related, combined product-process-related and process-related.

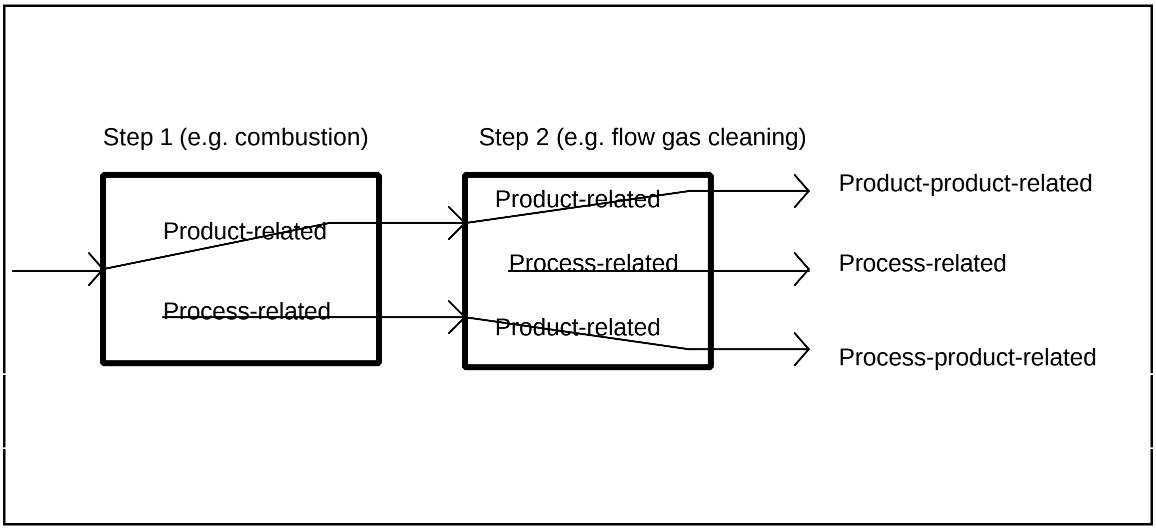

Figure 3.6 Combination of product-related and process-related emissions in a multi-process.

However, that is still a simplification. The real incinerator plant can be divided into several process steps, e.g. combustion, after-combustion, cooling, dust precipitation, and scrubbing or condensation. Each process step causes both product-related and process-related output. The combined processes give rise to several combinations of process- and product-related emissions. In this context also the operational control of emissions should be considered. Several of the emissions can be controlled, within some limits, by operational means in several of the incineration sub-processes. By this discussion follows that the whole problem is very complex. There is no simple way to correctly describe the relation between input and output.

3.2 *Time aspects in landfilling*

3.2.1 General

Wastes that are put into a landfill will cause emissions for a very long time in the future. During the other stages in the life time all other emissions will occur more or less instantaneously, or at least within a limited time period. A landfill may give emissions for thousands or millions of years. An important question is then how the future emissions shall be handled in LCA.

At the workshop *LCA and Treatment of Solid Waste* researchers working with LCA and waste were gathered and discussed different problems (Finnveden and Huppes, 1995). It was suggested that several of the time frame options could be relevant to use. The aims with the LCA, and decisions on system boundaries etc. during the phase ”goal definition and scooping” should govern which time frames that should be used. When time frames were discussed, there was a consensus that the emissions should be integrated over a special period, often called ”foreseeable” period. The suggested length of the period was varying and could be determined by different principles:

- A specific time period; different approaches varies from relatively short time period such as e.g. 15, 50 or 100 years, to long-term periods of, for example, 100 000 or 1 million years.
- Responsibility period for the landfill.
- Processes in the landfill (our approach).
- Period until the concentration of emittants in the leachate reaches an ”acceptable” level or background level, or until the concentration of contaminants in the landfill reaches an ”acceptable” level or background level.
- Period until all input have been output (infinite time).

These principles can roughly be arranged under two different time horizons:

1. A short time period, either a specific time as 15, 50 or 100 years, or the responsibility time (which usually should be 15 - 30 years), or by the processes (our surveyable time period approach, see below).
2. A longer period, either a specific time (e.g. 1 million years), or the period until the emissions reach an “acceptable” level or a “background” level, or the infinite time period.

One difficulty is how to define the different time periods and how to estimate the integrated emissions over each period. Definition of periods must be in relevance with the integration method and the data used for integration.

In our approach, described and discussed in Finnveden (1992), Sundqvist et al (1994), Finnveden et al (1995), and Sundqvist et al (1997), we have chosen to consider both a short-term period and a long-term period. The shorter time period is based on processes in the landfill. In several types of landfills there are consecutive processes. The first period is usually characterised by high internal activities and the formation of emissions is to a large extent depending on parameters governed by the processes in the landfill. Later the landfill will reach a kind of pseudo-steady-state and the formation of emissions is governed more by the external influence (percolation of water, diffusion of air oxygen, etc.).

We have called the shorter time period ***surveyable time period***. It is defined as the time period until the landfill reaches some kind of pseudo-steady-state. We have also stated that the surveyable time period should be of the magnitude one century.

The longer time period is called the ***hypothetical, infinite time***, when all landfilled material has been released to the environment. It is also a worst-case scenario.

For both the surveyable time period and the hypothetical, infinite time period the emissions are assumed to occur at the current moment, $t = 0$.

The surveyable time period is not related to the magnitude of the emissions. We have found that emissions from ash landfills, metal hydroxide sludges landfills and mine tailings were very small during the surveyable time perspective, compared with the emissions during the hypothetical infinite period. This have given rise to the objection that the surveyable time approach may give an underestimation of the total, potential environmental impact from landfills, for example if all other emissions after the surveyable time period are neglected and/or forgotten. Therefore, in stage two of our study we introduced the ***critical time period,*** which should be the time period until the major parts of emissions have occurred. In contrast to the surveyable time period, the critical time period is related to the magnitude of the emissions. The critical time is not essential for the inventory phase, it is more a tool to make the characterisation and valuation easier.

The total, integrated emissions during the surveyable time period and the hypothetical infinite time period can be compared with the emissions from the other stages in the LCA. The emissions during the surveyable period are a measure of the emissions caused by the landfilled waste during the closest foreseeable future (in the magnitude of one century). The emissions during the hypothetical infinite time are a “worst-case” scenario where all landfilled material has been released to the environment. By this distinction, the assessment of landfilled waste will be dependent on how different hazardous constituents are bound in the waste landfilled waste, and on different processes in the landfill, and not only the total composition of the waste.

The critical time period is more a deepened description of the leaching rate and when the major part of the leaching will occur. The critical time approach should usually not be used in initial or screening LCA:s, but rather in a deepened analysis when the screening analysis has shown that the landfilling is of importance, and that the time aspects are of importance for interpretation of the result.

An alternative to this critical-time approach can be to estimate the half times, the time when half of the emissions have occurred.

Our main approach for calculating the emissions from the surveyable time period and from the hypothetical, infinite time period is as follows:

1) The chemical, microbiological and physical processes in the landfill must be known. Information of the landfill processes can be obtained from a combination of mathematical models and experience from laboratory tests and from field data.

2) The composition and relevant information about physical-chemical and biological properties of the waste, or of the studied product, must be known.

3) The emissions during both surveyable time period and the infinite time period can be estimated from the analyses of the waste (ultimate analysis, proximate analysis and analysis of species) with knowledge of the landfill chemistry. Emission factors for some materials and for some different types of landfills are given below in this report.

The surveyable time period approach makes the emissions independent of site-specific data. The length of the surveyable time period may be different in e.g. the north of Sweden and the south of Italy, but the emissions during the surveyable time period will be equivalent for the same waste and the same type of landfill.

However, for calculating the critical time period emissions, also the kinetic rates of the leaching processes must be known.

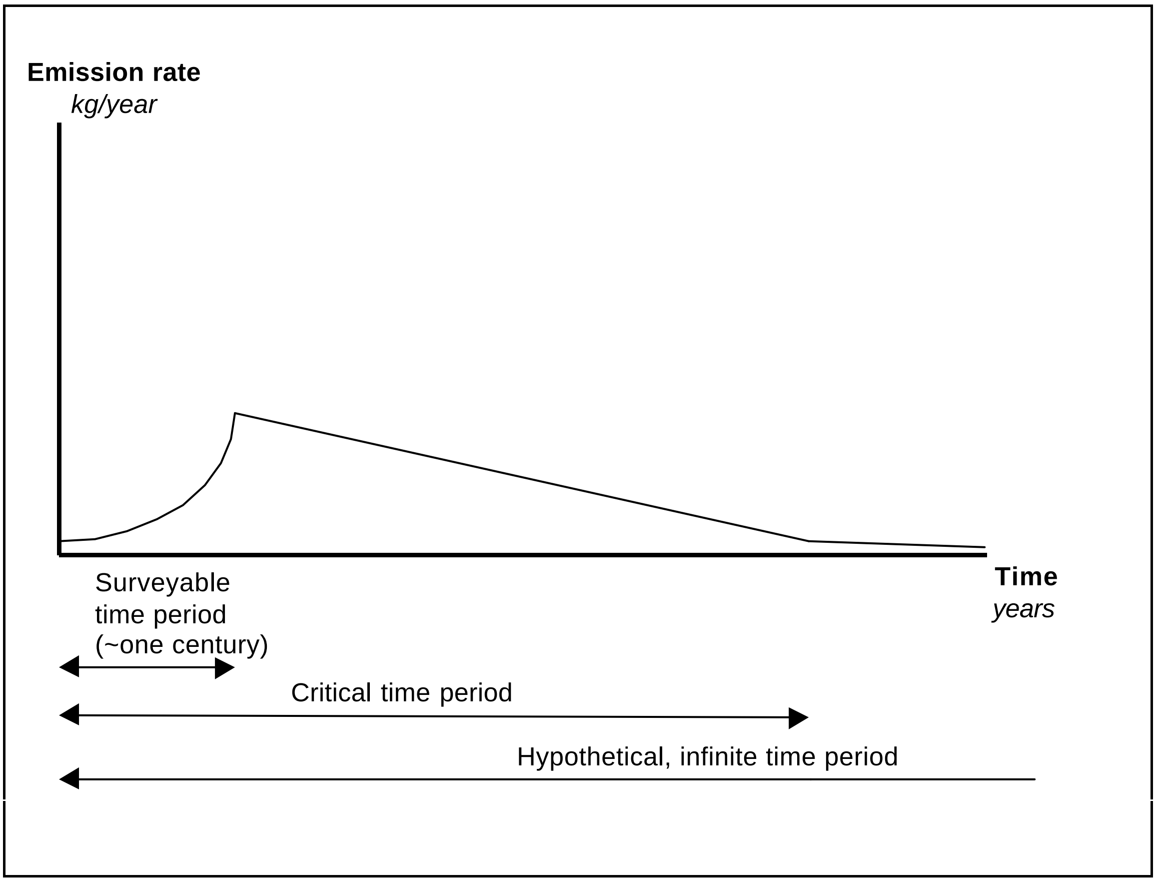

Figure 3.7. Illustration of surveyable time period, critical time period and hypothetical infinite time.

For municipal solid waste landfills we have in our earlier studies (Finnveden 1992, Sundqvist et al 1994) identified the surveyable time period to the period until the later part of the methanogenic phase.

For some industrial waste landfills we found it more difficult to identify the surveyable time for different reasons (Sundqvist et al 1994). For metal hydroxide sludge there is probably no true ”surveyable time period” according to our definition. Since the metal hydroxide sludge is almost water saturated when landfilled, it is expected that the landfilled masses will be in a kind of steady-state from the beginning. For sulphidic mine tailings the true surveyable time will be about 1000 years, estimated from the reactions: buffering, air diffusion into the landfill, and oxidation of sulphidic sulphur. This is to long according to the criteria “the magnitude of one century”. For both the metal hydroxide sludges and the mine tailings we based the proposed emission factors on a 100 year period, rather than the time until a pseudo-steady-state was reached.

In stage 1 of the project we also compared calculated emissions from landfills with emissions from other stages in a LCA for some materials (Sundqvist et al 1994, Finnveden et al 1995), see also Chapter 8 in this report. These comparisons showed that potential emissions from landfilling, in some cases will influence the results significantly. Thus, if landfilling is neglected - which it often has been in connection with LCA - misleading conclusions may be drawn.

3.2.2 Discussion of other approaches

As mentioned above in Section 3.2.1, there are several other possibilities to handle the time aspects, both in the short time perspective and the long time perspective.

Alternative short-term approaches

Other approaches considering shorter periods are generally based on a certain time period (a certain number of years). To estimate emission factors for these specific periods there are two possibilities:

1. Measure the emissions from a landfill with known composition of the waste during the specified period. However, this seems to be impossible. Modern landfill have existed for only 20 – 30 years, and the composition of the landfilled waste is usually very bad documented.
2. Theoretical modelling. The kinetics of the reactions must be known.

Alternative long-term approaches

The long-time perspectives are difficult to handle, because there is a lack of relevant emission models. Different leaching models may be assumed and tested, but there is a great risk that the models are irrelevant. In the longer periods we also have to consider other emission pathways than by leachate and gas. For example erosion can be of relevance in the long time perspective. Also natural deformation of the landscape caused by land elevation, earthquakes, flooding, or glacial ice will also influence the release of emittants, which has to be considered in modelling.

Acceptable level. It is possible to set up quantitative criteria for ”acceptable level”. Today’s acceptable levels can be obtained from e.g. different guidelines for risk assessment of contaminated land and contaminated ground water. These acceptable levels are different in different countries. The future acceptable levels are unknown. It may be irrelevant to use today’s acceptable levels, since the environment may be more sensitive in the future, depending on a slowly increasing background contamination. (On the other hand, sometimes the authorities state that the only acceptable level is zero.)

Background level. It is also possible to set up quantitative criteria for ”background level” or ”when the landfill becomes a part of the environment”. There is information available about the current background levels of metals and some organic compounds in soils, as well as in surface water and ground water.

Next glacial period. The next glacial period is a more interesting angle of approach to study, even if the glacial period is uncertain. Glacial periods have occurred in Scandinavia frequently. During the last glacial period the south of Sweden became free from glacial ice about 16000 years ago, and the north of Sweden about 9000 years ago. The glacial ice has reach down to the Alps in Europe. Glacial periods have occurred in also in North America during the Quaternary Age (the last million years). About 30 % of the land area of the Earth have been covered by glacial ice (today about 10 % is covered). From the history, it is to expect that next glacial period in Sweden will occur about 20 000 - 50 000 years from now. On the other hand, this historic tendency can be counteracted by the greenhouse effect. The temperature declination, that is a part of the natural climate cycles, may be neutralised by the temperature inclination caused by the greenhouse effect. We estimate that for several kinds of landfills the emission factors for metals and for a 20000 year period to the next glacial period can be roughly estimated to be within 0,001 - 0,5 kg/kg (see Chapter 4.3, 5, 6 and 7). Thus the major parts of the metal emissions have not occurred at time for the glacial period. However, after the glacial period all landfilled wastes have been released to the environment (in that aspect our hypothetical, infinite time period approach considers the emissions until next interglacial period). The time period until next interglacial period may be several hundred thousands to one million years.

From this discussion about the aspects of the longer time period we draw the conclusion that the hypothetical, infinite time period is most easy approach to use. There is a lack of reliable models for predicting the long-term behaviour of landfills. But if such models become developed criteria such as acceptable level or background level should be considered, at least in a deeper study. The hypothetical, infinite time period approach should still be good for a first estimate in a screening analysis.

The long-term perspectives should also be considered in a historical perspective. 1 million years is a very long time. The human being (Homo Sapiens) has not existed for that time yet, only about 100 000 years. When we are discussing a 1 million year perspective we are discussing a time period that is ten times longer than the human being has existed. In Chapter 4 the time aspects for incinerator ashes are discussed. The calculated time for complete leaching is 0,1 - 10 million years, different for different elements and for different landfill types. After 1 million year there may still be material in the landfill, but probably the major part of all emissions has occurred.

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4. Incineration of MSW

4.1 The incineration process

Incineration or combustion is a thermal treatment of the waste with excess of air. The organic matter is oxidised to mainly CO_2 and H_2O, which is discharged to the air recipient. Noncombustible, inorganic is discharged as a slag or ash. The energy deliberated during the combustion can be used for steam production, district heating production and eventually production of electric power. The process also gives unwanted emissions to the environment, such as nitrogen oxides, sulphur oxides, hydrochloric acid, heavy metals, polyaromatic hydrocarbons (PAH), chlorinated organic compounds (for example TCDD and other “dioxins”), etc.

A modern incinerator is a complex industrial process plant involving several process steps in order to optimise the energy production and to minimise the unwanted emissions. The process plant can be divided in several sub-plants of which the most important are (see also Figure 4.1):

- Combustion chamber, where the solid material is combusted.
- After-combustion chamber, where the gases from the combustion chamber are hold at high temperature and oxygen excess in order to oxidise unburned gases.
- Boiler, which recovers the energy from the flue gases.
- Flue gas cleaning system (there are several systems available)
- In cases of wet flue gas cleaning there is also a water treatment system.
- Ash handling system.
- Landfilling of slags and ashes.

The system borders used in this study is defined by Figure 4.1. In Section 4.2 is given emission factors for incineration. Landfilling of fly-ash and sludge is presented and discussed in Section 4.3.

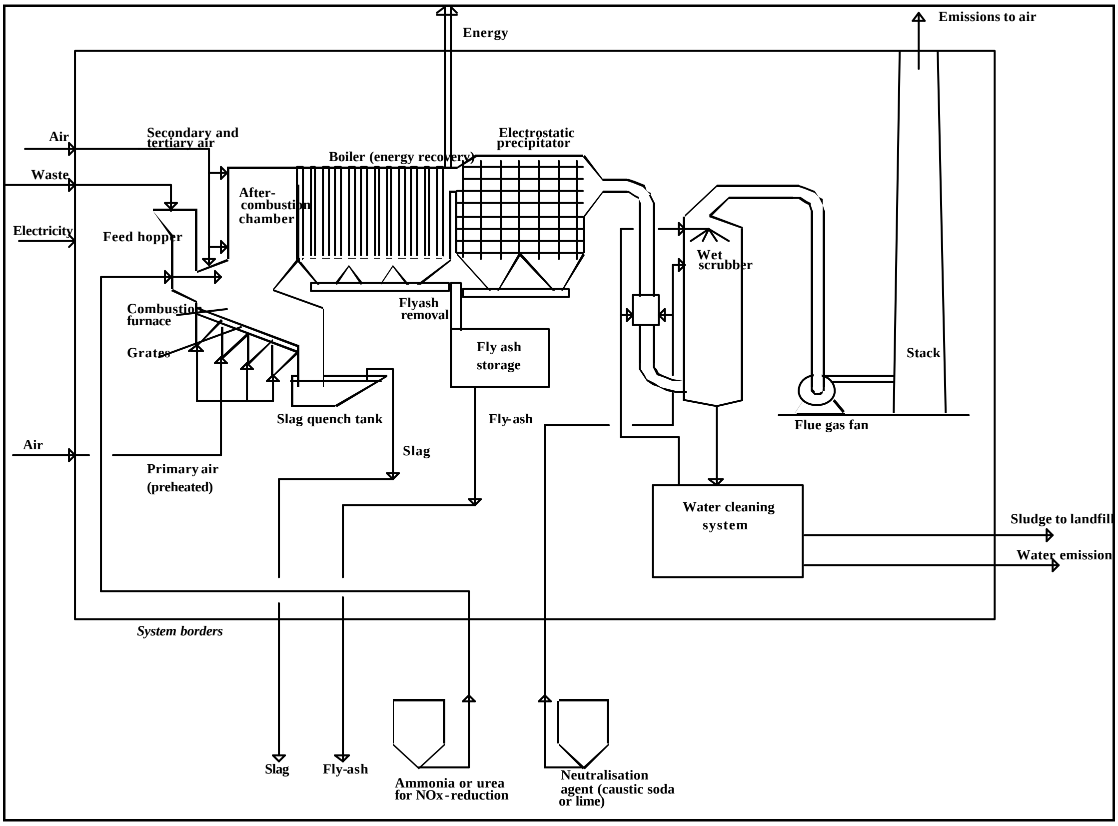

Figure 4.1. Illustration of an incinerator plant, and the system boundaries used in this report.

4.2 Emission factors for incineration

As described above in Chapter 3, there are two major classes of emissions:

- *Product-related emissions*, where the emitted components directly come from the waste and the quantity of the emissions can be calculated directly from the composition of the waste. Examples of such emissions are heavy metals, CO_2 and SO_2.
- *Process-related emissions*, for which the formation is more controlled by the process than by the composition of the combusted material. Typical examples are PAH, CO, dioxins, etc.

Some specific emissions can be formed in both ways, e.g. NO_x where a portion is formed from the nitrogen in the waste (fuel NO_x), and another portion is formed from the oxygen-nitrogen equilibrium (thermal NO_x). Still another portion is formed by reactions between air nitrogen and hydrocarbon radicals in the flame (prompt NO_x). The fuel NO_x is a product-derived emission, while the thermal NO_x and the prompt NO_x are process-derived.

In some cases it may be useful to work with *permission-controlled-emissions* or *operation-controlled emissions* (see Sections 3.1.2 and 3.1.3). Most of the emissions that are handled as product-related emissions are not product-related to 100 %. Both product-related emissions and process-related emissions can be controlled by the operation. The emission can be decreased by operational means, especially in the flue gas cleaning system. If a certain emission at a certain plant reaches a too high level, it can be lowered by changing of process parameters, e.g. increasing the water flow in the scrubber, decreasing the condensation temperature, etc.

The allocation principles of the product-related emissions are usually based on an elementary analysis of the waste component.

The allocation principles of the process-related emissions are more complex and different methods have been suggested. There are three main approaches:

1. Since there exist no unambiguous relations between the input and the formation of the emittant, a simple weight allocation can be used (see also Section 3.1.3). All materials will be accounted for the emission.
2. The emission is handled as a pseudo-product-related emission. E.g. CO can be related to the total carbon input, and thermal NO_x can be related to the energy content (heating value). Some emissions can be related to several input parameters, e.g. dioxins may be considered as being formed from both combustible material and from chlorine.
3. The emission is handled as permission-bound, which in most cases is the same as relating the emission to the total carbon input (see Section 3.1.3).

Emission factors for incineration have been derived from emission data from Swedish incinerator plants as reported by RVF (1997). The emission factors are in most cases related to average Sweden (data from 22 plants). The major emissions reported by RVF are according to Table 4.1. It should be observed that there is no detailed data about the material composition or elementary composition of the waste.

In Table 4.2 is given some proposed emission factors for incineration, mainly based on data from RVF (1997). Emission factors are presented for different allocation methods. The derivation of the emission factors is discussed in Sections 4.2.1 and 4.2.2.

Mass balances for metals are presented in Table 4.3. These are derived from data from the incinerator plants in Uppsala and Stockholm. These figures seem to vary from year to year and from plant to plant, but the presented figures seems to be reasonable relevant.

In Table 4.4 is the volatisation temperatur given for some heavy metal compounds. These temperatures have been used to estimate the fraction that is released from the plant with the slag (bottom ash)

Examples of emissions from different materials in the waste are given in Table 4.5. Results from different allocation methods are presented to illustrate the consequences of different choices.

4.2.1 Product derived emissions

Product derived emissions have been derived from material balance of incinerators. Different sources have used.

Carbon dioxide CO_2 to air. The carbon contents of waste and of different materials have been taken from several literature reports. The data used here is mainly from Sundqvist et al, 1997. The carbon content in MSW has been assumed to be 0,30 kg/kg. The oxidation of C to CO_2 has been assumed to be 96,7 % (the rest is carbon in slag and flyash). Carbon from fossil materials (synthetic rubber, plastics, synthetic textiles, etc.) are considered to produce antropogeneous carbon dioxide giving a contribution to the greenhouse effect, while wood, paper, vegetables and animalic materials are considered to produce biogenic carbon dioxide which are considered to be a part of the natural cycles and not give a contribution to the greenhouse effect.

Sulphur dioxide SO_2. Sulphur in waste will be oxidised and transferred to the slag, fly-ash and the raw gas. The sulphur removal depends on the flue gas cleaning process. Usually the neutralisation step is designed for only HCl removal, which will give a poor removal of SO_2. When the neutralisation step is designed for both HCl and SO_2, which requires two separate washing steps with different pH, both HCl and SO_2 can be removed efficiantly. The SO_2 is neutralised in the flue gas cleaning system and precipitated in the flue-gas cleaning residue. Sulphur will also be transfered to the slag and fly-ash. The S content has been estimated to be around 0,003 kg/kg in MSW. 1 kg of S in the waste will give the following SO_2 emissions to air:

- with a separate SO_2-absorption step: 0,05 kg SO_2/kg S
- without separate SO_2-absorption step: 0,5 kg SO_2/kg S
- average Sweden: 0,20 kg SO_2/kg S (based on figures in RVF(1997).

The rest is present in the flue gas cleaning residue, in the slag or in the fly-ash.

Hydrochloric acid HCl. Chlorine in the waste will be transferred to mainly the raw gas in the combustion chamber, but some amounts will also precipitate in the slag and fly-ash. The chlorine in the gas is mainly present as hydrochloric acid HCl, even if some alkali chlorides will be present . In the flue gas cleaning HCl is neutralised to $CaCl_2$ (with lime or limestone) or NaCl (with caustic soda). The content of Cl in waste is estimated to be approximately 0,005 kg Cl per kg waste. The emission reported by RVF (1997) corresponds to 0,0002 kg HCl per kg waste which gives an emission factor of 0,04 kg/kg (emission to air). All Swedish plants are equipped with flue gas neutralisation.

Dust emissions to air. Dust can be allocated in different ways.

a) The easiest way is to assume that the dust emission is proportional to the ash content (the same as non-volatile substance or inorganic content) from the proximate analysis. The ash in the waste material is then assumed to be distributed to
   - slag
   - fly ash and flue gas cleaning waste
   - dust (emitted to air)

   The ash content in the waste is estimated to be approximately 0,20 kg ash per kg waste. The dust emissions reported by RVF (1997) is 0,000 0179 kg dust emission per kg waste, which gives an emission factor of 0,000 092 kg/kg. However, this is probably not always true. A 1 kg solid piece of steel will probably give a lower dust emission than a 10 kg plastic product with 10 % inorganic filling material. So there should be a distinction between solid inorganic (metallic) products and products where the inorganic material is dispersed in a matrix of combustible material. We have not enough information to quantify this difference.

b) Because of the difficulties to get an appropriate allocation of dust, weight allocation is an alternative. With the data from RVF (1997) the dust emissions are 0,000 0179 kg dust emission per kg waste material (independent of ash content in the material).

Heavy metal emissions to air. Heavy metals are distributed between slag, fly-ash, flue-gas cleaning waste (sludge) and emissions to air (as a component in dust). In our first report (Sundqvist, 1994) we reported a material balance from an incineration plant, see Table 4.3.a. These figures have been updated with new analyses and the corresponding analyses from another incinerator plant, see Table 4.3.b-c. As can be seen there are large variations in the material balances, probably due to variability in the waste composition and insufficient sampling (the sampling of ash, slag, and fluegas does not cover the variations in the waste flow).

When using these figures in a LCA, it should be considered how the metal is present in the waste. The metals occur mainly in two forms: a) as metals or alloys, and b) as a compound dispersed in another medium, e.g. additives in plastics or paper, pigment in paints, etc. The transfer of metal from the solid phase to the gaseous phase during combustion is governed by mainly two mechanisms: a) physical entrainment of solid particles, and b) volatilisation. Physical entrainment is expected to occur when the metal is dispersed in another medium that is destroyed during combustion. Examples are a metal compound used as an additive in a plastic, or a metal compound used as pigment in a paint. The physical entrainment can occur irrespective of if the metal is volitilised or not. The volatilisation can occur when if the metal volatilises at the combustion temperature, or if the metal forms volatile compounds during the combustion (mainly oxide and chloride, eventually also sulphate). A non-volatile metal, e.g. iron, is expected to remain in the solid phase and be discharged in the slag. More volatile metals, can be expected to volatilize into the gas phase. When the flue gas is cooled in the boiler and in the flue gas cleaning step the metals will recondense (as metal, oxide, chloride, etc. depending on the chemical environment) and be discharged in the fly-ash, the flue-gas cleaning waste or as a dust emission to air. In the combustion chamber the temperature in the solid phase is 800 - 1000 ^{o}C, and the residence time of the solid phase is about 20 - 30 minutes. Larger metallic pieces will not react in the combustion chamber. Scrap

pieces will come out only slightly effected by the combustion process. For example, tin cans are often possibly to identify in the slag (however the tin layer usually seems to oxidise). Alloy metals are still bound in the metallic phase. Also the thickness of the metal pieces can be of importance. Thicker aluminium pieces will still be possible to identify (possibly the piece can be deformed by melting), while thinner pieces such as foils and beverage cans usually seem to be oxidised.
In Table 4.4 the volatilisation temperature is given for some metals and metal compounds.

When making a LCA where metal emissions are of importance, the actual metal properties and the geometry of the studied product should be considered:

- A larger part than indicated in Table 4.2 or 4.3. is expected to occur in the fly ash and in the dust emissions, when the metal or metal compound is dispersed in a combustible medium.
- For metallic pieces it is likely that most of the metal and alloy metals are intact and are discharged in the slag. In that case the transfer to the fly ash or to the air emissions is lower than indicated by Table 4.2 or 4.3.

4.2.2 Process related emissions

Process related emissions are difficult to relate to the composition of the input waste, since they are more dependant of the process conditions than of the composition of the waste. Here two main strategies are proposed for allocation of the process derived emissions:

1) Weight allocation. The process derived emissions are referred to all incoming material to the incinerator. Every material is sharing its part of the formation of these emissions, independent of its composition.
2) Total carbon allocation (C_{tot}). This is a modification of the weight allocation, where the emission from the waste is shared between the combustible materials, and incombustible materials are considered to not be involved in the formation. Equivalent alternatives are to allocate to the heating value or to the combustible material. However, the C_{tot} allocation has been preferred since this also is the basis for permission-controlled emissions.

The result of different allocation methods presented in Table 4.2 and 4.5. As seen, there are relatively low differences between the methods, see further the discussion in Section 4.2.8.

The total carbon allocation is not always satisfactory when analysing the causalities. According to the total carbon allocation model an increased carbon content will increase the emission. By analysing the combustion process, however, an increased carbon content (or increased heating value, or increased concentration of combustible material) is expected to give a better process condition, which reduces the formation of the actual emission. When considering this aspect it is found that an inverse carbon allocation (allocating factor = $1/C_{tot}$) eventually could be more appropriate in some cases.

Dioxin from waste incineration is a hot question. The role of chlorine for formation of dioxins has been discussed several times. There seems to be two different points of view:

1) There is a correlation between chlorine in waste and dioxin formation. This point has been emphasised by e.g. Greenpeace (Costner, 1997). Costner has made a literature survey, from which she claims that there is a correlation between chlorine in the waste and dioxin emission from incineration.
2) There is no correlation between chlorine in waste and dioxin emission. This point has been emphasised by e.g. Chandler (1997) and Rigo et al (1995). Their studies are based on 1900 published measurements of chlorine and dioxin, which have been assessed by statistical analyses, which showed no correlance.

Here, we have presented emission factors for dioxins, based on different allocation method:

- Weight allocation and carbon allocation are according to the main alternatives as for other process derived emissions.
- The heating value allocation is an alternative to the carbon allocation.
- Chlorine allocation is given as an example of consequence if only chlorine is used as allocation parameter.
- An alternative to weight allocation or carbon allocation, if the chlorine content is wished to be considered, is allocation according to both carbon and chlorine. The causal relation motivating this allocation is that dioxin is formed from both combustible material and from chlorine. The dioxin is formed in the combustion process due to the presence of both combustible material and chlorine. Then it is reasonable that both the combustible material (here counted as the carbon content) and the chlorine should share the guilt for dioxin formation. Dioxins are presented as 2,3,7,8-TCDD-equivalents. The weight ratio between carbon and chlorine is approximately 50:50 (actually 50,3 : 49,7) in 2,3,7,8-TCDD. In the carbon-chlorine allocation 50% of the dioxin formation is considered to come from the carbon content and 50% from the chlorine content.

Also for NO_X emission there are different allocation alternatives. According to the discussion from stage 2 of our study (Sundqvist et al, 1997) the main part (95%) of the NO_X comes from the nitrogen in the waste, and a smaller part (5%) from thermal NO_X. The combined allocation to N in waste and to heating value of waste seems to be the most appropriate way to allocate. The N in waste and total carbon in the waste is an equivalent alternative.

4.2.3 Energy

Consumption of energy

The incinerator plant consumes electric energy mainly for fans (in flue gas system) and cranes (for loading waste). Different sources report different information about the energy consumption. We suggest 0,25 MJ_{el} per kg waste as a reasonable compromise.

Production of heat and electricity

Produced energy is calculated as follows. The upper heating value and the lower heating value for the studied product or waste should be known. In Sweden all incineration plants (22 plants) produce district heating, 3 of them also produce electricity, see Table 4.2. In Europe, electricity generation seems to be more common. The thermal efficiency has been assumed to be 85% (related to the lower heating value) as an average, without flow gas condensation system. For plants with flue gas condensation the condensation heat should be accounted. Assume that 80 % of the water is condensed, and that the heat exchangers have an efficiency of 80%. The evaporation heat for water is 2,26 MJ/kg water, and the specific heat 4,19 kJ/kg,^{o}C. If the temperature of the fluegas to the condensation step is 100 ^{o}C, and the outlet temperature is 55C^{o}, this will give a net heat production of about 1,6 MJ/kg water in waste.

Literature data about heating values for different materials are often differing from each other because ash content and water content often are varying. The heating value can also be expressed in several ways, relating to organic content, to ash-free material or to ”raw” material with ash and moisture. The relation between the upper (calorimetric) and lower (effective) heating value can be calculated according to the following formulas (Härlin, 1943).

$$
\begin{aligned}
&\Delta H_{eff,b} = \Delta H_{cal,b} - 22{,}08 \cdot x_{H,b} \\
&\Delta H_{cal,t} = \Delta H_{cal,b} \cdot (1 - x_{ash,t}) \\
&\Delta H_{eff,t} = \Delta H_{cal,t} - 22{,}08 \cdot x_{H,t} = \Delta H_{cal,b} \cdot (1 - x_{ash,t}) \\
&\Delta H_{cal,raw} = \Delta H_{kal,t} \cdot (1 - x_{H2O,raw}) = \Delta H_{cal,b} \cdot (1 - x_{H2O,raw} - x_{ash,raw}) \\
&\Delta H_{eff,raw} = \Delta H_{cal,raw} - 22{,}08 \cdot x_{H,raw} - 2{,}451 \cdot x_{H20,raw} \\
&\Delta H_{eff,raw} = \Delta H_{cal,b} \cdot (1 - x_{H2O,raw} - x_{ash,raw}) - 2{,}451 \cdot x_{H2O,raw} \\
&\Delta H_{eff,raw} = \Delta H_{eff,t} \cdot (1 - x_{H20,raw}) - 2{,}451 \cdot x_{H2O,raw} \\
&x_{H,t} = x_{H,b} \cdot (1 - x_{ash,t}) \\
&x_{H,raw} = x_{H,t} \cdot (1 - x_{H20,raw}) \\
&x_{H,raw} = x_{H,b} \cdot (1 - x_{H2O,raw} - x_{ash,raw}) \\
&x_{ash,raw} = x_{ash,t} \cdot (1 - x_{H2O,raw})
\end{aligned}
$$

(*Equation 4.1)*

Where

H_{eff} = Lower heating value (effective heating value), *MJ/kg*
H_{cal} = Upper heating value (calorimetric heating value), *MJ/kg*
x_H = Hydrogen content, *kg/kg*
x_{ash} = Ash content, *kg/kg*
x_{H2O} = Moisture content, *kg/kg*

Indices (second lower index)

b	related to combustible (organic) material without water or ash
t	related to dry material with ash but without moisture
raw	related to raw material with ash and moisture

Results from laboratory analyses are often presented in relation to “dry material with ash”.

If heating values for the studied material is not available. it can be estimated from the elemental analysis by empirical formulas. For solid fuels (wood, peat, coal etc.) the following has been proposed (Härlin, 1943):

$$\Delta H_{eff,b} = 34{,}36 \cdot x_{C,b} + 117{,}3 \cdot x_{H,b} - 12{,}15 \cdot x_{O,b} + 0{,}84 \quad \textit{(Equation 4.2)}$$

where

x_C = carbon content, kg/kg
x_H = hydrogen content, kg/kg
x_O = oxygen content, kg/kg
with indici as above (index *b* refers to combustible (organic), dry, water-free material)
(the factor $34{,}36 \cdot x_{C,b}$ should not exceed 33,1)

4.2.4 Consumption of additives

Limestone $CaCO_3$, slaked lime $Ca(OH)_2$, or caustic soda (NaOH) is consumed for reducing the concentration of sulphur dioxide and hydrochloric acid. For stoichometric neutralisation of HCl from 1 kg Cl is required 1,4 kg limestone or 1,05 kg slaked lime. For stoichometric neutralisation of SO_2 from 1 kg S is needed 1,56 kg limestone or 1,15 kg slaked lime.

Usually ammonia NH_3 or urea NH_2CONH_2 is used for reduction of NO_x. Theoretically the stoichometric consumption for NO_X-reduction is approximately 0,48 kg ammonia per kg of reduced NO_X, or 1,5 kg ammonia per kg N in reduced NO_X. In case of urea. the stoichometric consumption is approximately 0,85 kg urea per kg reduced NO_X, or 2,65 kg urea per kg N in reduced NO_X. However, because of the complicity of the NO_X-formation process and the NO_X-reduction-process it is recommended to use empirical consumption. The consumption of ammonia seems to be around 0,0010 - 0,0015 kg/kg waste.

4.2.5 Slags, ashes and dust

Slag and ash are formed by inorganic materials in the waste. The bottom ash or slag is the solid residue from the grate in the combustion chamber. The fly ash is particles that have been transported by the flue gases through and is separated in boiler and flue gas cleaning equipment. Dust is the portion of flyash that is not collected, but emitted to the environment by the flue gas outlet. The slag and the fly ash quantities have been derived from RVF (1997), see Table 4.1. The dust emissions are discussed above in Section 4.2.1. Landfilling of slag and ashes are further presented in Section 4.3.

4.2.6 Emissions to water

In wet flue gas cleaning systems the absorption and cooling water is purified and neutralised. The major part of the water is recirculated in the process, but excess water has to be bled of. The impurities separated from the water is collected as a sludge and landfilled, usually the sludge is mixed with fly-ash. The amounts of heavy metals that are discharged to the water outlet are normally negligible.Water emissions of heavy metals are presented in Table 4.3.a.

4.2.7 Tables

Table 4.1. Data about quantities and emissions from MSW incineration in Sweden (RVF, 1997)

Parameter	Amount (1996)	*Unit*
Number of plants	22	
Input and residues		
Quantity of waste incinerated	1 853 214	*ton/year*
Quantity of slag	336 025	“
Quantity of fly ash and flue-gas-cleaning-residues	70 374	“
Recovered energy		
Heat (district heating)	4 802 722	*MWh/year*
Electricity (three plants)	433 781	“
Emissions to air		
Dust	33	*ton/year*
HCl	412	“
SO_2	1121	“
NO_X	1463	“
Dioxins	2	*g/year*
Hg	77	*kg/year*
Pb	214	“
Cd	8	“

Table 4.2 Emission factors and energy production for air emission from incineration, average Sweden if other not indicated.

Emission	*Calculation base*	Emission factor *kg/calculation base*	Remarks
Product related emissions			
CO_2, total carbon dioxide	*1 kg C_{tot}*	44/12*0,967 = **3,55**	96,7 % oxidisation is assumed, rest of carbon occurs in the flyash and slag
CO_2-antropogeneous carbon dioxide	*1 kg C_{tot}-antropogeneous*	**0 / 3,55**	From fossil fuels and products from fossil raw materials, e.g. plastic, rubber, synthetic textiles, etc.)
SO_2, sulphur dioxide, with separate SO_2-removal stage	*1 kg S*	0,8*0,05*64/32 = **0,08**	80% of S in waste is transferred to raw gas, from which 95% is separated in flue gas cleaning
- " - , without separate SO_2-removal stage	*1 kg S*	0,8*0,5*64/32= **0,8**	80% of S in waste is transferred to raw gas, from which 50% is separated in flue gas cleaning
- " - , average Sweden	*1 kg S*	**0,20**	
HCl, hydrochloric acid, average Sweden	*1 kg Cl*	0,8*0,05*36,5/35,5 =**0,04**	80% of Cl in waste is transferred to raw gas, from which 95% is separated in the flue gas cleaning
Dust, ash allocation	*1 kg inorganic*	**$8{,}1 \cdot 10^{-5}$**	
weight allocation	*1 kg*	**$1{,}78 \cdot 10^{-5}$**	
Hg, Mercury	*1 kg Hg*	**0,133**	See comments in Section 4.2.1 about metals, and Table 4.3.
Pb, Lead	*1 kg Pb*	**$2 \cdot 10^{-3}$**	- " -
Cd, Cadmium	*1 kg Cd*	**$5 \cdot 10^{-3}$**	- " -
Cu, Copper	*1 kg Cu*	**$1 \cdot 10^{-3}$**	- " -
Zn, Zinc	*1 kg Zn*	**$2 \cdot 10^{-3}$**	- " -
Ni, Nickel	*1 kg Ni*	**$1 \cdot 10^{-3}$**	- " -
Cr, Chromium	*1 kg Cr*	**$2 \cdot 10^{-3}$**	- " -

Table 4.2 (cont). Emission factors and energy producion for air emissions from incineration, average Sweden

Emission	*Calculation base*	Emission factor *kg/calculation base*	Remarks
b) Process related emissions			calculated as pseudo-product-bound
CO, carbon monoxide			
a. weight allocation	*a. 1 kg material*	**0,0015**	
b. carbon allocation	*b. 1 kg* C_{tot}	**0,0052**	
PAH, polyaromatic hydrocarbons			
a. weight allocation	*a. 1 kg material*	**$1{,}00 \cdot 10^{-8}$**	
b. carbon allocation	*b. 1 kg* C_{tot}	**$3{,}41 \cdot 10^{-8}$**	
Chlorinated dioxins (2,3,7,8-TCDD-equivalents)			
a. weight allocation	*a. 1 kg material*	**$1{,}0 \cdot 10^{-12}$**	
b. carbon allocation	*b. 1 kg* C_{tot}	**$3{,}4 \cdot 10^{-12}$**	
c. chlorine allocation	*c. 1 kg Cl*	**$2{,}2 \cdot 10^{-12}$**	
d. carbon + chlorine allocation.	*d. kg* C_{tot} *and kg Cl*	C: **$1{,}7 \cdot 10^{-12}$** Cl: **$1 \cdot 10^{-12}$**	
e. heating value allocation	*d. 1 MJ heating value*	**$9{,}1 \cdot 10^{-12}$**	Lower heating value
NO_x nitrogen oxides			
a. weight allocation	*a. 1 kg tot*	**0,00079**	
b. N_{fuel} allocation	*b. 1 kg N (in waste)*	**0,118**	
c. N_{fuel} + heating value all.	*kg N +MJ heating value*	N: **0,107** kg/kg E:**$3{,}6 \cdot 10^{-6}$**kg/MJ	95 % fuel NO_X and 5 % thermal NO_X are assumed in normal case
d. C_{tot} allocation	*kg* C_{tot}	**0,00269**	
Formation of slag and ash			
Fly ash, ash allocation	*kg ash (inorganics)*	**0,17**	
Slag, ash allocation	*kg ash (inorganics*	**0,83**	
Consumption			
$CaCO_3$ for neutralisation	*kg CaCO3/kg*	Cl: **1,4** S: **1,5**	

Table 4.2 (cont). Emission factors and energy production for air emissions from incineration, average Sweden

Production of energy		
a. Production of only heat, with flue gas condensation		
Primary heat recovery		**85% of lower heating value**
Heat recovery by condensation		**1,6 MJ/kg water in waste**
b. Production of both electricity and heat: (ideal plant)		
Primary heat	*MJ/kg (lower heating value)*	**57% of lower heating value**
Electricity	*MJ/kg (lower heating value)*	**28% of lower heating value**
Heat recovery by condensation		**1,6 MJ/kg water in waste**
c. Production of both electricity and heat: average Sweden		
Primary heat	*MJ/kg (lower heating value)*	**78% of lower heating value**
Electricity	*MJ/kg (lower heating value)*	**7% of lower heating value**
Heat recovery by condensation		**1,6 MJ/kg water in waste**

Table 4.3. a-d. Heavy metal balance from some incionerators
Table 4.3.a. Uppsala incinerator plant 1992 (Sundqvist 1994)

	Quantity in waste *kg/year*	Emission flue gas *% of input*	Emission water *% of input*	Landfill slag *% of input*	Landfill fly ash *% of input*	Landfill sludge *% of input*	Landfill total *% of input*
Cd	952	0,5	~0	42,3	56,3	0,9	**99,5**
Co	537	~0	~0	91,1	8,4	0,5	**99,5**
Cr	4232	0,2	0,1	87,6	11,6	0,5	**99,7**
Hg	412	13,3	0,7	4,6	4,1	77,2	**85,6**
Ni	3293	0,1	0,1	96,5	2,8	0,6	**99,8**
Pb	77055	0,2	~0	83,4	16,2	0,3	**99,8**
Zn	135500	0,2	~0	64,2	35,1	0,5	**99,8**

Table 4.3.b. Uppsala incinerator 1994 (Mingarini 1996)

Element	Emission to air	to water	To landfill slag	flygash and fluegas-cleaning residue	total to landfill
	weight-%	*weight-%*	*weight-%*	*weight-%*	*weight-%*
Major elements					
C, carbon	96,4	0	2,5	0,97	3,47
N, nitrogen	98,1	0	1,9	0	1,9
S, sulphur	7,8	1,6	21,5	69,1	90,6
P, phosphorus	0	23,5	21,6	54,9	76,5
Cl, chlorine	0,9	53,4	11,0	34,7	45,7
K, potasium	3,0	29,3	55	12,7	67,7
Ca, calcium	4,5	26	60	9,5	69,5
Heavy metals					
Pb, lead	0,0001	0,003	66,9	33,1	99,9969
Cd, cadmium	0,7	0,02	17,3	82	99,28
Hg, mercury	0,65	0,25	1,4	97,7	99,1
Cu, coppar	0,0005	0,0005	96,7	3,3	99,999
Cr, chromium	0,03	0,02	77,8	22,2	99,95
Ni, nickel	0,025	0,030	94,4	5,55	99,945
Zn, zink	0,01	0,0006	58,7	41,3	99,99

Tabell 4.3.c. Uppsala incinerator plant 1995 (Björklund 1998)

Element	Emission		To landfill		
	to air	to water	slag	flyash and fluegas-cleaning residue	total to landfill
	weight-%	*weight-%*	*weight-%*	*weight-%*	*weight-%*
Major elements					
C, carbon	97	0	2,0	1,0	3
N, nitrogen	73,5	0	2	24,5	26,5
S, sulphur	4,7	2,8	7	85,5	92,5
P, phosphorus	0,5	2,9	90	6,6	96,6
Cl, chlorine	0,2	11,8	9,0	79	88,0
K, potasium	0,3	48,7	41	10	51
Ca, calcium	0,2	26	60	13,8	73,8
Heavy metals					
Pb, lead	0,0004	0,0006	95	5	99,999
Cd, cadmium	0,003	0,08	16	83,9	99,9
Hg, mercury	3,3	0,1	1	95,6	96,6
Cu, coppar	0,001	0,004	95	5	99,995
Cr, chromium	0,05	0,02	76	24	99,93
Ni, nickel	0,1	0,06	92	7,8	99,84
Zn, zink	0,003	0,01	49	50,1	99,983

Table 4.3.d. Stockholm 1995 (Björklund, 1998)

Element	Emission to air	to water	To landfill slag	flygash and fluegas-cleaning residue	total to landfill
	weight-%	*weight-%*	*weight-%*	*weight-%*	*weight-%*
Major elements					
C, carbon	97,3	-	1,9	0,8	2,7
N, nitrogen	98,3	-	1,7	0	1,7
S, sulphur	19,9	-	20,5	59,6	80,1
P, phosphorus	0	-	21,1	78,9	100
Cl, chlorine	0,2	-	10,7	89,1	99,8
K, potasium	24,5	-	53,6	21,9	75,5
Ca, calcium	0,04	-	58,3	41,7	99,96
Heavy metals					
Pb, lead	0,002	-	82	18	99,998
Cd, cadmium	0,06	-	13,3	86,6	99,94
Hg, mercury	4,8	-	3,5	91,7	95,2
Cu, coppar	0,006	-	93,5	6,5	99,994
Cr, chromium	1,0	-	71,5	27,5	99,0
Ni, nickel	0,15	-	2,3	97,5	99,85
Zn, zink	0,01	-	45	55	99,99

Table 4.4. Volatilisation (boiling) temperature for some metal and metal compounds, controlling the fate of the metal during combustion

Metal	Melting point for pure metal	Volatilisation (boiling) temperature, °C				Remarks
		Metal	Oxide	Chloride	Sulphate	
Al, aluminium**)	669	2057	3500	183		non-volatile
Ca, calcium	842	1240	2850	>1600	anhydrite: 1193 gypsum: 163	non-volatile
Cd, cadmium	320	767	d.900-1000	960		semi-volatile
Cr, chromium**)	1890	2480		1300		non-volatile
Cu, copper*)	1083	2336	d.	d.993	d.650	non-volatile
Fe, iron*)	1535	3000		subl.		non-volatile
Fe, iron**)	1535	3000		315		non-volatile
Hg, mercury*)	-39	356	d.500	302	d.	volatile
K, potassium	62	760		subl.1500		semi-volatile
Na, Sodium	98	880	1413	subl.1275		semivolatile
Ni, nickel	1455	2900		973		non-volatile
Pb, lead*)	327	1620		950		non-volatile
Pb, lead**)	327	1620	d.		d	”
Zn, zinc	419	907	>1800	732	d.	semi-volatile

Remarks:

*)	Refers to metal compounds of II-valence
**)	Refers to metal compounds of III-valence
d.	Decomposes when heated
subl.	Sublimes at heating
non-volatile:	more than 80% of the metal is expected to be discharged in the slag
semi-volatile.	20 - 80 % of the metal is expected to be discharged in the slag
volatile:	less than 20 % of the metal is expected to be discharged in the slag

Table 4.5 Example of material composition and emission for some specific materials

Parameter	*Unit*	**Emission factor**	Raw MSW	Paper	Wood	PVC plastic	Polyethene plastic	Plastic mixture	Textiles	Rubber, leather	Organic degradable material	Metals	Glass	Other inorganics
Material data														
Input quantity to incinerator (normal composition)	*kg/kg*		1	0,29	0,01	0,005	0,07	0,02	0,025	0,03	0,45	0,03	0,03	0,04
Lower heat of combustion	*MJ/kg*		10,9	14	14	20	42	35	16	20	4,5	0	0	0
Upper heat of combustion	*MJ/kg*		12,8	15,4	15,6	21,4	44,4	37,8	17,3	21,6	6,9	0	0	0
C content	*kg C/kg*		0,29	0,33	0,30	0,38	0,67	0,75	0,36	0,4	0,25	0	0	0
H content	*kgH/kg*		0,049	0,054	0,051	0,064	0,11	0,125	0,06	0,067	0,042	0	0	0
S content	*kg S/kg*		0,0026	0,00075	0,00075	0	0	0,005	0,01	0,05	0,0012	0	0	0
N content	*kg N/kg*		0,007	0,0023	0,0015	0	0	0,001	0,036	0,006	0,012	0	0	0
Cl content	*kg Cl/kg*		0,005	0,0005	0,001	0,4	0	0,04	0,004	0,01	0,0037	0	0	0
Ash content (inorganics)	*kg ash/kg*		0,217	0,12	0,15	0,12	0,03	0,1	0,1	0,2	0,15	1	1	1
Moisture content	*kg H20/kg*		0,30	0,10	0,20	0	0	0	0	0,05	0,60	0	0	0
Emissions to air														
CO2														
Tot CO2	*kg CO2 /kg*	**3,55**	1,04	1,15	1,08	1,36	2,36	2,66	1,28	1,42	0,88	0	0	0
Antropogeneous	*kg CO2 /kg*	**3,55**	1,04	0,339	0,103	0,040	0,0263	0,020	0,0071	0,0028	0,00071	0	0	0
SO2, with separate SO2-absorption	*kg SO2/ kg*	**0,08**	0,000209	0,00006	0,00006	0	0	0,0004	0,0008	0,004	0,00010	0	0	0
" , without separate SO2-absorption	*kg SO2/ kg*	**0,8**	0,00209	0,0006	0,0006	0	0	0,004	0,008	0,04	0,00010	0	0	0
average Sweden	*kg SO2/ kg*	**0,202**	0,00053	0,00015	0,00015	0	0	0,0010	0,0020	0,010	0,00024	0	0	0
HCl	*kg HCl/ kg*	**0,04**	0,00020	0,00002	0,00004	0,016	0	0,0016	0,00016	0,0004	0,00015	0	0	0
Dust														
Ash allocation	*kg dust/ kg*	**$8,13 \cdot 10^{-5}$**	$1,8 \cdot 10^{-5}$	$9,8 \cdot 10^{-6}$	$1,2 \cdot 10^{-5}$	$9,8 \cdot 10^{-6}$	$2,4 \cdot 10^{-6}$	$8,1 \cdot 10^{-6}$	$8,1 \cdot 10^{-6}$	$1,6 \cdot 10^{-5}$	$1,2 \cdot 10^{-5}$	$8,1 \cdot 10^{-5}$	$8,1 \cdot 10^{-5}$	$8,1 \cdot 10^{-5}$
Weight allocation	*kg dust/ kg*	**$1,78 \cdot 10^{-5}$**	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$	$1,8 \cdot 10^{-5}$
CO														
Weight allocation	*kg CO/ kg*	**0,00153**	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015	0,0015
C allocation	*kg CO/ kg*	**0,0052**	0,0015	0,0017	0,00158	0,00120	0,0035	0,0039	0,0019	0,0021	0,0013	0	0	0
PAH														
Weight allocation	*kg PAH/kg*	**$1,00 \cdot 10^{-8}$**	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$
C allocation	*kg PAH/kg*	**$3,41 \cdot 10^{-8}$**	$1,0 \cdot 10^{-8}$	$1,1 \cdot 10^{-8}$	$1,0 \cdot 10^{-8}$	$1,3 \cdot 10^{-8}$	$2,3 \cdot 10^{-8}$	$2,6 \cdot 10^{-8}$	$1,2 \cdot 10^{-8}$	$1,4 \cdot 10^{-8}$	$8,5 \cdot 10^{-9}$	0	0	0

Parameter	*Unit*	**Emission factor**	Raw MSW	Paper	Wood	PVC plastic	Polyethene plastic	Plastic mixture	Textiles	Rubber, leather	Organic degradable material	Metals	Glass	Other inorganics
Dioxins														
Weight allocation	*kg TCDD/kg*	**$1,00 \cdot 10^{-12}$**	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$
C allocation	*kg TCDD/kg*	**3,41** $\cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,1 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,3 \cdot 10^{-12}$	$2,3 \cdot 10^{-12}$	$2,6 \cdot 10^{-12}$	$1,2 \cdot 10^{-12}$	$1,4 \cdot 10^{-12}$	$8,5 \cdot 10^{-12}$	0	0	0
Cl allocation	*kg TCDD/kg*	**2,00** $\cdot 10^{-10}$	$1,0 \cdot 10^{-12}$	$1,0 \cdot 10^{-13}$	$2,0 \cdot 10^{-13}$	$8,0 \cdot 10^{-11}$	0	$8,0 \cdot 10^{-12}$	$8,00 \cdot 10^{-13}$	$2,0 \cdot 10^{-12}$	$7,4 \cdot 10^{-13}$	0	0	0
C + Cl allocation	*kg TCDD/kg*	**C: $1,70 \cdot 10^{-12}$** **Cl: $1,00 \cdot 10^{-10}$**	$1,0 \cdot 10^{-12}$	$6,0 \cdot 10^{-13}$	$6,2 \cdot 10^{-13}$	$4,1 \cdot 10^{-11}$	$1,1 \cdot 10^{-12}$	$5,3 \cdot 10^{-12}$	$1,0 \cdot 10^{-12}$	$1,7 \cdot 10^{-12}$	$7,9 \cdot 10^{-13}$	0	0	0
Heating value allocation	*kg TCDD/kg*	**$9,13 \cdot 10^{-14}$**	$1,0 \cdot 10^{-12}$	$1,3 \cdot 10^{-12}$	$1,3 \cdot 10^{-12}$	$1,8 \cdot 10^{-12}$	$3,8 \cdot 10^{-12}$	$3,2 \cdot 10^{-12}$	$1,5 \cdot 10^{-12}$	$1,8 \cdot 10^{-12}$	$4,1 \cdot 10^{-13}$	0	0	0
NO_X														
N (fuel) allocation	*kg NOx/ kg*	**0,113**	0,00079	0,00025	0,00017	0	0	0,00011	0,0042	0,00068	0,0014	0	0	0
Heat allocation:	*kg NOx/ kg*	**$7,22 \cdot 10^{-5}$**	0,00079	0,001	0,001	0,0014	0,003	0,0025	0,0012	0,0014	0,00033	0	0	0
Ctot allocation	*kg NOx/ kg*	**0,0026922**	0,00079	0,00087	0,00082	0,001	0,0018	0,002	0,00097	0,001	0,00067	0	0	0
Weight allocation	*kg NOx/ kg*	**0,000791**	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079	0,00079
Formation of slag and ash														
Fly ash, ash allocation	*kg fly ash/ kg*	**0,17**	0,037	0,020	0,025	0,020	0,0051	0,017	0,017	0,034	0,026	0,17	0,17	0,17
Slag, ash allocation	*kg slag/ kg*	**0,83**	0,18	0,010	0,12	0,010	0,025	0,083	0,083	0,17	0,12	0,83	0,83	0,83
Consumption														
CaCO3 for neutralisation	*kg CaCO3/ kg*	**Cl: 2,82** **S: 3,12**	0,022	0,0038	0,0052	1,1	0	0,13	0,042	0,18	0,014	0	0	0
Production of energy														
a. Production of heat,														
boiler	*MJ/kg*	**85%**	9,3	11,9	11,9	17	35,7	29,8	13,6	17	3,8	0	0	0
condensation	*MJ/kg*	**1,60**	0,5	0,2	0,3	0	0	0	0	0,1	1,0	0	0	0
total	*MJ/kg*		9,8	12,1	12,2	17	35,7	29,8	13,6	17,1	4,8	0	0	0
b. Production of both electricity and heat: (ideal plant)														
heat (thermal energy)	*MJ/kg*	**57%**	6,3	8,0	8,0	11,4	23,9	20,0	9,1	11,4	2,6	0	0	0
electricity	*MJ/kg*	**28%**	3,1	3,9	3,9	5,6	11,8	9,8	4,5	5,6	1,3	0	0	0
condensation		**1,60**	0,5	0,2	0,3	0	0	0	0	0,1	1,0	0	0	0
total			9,8	12,1	12,2	17	35,7	29,8	13,6	17,1	4,8	0	0	0
c. Production of both electricity and heat: average Sweden														
heat (thermal energy)	*MJ/kg*	**78%**	8,6	10,9	10,9	15,6	32,76	27,3	12,5	15,6	3,5	0	0	0
electricity	*MJ/kg*	**7%**	0,8	1,0	1,0	1,4	2,94	2,4	1,5	1,4	0,3	0	0	0
total		**1,60**	0,5	0,2	0,3	0	0	0	0	0,1	1,0	0	0	0
			9,8	12,1	12,2	17,0	35,7	29,8	13,6	17,1	4,8	0	0	0

4.2.8 Discussion - permission controlled or operation-controlled emissions

In Section 3.2.3 the so called *permission controlled emissions* were introduced. A suitable synonym would be *operation controlled emissions*. They were introduced as a supplement to product related and process related emission, to take into consideration that several emissions can be controlled by operational control, by technical improvements, etc. The permitted emission often will set the framework for the operation and the technology used. E.g. an increased concentration of chlorine in the waste, or decreased permitted emission level, can be balanced by an increased water flow in the wet scrubber and thus keeping the HCl emission at the same level (kg/h, or mg/nm^3).

The permission-controlled emissions are often relatively easy to handle in waste incineration. Mostly the permitted emission is expressed in mg/nm^3 at CO_2 10 vol-%. This is actually the same as relating the emission to the carbon content of the waste. 1 nm^3 dry flue gas at 10 % CO_2 corresponds to 0,05355 kg elementary carbon input. Using the permission-controlled method is thus equivalent to use an allocation to the carbon content.

EU is just now discussing a new directive for incineration of waste. The directive will be adaptable on all wastes listed in the European Waste Catalogue (EWC) An amendment proposal for a Directive was introduced by the European Commission in July 1999. A preliminary proposal has been presented for discussion during the autumn 1997. The preliminary proposed permitted emissions are according to Table 4.5.

Table 4.6 Proposed air emission limits for waste incineration according to preliminary proposed EU directive (europe environment, 1999).

	Emission limit
NO_x (as NO_2), large plants (>3 ton/h)*	200 mg/m^3
-’’-, small plant (<3 ton/h)*	400 mg/m^3
Dust*	10 mg/m^3
HCl *	10 mg/m^3
HF*	1 mg/m^3
SO_2*	50 mg/m3
Organic carbon*	10 mg/m^3
Dioxins (TCDD-equivalents)**	0,1 ng/m^3
Cd and Tl***	0,05 mg/m^3
Hg***	0,05 mg/m^3
Sb+As+Pb+Cr+Co+Cu+Mn+Ni+V+Sn***	0,5 mg/m^3

* Daily average value

** Measured over a sample period of minimum 6 hours and amximum of 8 hours

*** Averages measured over the sample period of a minimum 30 minutes and a maximum of 8 hours

The use of permission controlled emissions are useful especially when studying larger changes. For the usual LCA of a product (or of a specific waste component) the product-related and process-related approach is reliable. However, when studying larger changes, the product-related and process-related approach can give overestimates of the emissions, since this approach is based on linear emission factors, and neglects possibilities to control the emissions by operation. Then the permission-controlled approach can be used as a check to avoid that kind of overestimates.

4.2.9 Discussion - other allocation models

The Dutch allocation model is presented in an appendix in our second report (Sundqvist et al, 1997)(the Appendix is a TNO-Report). The major characteristics of the Dutch model are:

- the product-related (called product-specific in the TNO-report) emissions are allocated on the basis of elemental composition.
- the process-related (called process-specific in the TNO-report) emissions are allocated on the basis on the volume on flue gas volume generated.

The approach for product-specific emissions is the same as our approach. The Dutch model is based on Dutch emission data, which differs from the Swedish emission data, so the emission factors are different. In most cases the data is comparable and the differences of minor importance.

The Dutch approach for process-specific emissions is somewhat different from our approach. However, the flue gas volume generated is theoretically depending on mainly the C-, H-, and O- content of the waste:

- The stoichometric consumption of air oxygen is calculated from the elemental C, H, O composition, assuming that carbon dioxide and water are formed.
- The air excess in waste incineration is normally 80 - 120 % (the real oxygen input is thus 1,8 - 2,2 times the stoichometric consumption).
- The dry flue gas volume is calculated from nitrogen in input air plus oxygen excess plus and carbon dioxide formed. The formed water is not included (dry basis).

This approach for process-specific emissions does not differ significantly from our approach, where the carbon content (or as alternatives also the organic content, or the heating value) is the base for allocation. There is an approximate relation between the heating value and the C, H, and O content, see Equation 4.2. The C content is for most materials the most dominant energy carrier. In Table 4.7.a is illustrated the result from different allocation principles for carbon monoxide, the calculations are based on a hypothetical Swedish waste. In Table 4.7.b the emission factors for some product-related (product-specific) are compared.

As seen in the Tables, the differences between our emission factors and the Dutch emission factors are ”relatively” small. According to LCA practice the normal uncertainty is one magnitude for most emission, and differences in emissions less than one order of magnitude should not be regarded as significant (Lindfors et al, 1995). In that aspect there is no significant difference between our model and the Dutch model. The differences depend on uncertainties in waste composition (due to the variety of the composition), and in different ways to calculate the emission factors. Our data and the Dutch data differs by a factor less than $2^{\pm 1}$ for most emissions, with the exception of weight allocation. For some product-related emissions the difference is larger, for example cadmium (Cd). The Swedish emission factor (0,005 kg/kg) was based on analyses of Cd in flue gas, in flyash, in slag, in sludge and in wastewater (however analyses were not made at the same occasions). The Dutch emission factor (0,11 kg/kg) was based on an assumed Cd concentration in waste of 3 mg/kg and a measured emission to air corresponding to 0,34 mg Cd per kg MSW.

Table 4.7.a Comparisons with the Dutch model: different allocation methods for process-specific emissions(here exemplified by CO)

	Material data					**CO emission**			
	Ash content *% of dry substance*	Moisture *% of total*	Total carbon C_{tot} *kgC/kg of material*	Lower heat. value, H_{eff} *MJ/ kg of material*	Flue gas volume, V *nm^3/ kg of material*	Weight allocation *kg CO per kg of material*	C_{tot} allocation *kg CO per kg of material*	Heating value allocation *kg CO per kg of material*	Flue gas volume allocation (Dutch approach) *kg CO per kg of material*
Food wastes	18	60	0,18	6,2	1,3	0,0153	0,00098	0,00086	0,00067
PE-plastic	5	0	0,81	41,7	20,2	0,0153	0,0044	0,0058	0,010
PET-plastic	5	0	0,54	19,4	9,6	0,0153	0,0030	0,0027	0,0050
PVC-plastic	20	0	0,31	14,9	6,0	0,0153	0,0030	0,0021	0,0031
Wood raw	5	40	0,25	8,2	2,7	0,0153	0,0017	0,0011	0,0014
Paper	10	10	0,36	12,7	5,4	0,0153	0,0014	0,0018	0,0028
Total MSW waste	30	30	0,29	11,0	3,0	0,0153	0,0020	0,0015	0,0015
Emission factor						0,0153 *kgCO/kg material*	0,0055 *kgCO/kgC_{tot}*	0,00014 *kgCO/MJ*	0,00052 *kgCO/nm^3*

Table 4.7.b Comparisons with Dutch model - product-specific emissions: metals, HCl and SO_2 emissions to air

Emission	Emission factor, according to this report *kg/kg*	Emission factor, according to the Dutch model (as described in Sundqvist et al, 1997) *kg/kg*
HCl	0,04	0,01
SO_2	0,16 / 0,8	0,034
As		0,014
Cd	0,005	0,11
Cr	0,002	0,003
Cu	0,001	0,001
Hg	0,13	0,55
Ni	0,001	0,003
Pb	0,002	0,008
Sn		0,01
Zn	0,002	0,01

4.3 Landfilling of slags and ashes

This Section is mainly from our report from the 2nd stage (Sundqvist et al, 1997).

The slag and ash from the incinerator are assumed to be landfilled. The landfill is handled according to the landfill approach described above in Chapter 3. The emissions are integrated over two different time periods:

- the surveyable time which is the time until the landfill reaches a kind of pseudo-steady-state
- the hypothetical infinite time period which is until all landfilled material has been released to the environment.

The emissions are assumed to occur at the current time (t=0).

The critical time period has been discussed in our second report (Sundqvist et al, 1997), and will not be further discussed here. Other approaches of longer time aspects are discussed in Section 4.3.3.

4.3.1 General description of incineration ashes

Owing to the large variety of components in municipal waste, also the ashes obtained from incineration will be complex and will exhibit large variations in composition with time. The major constituents will be different inorganic oxides and salts, but with a certain content of residual carbon and organic matter. The inorganic compounds are a mixture of residuals from the original waste that have been set free during the incineration and have been captured in the combustion chamber or in the flue gas cleaning equipment. Due to the high temperatures during the incineration also the inorganic compounds undergo chemical reactions and new solid phases may be formed. Compared with ashes from burning of coal and bio-fuels, the municipal waste incineration ashes usually have a significantly higher content of chloride and sulphate that may form soluble salts. The presence of soluble salts is an important factor influencing the definition of the surveyable time period for incineration ashes.

The municipal waste incineration ashes may, apart from inorganic compounds, also contain organic matter, which are thermostable or are formed during the incineration process. Examples of compounds that may be formed in trace amount in the incineration ashes are PAH:s, such as benso-(a)-pyrene, and TCDD (tetra chlorinated dibensodioxins). The mobilities of many such compounds are limited by low solubility and strong sorption on solid particles. Nevertheless, owing to their toxic properties the emissions of such compounds should be included in an LCA study. Since our study has focused on the methodological difficulties, no such estimates have been presented.

In Table 4.1 above is given amount of ashes, slags and flue gas residues from Swedish incinerator plants. The Table 4.8 gives a comparison of the chemical contents of selected fly ashes from some waste incineration plants.

Table 4.8 Average composition of municipal waste incineration ashes for some Swedish plants. (SNV Report 4192 and RVF report 1993:2:2).

	Slag	**Flyash**	**Unit**
Sulphur, S	1943	21300	*mg/kg*
Halogens			
Cl	2357	61300	*mg/kg*
F	146	2130	*mg/kg*
Br	21	189	*mg/kg*
Major components			
Na	23212	37627	*mg/kg*
Mg	13179	18003	*mg/kg*
Al	72151	78882	*mg/kg*
Si	196373	134054	*mg/kg*
P	5489	6612	*mg/kg*
K	15891	40914	*mg/kg*
Ca	84652	135811	*mg/kg*
Ti	8222	16786	*mg/kg*
Mn	1139	2500	*mg/kg*
Fe	37160	17881	*mg/kg*
Ba	1382	1804	*mg/kg*
Toxic organic compounds			
Chloro-phenols	36	2058	*mg/kg*
Chloro-benzene	4.6	1258	*mg/kg*
TCDD-equivalents	0.1	6.1	*mg/kg*
Trace elements			
As	26	207	*mg/kg*
Cd	2	160	*mg/kg*
Co	23	26	*mg/kg*
Cr	284	1119	*mg/kg*
Cu	720	1031	*mg/kg*
Hg	0.05	13	*mg/kg*
Mo	12	34	*mg/kg*
Nb	10	23	*mg/kg*
Ni	46	89	*mg/kg*
Pb	639	4443	*mg/kg*
Rb	73	205	*mg/kg*
Sr	136	361	*mg/kg*
U	9	14	*mg/kg*
V	14	31	*mg/kg*
W	13	40	*mg/kg*
Zn	1549	20386	*mg/kg*

Table 4.9. Some compiled data on the composition of MSW incineration fly ashes.

		Slag from MSW, Sweden	Fly ash from MSW, Sweden	Fly ash, USA	Fly ash, Canada	Fly ash, Denmark
	Unit	(SNV Report 4192 and RVF Report 1993:2:2)	(SNV Report 4192 and RVF Report 1993:2:2)	(Taylor et al, 1982)	(Eighmy et al, 1995)	(Yan and Neretnieks, 1995)
Sulphur						
S	*mg/kg*		21300	0		14000
Halogens						
Cl	*mg/kg*	2357	61300		232000	0
F	*mg/kg*	146	2 130			
Br	*mg/kg*	21	189	70	2380	
I	*mg/kg*					18500
Major components						
Na	*mg/kg*	23212	37 600		84000	26000
Mg	*mg/kg*	13179	18 000	4060	1100	18000
Al	*mg/kg*	72151	78 900	46400	20800	77500
Si	*mg/kg*	196373	134 000		38000	179000
P	*mg/kg*	5489	6 610	1770		8950
K	*mg/kg*	15891	40 900	11600	109000	37000
Ca	*mg/kg*	84652	136 000	21000	46300	109000
Ti	*mg/kg*	8222	16 800	2	6100	9400
Mn	*mg/kg*	1139	2 500	190	448	1050
Fe	*mg/kg*	37160	17 900	35000	1600	
Ba	*mg/kg*	1382	1 800	1000	2400	1000
Trace elements						
Ag	*mg/kg*				192	0
As	*mg/kg*	26	207	1250	960	60
Au	*mg/kg*				1	
Cd	*mg/kg*	2	160	3	1660	250
Ce	*mg/kg*			79	13	
Co	*mg/kg*	23	26	100	13	45
Cr	*mg/kg*	284	1 120	70	494	200
Cs	*mg/kg*			10	14	
Cu	*mg/kg*	720	1 030	100	2220	1095
Dy	*mg/kg*			11	10	
Eu	*mg/kg*			2	1	
Ga	*mg/kg*			80	0	
Hf	*mg/kg*			4	1	
Hg	*mg/kg*	0.05	13		10	2
La	*mg/kg*			41	5	
Mo	*mg/kg*	12	34	20	47	28
Nb	*mg/kg*	10	23			
Nd	*mg/kg*			35	25	
Ni	*mg/kg*	46	89	1000	70	115
Pb	*mg/kg*	639	4 440	500	27000	7600
Rb	*mg/kg*	73	205	82	206	
Sb	*mg/kg*			17	2073	
Sc	*mg/kg*			30	2	
Se	*mg/kg*			62	17	9
Sm	*mg/kg*				1	
Sn	*mg/kg*				5900	
Sr	*mg/kg*	136	361	1090		250
Ta	*mg/kg*			3	1	
Tb	*mg/kg*				1	

Th	*mg/kg*			19		
U	*mg/kg*	9	14	9	3	
V	*mg/kg*	14	31	800	35	42
W	*mg/kg*	13	40			
Zn	*mg/kg*	1549	20400	560	104400	23500
Zr	*mg/kg*	86	176	250	600	
Toxic organic compounds						
Chlorophenols	*µg/kg*	36	2 060			
Chlorobenzene	*µg/kg*	4.6	1 260			
TCCD-equivalents	*µg/kg*	0.1	6			

4.3.2 Calculations for municipal waste incineration fly ashes

Emission factors have been calculated by a simplified theoretical chemical model, described in Sundqvist et al (1997). The calculations have been focused on fly ashes, which can be expected to give the major contribution to the release of heavy metals. The following chemical elements have been considered:

Major components: Na, K, Ca, Si, Cl, Al and Fe.

Trace elements: Pb, Zn, Cr, Cd and Cu.

The initial assembly of minerals that are expected to control the solubility of the heavy metals is given in Table 4.10. Table 4.11 show the chemical conditions that have been considered in the different parameter variations. The different conditions reflects both possible variations in the ash composition and in landfill internal environment. The different cases illustrated in Table 4.11 illustrates the uncertainty in the model. The conditions can vary depending on the landfill design (oxidiced resp. reduced conditions) and on the exact composition of the ash – e.g. pH-buffering capacity and redox buffering capacity. In a real landfill all conditions may be present at the same time in different parts.

Table 4.10. Initial mineral assembly assumed in the calculations, including also soluble components and potential secondary minerals that may be formed at later stages of the leaching.

Element	**Mineral phase**	**Remark**
Al	Gibbsite	Initial assembly (only considered in some variations)
	Magnesium-silicates	Secondary mineral
Ca	Calcite	Secondary mineral, Initial assembly in variations
Cl	-	Assumed soluble
	Pb_2OH_3Cl	Secondary mineral (only considered in some variations)
Fe	$FeCO_3$	Initial assembly
	$Fe(OH)_3$	Secondary mineral
K	-	Assumed soluble
Na	-	Assumed soluble
Si	Magnesium-silicates	Initial assembly (only considered in some variations)
	SiO_2	
Cd	$Cd(OH)_2$	Initial assembly (only considered in some variations)
	$CdCO_3$	Secondary mineral (only considered in some variations)
Cr	$Cr(OH)_3$	Initial assembly
	$PbCrO_4$	Secondary mineral
Cu	$Cu(OH)_2$	Initial assembly (only considered in some variations)
	Malachite	Secondary mineral (only considered in some variations)
Pb	PbO	Initial assembly
	Pb_2OH_3Cl	Secondary minerals (only considered in some variations)
	Cerrussite	- ” -
	Hydrocerrussite	- ” -
	$PbCrO_4$	- ” -
	$PbSiO_3$	
Zn	ZnO	Initial assembly
	$ZnCO_3$	Secondary mineral

Table 4.11. Chemical conditions considered in the different parameter variations for leaching of municipal solid waste incineration ashes.

Case number	**General characteristics**	**pH- and redox-control**	**Heavy metals considered**
1a	Oxidised ashes	Neutral pH, buffered by silicates, low content of redox sensitive components	Pb, Zn, Cr, Fe
1b	Oxidised ashes	Buffering by calcite, low content of redox sensitive components	Pb, Zn, Cr, Fe
1c	Oxidised ashes	Buffering by calcite low content of redox sensitive components	Pb, Zn, Cr, Fe, Cd, Cu
2a	Reduced ashes	Neutral pH, redox buffering by $FeCO_3$	Pb, Zn, Cr, Fe
2b	Reduced ashes	pH-buffering by calcite, redox buffering by $FeCO_3$	Pb, Zn, Cr, Fe
2c	Reduced ashes	pH-buffering by calcite, redox buffering by $FeCO_3$	Pb, Zn, Cr, Fe, Cd, Cu

A proposed new regulation for Swedish landfills gives limits for maximum percolation rate for different kind of landfills. Landfills for the most hazardous wastes should have a maximum percolation rate of less than 5 l/m^2/year, and for the intermediate hazardous wastes the percolation rate should be less than 50 l/m^2/year. We have simply applied the latter value. Thus, for a landfill height of 10 m and with an assumed porosity of the ashes of 50%, the pore water exchange rate corresponding to an annual percolation rate of 50 l/m^2/year, is once in 100 years. Consequently, a cut-off time at 100 years for the surveyable time period, would reflect a very early stage of the leachate represented by the first points in the plotted results. At this stage the chemical conditions in the landfilled waste is not likely to be near a pseudo steady-state. This brief estimate indicates that the time scales of interest will be very long, many thousands of years. If this is coherent with the intentions of an LCA methodology is still to be discussed and the decision must remain open for the time being.

By selecting a suitable period of time for the integration, the releases corresponding to the surveyable time period can be obtained. Figures 4.3 and 4.4 show an example of some plotted results for a hypothetical case (1c) where the concentrations, respective released fractions of different heavy metals are followed as a function of time.

The calculated emission factors for the surveyable time period are given in Table 4.12. In the same Table also the calculated time for complete leaching is given.

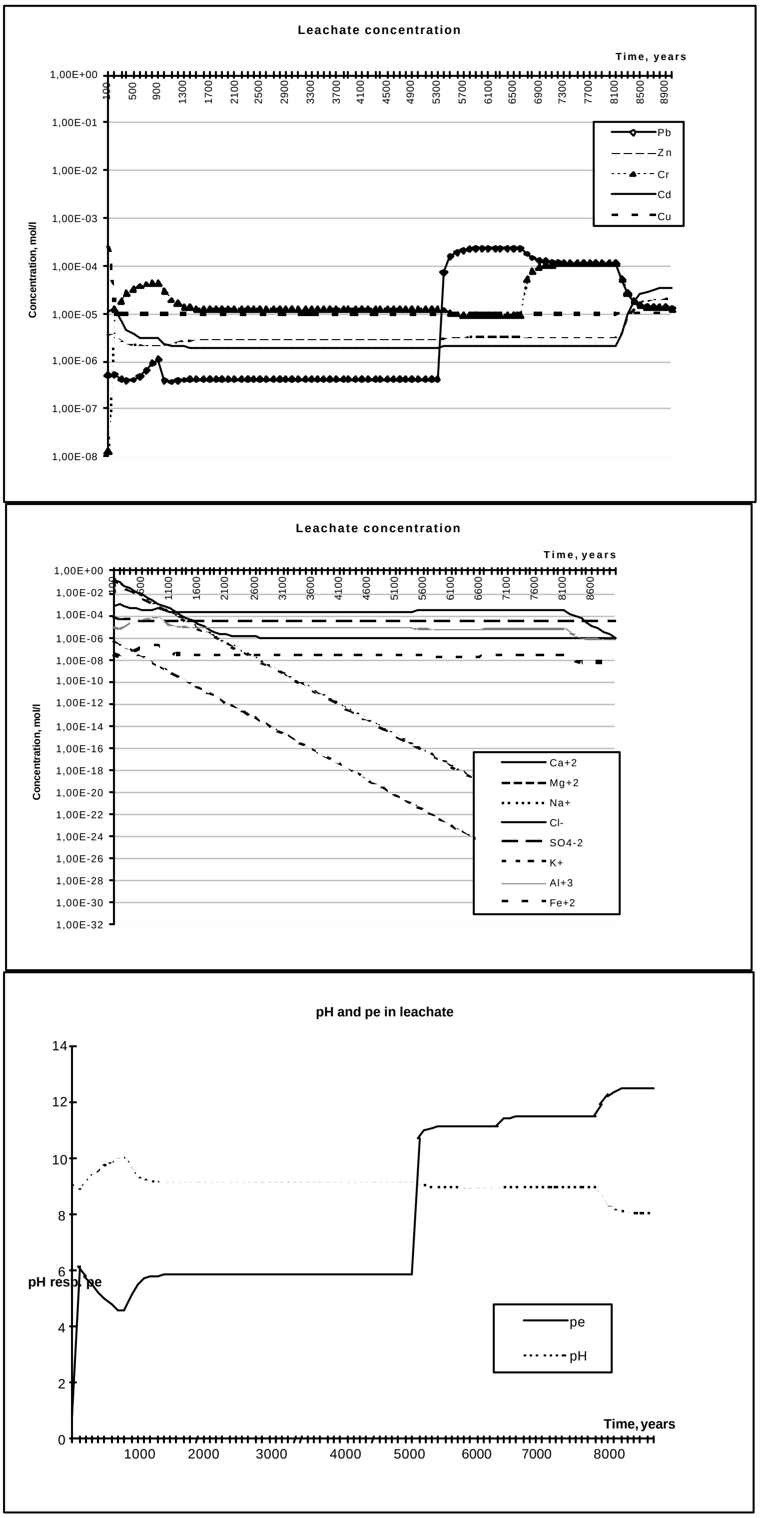

Figure 4.3 Concentrations in leachate from landfilled incinerator ashes. Case 1c: Oxidised ashes buffered bycalcite.

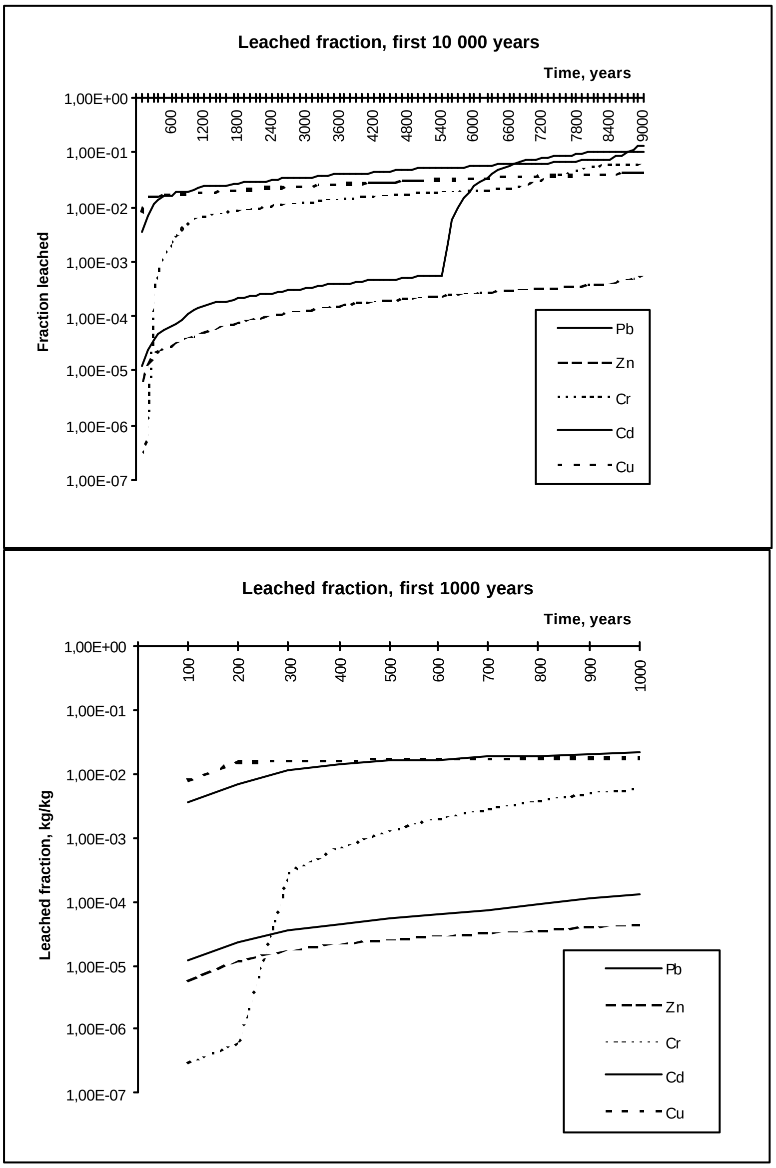

Figure 4.4 Leached fraction during 1000 resp 10 000 years. Case 1c: oxidised ashes, buffered by calcite

Table 4.12 Emission factors for landfilling of MSW incineration ashes..

Case	1. Oxidised ashes			2. Reduced ashes		
	1a pH 7, silicates	1b buffering by calcite	1c buffering by calcite	2a Neutral pH, redox buffering by $FeCO_3$	2b pH-buffering by calcite, redox buffering by $FeCO_3$	2c pH-buffering by calcite, redox buffering by $FeCO_3$
Emission factor, surveyable time period (200 years), ***kg emitted of metal per kg landfilled of metal***						
Cd	-	-	$6{,}3 \cdot 10^{-3}$	-	-	$1{,}1 \cdot 10^{-2}$
Cr	$1{,}8 \cdot 10^{-5}$	$4{,}3 \cdot 10^{-5}$	$6{,}6 \cdot 10^{-4}$	$6{,}3 \cdot 10^{-7}$	$2{,}1 \cdot 10^{-7}$	$9{,}6 \cdot 10^{-7}$
Cu	-	-	$4{,}3 \cdot 10^{-3}$	-	-	$1{,}4 \cdot 10^{-1}$
Pb	$3{,}3 \cdot 10^{-5}$	$1{,}1 \cdot 10^{-5}$	$2{,}1 \cdot 10^{-5}$	$3{,}4 \cdot 10^{-5}$	$1{,}2 \cdot 10^{-5}$	$4{,}3 \cdot 10^{-5}$
Zn	$2{,}6 \cdot 10^{-5}$	$6{,}4 \cdot 10^{-6}$	$1{,}0 \cdot 10^{-5}$	$2{,}9 \cdot 10^{-5}$	$9{,}0 \cdot 10^{-6}$	$3{,}9 \cdot 10^{-4}$
Estimate of time period required for complete leaching ***years***						
Cd	-	-	60 000	-	-	41 000
Cr	53 000	890 000	140 000	71 000	22 000 000	980 000
Cu	-	-	260 000	-	-	28 000
Pb	250 000	920 000	82 000	9 300 000	15 000 000	1 900 000
Zn	12 000 000	62 000 000	15 000 000	7 700 000	7 800 000	420 000

4.3.3 Bottom ashes

The above result was derived for fly ashes. The figures can be applied on bottom ashes as well. We have not done any calculations on bottom ashes (slag). However, leaching test on fly ash and bottom ash shows that the leaching of metals are of the same magnitude in fly ash and bottom ash (the emission factor is expressed in kg leaches metal per kg of metal in ash. Some elements have a slightly lower leaching factor, but still within the uncertainties. Considering the observed differences we suggest the following emission factors for bottom ashes.

Table 4.13 Estimated emission factors for bottom ash (slag)

Metal	**Bottom ash** Leaching during surveyable time period Oxidised ashes *kg/kg*	**Bottom ash** Leaching during hypothetical, infinite time *kg/kg*
Cd	$4 \cdot 10^{-3}$	1
Cr	$3 \cdot 10^{-4}$	1
Cu	$4 \cdot 10^{-2}$	1
Pb	$1{,}5 \cdot 10^{-5}$	1
Zn	$1 \cdot 10^{-5}$	1

4.3.4 Discussion of the time aspects

As noticed above, the surveyable time period was roughly set to about 200 years, corresponding to two water renewals in the landfill. According to the processes in the landfill there are several reactions occuring during the first 1000 years. A some kind of steady-state seems to be obtained first from about year 1000. This seems to be a rather long time for a surveyable period (by the definition the length of the surveyable period should be of the magnitude one century). The surveyable time period for the ash landfill is larger than the surveyable time period identified for MSW landfills, see Chapter 5. In a study where landfilling of raw MSW is compared with incineration of MSW, inclusive landfilling of ash and slag, this diversity can be offending, when different time scales (in real time) are used. However, as can be noticed in Chapter 5, the integrated emissions during a 50 - 100 year period should be of the same order of magnitude as the emissions during a 200 period, because there are no major emissions to expect immediately after the end of the methane stage.

The time aspects can also be discussed in the longer time perspective, see discussion in Section 3.2.2. In Figure 4.5 the long term emissions are illustrated for oxidised ashes buffered by calcite. The following conclusions can be drawn from the Figure:

- The time for complete leaching is less than 1 million years for Pb, Cr, Cd and Cu but 15 millions years for Zn. The difference between the infinite period and 1 million years is of minor importance in that perspective, except for Zn.
- By the beginning of next glacial period in Scandinavia about 10 - 50 % of landfilled Pb, Cr, Cd and Cu and 0,2 % of the Zn would have been released to the environment. Thus, only a minor part of the potential emissions has occurred at the start of the glacial period.
- The difference in emissions between 100 years and 200 years for the surveyable time period is about a factor 2. Thus, the emissions are more sensitive for the landfill performance as indicated by the differences between the different cases 1a, 1b, 1c, 2a, 2b, and 2c in Table 4.13.

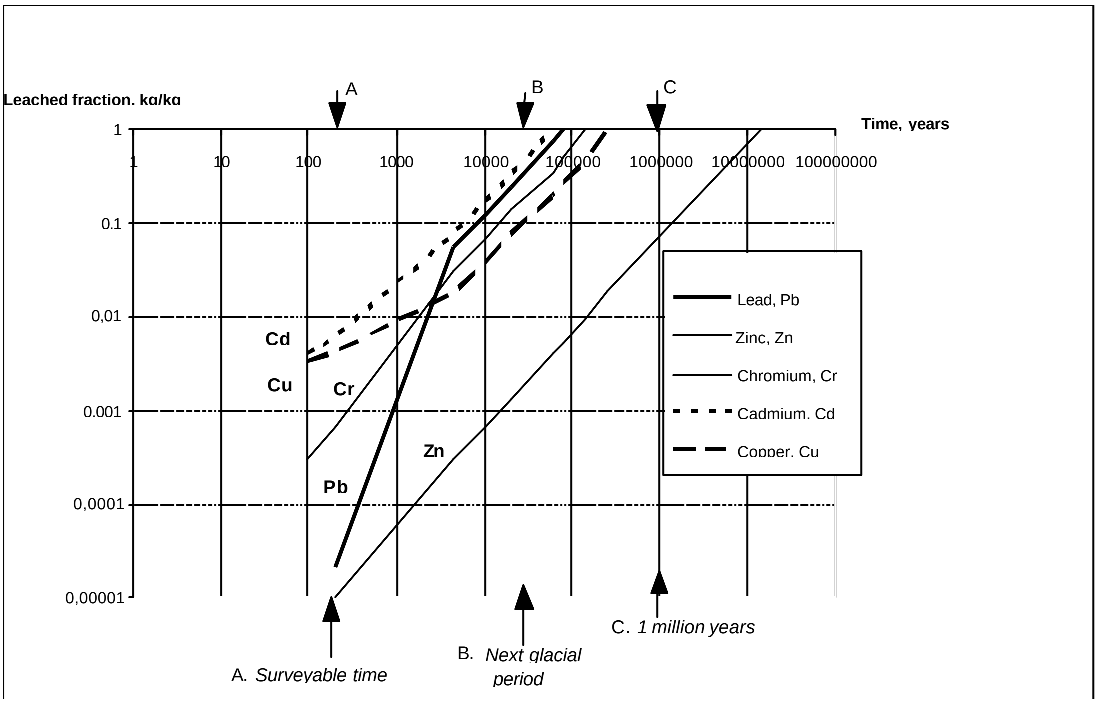

Figure 4.5 Illustration of time aspects. The figures are based on case 1c (Table 4.11) with oxidised ashes buffered by calcite.

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5. Conventional Landfilling of Municipal Solid Waste

5.1 General

5.1.1 Municipal Solid Waste

Here, Municipal Solid Waste (MSW) means the wastes that are disposed at municipal waste facilities, mostly incineration plants and landfills. The origin of the MSW is both domestic and industrial and business. MSW is characterised as a mixture of several materials: paper, plastic, wood, food and cooking wastes, garden wastes, glass, metals, ceramics, electronic scrap, etc. The composition is approximately according to Table 5.1.

Table 5.1. Composition of MSW in Sweden (Sundqvist, 1995)

Material	Weight-%
Paper (newsprint, packages, cardboard, etc.)	30 - 35
Plastics	5 - 10
Textile, rubber, leather	2 - 4
Kitchen and garden refuse	35 - 45
Glass	3 - 5
Metal	3 - 5
Others	5 - 8

As averages the moisture contents use to be 25 - 35 % and the ash content 20 - 25 %.

5.1.2 Approaches for landfilling

The time aspects in our landfill model for organic materials are based on the two time horizons, as described in Chapter 3:

- the **surveyable time** period, which is the time until some kind of pseudo-steady-state is obtained. The period is of the magnitude one century.
- the **hypothetical, infinite time** period, which is the period until the landfilled material is completely released to the environment.

All emissions are handled as product-related, see Chapter 3. The potential emissions are estimated from the elemental and chemical composition of the material. The model is formulated from both theoretical considerations and from results from laboratory studies and field studies. The model and data below have to a large extent been presented in earlier reports Sundqvist et al (1994), Finnveden at al (1996), Sundqvist (1995), Finnveden and Huppes (1995), Finnveden (1996), Sundqvist et al (1997).

5.2. *Characteristics of a MSW landfill*

5.2.1 Processes in the landfill

The behaviour of the landfill can be better understood if the degradation process is described by series of consecutive reactions. There are several reactions involved, but the most important is as follows, see also Figure 5.1.

1. First, larger organic molecules are split into simpler molecules. Hydrolysis is one example of reaction taken place. For example, hydrolysis of cellulose gives different sugars:
   $(C_6H_{10}O_5)_n$ + n H_2O --> n $C_6H_{12}O_6$

   cellulose *sugar*

2. The smaller molecules are then microbially degraded to intermediate compounds, such as fatty acids, ketones, aldehydes, alcohols. From the beginning some oxygen is present that give aerobic condition during a short period, but the major part of the degradation will occur during anaerobic conditions.
3. These formed intermediates are then further degraded during anaerobic conditions, first to acetate and hydrogen, which both will form methane and carbon dioxide.
4. When the organic material is degraded there is humus, lignin, plastics and other relatively stable material left, which may degrade very slowly. The degradation can occur in aerobic conditions if ambient air oxygen diffuses into the waste or if solved oxygen in the percolating rainwater is transferred to the waste. However, the anaerobic conditions will probably remain in the major part of the landfill for a very long time.

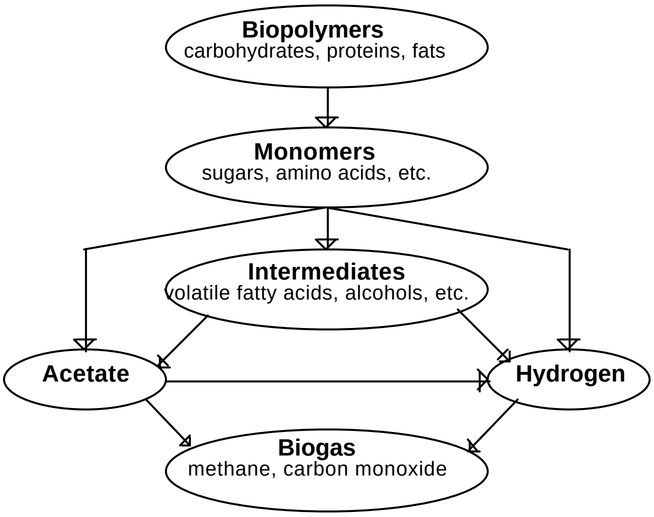

Figure 5.1. Conversion paths in biogas formation from organic

In the landfill several of these reactions occur parallel to each other. Studying the whole landfill, the emissions from the landfill will have different characteristics, depending on which reactions are dominating. Usually it is possible to identify several stages in the landfill behaviour. The stages that normally can be identified are (Öman, 1991; Sundqvist et al, 1994; Sundqvist et al, 1997):

1. Initial stage, until the processes begin. The length of the initial stage may be from a week up to one year.
2. Oxygen and nitrate oxidation stage: the oxygen and nitrate in the landfilled waste are oxidising organic material. Since there are limited amounts of free oxygen and nitrate this stage will be very short, often a few weeks. The main degradation products are water and carbon dioxide.
3. Acid anaerobic stage, characterised by a large formation of volatile fatty acids and acetic material. Some gas containing hydrogen and carbon dioxide is formed. The high content of fatty acids will decrease the pH, which can cause dissolution of metals that will give high levels of metals in the leachate. The length of the period is from one year up to 10 years.
4. Methane stage: methane and carbon dioxide are formed, mainly from the earlier formed fatty acids. Sulphur is transformed to sulphide that will bound metals. The pH is stable about neutral. The metal leachage is very low.
5. Humic stage. The relatively stable humic products are slowly degraded or mineralised. Ambient air oxygen will begin to diffuse into the landfill. Oxygen can also be transferred dissolved in the percolating rainwater. The oxygen inlet will cause an increase in the redox-potential, which can cause an oxidation of sulphides, and lead to an increased leachage of metals that have been bound as sulphides.

5.2.2 Identification of the surveyable time period and the hypothetical, infinite time period

For the municipal solid waste landfill, we have earlier (Finnveden 1992, Sundqvist et al 1994, Finnveden et al 1995, Sundqvist et al 1997) suggested that *the surveyable period should correspond to the period until the later part of the methane stage*, thus including the initial stage, the oxygen and nitrate reducing stage, the acid anaerobic stage and the larger part of the methane stage. These stages are characterised by a high internal activity, which is decreased at the end of the methane stage, when the external influence (e.g. oxygen diffusion) increases. At the end of the methane stage the landfill has reach some sort of pseudo-steady-state.

After the methane stage, atmospheric oxygen may be transported into the landfill by the rain water or by diffusion. The oxygen may cause an oxidation of the sulphides to which metals may be bound. The major part of the metals may then leach out during a certain period, occuring after the surveyable time period.

The hypothetical, infinite time period is defined as the time until the landfilled material is completely released to the environment. This means that all inorganic compounds in the landfill will be released by leachate, by gases or by erosion. The organic compounds, to a large part humus, will be totally degraded to simple compounds and released by leachate, gas or erosion.

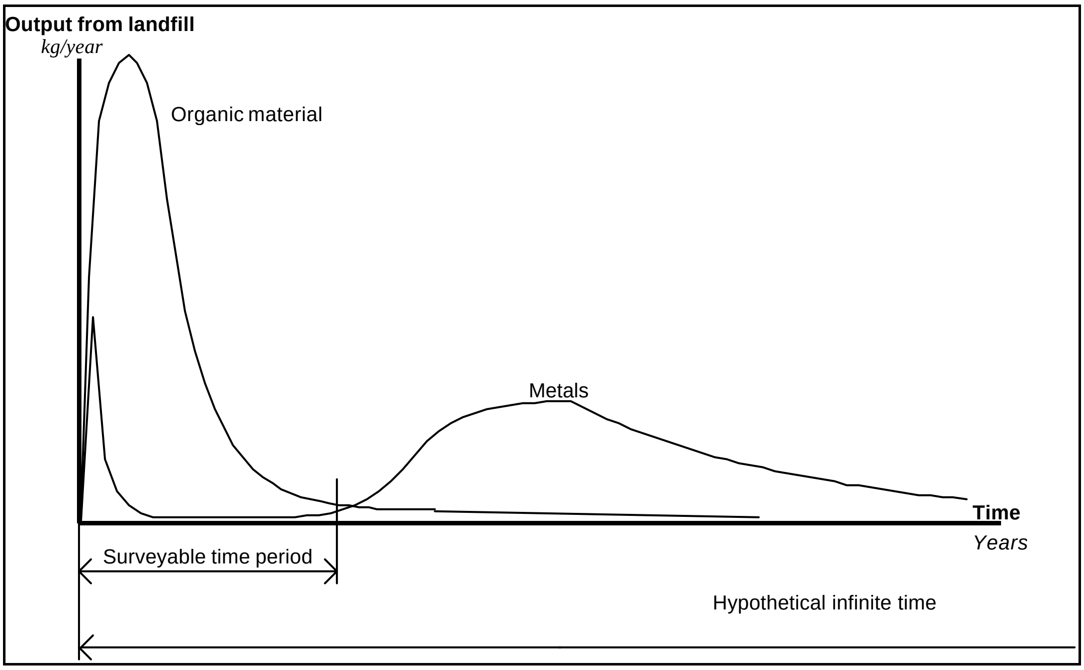

Figure 5.2 Schematic illustration on surveyable time, critical time and hypothetical infinite time periods.

Another aspect that we have taken into consideration is emissions from landfill fires. In Sundqvist et al (1997) we discussed emissions from landfill fires and found that they could be of major importance. The landfill fires are further discussed in Section 5.4.3 and Appendix A.

5.3 Emissions from landfills

5.3.1 Overview

The emissions from MSW landfills are:

- Emissions of landfill gas. The major emittant is methane gas, but different volatile constituents in the waste, as well as volatile degradation products may occur in the emitted gas. The gas is mainly a procuct from the degradation of organic material.
- Emission of leachate water, polluted by both organic compounds and metals. The leachate water is produced from excess precipitation water.

<u>*Carbon emissions to water*</u>

Mass-balances from field studies have indicated that approximately 1 weight-% of the degraded carbon will outflow via leachate mainly as fatty acids and similar, and 99 weight-% via the landfill gas as CH_4 and CO_2 (Baccini et al 1987). As a thumb rule 1 g of organic carbon (TOC) corresponds to 3 g COD. The ratio BOD/COD varies during the lifetime of the landfill. During the acid, anaerobic stage the ratio will be high, 0,1 - 0,8, and during the methane stage low, <0,05 (Öman, 1991). The average ratio

BOD/COD is assumed to be about 0,25 during the surveyable time period, see Sundqvist et al (1997).

Carbon emissions in landfill gas

The formed landfill gas contains mainly CH_4 and CO_2. Different organic materials give different ratios between CH_4 and CO_2, see further the discussion below and Table 5.3. Part of the gas can be recovered as fuel gas. The collection yields have often been very low. Collection efficiencies between 10 and 50 % have been reported (Suflita et al, 1994). There is research and development projects in e.g. Sweden and Great Britain with the aim to increase the recovery yield, see also Section 6.2 below.

The non-recovered gas will migrate through the soil cover, and methane-oxidising micro-organisms will oxidise a part of the methane to carbon dioxide. According to Gardner (1993) about 15 % of the methane may be oxidised. There are development projects where higher oxidation efficiency has been obtained. In the future it should be possible to obtain a higher degree of methane oxidation in the soil cover, see also Section 6.2 below.

The carbon balance in the landfill is shown in Figure 5.3.

Nitrogen, phosphorus and sulfur emissions

All nitrogen in the organic material is assumed to be transformed to ammonia or ammonium. The formed ammonia will be transferred to the leachate and emitted as NH_3/NH_4^+ in the leachate water. It is to expect some ammonia in the landfill gas. However, usually the analyses of ammonia in air are disturbed by interference from air nitrogen, so there is a lack of knowledge about ammonia in lanfdfill gas.

Almost all phosphorus is expected to remain in the landfill during the surveyable time periiod. The loss of phosphorus is estimated to be ca. 2 % to the leachate. The rest of the phosphorus will be emitted during the hypothetical, infinite time period.

All sulphur in the organic material is assumed to be transformed to hydrogen sulphide or metal sulphides. The hydrogen sulphide and metal sulphides will to a large extend be precipitated as solid metal sulphides and retained in the solid phase, thus immobilising metals.

Metal emissions

Metals in the waste will be dissolved by the percolating water and thus transferred to the leachate water. Several mechanisms, e.g. sulphide precipitation, will decrease the metal emission. The metal emission level can be relatively high during the acid anaerobic stage, but very low during the methane stage. Mercury is also supposed to evaporate to the ambient air from the landfill.

Metals in municipal solid waste can be divided into metallic materials and metal compounds. The latter may for example be present as additives in different materials. Before metals can be emitted via leachate they must be released from the solid waste matrix into the percolating water. In the case of metallic materials, the release process is corrosion. In case of additives the release can be governed by the decomposition of the solid matrix or by diffusion in and from the solid matrix. Once released to the water, the metals may be precipitated or sorbed to the solid phase. However, once released and dissolved in the leachate, the further fate is independent of the origin of the metal.

Emissions of specific organic compounds

Organic compounds are used in several products that occurs in the waste. Some compounds are used as solvents and similar. Other compounds are used as additives, e.g. in plastic, rubber or paper. These compounds may give rise to emissions in the leachate or in the landfill gas. Organic compounds are also formed during the degradation of organic material, especially volatile fatty acids and alcohols, ketones and aldehydes. Emissions of specific organic compounds from landfills are relatively poorly analysed. It has been suggested that the antropogenic specific organic compounds constitute the most significant group of leachate pollutants (Christensen et al, 1994).

The actual emissions of organic compounds from landfills depend on the fate of the compounds inside the landfill, due to processes that are specific for each compound. A conceptual model for prediction of emissions of organic compounds from landfill has been suggested by Öman (1995). This model has been verified in a pilot study (Öman and Wennberg, 1997; Öman et al, 1997). The conceptual model is shown in Figure 5.3. In the model the residence times of specific organic compounds inside landfills are primarily dependant on the sorption of the compounds to solid material and on the water transport. The residence times are then crucial for whether compounds will be transformed or evaporated during this time. The model predicts the compound's potential to be a) emitted with the leachate, b) emitted with the landfill gas, and c) mainly retained in the landfill, see Figure 5.3. The model is further described in Appendix B.

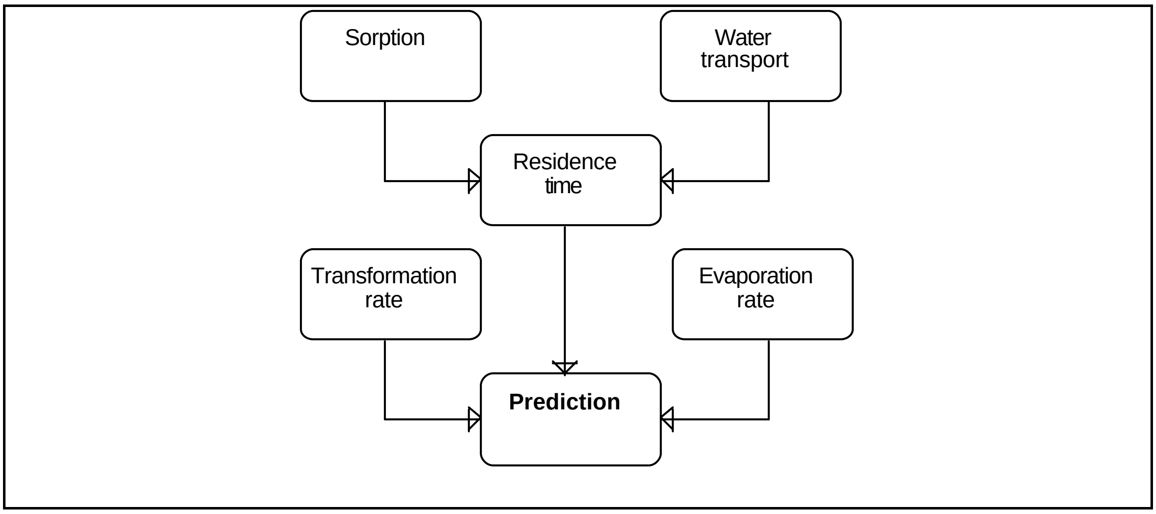

Figure 5.3. The conceptual fate model for specific organic compounds (Öman, 1995)

5.3.2 The landfill model - system boundaries

The landfill modelled in this study is a conventional municipal solid waste landfill, where mixed waste materials (exclusive hazardous waste) are deposited, see Figure 5.4. The landfill has a bottom sealing, e.g. plastic, rubber, bentonite or clay, and a top cover of soil. Besides waste, the inputs consist of soil for covering the waste, and energy (diesel oil) for the compactor. Another input is water from precipitation (rain and snow). The precipitated water will evaporate, run off the cover or percolate through the landfilled waste.

Leachate is collected and may be treated. The treatment is not included in the model described below. The landfill may be equipped with a gas extraction system.

The physical boundaries of the model are considered to be ~1 mm under the bottom sealing, ~1 mm above the soil cover, and the immediate outlet from the leachate collection pond.

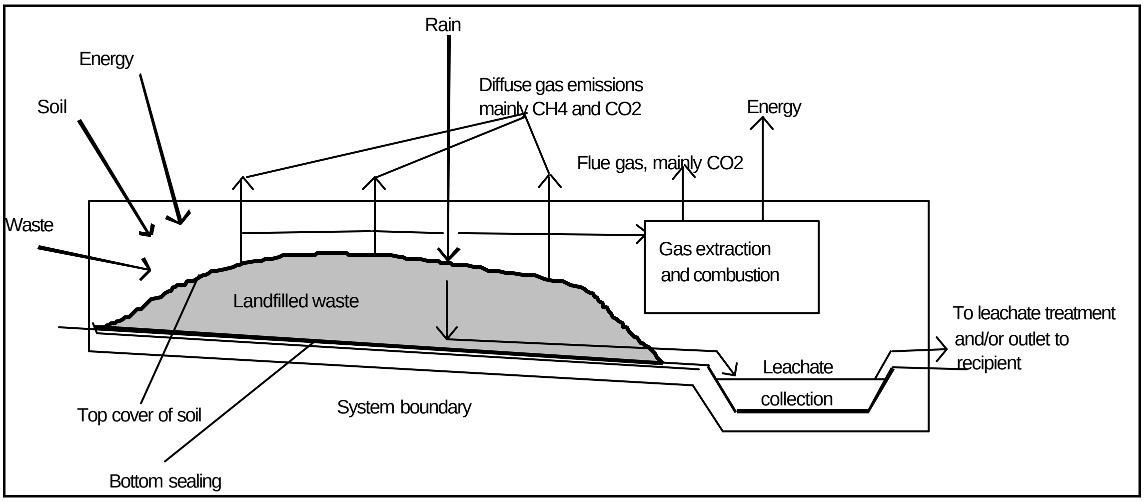

Figure 5 .4. System boundaries of the landfill model.

5.4 Emission factors

5.4.1 Resource consumption

Diesel for the compactors.

The waste is compacted on the landfill. Raw municipal solid waste may have a volume weight of about 150 - 200 kg/m^3. With several passages by the compactor the volume weight is increased to 700 - 800 kg/m^3 (Meijer, 1995). At the same time the waste pieces are torn down into smaller pieces, bags and sacks are torn and opened, etc. The energy consumption of the compactor is about 1 litre of diesel oil per ton (40 kJ/ton) (Persson 1995, Eggels and van der Ven 1995). Allocation of the diesel consumption can be discussed. There are at least two possibilities in accordance with the causality principle:

- Allocation by volume. Since one of the aims with compaction is to decrease the volume, it is practical to suggest allocation by volume. The volume base should be the volume of the studied product plus the void volume (the void volume between the waste items can be assumed to be 30 - 40 % of the volume). If 1 litre diesel oil is consumed per ton of waste, it will correspond to 0,23 l/m^3 (9,7 MJ/m^3) or 0,00023 l/dm^3 (0,0097 MJ/dm^3) of product. A dilemma with this allocation method is that two-dimensional pieces, e.g. a piece of plastic film or a sheet of aluminium foil, has no distinct volume. On the other hand two-dimensional pieces will not need any compaction, since they already have a dense volume.
- Allocation by weight. Another aim with compaction is to tear the waste materials to smaller pieces. This function is best satisfied with weight allocation since all materials actually will be affected by the compactor. The energy consumption for compacting is then 1 l/ton or 0,001 l/kg (40 MJ/ton respectively 0,04 MJ/kg).

Soil cover

The waste is frequently covered with soil. At many landfills a daily cover with soil is made, i.e. to make the waste more inaccessible for birds and rats and to decrease odours. Often some kind of inert wastes (concrete, excavating masses, etc.) or stabilised wastes (compost, sludge) are used for cover. After the landfill is finished, an extra cover of clay or other soils with low hydraulic gradient will be put on the landfill, i.e. to decrease the leachate production. Often the landfill is built in 2 m layers of waste (volume weight 600 kg/m^3), with a daily cover of 0,1 m depth (Meijer, 1995). This makes the daily need of cover material to about 0,08 m^3/ton waste. Most of this is usually inert or stabilised wastes and should not be accounted as resource depletion.

The final cover should be of 1 - 2 m depth. In an average landfill this will correspond to about 0,125 m^3 per ton landfilled waste (see Sundqvist et al 1997). This soil should be noted as a resource depletion in the LCA.

It can be discussed if the soil consumption should be allocated by mass or by volume. The final cover is made on compacted and partly degraded waste, and the original products have lost their identities when the landfill is finished and the cover is put on the landfilled wastes. In the compacted waste the density of different waste particles is more uniform than in raw waste. Therefore, allocation by weight is preferable, with a suggested soil consumption of 0,10 - 0,15 m^3 per ton of waste (or 0,0001 - 0,00015 m^3 soil per kg product under study).

5.4.2 Resource recovery

Energy from landfill gas

Landfill gas with a high methane concentration can be recovered from the landfill. In the Section 5.4.3 below is described how the theoretical available methane can be calculated, see also Table 5.3 where the theoretical gas yield for different compounds is given. The energy recovered can be calculated from the heating value for methane and the thermal efficiency of the combustion equipment (the upper heating value or calorimetric heating value for methane is 891,6 kJ/mole or 55,7 MJ/kg).

Leachate as fertiliser

Leachate has sometimes been used for irrigation of energy crops. Especially the nitrogen in the leachate is utilised.

5.4.3 Emissions

This basic emission model was described in Finnveden (1992), Sundqvist et al (1994), Finnveden et al (1995), and Sundqvist et al (1997). From the chemical formula of the organic material the following formula can be used for estimating the basic formation of potential emittants (all units are in molar units):

$$C_aH_bO_cN_dS_e(s) + (a - b/4 - c/2 + 3d/4 + e/2)\, H_2O \;\text{-->}$$

$$\text{-->}\; (0.99\, a/2 + b/8 - c/4 - 3d/8 - e/4)\, \mathbf{CH_4}(g) +$$

$$+ (0.99\, a/2 - b/8 + c/4 + 3d/8 + e/4)\, \mathbf{CO_2}(g) +$$

$$+ 0{,}01\, \mathbf{CH_2O}(aq, leachate) +$$

$$+ d\, \mathbf{NH_3}(aq, leachate) +$$

$$+ e\, [0.01\, \mathbf{H_2S}(g) + 0{,}05\, \mathbf{H_2S}(aq, leachate) + 0{,}94\, \mathbf{S^{--}}(s) + 2\, \mathbf{H^+}(aq)]$$

..........*(Equation 5.1)*

However, not all of the formed reactants will be emitted. The landfill gas emissions can be reduced by recovering the gas as a fuel. The methane emission is also reduced by methane oxidation in the soil cover. Leachate may be treated in different ways: adsorption, chemical precipitation, aeration, etc.

A calculation scheme is given in Figure 5.5. below.

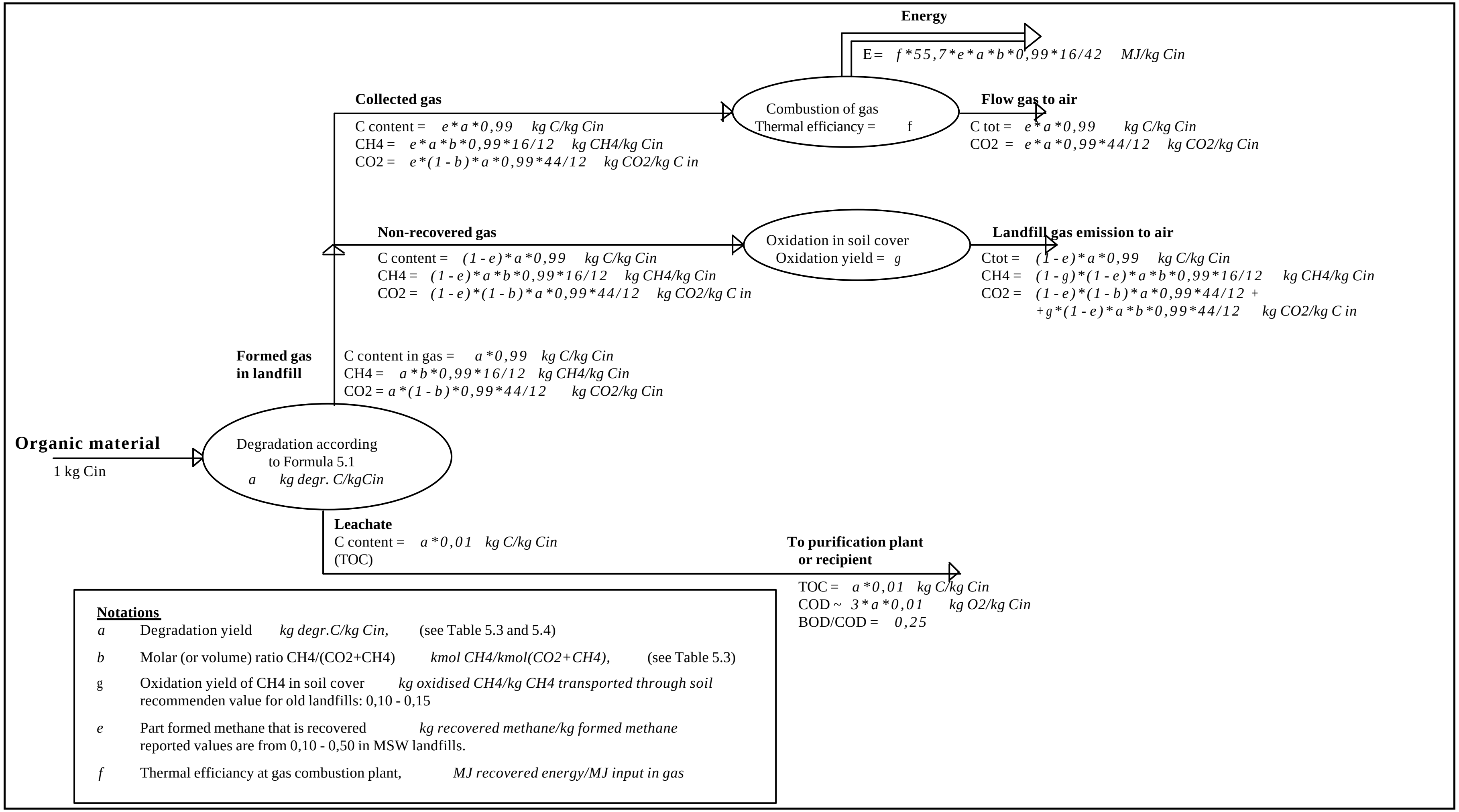

Figure 5.5. Carbon balance of a MSW landfill. The factors α, β, γ, ϕ, and ε are described in Table 5.3 and 5.4 and in the text.

Landfill gas

The main constituents of the landfill gas are methane and carbon dioxide. The ratio between methane and carbon dioxide depends on the material, and can be calculated by the Equation 5.1. In Table 5.3 is given the ratio $CH_4/(CH_4+CO_2)$ for some organic substances.

It is assumed that a part of the landfill gas is collected and recovered. In a conventional landfill, about 50 % of the formed landfill gas is assumed to be collected (if there is any collection system). In a biocell landfill the recovery rate is expected to be higher, see Chapter 6. The rest will migrate through the soil cover, where a part of the methane will be oxidised by methane-consuming micro-organisms. At a conventional landfill about 15 % will be oxidised, see Figure 5.5. In a modern biocell landfill a larger part should be oxidiced.

Table 5.3 Methane formation and concentrations (β) in landfill gas during the surveyable time periods. All figures are related to dry, organic (ashfree) substance. For degradation yield see also Table 5.4.

Compound	Degradation yield during surveyable time period, α *kg degraded C per kg C*	Methane ratio in gas, β $CH_4/(CH_4+CO_2)$ *kmol/kmol or nm^3/nm^3*	Quantity of formed methane, surveyable time period *kg CH_4 per kg material*	Remarks
Protein	100%	0,52	0,361	
Fat	100%	0,82	0,75	
Cellulose and hemi-cellulose	70%	0,50	0,227	
Degradable carbohydrates, starch, sugar	100%	0,50	0,324	
Humus, lignin, etc.	0%	0,49	0	
Polyethene, PE	3%	0,75	0,026	
Polystyrene, PS	3%	0,62	0,023	Also ~0,02 kg styrene in gas
Polyethyleneterephtalate, PET	2%	0,50	0,0076	
Poly vinyl chloride, PVC	3%	0,69	0,024	Also 0,0175 kg HCl. Also some vinyl chloride monomer in gas and leachate

Leachate treatment

Leachate emissions may be reduced by treatment of the leachate water. Some Swedish landfills have transferred the leachate to municipal sewage water treatment plants. Some landfills have local treatment. Several local treatment methods are available, e.g. biological treatment, chemical precipitation, sand filtration, activated carbon adsorption, membrane separation processes, etc. Especially BOD, COD, TOC and nitrogen can be reduced. Leachate treatment is not further discussed here, but if the water emissions are of importance in a LCA, leachate treatment should be considered.

Degradation rates during surveyable time period

Different substances degrade with different rates. In Tables 5.3 and 5.4 the degradation degree during the surveyable time period is suggested for some organic substances (see Sundqvist et al 1997). The amount of the material that is not degraded, is considered to remain unreacted.

Table 5.4. Degradation yields (α) of some organic materials during surveyable time period

Material	*Degradation during the surveyable time period*
Biomass:	
Lignin, humus and stable compounds	0%
Cellulose and hemicellulose (average)	70 %
Other carbohydrates (starch, etc.)	100 %
Fats	100 %
Proteins	100 %
Paper and carton:	
Bleached chemical pulps	90%
Unbleached chemical pulps	80%
Chemi-mechanical pulps	70%
Mechanical pulps	60%
Plastics	
Polyethene, PE	1 - 5 %
Polystyrene, PS	1 - 5 %
Polyvinyl chloride, PVC	1 - 5 %
Polyetylentereftalate, PET	1 - 5 %

Metals in MSW landfills

From a study presented in Finnveden (1996) and Sundqvist et al (1997) the emission factors for metals according to Table 5.5 have been proposed as “best available data”.

Table 5.5. “Best available data” for emission factors [kg emitted/kg landfilled].

Element	The surveyable time period	The hypothetical, infinite time period	Remarks
Fe	$1 \cdot 10^{-4}$	1	To leachate
Cu	$7 \cdot 10^{-5}$	1	To leachate
Zn	$2 \cdot 10^{-4}$	1	To leachate
Cd	$5 \cdot 10^{-4}$	1	Approximately 10 % by gas, the rest by leachate.
Hg	$1 \cdot 10^{-4}$	1	Approximately equal amounts by gas and by leachate
Pb	$6 \cdot 10^{-5}$	1	To leachate
Ni	$5 \cdot 10^{-3}$	1	To leachate
Cr	$7 \cdot 10^{-4}$	1	To leachate
As	$2 \cdot 10^{-3}$	1	To leachate

The calculation of these emission factors is based on the assumption that the release rate (corrosion, diffusion or decomposition) is not the rate determining step. If release rates can be estimated, these can be compared with the emission factors. If the release rate is equal to or lower than the emission factors, the release process may be the rate determining step. In Finnveden (1996) and Sundqvist et al (1997) some case is discussed. Considering this discussion, it seems reasonable to assume that the release process will not be the rate determining step in general, with the possible exception of Cr and Ni. For specific products, constructed in a special way, it may of course still be possible that the release process is slow enough to be the rate determining step. In these cases, the emission factors will be over-estimates and can thus still be used for conservative calculations.

The presented emission factors are uncertain. This is largely due to the variations in the data used for the calculation (i.e. concentrations in leachate and in the solid waste and the amount of leachate). Since these data vary geographically and temporally, one way of reducing the uncertainty may be to define the area and time more precisely. It is estimated that the emission factors are uncertain by one or two orders of magnitude. This is somewhat larger than the rule-of-thumb suggested by Lindfors et al (1995), where it is suggested that if nothing else is known, differences in emissions less than one order of magnitude should not be regarded as significant.

When calculating the emission factors, the landfill was regarded as a “black box”. Nothing needed to be known about the processes inside the landfill. The emission factors should therefore only be used for small changes in the composition of the solid waste. The presented emission factors are not relevant for large changes.

It is important to note that the emission factors for the surveyable time are quite small compared to the emission factors for the hypothetical, infinite time period. This implies that the major part of the emission will occur after the surveyable time-period. It has been suggested that the metal concentration may rise again after the methane-phase, (Lagerkvist, 1992, Lindfors, 1989). This implies that the environmental impacts after the surveyable time period may be more severe than those occurring in the near future. Both the surveyable time perspective and the hypothetical, infinite time perspective are thus of interest.

Specific organic compounds

Above in Section 5.3, and in Appendix B. a conceptual model for estimating the fate of organic compounds is presented. However, available data is not sufficient to calculate emission factors for specific compounds. The Table B.1 in Appendix B gives some indications of the fate of some compounds. A preliminary model to roughly estimate emission factors is given as follows:

- Compounds with Henry's constant $H > 300\ P \cdot m^3/mole$ will likely be emitted as gas in the landfill gas.
- Compounds with the octanol-water distribution factor $log\ K_{OW} < 3,4$ will likely be emitted in the leachate.
- Slowly or easily biodegradable compounds can be assumed to be microbially degraded, according to Equation 5.1. Since the surveyable time periods comprise a longer period, it is likely that several of the more or less persistent compounds will be more or less degraded. However, there is a risk that toxic intermediates are formed that can be emitted by the leachate.

Additives in paper and plastic

Additives are used in paper, as well as plastic in order to impove processability and material properties. Both paper and plastic can contain considerably amounts of additives taht should be consideed in a LCA. The roles of additives in paper and in plastic are discussed in Appendix C. Some indicative emissions factors are also given in the Appendix C.

5.4.4 Landfill fires

Landfill fires are further discussed in Appendix A in this report. Some aspects of landfill fires were discussed in our second report (Sundqvist et al, 1997), but the result presented in this report is new. Landfill fires can be of importance in a life cycle perspective. Landfill fires seem to occur so frequently that they could be considered as ”normal” degradation pathway, parallel to the normal biodegradation.

Spontaneous fires on the landfills occur by accident on most landfills. There are usually two kinds of fires: ***surface fires*** with materials burning on the surface of the landfill, and ***deep fires*** with material burning down in the landfill masses. The frequency of landfill fires seemed to be 0,5 - 1 fire per year and landfill. The amount burnt uncontrolled was estimated to be 25000 tons/year (Sundqvist et al, 1997; Bergström and Björner, 1994).

Different kind of wastes seems to have different affinity to landfill fires. Bulky domestic waste seems to have the largest affinity, and ordinary domestic waste the lowest. We estimated that the following amounts of landfilled waste could be assumed to be burnt in landfill fires were:

- domestic waste 0,2 % of landfilled amount
- bulky domestic waste 4 - “ -
- industrial waste 0,8 - “ -
- construction/demolition waste 0,6 - “ -
- other wastes 5 - “ -
- average 0,7 - “ -

In a recent Finnish study (Ettala et al, 1996) a survey of landfill fires was done in Finland. The Finnish study confirms that there is considerably amount of wastes that are burnt uncontrolled in landfill fires.

The formation of different contaminants has been studied by Bergström and Björner (1994), and Pettersson et al (1996), see Appendix A. Emission factors for landfill fires have been estimated with data from these studies, see Table 5.6. We have chosen to allocate the emissions to the organic material (combustible material), since that is the portion that is burned.

Table 5.6. Formation of potential contaminants in landfill fire

	Formation of emittants per kg of landfilled dry organic material					
	Ordinary domestic waste	Bulky domestic waste	Industrial waste	Construction and demolition wastes	Other wastes	Average
	kg/kg	*kg/kg*	*kg/kg*	*kg/kg*	*kgIkg*	*kg/kg*
Chlorobenzens	$4 \cdot 10^{-9}$	$8 \cdot 10^{-8}$	$1{,}6 \cdot 10^{-8}$	$1{,}2 \cdot 10^{-8}$	$1{,}0 \cdot 10^{-9}$	$1{,}4 \cdot 10^{-8}$
Dioxins (TCDD-eq.)	$1{,}8 \cdot 10^{-12}$	$3{,}6 \cdot 10^{-11}$	$7{,}3 \cdot 10^{-12}$	$5{,}5 \cdot 10^{-12}$	$4{,}5 \cdot 10^{-11}$	$6{,}4 \cdot 10^{-12}$
PAH	$5{,}4 \cdot 10^{-8}$	$1 \cdot 10^{-6}$	$2{,}2 \cdot 10^{-7}$	$1{,}6 \cdot 10^{-7}$	$1{,}4 \cdot 10^{-6}$	$1{,}9 \cdot 10^{-7}$
PCB	$9{,}3 \cdot 10^{-11}$	$1{,}8 \cdot 10^{-9}$	$3{,}7 \cdot 10^{-10}$	$2{,}8 \cdot 10^{-10}$	$2{,}33 \cdot 10^{-9}$	$3{,}3 \cdot 10^{-10}$
Hg	$9{,}2 \cdot 10^{-10}$	$1{,}8 \cdot 10^{-8}$	$3{,}7 \cdot 10^{-9}$	$2{,}75 \cdot 10^{-9}$	$2{,}3 \cdot 10^{-8}$	$3{,}2 \cdot 10^{-9}$

It should be noted that mercury has been treated as a ”process-related emission”, formed by the process ”landfill fire”. This is because the mercury emission would not have occurred if the organic material was not burnt.
Beside these emissions, there will also be formation of several others, e.g. sulphur dioxide, nitrogen oxides, hydrochloric acid, heavy metal dusts, etc. Those landfill fire emissions may be calculated according to the Chapter 4, and there be regarded as material-related emissions and assuming no cleaning of flue gas (in this case fire gas).

The Table 5.6 presents the amounts of different pollutants that can be formed during landfill fires. However, note that these amounts do not show the emission from the landfill - the larger parts of the pollutants are supposed to be imitted on the landfill surface, thus not leaving the system border as defined in Section 5.3.2 above. The most important emittants from the fire (PAH, PCB, dioxin) are expected to be bound to airborne particles, e.g. soot particles. It is likely that that these particles will fall down to the ground. We have made some simple calculations of the dispersion of the smoke plume from the fire, and of falling velocities of smoke particles, see Appendix A. From these calculations we estimate that that about 60 - 90 % of the formed emissions have a fall-down within 100 m from the fire. **Only 10 - 40 % of the quantities of formed emittants should be counted as emissions.** However, this estimate is very uncertain.

Another aspect that must be taken into account is that landfill fires occur accidentally, but the generation of the fires are possible to control within some limits. Ettala et al (1996) reported that they found a statistical dependence between landfilling technology and fire frequency. Insufficient covering and compacting, ash disposal and deliberate fire starting were reported to be the most typical reasons for waste ignition. Improved landfill technology reduces the frequency of landfill fires.

5.5 Discussion of alternative approaches

5.5.1 Allocation model

In the above model the emissions (except emissions from landfill fires) have been treated as product-related. The model requires detailed knowledge of the chemical composition of the studied material. In principle, it is possible to choose other allocation models. In digestion studies the biogas yields sometimes are correlated against the organic content of the waste (organic content in this case is equal to V.S. = Volatile Substance). For a typical organic waste the formation of methane and COD is according to Table 5.7.

Table 5.7 Alternative approach for lanfill. Simplified model for methane and COD generation (surveyable time period), based on organic material.

Emittant	Formed amount during surveyable time period, *kg per kg organic (dry, ash-free) substance*
Methane, CH_4	0,3
Carbon dioxide, CO_2	0,73
Ammonium, NH_3/NH_4^+	0,0005
COD (in leachate)	0,012
BOD, (in leachate)	0,003

5.5.2 Alternatives to surveyable time approach

In Figure 5.7 a hypothetical case for a municipal solid waste landfill is illustrated, where different time alternatives are marked.

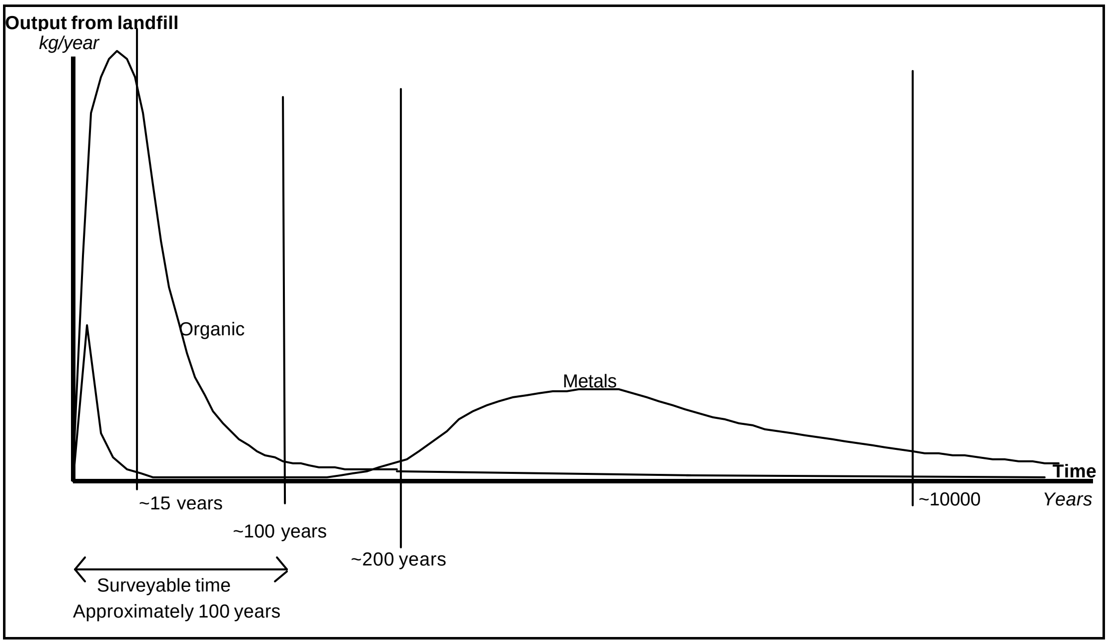

Figure 5.7. Illustration of different time horizons.

Our surveyable time approach corresponds to a period of about 1 century. The integrated emissions over a 50 year period are not expected to differ largely from the emissions over a 200 year perspective, see Figure 5.7. During the period 50 - 200 years the landfill is presumed to have low activity and low levels of emissions. However, the period before 50 years is characterised by a high biological activity causing a lot of methane generation and also relatively high leachage of organic materials. After some hundred years an increased metal leachage may be possible, especially if ambient air is diffusing into the landfill and increases the redox potential.

Shorter periods than 50 years can give other results. The Dutch approach is based on a 15 years period (Eggels and van der Ven, 1995). This period is significantly shorter than our surveyable time. A short comparison between the Dutch data and our data are given in Table 5.8.

Table 5.8 Comparisons between the Dutch approach and our approach

Emittant		Dutch approach, emissions during 15 years (Eggels and van der Ven, 1995)	Our approach emissions during surveyable time period
Assumed MSW composition			
Total carbon, C_{tot}	*kg/kg*	0,30	0,30
Mercury, Hg	*kg/kg*	$4 \cdot 10^{-7}$	$4 \cdot 10^{-7}$
Lead, Pb	*kg/kg*	$5 \cdot 10^{-5}$	$5 \cdot 10^{-5}$
Cadmium, Cd	*kg/kg*	$3 \cdot 10^{-6}$	$3 \cdot 10^{-6}$
Emissions			
Methane, CH_4	*kg/ton MSW*	62,5	127
COD	*kg/ton MSW*	2,36	5,5
Mercury, Hg	*kg/ton MSW*	$4 \cdot 10^{-7}$	$6 \cdot 10^{-8}$
Lead, Pb	*kg/ton MSW*	$5 \cdot 10^{-5}$	$2 \cdot 10^{-5}$
Cadmium, Cd	*kg/ton MSW*	$3 \cdot 10^{-6}$	$1,5 \cdot 10^{-6}$

As seen in the Table, the organic emissions differ between the Dutch approach and our approach with a factor about 2. This difference is to expect when considering the time aspects, see illustration in Figure 5.7. The Dutch 15 year period is not enough to reach the end of the methane period, the Dutch landfill will still produce methane and TOC after the 15 year period. There is also a difference in metal emissions, but these differences are probably more a consequence of use of other data when calculating the emission factors. The difference is less than one magnitude. When we presented the metal emission factors (Finnveden 1995; Sundqvist et al, 1997) we noted that the emission factors are uncertain by one or two orders of magnitudes. In that aspects the Dutch data and our data do not differ significantly.

5.5.3 Alternative approaches to the hypothetical, infinite time period

Time to reach background level or acceptable level

It is possible to set up quantitative criteria for ”acceptable level”. Today’s acceptable levels can be obtained from e.g. different guidelines for assessment of contaminated land and contaminated ground water. These acceptable levels are different in different countries. However, the future acceptable levels are unknown. The the acceptable level may be lower in the future.

It is also possible to set up quantitative criteria for ”background level” or ”when the landfill becomes a part of the landfill”. There is information available about the current background levels of metals and some organic compounds in soils, as well as in surface water and ground water.

In Table 5.9 the background levels in soil and "acceptable" limits for concentration in soil (for non-restricted land use in Sweden) are compared with the concentrations in MSW.

Table 5.9. Background level and acceptable level of contaminants

Metal	Background level 1)	Acceptable level, generic guideline values for contaminated soil 2)	Initial level in MSW 3)	Level in landfill after surveyable period 4)
	mg/kg dw	*mg/kg dw*	*mg/kg tot*	*mg/kg dw*
As	7	40		
Cd		12	5	15
Co	19	250		
Cr	52	250		
Cu	15	200	1500	4600
Ni	17	200		
Pb	24	300	1200	3700
V	56	200		
Zn	51	700	1200	3700

Remarks:

1) Background level in forest soil in Sweden (Naturvårdsverket 1996)
2) Generic guideline values for contaminated soils in Sweden, land with less sensitive use (Naturvårdsverket, 1997)
3) Metal analyses in solid waste (Rylander, 1985)

After the surveyable time period it is to expect that both metals and organic material will be released. It is difficult to estimate the future long-term release rates for both organics and metals. Some comparisons can be done with e.g. peat bogs, archaeological remainings in soil, etc. There is a possibility that the release of organic material will exceed the release of metals, thus causing the concentration of metals in the landfill to rise. In that aspect the landfill will never reach the background level, except when all metals and all organic material has been released (that is equivalent to the hypothetical, infinite time period). On the other hand, if the metal leachage is higher than the release of organic material, then the metal concentration will decrease and some time be of the same level as the background, or any other ”acceptable level”.

A preliminary, simplified illustration of the time to reach acceptable level and background level has been made according to the following simplifications and assumptions:

- During the surveyable time period there is a considerable loss of organic material due to the degradation process. However, the rate of transport of organic material from the landfill has been assumed to be negligible after the surveyable time period.
- The inorganic material amounts in the landfill is almost constant during the surveyable time period. Negligible amounts of metal have leached out (10^{-5} to 10^{-3} kg/kg of heavy metals have leached out). Some chlorine and soluble salts have been released, but more than 95 % of the inorganic material is estimated to remain in the landfill.
- The rates of metal emissions (emission per year) have been assumed to be same as during the surveyable time period (Table 5.5). The surveyable period was set to 100 years.

With these assumptions, it was found that the time until the metal in the landfill matrix reaches the acceptable level is about 10^7 - 10^8 years, and the time until the background level 10^9 - 10^{10} years.

Another variant of the background approach is to compare the concentration of the leachate water with the concentrations in natural waters. Table 5.10 presents background levels in Sweden (Naturvårdsverket 1990), and concentration measured in leachate waters (samples taken close to the landfill body) in a recent study (Öman and Wennberg, 1997).

Table 5.10 Background level in surface waters, and concentration in leachate

Metal	Background level surface water (”moderately high”) (Naturvårdsverket 1990) *µg/l*	Concentration in leachate (five landfills) (Öman & Wennberg 1997) *µg/l*
Cd	0,05 - 0,1	0,09 - 1,4
Pb	1 - 2	1 - 15
Cr	1 - 2	7 - 45
As	1 - 2	<2,5 - 7
Cu	1 - 2	8 - 45
Ni	1 - 2	11 - 91
Zn	5 - 15	19 - 342

As seen in the Table, the quotient between the concentration in leachate and in background level varies from about 4 to 30 for different metals. The release of metals can be estimated by different models. Two simple models are as follows:

- Constant leaching rate. The leachate production and the concentration are constant. The solubility in water is the rate-determining stage. In this case the background level is reached first when the leaching is complete.
- The leaching rate is proportional to the amount of each metal in the landfill. This can be the case when the release is the rate-determining stage. In this case the concentration will slightly decrease with time and reach the background level some time in the future.

Leachate concentrations and background concentrations are assumed to be according to Table 5.10, and the metal composition in MSW is assumed to be according to Table 5.9. The landfill characteristics are assumed as: landfill height 10 m, volume weight in landfill 0,7 ton/m^3 and a leachage production of 0,2 m^3/m^2,year. Simple calculations then give the following results:

- The times for complete leaching, according to the first model (constant concentration in leachate) vary from about 10^4 to 10^7 years for different metals. It should be observed that this approach gives higher leaching rate than the use of emission factors used on the previous page.
- The second model (leaching rate proportional to the amount in landfill) gives times for metal concentration in leachate to reach the background level between 10^3 and 10^7 years. However, there are still considerable amounts of metals left in the landfill when the concentration in leachate has reached the background level. The concentration in landfill is up to 70 times higher than the background level in soil (assuming that there is no loss of organic material after the surveyable time period). This demonstrates the necessity to study the concentrations both in water (leachate) and in the remaining solid phase in the landfill.

In these simple calculations, it was presumed that the landfill is in the methane phase (redox below zero, and neutral pH) during the whole studied period. However, this is probably not the case in reality. Probably parts of the landfill will be aerobic after several hundred years, and this will give other leaching conditions, not considered in the calculations above.

It should also be noticed that the use of concentration levels for assessment of landfilling is not in accordance with conventional LCA practice. Usually in LCA (of products) concentrations are not considered, only flows. It is the amount of the pollutant that is studied, independent of the concentration. However, in other kind of studies, such as environmental impact assessment of a waste landfill, the discussion about background levels could be more relevant.

The 1 million year period and the time to complete leaching

In Chapter 4 we discussed the time aspects for incinerator ashes, and found that the time for complete leaching is 1 - 10 million years, different for different elements and for different of landfill types. We can roughly assume that the time for complete leaching from a MSW landfill may be of the same magnitude, or possibly longer. After 1 million years there may still be material in the landfill, but probably the major part of all emissions has occurred. The emission factors for the 1 million year period can then be roughly estimated to be 0,1 - 1 kg/kg for most heavy metals.

Next glacial period

The next glacial period is a more interesting approach to discuss, even if the glacial period is uncertain. From the history, it is to expect that next glacial period in Sweden will occur about 20 000 - 50 000 years from now. On the other hand, this historical tendency can be disturbed by the greenhouse effect. But assuming that the glacial period will occur, the emission of metals can be roughly estimated to be about 0,001 - 0,1 kg/kg for most metals for a 20 000 year period until next glacial period. Thus the major part of the metal emissions has not occurred at time for the glacial period (assuming that the anaerobic, and pH neutral environment in the landfill will last). However, after the glacial period all landfilled wastes have been released to the environment (in that aspect our hypothetical, infinite time period approach considers the emissions until next inter-glacial period). The time period until next interglacial time may be several hundred thousands to one million years.

Conclusions

From this discussion about the aspects of the longer time period, we draw the conclusion that the hypothetical, infinite time period is most easy approach to use. There is a lack of reliable models for the long-term behaviour of MSW landfills. But if such models are developed, criteria such as acceptable level or background level should be considered, at least in a deeper study. The hypothetical, infinite time period approach should still be good for a first estimate in a screening analysis.

The relevance of using the next glacial period as a guideline for LCA can also be discussed. Since the movements of the glacial ice will transport all landfilled material to the environment, it is - in a screening analysis - of little interest how much that has been released when the period starts, when anyhow all material has been released when the next interglacial period starts. However, in a more detailed analysis also the glacial period aspects can be of interest.

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6. New methods of landfilling of MSW

6.1 General aspects

The MSW landfill described in Chapter 5 is characterised by its input of mixed wastes. Different materials and different wastes are mixed when put into the landfill. However, that kind of landfilling will be restricted in the future. In Sweden there is a ban for landfilling of organic wastes from year 2005 and of combustible wastes from year 2005. From year 2000 there is a landfill landfill tax of 250 SEK/ton. A national landfill directive is expected, based on the EU Landfill Directive. The flows of wastes to landfills are thus expected to be changed in a close future. Different studies by Swedish authorities have shown that the landfilled amount will decrease by 50 % between year 1995 and year 2005.

This tendency is general in whole Europe. The current EU landfill directive will force the European countries to change the landfill policies. Landfill taxes are existing in other countries such as Denmark, Great Britain and Finland. Different kinds of national landfill directives that restricts the use of landfills are in action in Germany, France and other countries.

The future waste management will probably be based on source separation, where different waste fractions are collected separately. A possible separation scheme may be based on the following preseparated fractions (Swedish EPA, 1996):

- hazardous domestic waste, e.g. paints, oils, hazardous batteries, electronic scrap, etc., for special treatment at authorised treatment plants
- recyclable materials, e.g. paper, glass, metals, plastics
- biodegradable material, garden wastes, food wastes, etc. which are treated by anaerobic digestion or composting. For the anaerobic digestion a digestion reactor can be used, or special ”biocells” can be constructed a landfill, see below.
- combustible materials, which are treated in an incineration or combustion plant where its energy contents are recovered
- the residue will be put in a separate landfill

There are especially two kinds of wastes that can be of interest for landfilling in the future:

- Organic, degradable wastes can be put into special designed cells and undergo an accelerated digestion in order to rapidly recover the methane content. These cells are called biocells, biofills or digestion cells. The waste is put on a tight bottom and sealed with gas-tight material. Leachate is collected. The generated gas is collected.
- The residue consisting of material that cannot be recycled, combusted or treated by biological methods. Probably, to a large extend this fraction will consist of discarded composite products which is assembled by several different materials. This waste is put is special cells with tight bottom and a tight cover to reduce the leachate generation.

In this Chapter we will discuss these two kinds of future landfills.

6.2 The biofill

6.2.1 General

Landfilling of biodegradable wastes in separate cells was introduced in Sweden in the end of the 1980:s. The most common form of this system is the use of ”biocells” or ”biofills”. A biofill may be characterised as a landfill designed for landfill gas production and involves such features as

- clay embankments shielding the cells from their surroundings
- installation of horizontal gas extraction systems
- improved operation.

Only organic degradable materials are assumed to be put into the biofill. Paper and wood may be a part of the organic waste. Plastics, metals, glass and similar is assumed to not occur in the biofill.

The biofill is constructed as an isolated cell, with a minimum of precipitation water percolating the waste and with a minimum leakage of biogas to the environment. There is an effective collection of the generated biogas, and an effective leachate collection system. The principal construction is actually as the described MSW landfill, see Section 5.2, but the gas collection system is more efficient. The process is better controlled than in the mixed MSW landfill, and the rate of the degradation is faster.

The organic material undergoes the same processes as in the mixed MSW landfill, however the degradation rate is fasterr. In the mixed MSW landfill the methane stage ends about 50 - 100 years after the waste has been put into the landfill. In the biofill the end of the methane stage is expected to be reached after 10 - 20 years. With improved technologies the rate can be ever faster.

6.2.2 Emissions from biofills

The processes in the biocell are the same as in the mixed MSW landfill. The same products are formed. However, there are at least two important improvements that must be considered:

- The gas collection is more efficient. In the mixed MSW landfill only 10 - 50 % of the formed methane is able to be recovered. In the biocell there is indications that 75 - 80 % of the formed methane can be collected.
- The soil oxidation techniques are being improved. There are indications that a major part of the non-collected methane may be oxidised in the soil cover if it is designed in a proper way. Lagerkvist and Maurice (1996) have measured the methane profile

in a soil cover and found that the methane concentration in the soil air was not detectable in the upper zone of the cover. Methane leakage may also occur from cavities in the cover and from leachate channels. We suggest that perhaps 50 - 75 % (or even more) of the non-recovered methane can be oxidised, if the cover is designed in a proper way, and other leakage pathways is eliminated.

The partition between emitted carbon in the leachate and in the gas seems to be the same for biocells and mixed MSW landfills. In Chapter 5 it was noted that the ratio between the carbon in the leachate and the carbon in the produced gas (methane plus carbon dioxide) was about 0,01. Studies by Åkesson (1997) indicate about the same ratio (about 0,01) for six different test cells.

The surveyable time aspects may be discussed. For the mixed MSW landfill, we defined the surveyable time period as the period until the end of the methane stage. That should correspond to a period of 50 - 100 years. In the biocell the end of the surveyable time period is reached after perhaps 10 - 20 years. However, if comparing a biocell with a mixed MSW landfill over a 100 year period, the differences in emission (related to per weight of landfill organic waste) will be small, see discussion in Section 5.5.2. The time directly afterwards the methane stage is characterised by low activities: very low organic degradation and the metal will still be bound in the remaining organic matrix. So it seems appropriate to use the same process definition on the surveyable period. And the difference between a 100 year perspective and a 10 - 20 year perspective will probably be negligible – the biocell will give only slightly more emissions during a 100 year period than a 10 – 20 year period. There is also discussion about using the biofill residue as fertilser or soil material, instead of laeving it in the landfill.

The following procedure is recommended to use for calculation of the emissions from the biocell during the surveyable time period.

- The ultimate analysis of the waste should be known (C, H, O, N, S, Cl, moisture, plus heavy metals of interest). Also the contents of fat, proteins, cellulose, lignin+humus (”non-degradable”), sugar+starch (easy degradable”) should be known.
- The formed degradation products can be calculated from the Equation 5.1 in Chapter 5.
- 75 - 80 % of the formed methane will be recovered as fuel gas.
- 50 - 75 % of the remaining methane will be oxidised in the soil cover.
- 1 % of the degraded carbon will be let out in the leachate as TOC. The BOD and COD should be calculated in the same way as for mixed MSW landfills. Treatment of leachate should be considered.
- The ammonia formed by the nitrogen in the waste is emitted in the leachate as ammonium. The hydrogen sulphide formed from the sulphur in the waste will be partitioned between the solid phase, the leachate and the gas according to the Equation 5.1.
- Only minor quantities of metals are assumed to be in the organic waste. There is insufficient data available to determine metal emission factors, but as a first estimate the emissions factors presented in Section 5.4 should be relevant to use.

- If plastics are present in the waste it is likely that the plastics will undergo some degradation. There is insufficient data about plastic behaviour in biofills, but probably the degradation during the surveyable time will be slightly slower than in a mixed MSW landfill (relatively to other organic materials). As a first estimate the figurs from Table 5.3 or 5.4 can be used.
- After the surveyable time period the processes in a biocell should be equivalent to the processes in a mixed MSW landfill.

6.3 Cell deposits - residue waste

The residual waste can be defined as the residue when hazardous materials, recyclable materials, combustible wastes and biodegradable wastes have been separated. The content should be mainly inorganic, such as metals, glass, ceramic, leather and similar material, plus non-recyclable plastics. A large portion is expected to be composite components. Some quantities of organic impurities are also to be expected.

Landfill cells for residual waste have been tested in the Sofielund landfill plant outside Stockholm. However, the tests have only run for a few years and it is difficult to interpret the data collected.

Leaching tests of residues have also been carried out in Germany. The German separation system is based on the residue when hazardous materials, DSD-material (packages and newsprint paper) and organic wastes have been collected. The leaching tests often show low leachability, depending on that the tests are only short-term (one day) and do not consider the possibility for leaching of corrosion products. For example, some characterisations of residual wastes are example described by Brinkmann et al (1996) and Blume (1996).

Composition of residual waste

The residual waste at the Sofielund plant has been analysed (Sundqvist, 1995). The material composition was found to be according to Table 6.1.

Table 6.1 Composition of residual waste at the Sofielund plant (Sundqvist, 1995). Note: The original analysis was made in the start-up of the new source separation system. The waste contained some combustible material and some recyclable materials that has been excluded in this presentation. The analyses presented here should be more relevant for the future situation.

Component	Weight-%
Hazardous wastes	1,0
Glass, recyclable	6,1
Paper, recyclable	0,2
Plastic packages, recyclable	3,0
Tin cans, recyclable	17,8
Metal packages (other than tin cans)	0,6
Waste bags	0,9
Combustible material, excl. plastics	6,1
Organic material (biodegradable)	1,2
Plastic objects (non-packages)	7,7
Home kitchen appliances	3,2
Electronic devices	2,2
Electric devices	0,4
Cables, flexes	2,1
Bulbs, fluorescent tubes	0,5
Computers	0,1
Cableless o flexless e/e-devices	0,2
Batteries	0,1
Metal scrap	21,8
Aluminium	1,9
Copper	0,8
Lead	0,2
Leather and rubber	10,6
Ceramics	6,1
Ornament glass	0,6
Others	4,2

Estimates of the content of heavy metals in residual waste have been made (Sundqvist, 1995), see Table 6.2.

Table 6.2 Metal and heavy metal in residual waste at Sofielund (Sundqvist, 1995)

Metal	*mg/kg waste*
Aluminium, Al	19 000
Cadmium, Cd	30
Chromium, Cr	400
Copper, Cu	13 000
Iron, Fe	20 000
Lead, Pb (excl. PbO)	1900
Mercury, Hg	1
Nickel, Ni	150
Zinc, Zn	7000

Preliminary estimation of emission factors

There is a lack of relevant field data for emissions from residual waste landfill cells. There is also a lack of characterisation of the waste that make it difficult to derive a mathematical model. However, some simple and preliminary considerations have been done below.

As mentioned in Chapter 5 leaching of metals can be seen in two steps: 1) release from the solid phase, and 2) further transport in the landfill. The release rate is governed by, for example, corrosion of metal pieces and diffussion metal compounds in plastics. The transport rate is assumed to be governed by the solubility (which depends on pH, redox and presence of certain ions).

In the first stage of our study (Sundqvist et al., 1994) we made some calculation of corrosion rates and solubilities in water during different stages of the mixed MSW landfill. The ”aerobic oxygen and nitrate reducing stage” can to some extent be representative for the conditions in the residual waste landfill: a slightly positive redox potential and a slightly acidic pH. In Table 6.3 corrosion rates and solubilities is given:

Table 6.3. Corrosion rate and solubilities in the residual waste landfill. Positive redox potential and slightly acidous pH is presumed (adapted from Sundqvist et al, 1994).

Metal	Corrosion rate (metallic surfaces) *µm/year*	 *kg/m^2,year*	Solubility *mg/l*
Iron	25	0,2	$5,6 \cdot 10^{-6}$
Aluminium	5	0,013	2,7
Copper	5	0,045	$60 \cdot 10^{-9}$
Lead	25	0,28	$<2 \cdot 10^{-5}$
Cadmium	*		soluble
Mercury	-	-	$2 \cdot 10^{-5}$
Zinc	*		soluble

Remarks:

* Both Cadmium and Zinc are subject to corrosion, but the corrosion rate was not estimated.

soluble: solubility $>10^2$ M

The corrosion rate expressed in kg/kg (kg corroded per kg of metal) depends on the geometry of the studied metal object. The geometrical shape can be plate, wire, tube, etc. Different alternative shapes of the metal, and different scenarios of the leachate production (see below) show that the corrosion rate according to the table 6.3 is several magnitudes higher than the leaching rate (if the solubility is rate determining for the leaching). E.g. a corrosion rate of 5 µm/year, m^2 applied on a 1 mm thick plate corresponds to a period of 200 years for complete corrosion. Thus, for metallic objects it is the transport that is rate determining.

The release of additives of metallic compounds from plastics has not been studied in this project. However, some rough estimates indicate that the diffusion of metals from plastic may be faster than the transport rate. A very simple, hypothetical case with an assumed diffusivity of 10^{-15} m/s, a plastic object with a thickness of 2 mm (1 mm diffusion pathway), a metal concentration in the plastic of 1 weight-%, and a hypothetical molar weight of 100 g/mole of the metal, gives a diffusion rate of about 0,02 gram per year and m^2 of plastic surface. With a surveyable time period of about 200 years this corresponds to an emission factor of around 0,4 kg/kg. The above preliminary estimate of emission factors should be valid for metal compounds used as additives in plastics.

The surveyable time period can be discussed. This period has been studied from two perspectives:

- The residence time of the water in the landfill is approximately 120 years. Probably there should be several water renewals to achieve a kind of pseudo-steady-state.
- The first period seems to be characterised by a high corrosion rate, while the transport from the landfill is low. A corrosion rate of 5 µm/year, m^2 applied on a 1 mm thick plate corresponds to a period of 200 years for complete corrosion.

Thus we define the surveyable time period as the period when corrosion is of importance. The length of this period may be several hundred years. For the calculation below, the time for two water renewals, 240 years has been chosen.

For estimating emission factors, we assume a model landfill with a height of 10 m, a bulk density 800 kg/m^3, and the porosity 60 %. The leachate production is assumed to be 50 l/m^2,year according to the proposed national landfill directive (Swedish EPA, 1996). The solubilities according to Table 6.3 is governing the transport of metals from the landfill.

From these assumptions, preliminary emission factors for some metals have been calculated. Preliminary emission factors from these assumptions were calculated, see Table 6.4.

Table 6.4 Preliminary estimates of emission factors for metals in residual waste landfill.

Metal	Emission factor, surveyable time period *kg/kg*	Emission factor, hypothetical, infinite time period *kg/kg*
Aluminium, Al	10^{-4}	1
Cadmium, Cd	0,1 - 1	1
Copper, Cu	10^{-12} - 10^{-11}	1
Iron, Fe	10^{-10} - 10^{-9}	1
Lead, Pb	10^{-8}	1
Mercury, Hg	10^{-5} - 10^{-4}	1
Zinc, Zn	10^{-2}	1

References Chapter 6

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7. Industrial production wastes

7.1 Coal ashes

The Sections considering coal ashes are from both our reports (Sundqvist et al, 1994; Sundqvist et al, 1997).

7.1.1 General

Coal is a fossil fuel, sometimes used for production of electricity. In LCA there are two approaches to handle electric energy production: a) average energy production, and b) marginal energy production. In Sweden the ”average energy production” is based on mainly hydropower and nuclear power with a small addition of biofuel and fossil fuel. The ”marginal energy production” is based on the assumption that the electricity used in the studied system is taken from a marginal energy source. The most common marginal electricity source used in Sweden is imported electricity from Denmark, where the electricity is produced from coal combustion. The coal combustion generates coal ash as a waste. The coal ash is disposed by landfilling.

Coal belongs to the sediments of organic origin. Except organic substances, coal also contains minerals, such as clay minerals, quartz, sulphides and calcite. During combustion of coal the organic fraction is mainly converted into CO_2 and H_2O, while most of the minerals melt out of the coal. During the combustion and the rapid cooling in the flue gas duct, there is a large formation of glasses. Metal sulphides in the coal are mainly converted to sulphur dioxide and metal oxides. These metal oxides, together with various metal salts, occur mostly as discrete particles, but are also integrated with the glass. The glass phases consist of a resistant internal matrix and a reactive surface layer.

7.1.2 Coal ash landfilling

The processes in the coal ash landfill is discussed in our first report (Sundqvist et al, 1994). The most important processes in the initaial stage which governs the identification of the surveyable time period is:

- water uptake in cover material and ashes
- consolidation effects
- hydratisation of various minerals in the ashes
- impact of atmospheric oxygen and carbon dioxide
- devitrification of clay minerals in the ashes
- swelling effects
- cementation of ashes
- leaching of soluble salts
- subsidence due to leaching and consolidation
- late transformations due to changed chemical conditions.

Leaching of soluble salts will gradually change the chemical conditions in the ashes. Such changes in the chemical conditions may influence the stability of some hydrated minerals formed when the ashes were first exposed to water in the landfill. New interactions between components in the ashes may take place, when the chemical conditions in the ashes have been changed due to leaching of soluble salts.

7.1.3 Emission factors for coal ash landfills

Emission factors for coal ash have been calculated from a theoretical model. The model is described in detail in our report from the first stage of the study (Sundqvist et al, 1994). It has been assumed that the ashes are covered by soil. The ashes have been assumed to be brought into the landfill in a partly water saturated form, since water is added to prevent problems with dust during handling. It has also been assumed that the ashes are compacted by suitable equipment to ensure the mechanical stability of the landfill. Further, it has been assumed that the cover is applied during the first ten years after disposal of the ashes. As a first estimate, it has been assumed that consolidation of the ashes is most pronounced during the initial stage before the cover is applied, and that sufficient allowances are made in the design work of the cover for remaining consolidation effects. Therefore, disruptive events, such as faulting, have not been considered in the model.

The model considers the partitioning of water between percolation and water diverted laterally to the side. To account for expected heterogeneities in the cover material (i.e. by other causes than disruptive events) and in the ashes, it has been assumed that a minor part of the surface area will have increased hydraulic conductivity and allow water to penetrate easily. The percolating water has been assumed to become fully saturated with contaminants when passing through the ashes. The lateral runoff water in the soil cover has been assumed to accumulate dissolved contaminants that diffuse from the ashes into the soil cover. Depending on the partitioning coefficient for the infiltrating water, the distribution between the two release mechanisms will be different, and in certain cases the surface runoff may be the predominant release mechanism.

The ashes are initially partly water saturated due to wetting. During compaction and consolidation the porosity will decrease, thereby increasing the degree of water saturation. As a result of capillary forces, water will be retained in the ashes until the pores are completely water filled (this is true for dense materials with small particle sizes). Consequently, only small amounts of drainage water can be expected during the initial period before the ashes are water saturated. Assuming a porosity of 30% after compaction and consolidation, and an initial water content of 10% by weight, the initial water saturation can be calculated to about 60%. In a 10 m high landfill the amount of water required to attain water saturated conditions can be estimated to 1,2 m^3/m^2 of landfill. Assuming an infiltration rate of 0,4 m^3/m^2,year before the soil cover is applied, we find that water saturation can be reached after about 3 years. The model has been formulated assuming water saturated conditions. The importance of an early period with uncovered ashes has been demonstrated in separate calculations assuming increased percolation rates.

The chemical behaviour of the studied chemical elements has been assumed to resemble that of their respective oxides or hydroxides. Solubilities of the metals have been judged based on measured data for different types of coal ashes but also theoretical estimates.

The emissions have been exemplified by calculations for copper, nickel, zinc (Sundqvist et al, 1994) and chromium and cadmium (Sundqvist et al, 1997).

A number of difficulties have been identified when trying to define objectively the extension of the surveyable period. It has been realised that the developed model is presently too simple to address these questions. Therefore, it has been decided to present the emissions over a fixed period of 100 years. Consequently, the results should not be regarded as representative for a true surveyable period, the quantification of which will require further development.

The amount of water infiltrating the covered landfill has been varied using an infiltration rate of 0,125 m^3/m^2,year as a central case. The partitioning coefficient between percolating water and surface runoff through the soil cover has also been varied, using a factor of 0,5 as a central case.

The content of contaminants in the solid ashes has been assigned an average value estimated from compiled analyses of coal ashes (Wu and Chen 1987; Straughan et al. 1978; Taylor et al. 1982, SOU 1978:49). The used average values for the studied metals are presented in Table 7.1.

The aqueous solubility has been varied for each of the studied metals, using the average value from analysed leach water samples from leach ponds downstream ash landfills as a central case (Straughan et al. 1978; KHM). Extreme value variations have been made using theoretically derived solubilities in highly alkaline environments, pH 12, representative for the early period with fresh ashes (Conner 1990; Parkhurst 1980).

In Table 7.1 the results are summarised. The results are given as the integrated emissions of the individual metals over a fixed period of 100 years, respectively the hypothetical, infinite time. In Figure 7.1 the emission over an extended period is presented. The results have been presented as the emissions related to the production of 1 kWh of electric energy. In our first report (Sundqvist et al, 1994) a sensitivity analysis was made, where the influences of infiltration (from 80 to 400 mm per year), solubility (varied with a factor $10^{\pm 1}$) and covering (uncovered resp. covered) were studied. The figures presented below is referred to the ”central case”. The sensitivity analysis showed that the emission factor presented varied less than one magnitude.

Table 7.1 Summary of calculated emissions from a landfill for coal ashes during a 100 years period resp. the hypothetical infinite time period

Element	Ash content *mg/kg*	Solubility *mg/l*	Integrated emissions during 100 years $\mathit{kg/kWh}_{el}$	Integrated emissions during infinite time $\mathit{kg/kWh}_{el}$	Emission factor 100 year period *kg/kg*	Emission factor infinite time *kg/kg*
Copper, Cu	342	0,06	$2{,}1\cdot10^{-9}$	$2{,}1\cdot10^{-5}$	$1\cdot10^{-4}$	1
Nickel, Ni	295	0,06	$2{,}1\cdot10^{-9}$	$1{,}8\cdot10^{-5}$	$1\cdot10^{-4}$	1
Zinc, Zn	676	0,6	$2{,}0\cdot10^{-8}$	$4{,}1\cdot10^{-5}$	$5{,}5\cdot10^{-4}$	1
Chromium, Cr	135	0,2	$1{,}8\cdot10^{-8}$	$8{,}1\cdot10^{-6}$	$2\cdot10^{-3}$	1
Lead, Pb	185	0,02	$1{,}3\cdot10^{-9}$	$1{,}1\cdot10^{-5}$	$1\cdot10^{-4}$	1
Mercury, Hg	3	0,002	$8\cdot10^{-9}$	$1{,}8\cdot10^{-7}$	$5\cdot10^{-2}$	1

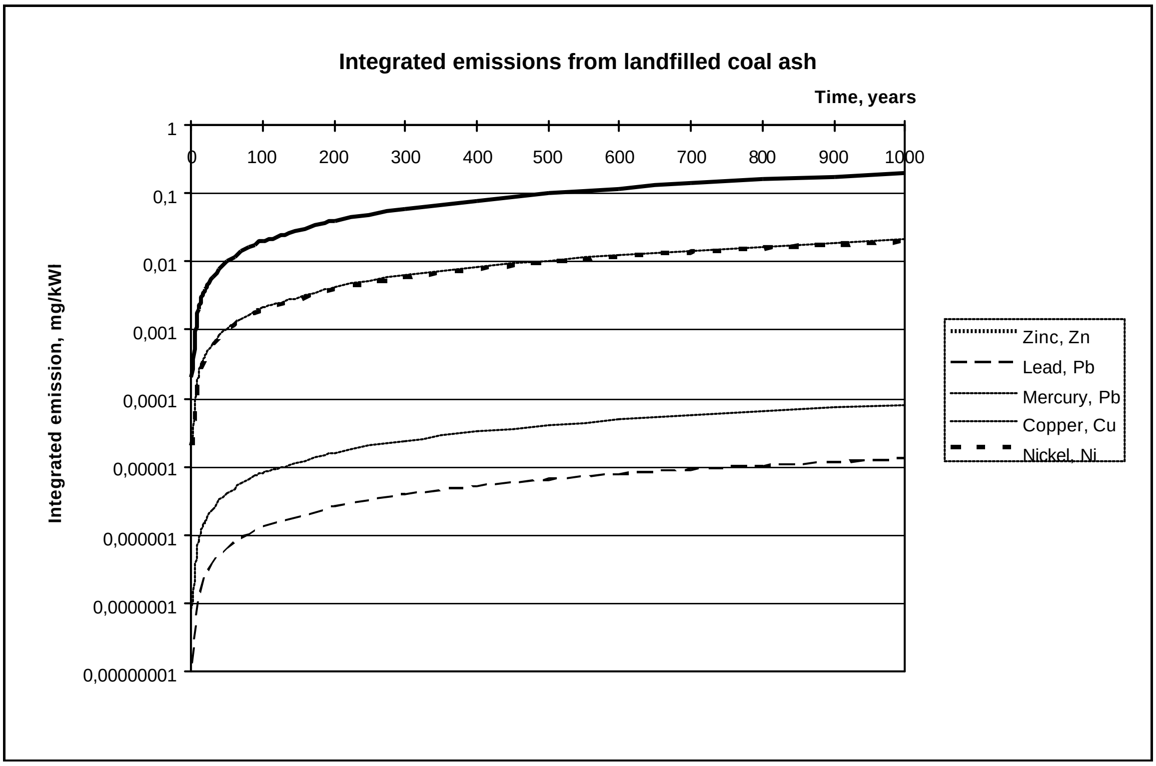

Figure 7.1 Cumulative emissions from landfilled coal ashes.

7.1.4 Applicability of the coupled chemistry- and transport model

The general chemical model concept developed for the MSW incineration ashes, referred to in Section 4.3, and presented in our second report (Sundqvist et al, 1997), would be quite suitable also for landfilled coal ashes. The general characteristics of coal ashes are in certain respects similar to the MSW incineration ashes. In a comparative study Taylor et al. (1982) noted that when different mixtures of coal ashes and MSW incineration ashes where subjected to leach tests, the leachability in general increased with an increasing fraction of MSW incineration ashes in the mixtures. This indicates that coal ashes can be expected to be leached to a lesser extent than the estimates presented for the MSW incineration ashes.

7.2 Landfilling of mine tailings

7.2.1 General

Mine tailings are of different kinds, e.g. iron ores and sulphidic ores. Iron ores generally pose fewer problems. The iron oxides/hydroxides are quite stable and have an ability to retain other heavy metals by adsorption or co-precipitation. Sulphidic ore tailings are more problematic since oxidation of the sulphide traces to sulphate will result in lower pH and increased solubility of different trace metals. The low pH of the leachate may also rapidly (in the order of tens to hundreds of years) deplete the buffering capacity of the tailings. In the present study only sulphidic ore tailings will be considered.

It should be noted that mine waste is not only tailings sand, but also waste rock, which in volume may be the predominant waste type. Waste rock has not been considered in the present study.

It has been assumed for the study that the tailings are equipped with an efficient cover to minimise the release of heavy metals from the landfill. A cover design representative of current projects in Sweden has been assumed, i.e. a tight clayish till or Cefyll layer in combination with a protective layer of ordinary till.

The sulphidic tailings are usually quite inert during an initial phase. The oxidation of sulphides may require an induction period, possibly connected with the in-growth of bacteria that can catalyse the oxidation of sulphidic minerals. If the tailings contain lime, they may have an initial pH buffering capacity. The presence of lime in the material restricts the leachability of many heavy metals, possibly due to secondary precipitation of metal hydroxides or metal carbonates.

When a cover is applied to the landfill, a certain time is required to establish stationary moisture conditions in the waste heap. Studies for Swedish climatic conditions have shown that stationary conditions may be reached after a few years. The way the covers are designed, the oxygen penetration is significantly reduced. By optimising the cover design so that the tight layer remains essentially water saturated for the major part of the year, both the oxygen penetration and the water percolation can be kept low.

The rate of oxygen transport into the landfill determines the time schedule for the depletion of buffering components in the waste heap. Once acidic conditions are established throughout the landfill, increased metal concentrations can be expected. Depending on the content of buffering components, the acid production potential and the rate of oxygen transport into the landfill, the extension of the buffered period may be in the order of a few years to many hundreds of years.

Due to weathering reactions in the tailings, small size particles will be washed out from the landfill. It is quite possible that this material is responsible for the acidic drainage waters identified at present day. If this is the case, there is a potential for significantly increased emission rates from tailings in the future. However, by application of covers, the potentially increased emission rates can be significantly reduced.

It is desirable that the surveyable period should consider the following effects, i.e.:
- induction period for the sulphide oxidation,
- buffering capacity of the tailings,
- establishment of stationary conditions,
- ability of cover materials to decrease oxygen intrusion and water percolation, and
- washout of small-size weathering products from the landfill.

7.2.2 Emission factors for landfilling of mine tailings

The behaviour of mine tailings requires a special treatment, and a different approach than used for ashes and sludges has been taken to model the emissions. It has been recognised that the emissions of metals from mine tailings are governed by oxidation of the sulphidic traces in the tailings sand. The processes taking place in the tailings are very complex and the estimates given by the model must therefore be regarded uncertain. However, more sophisticated modelling exercises have been used as a background for the present calculations, sometimes by using results from more complex calculations than would have been possible to perform within the present study. The study has been restricted to two metals, copper and zinc.

As for the other types of waste, a production quotient has been defined as the mass of sand tailings created divided by the mass of produced metal. Production quotient has been estimated for a number of Swedish sites using partly historical data for the production. The production quotient has been calculated from available production data for different Swedish mining areas (SCB Statistical Abstract of Sweden 1984; Qvarfort 1989; SCB Naturmiljön i siffror 1990). Due to varying composition of the ores, the calculated production quotients differ significantly. A general trend can be observed, that the production quotients are higher for today's mining sites than for historical sites, owing to the lower grade of the ores mined today. On the other hand, the metal content of the tailings is usually lower today, since extraction of metals is more efficient than previously. A summary of calculated values are presented in Table 7.2.

Table 7.2 Calculated values of the production quotient between produced sand tailings and produced metal

	Production quotient *kg tailings/kg refined metal* Copper, Cu	 Zinc, Zn
Average	110	130
Max.	270	550
Min.	67	10

The weathering of pyritic mine tailings is governed by the supply of oxygen from the atmosphere, the content of sulphidic sulphur in the tailings and the oxidation kinetics of the sulphides. In covered tailings the supply of oxygen is expected to determine the overall weathering rate (Magnusson and Rasmuson, 1983).

The model used for the calculation was presented in our first report (Sundqvist et al, 1994). The model is based on oxygen penetrating into the tailings and oxidising the sulphides. The oxygen penetration through the soil covered was estimated to average 0,5 mole/m^2,year, with a variation from 0,05 mole/m^2,year as a minimum penetration case and 5 mole/m^2,year as maximum penetration case. The so calculated values have been presented as the surveyable time period emissions (corresponding to a period of 100 years) for the production of 1 kg metal.

Considering available information for sand tailings in different Swedish locations, the values presented in Table 7.3 have been calculated. An interesting notation is that the production of copper will lead to production of waste containing also other contaminants, such as zinc and arsenic. The results in Table 7.3 are calculated considering only the primary minerals, e.g. zinc has been neglected in mining areas where only copper is produced commercially and vice versa. The results for the different cases are also shown in Figure 7.2. Table 7.3 also shows the estimated emissions during the hypothetical infinite period.

Table 7.3 Emission factors for for mine tailings. Results refer to data from 8 different mining sites in Sweden.

Studied sites	Emissions during surveyable period (100 years) *kg emitted per kg of produced metal*						Total emission during infinite period *kg emitted per kg of produced metal*	
	Copper, Cu Weathering rate			Zinc, Zn Weathering rate			Copper	Zinc
	Average	High	Low	Average	High	Low		
Average value	$2\cdot10^{-4}$	$4\cdot10^{-3}$	$8\cdot10^{-6}$	$9\cdot10^{-3}$	$1\cdot10^{-1}$	$3\cdot10^{-5}$	$7\cdot10^{-2}$	$4\cdot10^{-1}$
Maximum any site	$5\cdot10^{-4}$	$5\cdot10^{-3}$	$1\cdot10^{-5}$	$4\cdot10^{-2}$	$4\cdot10^{-1}$	$4\cdot10^{-5}$	$1\cdot10^{-1}$	$7\cdot10^{-1}$
Minimum any site	$2\cdot10^{-5}$	$4\cdot10^{-3}$	$2\cdot10^{-6}$	$2\cdot10^{-4}$	$8\cdot10^{-3}$	$2\cdot10^{-5}$	$2\cdot10^{-2}$	$5\cdot10^{-2}$
	Emission factors during surveyable period Case: average weathering rate *kg emitted per kg of metal in tailings*						Total emission during infinite period *kg emitted per kg of metal in tailings*	
	Copper			Zinc			Copper	Zinc
Average value	$5\cdot10^{-3}$			$5\cdot10^{-2}$			1	1

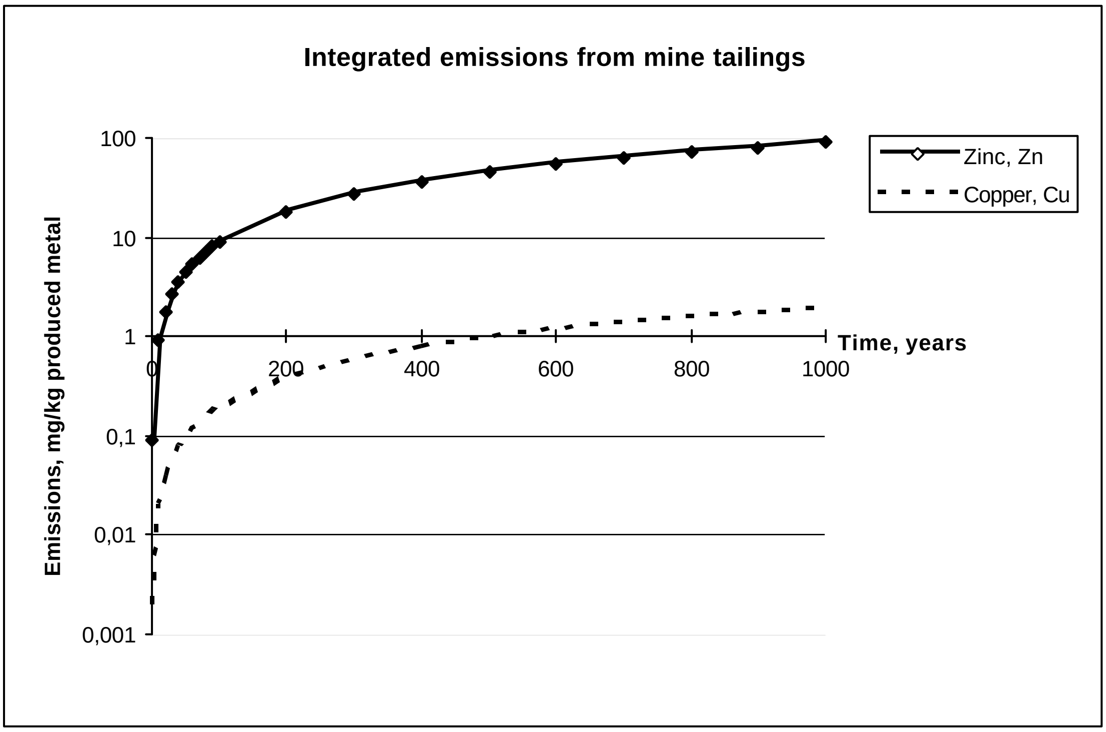

Figure 7.2 Cumulative emissions from mine tailings.

7.2.3 Discussion

Buffering period as measure for suveyable time period

The period of buffered conditions can be a possible limiting process for the surveyable period for mine tailings. The relevance of using the buffered period as a measure for the surveyable time may be questioned. For most metals, e.g. copper and zinc, buffered conditions would mean a very limited release, whereas, depletion of buffering minerals would result in increased emission rates. In this sense, the surveyable period would mark the end of the well-controlled period. Nevertheless, application of the buffered period complies with the proposed definition of the surveyable and infinite periods. Further discussion is encouraged on this topic.

The buffering capacity is in the first hand related to the presence of calcite in the tailings sand. A typical value of the calcite content is in the order of 0,1 - 5 %. For a 10 m high landfill with a solid density of 2900 kg/m^3, a porosity of 0,5 m^3/m^3, the calcite content is calculated to 0,145 kmol/m^2 surface area of the landfill. Assuming the simplified overall reactions:

$FeS_2 + 3,5\, O_2$ --> $FeSO_4 + SO_4^{2-} + 2\, H^+$
$CaCO_3 + 2\, H^+$ --> $Ca^{2+} + H_2CO_3$

We find a stoichiometric relation between supplied oxygen and consumed calcite of 3,5:1. With the estimated oxygen penetration rate of 0,5 mole/m^2,year, an estimated extension of the buffered period of 1015 years can be calculated. If the effects of acidic rain are considered the buffered period will be decreased. We may conclude that the surveyable period, based on an estimate of the extension of the buffered period, would be in the order of a few thousands of years.

General aspects on the applicability of the coupled chemistry and transport model

Landfilled mine-tailings, e.g. from iron ores and sulphidic ores, relate to metal production, which is relevant for most consumer products containing metal parts. Iron ores generally pose fewer problems. The iron oxides/hydroxides are quite stable and have an ability to retain other heavy metals by adsorption or coprecipitation. Sulphidic ore tailings may be more problematic since oxidation of the sulphide traces to sulphate will result in lowering of the pH and an increased solubility of different trace metals. The low pH of the leachate may also rapidly (in the order of tens to hundreds of years) deplete the buffering capacity of the tailings.

It should be noted that mining waste is not only tailings sand, but also waste rock, the volume of which may be the predominant waste type. Waste rock has not been considered in the present study but current research studies may give important information for future investigations (Eriksson N., licentiate treatise 1995; Strömberg and Banwart, 1994).

To estimate the emissions from landfilled mining wastes over longer periods of time, it must be assumed that the tailings are equipped with an efficient cover to minimise the release of heavy metals from the landfill. A cover design representative of current remediation projects in Sweden would imply a tight clayish till in combination with a protective layer of ordinary till. The way the covers are designed, the oxygen penetration is significantly reduced. By optimising the cover design so that the tight layer remains essentially water saturated for the major part of the year, both the oxygen penetration and the water percolation can be kept low.

The sulphidic tailings are usually quite inert during an initial phase. The oxidation of sulphides may require an induction period, possibly connected with the in-growth of bacteria that can catalyse the oxidation of sulphidic minerals (Ledin and Pederssen, AFR 1995). If the tailings contain lime, they may have an initial pH buffering capacity. The presence of lime in the material restricts the leachability of many heavy metals, possibly due to secondary precipitation of metal hydroxides or metal carbonates.

The rate of oxygen transport into the landfill determines the time schedule for the depletion of buffering components in the waste heap. Once acidic conditions are established throughout the landfill, increased metal concentrations can be expected. Depending on the content of buffering components, the acid production potential and the rate of oxygen transport into the landfill, the extension of the buffered period may be in the order of a few years to many hundreds of years.

Experience from other modelling work concerning landfilled mine tailings indicates that in many cases additional processes may play an important role in the leaching of heavy metals. Most important are probably kinetic effects, which may significantly lower the emission rates, at least when the availability of oxygen is high during the first period. Kinetic effects may significantly influence the general chemical conditions in the landfill, in that partly oxidising conditions may be created despite the sulphidic tailings. This may furthermore lead to secondary mineralisations in the landfill of oxidised mineral that may act as temporary deposits for weathered heavy metals. Under certain conditions such secondary mineralisations may be redissolved and release the metals after covering the tailing.

From the above it is apparent that the chemical processes in mine tailings are fairly complex and would certainly require significant investigations in order to derive the results desired for a LCA study.

7.3 Metal hydroxide wastes

7.3.1 General

As a basis for the present work it is assumed that the hydroxide sludges consist of metal hydroxides containing copper, zinc and nickel. This can be assumed representative for waste streams from mechanical industry or plating industry. Many other heavy metals and complexing agents, e.g. fluoride, cyanide, sulphate, etc., may also be present in different sludges, but have not been considered here.

Similar to coal ashes, various consolidation processes may initially take place in a sludge landfill. There is, however, one important difference between the two in that the metal hydroxide sludges have never been dried and dehydrated as happens with coal ashes. Consequently, the hydroxide sludges can be assumed to contain some capillary water and to be in a fully hydrated state when disposed.

To meet the requirements it must be assumed that some type of cover is applied over the sludge landfill. Therefore, a certain time is required to obtain pseudo stationary conditions in a landfill of this kind. Mathematical tools to treat this type of problem are available and operable, although not practical to set up in a life-cycle assessment study.

The use of tight cover materials, such as plastic liners, may significantly influence the onset of stationary conditions in the landfill. Facing the possibility that the longevity of plastic liners may be restricted (in the order of a hundred years), it must be assumed that the primary long-term effect of such construction elements in the landfill design will be to postpone the establishment of stationary conditions in the landfill.

The sludges may contain certain amounts of buffering materials, e.g. calcite, mainly added in the process to precipitate the sludges. This buffering may influence the leaching characteristics of the hydroxide sludges due to suppression of the solubilities during the early period. The extension of the buffered period can be estimated from the content of buffering components in the sludges. An increased solubility of metal hydroxides can be expected when buffering components have been depleted.

It is desirable that the surveyable period should include the above mentioned effects, i.e.:

- consolidation and possible subsidence,
- longevity of tight cover materials,
- establishment of stationary conditions,
- the choice of landfill design and treatment method, and
- impact of atmospheric oxygen and carbon dioxide,
- depletion of buffering components in the sludges.

However, for the purpose of the present introductory study, quite drastic simplifications need to be made in the model.

7.3.2 Emission from hydroxide sludges landfills

Using the same model as for coal ashes the emissions from a landfill for metal hydroxide sludges have been estimated. Two different types of hydroxide sludges have been investigated: hydroxide sludges from electroplating industry and pickling sludges from steel plate production. Different production quotients have been derived for pickling sludges from steel plate production and for electroplating industry. The production quotients have in both cases been defined as the amount of sludges formed by treatment of 1 m^2 of metal surface.

The production quotients have been estimated based on information obtained by interviews with about 15 different industries. Compiled results are presented in Table 7.4.

Table 7.4 Produced sludges per m^2 of treated metal surface. Compiled information obtained from interviews with 15 Swedish industries (Sundqvist et al, 1994).

	Production quotient *kg sludge/m^2 metal surface*	Solid content *% by weight*
Electroplating sludges		
Average	0,5	30 %
Max.	0,7	30 %
Min.	0,3	25 %
Steel plate pickling sludges		
Max.	2,6	45 %
Min.	0,3	60 %

Also for the metal hydroxide sludges the emissions have been exemplified by calculations for copper, nickel and zinc. It must be stressed that these metals are merely selected as examples to demonstrate the methodology. A full LCA study would certainly have to account for many other chemical substances, e.g. chromium, cadmium, lead etc.

The amount of water infiltrating the landfill has been varied using an infiltration rate of 0,125 m^3/m^2,year as a central case. The partitioning coefficient between percolating water and surface runoff through the soil cover has also been varied, using a factor of 0,5 as a central case.

The content of contaminants in the solid sludges has been assigned a fixed value estimated from compiled analyses of hydroxide sludges. The used values for the three studied metals are presented in Table 7.5.

The aqueous solubility has been varied for each of the three studied metals, adapting theoretically derived solubilities calculated by the chemical equilibrium program PHREEQE (Parkhurst 1980). The calculations include leaching in acidic rain for prolonged periods and are expected to give a reasonable estimate for the behaviour of the three studied metals. The more acidic conditions are expected to represent the behaviour of the hydroxide sludges after the initial period where the pH may be buffered by traces of lime from the processes. During the investigation the possible presence of substances such as chloride, ammonia and cyanide, but also different organic complexing agents, in the sludges were identified as important factors which may increase the solubility of some metals, but which may also be emitted themselves.

The importance of the initial phase, before the soil cover has been applied, and possibly before consolidation has reduced the hydraulic conductivity of the sludges, has not been investigated within the study.

In Table 7.5 the different calculations are summarised. The results are given as the integrated emissions of the individual metals over a period of 100 years. The extension of the surveyable period has not been possible to quantify within the study. A number of phenomena need to be implemented in the model to enable a justified quantification of the surveyable time period. As a first step, it has been decided to present the emissions over a fixed period of 100 years. Also the total emissions for the hypothetical infinite period are given. The results have been presented as the emissions related to the treatment of 1 m^2 metal surface. The results are also given in Figure 7.3, where the release is presented over an extended period.

Table 7.5 Compiled results for metal hydroxide sludges. Assumed sludge content: copper 10000 mg/kg, nickel 5000 mg/kg, zinc 5000 mg/kg

		Emission during surveyable time period (100 years) *kg/m^2 metal surface*	Emission during hypothetical infinite time *kg/m^2 metal surface*
Electroplating sludges			
Central case	Cu	$4{,}9 \cdot 10^{-7}$	$1{,}5 \cdot 10^{-3}$
	Ni	$4{,}8 \cdot 10^{-7}$	$7{,}5 \cdot 10^{-4}$
	Zn	$7{,}0 \cdot 10^{-7}$	$7{,}5 \cdot 10^{-4}$
Maximum case*	Cu	$9{,}6 \cdot 10^{-7}$	$1{,}5 \cdot 10^{-3}$
	Ni	$9{,}5 \cdot 10^{-7}$	$7{,}5 \cdot 10^{-4}$
	Zn	$1{,}4 \cdot 10^{-6}$	$7{,}5 \cdot 10^{-4}$
Minimum case**	Cu	$7{,}3 \cdot 10^{-8}$	$1{,}5 \cdot 10^{-4}$
	Ni	$7{,}3 \cdot 10^{-8}$	$7{,}5 \cdot 10^{-4}$
	Zn	$2{,}1 \cdot 10^{-7}$	$7{,}5 \cdot 10^{-4}$
Steel plate pickling sludge			
Central case	Cu	$3{,}9 \cdot 10^{-6}$	$1{,}2 \cdot 10^{-2}$
	Ni	$3{,}7 \cdot 10^{-6}$	$5{,}9 \cdot 10^{-3}$
	Zn	$5{,}5 \cdot 10^{-6}$	$5{,}9 \cdot 10^{-3}$
Maximum case*	Cu	$7{,}5 \cdot 10^{-6}$	$1{,}2 \cdot 10^{-2}$
	Ni	$7{,}4 \cdot 10^{-6}$	$5{,}9 \cdot 10^{-3}$
	Zn	$1{,}1 \cdot 10^{-5}$	$5{,}9 \cdot 10^{-3}$
Minimum case**	Cu	$5{,}7 \cdot 10^{-7}$	$1{,}2 \cdot 10^{-2}$
	Ni	$5{,}7 \cdot 10^{-7}$	$5{,}9 \cdot 10^{-3}$
	Zn	$1{,}6 \cdot 10^{-6}$	$5{,}9 \cdot 10^{-3}$

Remarks:

* Maximum case: high solubility and high infiltration

** Minimum case: low solubility and low infiltration

The emission factors, kg leached metal per kg of landfilled metal in sludge are according to the Table 7.6.

Table 7.6. Emission factors, kg leached metal per kg of landfilled metal in sludge, for metal hydroxide sludges.

	Emission factor during surveyable time period		Emission factor during hypothetical infinite time period	
	kg leached metal per kg of landfilled metal in sludge		*kg leached metal per kg of landfilled metal in sludge*	
Metal	Electroplating sludges	Steel plate pickling sludges	Electroplating sludges	Steel plate pickling sludges
Cu	$3 \cdot 10^{-4}$	$3 \cdot 10^{-4}$	1	1
Ni	$6 \cdot 10^{-4}$	$6 \cdot 10^{-4}$	1	1
Zn	$9 \cdot 10^{-4}$	$9 \cdot 10^{-4}$	1	1

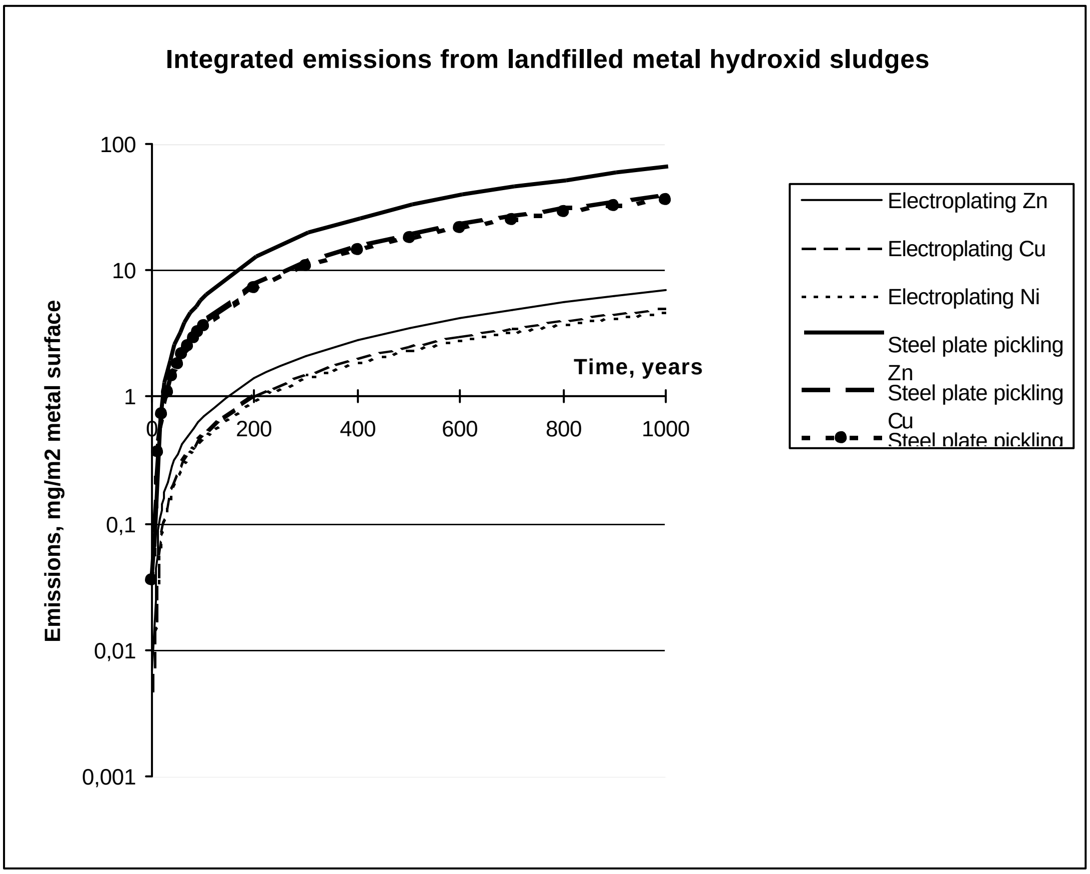

Figure 7.3. Cumulative emissions from a landfill for metal hydroxide sludges.

7.3.3 Discussion

The metal hydroxide sludges may initially contain substantial amounts of other salts than metal hydroxides, e.g. sulphate, chloride, fluoride, etc. Soluble salts such as chlorides can in a first attempt be considered soluble and be transported without retention by percolating water. The water residence time in the landfill can be estimated to some 20-50 years. Consequently, the contained chlorides may be released during the same period. Sulphate may form precipitates such as gypsum but may also be retained by adsorption onto metal hydroxides, mainly ferric hydroxides. As an example, we may assume sulphate to be adsorbed onto ferric hydroxide corresponding to a distribution coefficient of 0,01 m^3/kg. Assuming the sludges to contain 5% of $Fe(OH)_3$, the porosity to be 0,3 m^3/m^3, the density to be 2800 kg/m^3, the retention factor, R, can be calculated to: $R = 1+ (1-0,3) / 0,3 \cdot 0,05 \cdot 0,01 \cdot 2800 = 4,3$

Thus, if the water residence time is 50 years, the desorption of sulphate will be completed after approximately 215 years.

The presence of redox sensitive elements in the sludges requires a special treatment and is suggested as a continuation of the present study. To demonstrate the importance of redox sensitive components of the sludges we may consider the presence of an oxidizable metal present in the sludges at a concentration of 1000 mg/kg, or roughly 0,3 kmol/m^2 of surface area of landfill, which is exposed to oxidation by atmospheric oxygen leaking into the landfill. Assuming the average value of 0,5 mol/m^2,year (as in the case of mine tailings) as an estimate for the oxygen that enters the landfill, the time for oxidation of the metal is calculated to 300 years.

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8. Some examples

In this Chapter some examples are given, where some of the results from this report have been used to examplify the use of the data given in the report. The examples are only for illustrative use, and too far conclusions should not be drawn. But some examples may lead to some reflections.

Example 1. The importance of the waste stage compared to the other stages in the life cycle

These examples were also presented in the report from the first stage (Sundqvist et al, 1994). Results from some LCA studies for plastic and corrugated cardboard are summarised in Tables 8.1.a and 8.1.b. The emissions from raw material acquisition, manufacture and distribution are accounted are from the referred LCA studies, and these emissions are compared with the emission from landfilling, calculated by our method.

It should be noticed that the referred studies have different system borders, and are based on different methods, which make them difficult to compare. However, the comparisons clearly show that the emissions from landfilling of cardboard and plastic are of importance in the total life cycle.

It can be noted that the potential emissions of methane (CH_4) from landfilling during the surveyable time-period will be approximately 500 times higher than all the other emission sources together according to the study from the Tellus Institute. The potential emissions of methane are also higher than the total amount of hydrocarbons (CH) according to the studies by Habersatter and Tillman et al. Thus, even if the estimated degradation of PE is uncertain, it can be concluded that landfilling is of importance compared to other stages in the life-cycle concerning emissions of methane. Landfilling may also be of significance concerning total emissions of hydrocarbons and volatile organic compounds during the surveyable time-perspective.

Table 8.1.a LDPE plastic: Comparison between emissions during the life-cycle except landfilling potential emissions from landfilling.

	Emissions from life cycle except landfilling			Emissions from landfilling (surveyable time period)
	Study A	Study B	Study C	Our results
	g/kg LDPE	*g/kg LDPE*	*g/kg LDPE*	*g/kg LDPE*
CO_2, carbon dioxide	-	1023	-	24
CH, hydrocarbons (inclusive, CH_4)	8,8	11		>26
VOC, volatile organic compounds (incl. CH)	-	-	27	>26
CH_4, methane	-	-	0,043	26
COD (chemical oxygen demand)	0	-	13	0,77

Remarks.: Study A: Habersatter, (1991)
Study B: Tillman et al (1991)
Study C: Tellus Institute (1992)

Table 8.1.b Corrugated cardboard: Comparison between emissions during the life-cycle except landfilling potential emissions from landfilling.

	Emissions from life cycle except landfilling		Emissions from landfilling, (surveyable time period) our results	
	Study A	Study B	without gas recovery	with gas recovery
	g/kg	*g/kg*	*g/kg*	*g/kg*
CO_2, carbon monoxide	-	310	0^*	0^*
CH_4, methane	-	-	130	67
CH, hydrocarbons (incl. CH_4)	3,5	0,18	>130	>67

Remarks.:* Carbon dioxide from renewable energy sources (biomass) is not included.
Study A: Habersatter (1991)
Study B: Tillman et al, (1991)

Example 2. What happens with 1 kg of food wastes at landfilling and at incineration.

The food waste is assumed to consist of 50% dry material and 50% moisture. The composition of the dry material is 24% fat, 13% protein, 18% degradable carbohydrates, 21% cellulose and 6% “nondegradable” organic material. The chlorine content is 0,4% of the dry material. The contents of eventual heavy metals are negligible.

Landfilling in a MSW landfill gives emissions according to Table 8.2.a. Emissions from incineration are given in Table 8.2.b. (Note: There should not be any conclusions drawn from this of which method that is preferable).

Table 8.2.a. Emissions from surveyable time period at landfilling of food wastes.

		Emission from conventional MSW landfilling during surveyable time period		Emissions from biofilling during surveyable time period
		with gas recovery*)	without gas recovery	with effective gas recovery**)
Emissions to air				
Methane, CH_4	*g/kg raw waste*	67	134	15
Ammonia, NH_3	*g/kg raw waste*	~0	~0	~0
Hydrogen sulphide, H_2S	*g/kg raw waste*	~0	~0	~0
Emissions to water (exclusive leachate treatment)				
Biological oxygen demand, BOD	*g/kg raw waste*	1,4	1,4	1,4
Chemical oxygen demand, COD	*g/kg raw waste*	5,6	5,6	5,6
Ammonia/ammonium NH_3/NH_4^+	*g/kg raw waste*	0,6	0,6	0,6
Recovered energy	*MJ/kg raw waste*	3,8	0	5,7

Remarks:

*) 50% of produced methane are collected. 10% of methane through soil cover are oxidised.

**) 75% of produced methane are collected. 60 % of methane through soil cover are oxidised.

Table 8.2.b Emissions from incineration of food wastes (emission factors according to Table 4.2)

		Emission to air during incineration
Emissions to air		
Sulphur dioxide, SO_2	*g/kg raw waste*	0,16
Hydrochloric acid, HCl	*g/kg raw waste*	0,08
Carbon monoxide, CO	*g/kg raw waste*	1,2
Polyaromatic hydrocarbons, PAH	*g/kg raw waste*	$7 \cdot 10^{-6}$
Nitrogen oxides, NO_x	*g/kg raw waste*	1,3
Dust	*g/kg raw waste*	0,008
Recovered energy	*MJ/kg raw waste*	*7,2*

Example 3. Comparison between incineration and landfilling of metal containing wastes.

Domestic metal wastes can be disposed by incineration or by landfilling. Here is a comparison between the two methods. It can be observed that incineration gives more metal emissions during surveyable time period than landfilling.

Table 8.3. Comparison between incineration and conventional landfilling of metal wastes. Calculation base is 1 kg of each metal.

Metal	Conventional MSW landfilling	Incineration plus landfilling of ashes		
	Emitted during surveyable time period *g per kg of disposed metal*	Emission at incineration *g per kg of disposed metal*	Emission from landfill (surveyable time period)*) *g per kg of disposed metal*	**Sum** *g per kg of disposed metal*
Cadmium, Cd	**0,5**	5	6	**11**
Chromium, Cr	**0,7**	2	0,7	**2,7**
Copper, Cu	**0,07**	1	4	**5**
Lead, Pb	**0,06**	2	0,02	**2**
Zinc, Zn	**0,2**	2	0,01	**2**

Remark:

*) Landfilled oxidised ashes buffered by calcite

Example 4. Wastes that are associated with a copper tube

1 kg of a copper tube has at the raw material acquisition stage caused a waste of 110 kg mine tailings. After several years of use the copper tube is discarded in the MSW and landfilled in a conventional MSW landfill. The copper emissions from the mine tailings resp. from the MSW is according to Table 8.4. It can be observed that during the surveyable time period the waste from the mine tailings dominates over the MSW. However, in the infinite perspective the emissions from the MSW are dominating.

Table 8.4. Emissions from wastes associated with 1 kg copper tube.

		Emission of Cu during surveyable time period	Emission of Cu during hypothetical infinite time period
Mine tailings	*g Cu/kg copper tube*	0,2	70
Other waste from production	*g Cu/kg copper tube*	~0	~0
MSW landfill	*g Cu/kg copper tube*	0,07	1000
SUM	***g Cu/kg copper tube***	**0,27**	**1070**

Example 5. Wastes associated with a galvanised plate

A galvanised plate consists of an steel plate where both sides are plated with zinc. The amount of zinc is cirka 70 g/m^2 (on each side). The emissions from landfilling of wastes from production and final (MSW) disposal of the galvanised plate is shown in Table 8.5. As can be noticed, the emissions from the final MSW disposal are dominating, while the emissions from landfilling of mine tailings are of some importance. The importance of emissions from landfilling of metal hydroxide sludges is very small.

Table 8.5. Emissions from wastes associated with a galvanised steel plate

		Emission of Zn during surveyable time period	Emission of Zn during hypothetical, infinite time period
Mine tailings	*g Zn/m^2 plate*	1,3	56
Metal hydroxide sludges (electroplating)	*g Zn/m^2 plate*	0,0014	1,5
Final disposal (MSW)	*g Zn/m^2 plate*	2,8	140
SUM	***g Zn/m^2 plate***	**4,1**	**197,5**

Example 6. Comparisons of emission factors for metals in different of wastes and landfills

In this report are emission factors presented for different wastes and different landfills. They have been summarised in Table 8.6.

As can be seen, the emission factors can vary a lot for the same metal in different kind of wastes. One common characteristic is that the emissions during the surveyable time period is relatively low. The geometric average is about $9 \cdot 10^{-4}$ kg/kg.

Table 8.6 Emission factors for metals in different wastes and landfills

Metal	Emission factor, kg emitted per kg of landfilled metal during surveyable time period						
	Municipal Solid Waste, mixed landfill *kg/kg*	MSW residual fraction, cell deposit *kg/kg*	Incinera-tion ashes, oxidsed *kg/kg*	Incinera-tion ashes, reduced *kg/kg*	Coal ashes *kg/kg*	Metal hydroxid sludges *kg/kg*	Mine tailings *kg/kg*
Arsenic, As	$2 \cdot 10^{-3}$						
Cadmium, Cd	$5 \cdot 10^{-4}$	10^{-1}-1	$6 \cdot 10^{-3}$	$1 \cdot 10^{-2}$			
Chromium, Cr	$7 \cdot 10^{-4}$		$7 \cdot 10^{-4}$	$1 \cdot 10^{-6}$	$2 \cdot 10^{-3}$		
Copper, Cu	$7 \cdot 10^{-5}$	10^{-12}-10^{-11}	$4 \cdot 10^{-3}$	$1 \cdot 10^{-1}$	$1 \cdot 10^{-4}$	$3 \cdot 10^{-4}$	$5 \cdot 10^{-3}$
Lead, Pb	$6 \cdot 10^{-5}$	10^{-8}	$2 \cdot 10^{-5}$	$4 \cdot 10^{-5}$	$1 \cdot 10^{-4}$		
Mercury, Hg	$1 \cdot 10^{-4}$	10^{-5}-10^{-4}			$5 \cdot 10^{-2}$		
Nickel, Ni	$5 \cdot 10^{-3}$				$1 \cdot 10^{-4}$	$6 \cdot 10^{-4}$	
Zinc, Zn	$2 \cdot 10^{-4}$	10^{-2}	$1 \cdot 10^{-5}$	$4 \cdot 10^{-4}$	$5 \cdot 10^{-4}$	$9 \cdot 10^{-4}$	$5 \cdot 10^{-2}$

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9. Some concluding remarks

As mentioned in Chapters 2 and 3, the major problems when handling waste disposal in connection with LCA are time aspects in landfilling and allocation of emissions in treatment processes. In this report landfilling of both municipal solid waste (MSW) and different industrial production wastes have been studied. Also incineration of MSW has been studied, with consideration of landfilling of the ashes. Different allocation methods and different approaches for handling the time aspects have been studied.

In Chapter 4 different allocation methods were compared. It was found that different allocation principles gave different results. But as long as the allocation principle is fairly relevant, the results differ often by a factor less than $2^{\pm 1}$. According to LCA practice the normal uncertainty is one order of magnitude for most emissions, and differences in emissions less than one order of magnitude should not be regarded as significant (Lindfors et al, 1995). There are large uncertainties in the data available about incineration emissions. Different data sources can give larger differences than different allocation methods. Thus, too much work should not be done on elaborating allocation models. It is better to choose a reasonable allocation model and concentrate the work upon collecting better data or upon interpretation of the result.

In Chapter 4.4 and in Chapter 5.5 the time aspects in landfilling were discussed, and different approaches were compared. It was found that different short-term approaches do not differ significantly from each other. Often the difference is less than a factor $2^{\pm 1}$, while the uncertainties in the used data may be a factor $10^{\pm 1}$. Thus, it is not worth to much effort to elaborate landfill models. It is better to choose a reasonable time model and concentrate the work upon collecting better data or upon interpretation of the result.

The long-term aspects can differ a lot depending on the time horizons. For Scandinavian conditions the next glacial period is of interest as an upper time limit. The next glacial period is expected to occur 20 000 to 50 000 years from now. In this aspect all other time horizons beyond this are irrelevant. When the glacial period has come all landfilled material will be released to the environment anyhow.

All this long-term aspects should also be considered in the historic perspective. When we are talking about, for example, a 1 million years perspective for a landfill, we also should consider that the human being has existed only in 100 000 years. Sweden has been populated for 8000 years. North America was discovered by the white man only 500 years ago. The industrialised era began just about 200 years long.. Even if we in our model consider the future as foreseeble, we have to realise that a lot of unforeseeble things can happen in the longer perspectives.

One important uncertainty, when considering the time aspects, is that the emission rate (mainly leaching) must be modelled. The kinetics of the landfill must be known, as well as the processes in the landfill. The longer times horizons that are considered, the larger is the uncertainties in the models. However, our approach with the surveyable time period, and hypothetical, infinite time period is rate-independent. We have to know the processes in the landfill for identifying the surveyable time, but we do not have to know the rate of the processes. The hypothetical, infinite time is also time-independent. It is simply a worst-case for a scenario where all landfilled materials have been released to the environment. However it can be discussed how the different components are released to the environment, but this is more a question of knowing the processes, than of knowing the rate of the processes. Thus, our approach with the surveyable time period and the hypothetical time period seems to be the easiest models to handle.

One weakness with our approach is that the emissions of metals during the surveyable time period are relatively low, often 10^{-3} - 10^{-6} kg/kg, while the emissions during the hypothetical infinite time are 1 kg/kg (by definition). It should be helpful to have some intermediate time period somewhere in between these two periods. A development of the critical time period, as discussed in our second report (Sundqvist et al, 1997) should be useful. There the critical time period was defined as the period until the major part of the emissions has occurred. Normally the critical time approach is not useful in a screening LCA, but can be useful when interpretting the inventory or the charakterisation pahses of a deepened LCA. However, the critical time approach needs more development to be operative.

Concerning the surveyable time period, it can be seen that there is a distinction between organic landfills (MSW landfills) and monofills (ash landfills, mine tailings, etc.). The MSW landfill has a relatively short surveyable time period, governed by the degradation of organic material. The length of the surveyable time period seems to be 25 - 100 years. In the monofills the reactions are slower, and the length of the surveyable time period is longer, perhaps several hundred years. In a study where both organic landfills and monofills are concerned, it could be more practical to use the same real time. As indicated in the discussions in Chapters 4.4 and 5 there is only small differences between different short-term periods - the uncertainties in the data and in the models are larger than the difference between different time approaches.

It should also be admitted that there have been difficulties with the surveyable time period approach. The criterion for the period is that a kind of pseudo-steady-state should be obtained and that the period should be of the magnitude one centuy.. For organic landfills does the surveyable time approach work well. There is a noticable pseudo-steady-state after 50 - 100 years. For monofills (incineration ashes, coal ashes, mine tailings, metal hydroxid sludges) there were difficulties to identify the surveyable time period. The searched steady-state period seems to occur after several hundred or thousands years. For incineration ashes we noticed a very slightly pseudo-pseudo-steady-state in the beginning (200 years), while a more stable steady-state occurred after perhaps 1000 years. Also the mine tailings indicated a better steady-state after about thousand year. For the difficult wastes we made the projection that the period was 100 or 200 years. The criteria ”the magnitude of one century” seems to be more important than the ”pseudo-steady-state” criterion, and has been given priority in our studies.

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Appendix A. Emissions from landfill fires for use in LCA.

Review

This Appendix is a development of the Section about ”Tip fires” in our second report (Sundqvist et al, 1997). Parts of the text are copied from the report, but the data presented here is updated and developed.

Spontaneous fires on the landfills (”tip fires”) occur by accident on most landfills. There are usually two kinds of fires: ***surface fire***s where material is burning on surface of the landfill, and ***deep fires*** where material down in the landfill body is burning.

The fires may be initiated in the following ways (Bergström and Björner, 1994):

- ash and slag from incineration with hot, unextinguished spots
- chemical and biological reactions that induce combustible gases, combined with exothermic reactions that rises the temperature above the autoignition temperature.
- catalytic reaction, e.g. between bark ash and lignin
- some gases, e.g. hydrogen sulphide can be ignited in contact with rust and other metal oxides.

Frequency of landfill fires

Bergström and Björner (1994) made an inquiry to a majority of the Swedish landfills. In the inquiry they asked both the landfill operating companies and the municipal fire departments of experience from landfill fires. The inquiry was made for two years 1988 - 89, and data from 270 landfills were gathered. The conclusions they draw from the study are:

- During the two years there were 384 fires on 127 landfills. (More fires were reported by the fire department than by the landfill operators).
- Several of the fires were possible to extinguish within a few hours. The average extinguishing time for surface fires was 9 hours and for deep fires 4,5 days.
- 23 % of the fires were classified as deep fires.
- The primary causes to the fires were according to Table A.1.

Table A.1 Primary causes to landfill fires

Answer in inquiry	%
Fire source unknown	65
Incendiary	7
Glowing material in waste	17
Highly inflammable material	5
Biological process	7

Wastes and materials that were considered to be connected with fire hazards were ashes, tires, grain, corn, cereals, straw

- Only a few of the landfills with landfill gas extraction think that the fire frequency has decreased since the gas extraction plant was taken into operation.
- The types of waste that were on fire are according to Table A.2.

Table A.2 Fires in different types if waste

Type of waste	Number of fires %
Domestic waste	10
Industrial waste	32
Construction and demolition wastes	25
Bulky wastes from households	22
Other wastes	11

- The age of the waste that was on fire was in 47 % of the cases less than 2 weeks and in 32 % of the cases between 2 and 26 weeks.
- 75 % of the fires were on, or close to, the landfill slope, 21 % on a central place on the landfill and 4 % on other places.

They also estimated that there are around 250 tip fires per year [5] in Sweden, and about 25 000 ton of waste material are burned in the fires.

In a recent Finnish study (Ettala et al, 1996) a survey of landfill fires was done in Finland. On average, there were 633 sanitary landfills in operation 1990 - 92 in Finland. Annually, 380 landfills occurred, one-quarter of which were deep fires. The total amount of waste burnt in landfill fires were estimated to be 84 000 tons/year. The Finnish study does not present information about landfilled quantities, but roughly

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[5] There are about 300 municipal solid waste landfills in operation in Sweden.

estimated, the landfilled waste in Finland does not exceed the Swedish landfilled waste. As seen, the results from Finland and Sweden are very similar to the results. The Finnish study possibly indicates that the estimate of 25 000 burnt wastes in Sweden may be an underestimate.

Wastes burnt in landfill fires

At Swedish municipal solid waste landfill about 3,5 Mton/year of households and industrial wastes are landfilled. A quantity of 25 000 ton/year uncontrolled burned waste corresponds to about 0,7% of the landfilled quantity. Also to take into account is that different wastes have different affinity to fires. Industrial wastes and construction and demolition wastes are more fire hazardous than household waste. If the portion 25 000 ton is apportioned according to the fire frequencies, the total amounts of burned wastes are according to Table A.3.

Table A.3. Calculation of the portion of different wastes that are burned in landfill fires.

Waste category	Estimated quantity to landfill *Mton/year*	Number of fires %	Quantity burned *ton/year*	Part of resp waste category that is burned %
Domestic waste	1.1	10	2 500	0.2
Bulky domestic waste	0.1	22	5 500	4
Industrial waste	1.0	32	8 000	0,8
Construction waste	1.0	25	6 250	0,6
Others	0.3	11	2 750	5
TOTAL	**3.5**	**100**	**25000**	**~0.7 %**

If the discussion is restricted to domestic waste (post-consumer waste), and the potential contribution from different products in the post-consumer waste (it is understood that we make an product LCA), it is to expect that about 0,2 % of the ordinary household waste and 4 % of the bulky waste will be subject to a fire.

Another discussion question is allocation. Above, when we compared the landfill fire affinity of different wastes, we simply allocated by total weight. However, when applying on a special product that are studied in a LCA, it seems more appropriate to allocate by the content of organic material, combustible material or total carbon. Allocation by organic content is assumed in the following. Other allocation principles could be used, but here we have preferred allocation by organic content for simplicity.

Domestic wastes usually have a moisture content of 30 - 35 % and an inorganic (”ash”) content of about 20 - 25 % (Sundqvist 1995). This gives an organic portion of 40 - 45 % of the total weight. Bulky wastes usually have some lower contents of moisture but higher contents of inorganics. The organic portion of bulky waste can also be assumed to be 40 - 45 %. The ”average waste”, for which the production of contaminants has been studied, see Table A.3., can also be assumed to have around 40 - 45 % dry organic contents. For a LCA, we can calculate that 1 kg of dry organic material in a product will give production of pollutants according to Table A.5:

Formation of contaminants in landfill fires

Emissions from landfill fires have been discussed by Bergström and Björner (1994). Testes with fires burnt in containers showed that there was produced 1700 Nm^3 fire gas for every ton waste that were burned. The fire gases were analysed. Considerably amounts were found of tars (PAH) and other contaminants, see Table A.4.

In a recent study by Pettersson et al (1996) landfill fires were studied ”on-site”. Both surface fires on the landfill and deep fires were measured. The surface fires were obtained by setting fire on a heap of fresh waste. The amount of formed emittants is presented in Table A.4.

Table A.4. Emissions from landfill fires

	Formed amount of contaminants per ton burnt waste		
Compound	Deep fire (Pettersson et al, 1996)	Surface fire (Pettersson et al, 1996)	Simulated surface fire in container (Bergström and Björner, 1994)
Chlorobenzens	0,5 - 1,5 g	0,1 - 1,5 g	4 g
Dioxins (TCDD-eq.)	3 - 8 µg	50 - 900 µg	1200 µg
PAH	-	1,2 - 26 g	~250 g
PCB	1 - 4 mg	10 - 40 mg	0,3 mg
Hg	9 mg	0,1 - 0,4 g	-

As seen in the Table, there are large amounts of contaminants formed during the fires. However, it should be noted that a lot of the formed contaminants will be imitted on the surface of the landfill, and will not be released to the environment. The formed contaminants will to a large part remain on the landfill. In that aspect the landfill fire may be regarded as a parallel degradation pathway, where a part of the organic fraction will be transformed to e.g. PAH, PCB and TCDD (dioxin), which will further be subject to degradation, evaporation, leachage, etc., see discussion about specific organic compounds in Appendix B.

From Tables A.3 and A.4 and the information presented above emission factors for landfill fires can be calculated. We have chosen to allocate the emissions to the organic material (combustible material) since that is the portion that is burned.

Table A.5. Formation of potential contaminants in landfill fire

	Formation of contaminants per kg of landfilled dry organic material					
	Ordinary domestic waste *kg/kg*	Bulky domestic waste *kg/kg*	Industrial waste *kg/kg*	Construction and demolition wastes *kg/kg*	Other wastes *kgIkg*	Average *kg/kg*
Chlorobenzens	$4 \cdot 10^{-9}$	$8 \cdot 10^{-8}$	$1,6 \cdot 10^{-8}$	$1,2 \cdot 10^{-8}$	$1,0 \cdot 10^{-9}$	$1,4 \cdot 10^{-8}$
Dioxins (TCDD-eq.)	$1,8 \cdot 10^{-12}$	$3,6 \cdot 10^{-11}$	$7,3 \cdot 10^{-12}$	$5,5 \cdot 10^{-12}$	$4,5 \cdot 10^{-11}$	$6,4 \cdot 10^{-12}$
PAH	$5,4 \cdot 10^{-8}$	$1 \cdot 10^{-6}$	$2,2 \cdot 10^{-7}$	$1,6 \cdot 10^{-7}$	$1,4 \cdot 10^{-6}$	$1,9 \cdot 10^{-7}$
PCB	$9,3 \cdot 10^{-11}$	$1,8 \cdot 10^{-9}$	$3,7 \cdot 10^{-10}$	$2,8 \cdot 10^{-10}$	$2,33 \cdot 10^{-9}$	$3,3 \cdot 10^{-10}$
Hg	$9,2 \cdot 10^{-10}$	$1,8 \cdot 10^{-8}$	$3,7 \cdot 10^{-9}$	$2,75 \cdot 10^{-9}$	$2,3 \cdot 10^{-8}$	$3,2 \cdot 10^{-9}$

It should be noticed that mercury has been treated as a ”process-related emission”, formed by the process ”landfill fire”. This is because the mercury emission would not have occurred if the organic material was nor burnt.

The Table A.5 presents the amounts of different contaminants that can be formed during landfill fires. However, note that these figures do not show the emissions - the larger parts of the pollutants are supposed to be imitted on the landfill, thus not leaving the system border.

The estimated amounts were based on the assumption that there are around 250 landfill fires per year in Sweden, where about 25 000 ton of waste material are burned. Bulky wastes, construction and demolition wastes, and industrial wastes seem to have larger affinity to landfill fires than domestic waste. The landfill fires are allocated to the dry organic content of the waste. For a LCA, we can calculate that 1 kg of dry organic material in a product will give formation of contaminants according to Table A.5.

Beside these contaminants, there will also be formation of several other contaminants, e.g. sulphur dioxide, nitrogen oxides, hydrochloric acid, heavy metal dusts, etc. Those landfill fire emissions may be calculated according to the Chapter 4, and there be regarded as product-related emissions and assuming no cleaning of flue gas (in this case fire gas).

Emissions from landfill fires

It has been pointed out above that there are relatively large amounts of wastes that are burnt in landfill fires, and that relatively large amounts of emittants are formed. However, not all of the formed emittants will be released to the environment, but will be imitted on the surface of the landfill.

The most important emittants from the fire (PAH, PCB, dioxin) are expected to be bounded to airborne particles, e.g. soot particles. It is likely that that these particles will fall down to the ground.
We have made some simplified calculations of dispersion of the smoke plume from the fire by the Gauss dispersion model (Frennemo, 1986):

$$c(x,y,z) = \frac{Q}{2\pi \cdot \sigma_y \cdot \sigma_z \cdot u} \cdot e^{-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2} \cdot \left\{ e^{-\frac{1}{2}\left(\frac{z-h}{\sigma_z}\right)^2} + e^{-\frac{1}{2}\left(\frac{z+h}{\sigma_z}\right)^2} \right\}$$

$$\sigma_y = 0{,}32 \cdot x^{0{,}78} \text{(at neutral atmospheric stability)}$$

$$\sigma_z = 0{,}22 \cdot x^{0{,}78} \text{(at neutral atmospheric stability)}$$

(Equation A.1)

where

$c(x,y,z)$ = concentration on the co-ordinates (x,y,z), *mg/m*^{3}
x = distance in the wind direction, *m*
y = horizontal distance from the x-axis, *m*
z = height distance from the ground level, *m*
Q = the flow of emittant, *mg/s*
σ_y = parameter that shows the horizontal dispersion (y-direction)
σ_z = parameter that shows the vertical dispersion (z-direction)
h = height of plume at the release point, *m*
u = wind speed, *m/s*

In the calculations we have assumed a fictive contaminant flow Q=1 at the outlet. The wind velocity has been set to 2 m/s, and the height of the plume at the release point (the fire) has been assumed to be 10 m. The results are given in Figures A.1 and A.2, where the concentration in the plume in the wind direction is given.

For interpretation of the result we have also assumed that the distance from the fire to the border of the landfill is 100 m. If the fire occurs in the central part of the landfill this corresponds to a landfill area of about 30 000 - 40 000 m^2.

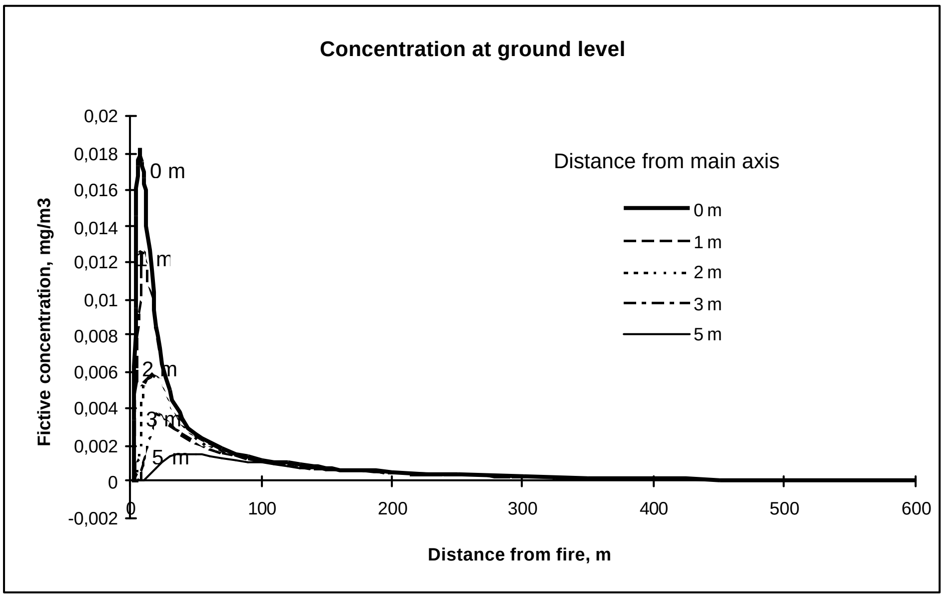


Figure A.1 Calculation of concentration in smoke plume at ground level

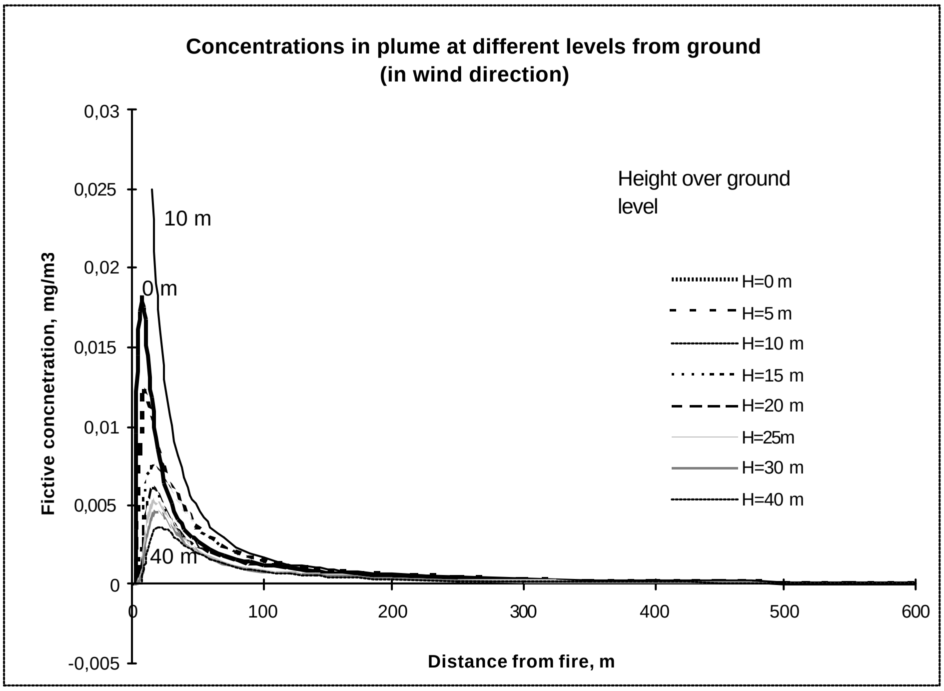


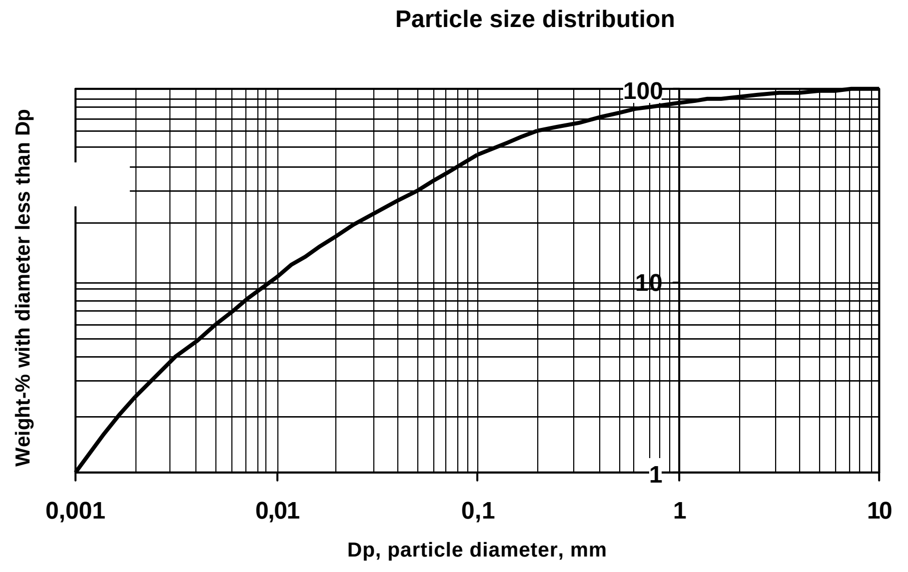
Figure A.2 Calculation of concentration in smoke plume in wind direction at different height levels.

Another alternative way to roughly estimate the imission within the borders of the landfill is to study the falling velocities of the particles in the smoke, which most of the PAH, PCB, dioxins, etc. are bound to. A simple model can be described as the following. The smoke from the fire reaches the height of 10 m above the fire. The smoke is than transported by the wind in horizontal direction. The particles fall down, and their velocity is depending on the particle diameter (see Equation 2 below). Some of the particles have enough vertical velocity to reach the ground level, while other will have their fall-down outside the border of the landfill. We have assumed a particle size distribution like a typical waste incinerator fly ash particle size distribution (ENA, 1986), see Figure A.3. The falling velocity is calculated by the equation (Perry et al, 1984):

$$w = (r_p - r_l) \cdot g \cdot D_p^2 / (18 \cdot h) \qquad \textit{(Equation 5.3)}$$

where

r_p = particle density, *kg/m³*, (assumed to be 1000 kg/m³)
r_l = air density, *kg/m³*, (1,29 kg/m³)
D_p = particle diameter, *m*
h = viscosity, Ns/m²



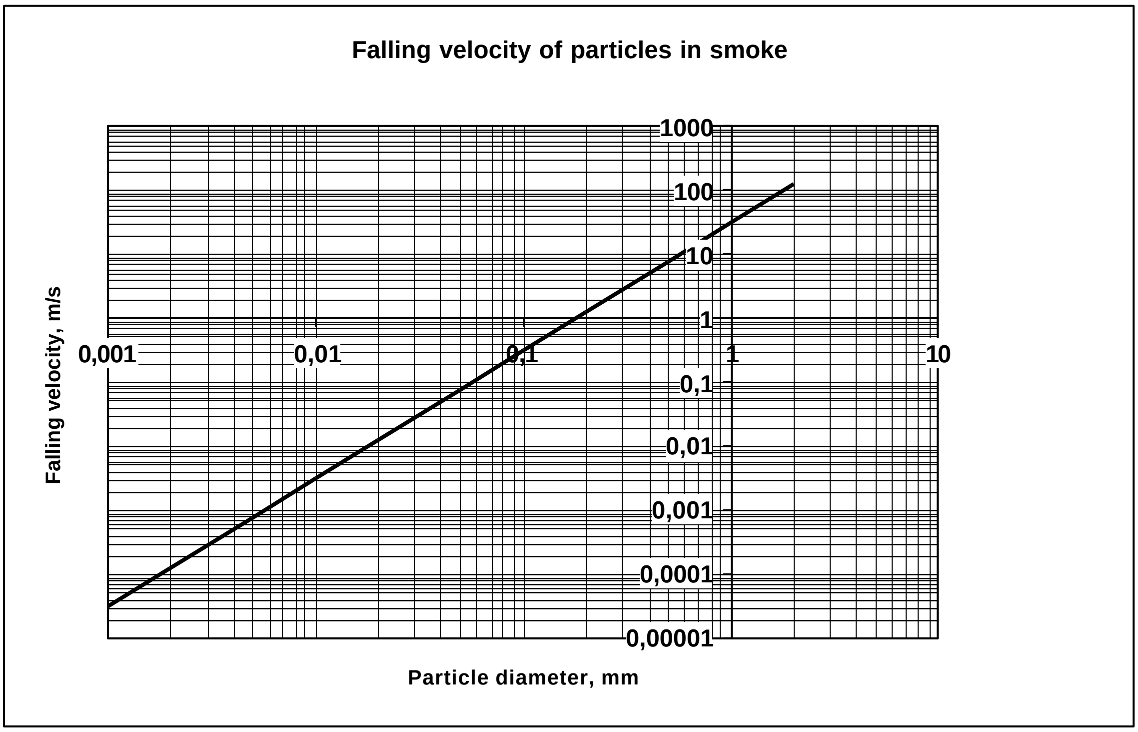


Figure A.3. Assumed particle size distribution and calculated falling velocities for particles in smoke from landfill fire.

The same case as above (wind speed 2 m/s, fire 100 m from landfill border) gives a residence time within the landfill borders of 50 s . About 60 weight-% of the particles have falling velocities enough to fall 10 m or more during these 50 s. As seen by Figure A.3, the contaminant concentrations are large at lower height levels relatively close to the fire. Still another 10 weight-% of the particles have a falling velocity enough to fall between 5 and 10 m during the smoke transport over the landfill. Further, as seen by the Figures A.1 and A.2 a large part of the smoke will have a fall-down relatively close to the fire. The maximum concentration in the air at the ground level (z = 0) is at about 10 m distance from the fire. The dispersion of the smoke will probably give larger imissions than estimated from the falling densities. As a very preliminary estimate it is assumed that about 60 - 90 % of the formed emissions have a fall-down within the borders of the landfill. **Only 10 - 40 % of the quantities of formed contaminants should be counted as emissions, when calculating the emissions from landfill fires in a LCA.** (However, this estimate is very uncertain).

Another aspect that must be taken into account is that landfill fires occur accidentally, but the generation of the fires are possible to control within some limits. Ettala et al (1996) reported that they found a statistical dependence between landfilling technology and fire frequency. Insufficient covering and compacting, ash disposal and deliberate fire starting were reported to be the most typical reasons for waste ignition.

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Appendix B. Emissions of specific organic compounds from MSW landfills

Emissions of specific organic compounds

Organic compounds are used in several products that occurs in the waste. Some compounds are used as solvents and similar. Other compounds are used as additives, e.g. in plastic, rubber or paper. These compounds may give rise to emissions in the leachate or in the landfill gas. Organic compounds are also formed during the degradation of organic material, especially volatile fatty acids and alcohols, ketones and aldehydes. Emissions of specific organic compounds from landfills are relatively poorly analysed.

Usually the emissions are measured as sum parameters, for example BOD, COD, TOC, etc. Öman (1993) has made a survey of organic analyses of leachates. In 21 published studies totally 265 specific organic compounds have been identified. The following 16 compounds were reported in at least five of these studies:

- volatile aromatics: benzene, toluene, xylene, ethyl benzene, phenol, cresol
- chlorinated volatile compounds: dichloromethane, dichloroethane, trichloroethene, tetrachloroethene
- bicyclic aromates: naphthalene
- volatile fatty acids: acetic acid, propanoic acid, butanoic acid, pentatonic acid (valeric acid).

Of these compounds the last category origins from degradation of organic material, while the other are expected to be of antropogeneous origin from usage as solvents or additives in different materials.

It has been suggested that the antropogenic specific organic compounds constitute the most significant group of leachate pollutants (Christensen et al, 1994). The actual emissions of organic compounds from landfills depend on the fate of the compounds inside the landfill, due to processes that are specific for each compound. Examples of important processes are (Öman, 1995; Öman and Wennberg, 1997):

i) the initial release of the compounds from the waste.
ii) sorption of the compounds to stationary or mobile matrices.
iii) transport of dissolved compounds with the leachate.
iv) transport of the compounds, with or as colloids or particles.
v) transformations of compounds, e.g. by microbial degradation, abiotic hydrolysis or abiotic redox transformations.
vi) aggregations to humic-like material.
vii) evaporation and transport of compounds in the gas phase.

A conceptual model for prediction of emissions of organic compounds from landfill has been suggested by Öman (1995). This model has been verified in a pilot study (Öman et al, 1997). The conceptual model is shown in Figure B.1. In the model the residence times of specific organic compounds inside landfills are primarily dependant on the sorption of the compounds to solid material and on the water transport. The residence times are then crucial for whether compounds will be transformed or evaporated during this time. The model predicts the compound's potential to be a) emitted with the leachate, b) emitted with the landfill gas, and c) mainly retained in the landfill. The model is based on some fundamental material properties:

- The octanol-water distribution coefficient K_{OW} is often used in environmental science for prediction of bioaccumulation. K_{OW} gives a measure the solubility in fat (octanol is a model substance for fat). K_{OW} is determined from a standardised laborative procedure by

$$K_{OW} = \frac{Concentration\ in\ octanol\ phase,\ mole / litre}{Concentration\ in\ water\ phase,\ mole / litre}$$

or can also be estimeted by empirical formulas. Often the log K_{OW} is given. There is also a connection between K_{OW} and the affinity for sorption on organic particles. Öman (1995) found that compounds with log K_{OW}<3,4 often occurs in leachates.

- Henry's constant H gives the partial pressure of the vapour in a water solution. H is defined by

$$H = \frac{Partial\ pressure\ (Pa)}{Solubilty\ in\ water\ (mole / m^3)}$$

Öman (1995) found that compounds with $H > 300$ Pa $\cdot$ m^3/mole occurs in the landfill gas.

- The acid dissociation constant K_a gives the strength of acidous compounds. In literature often the negative logarithm of K_a is given as pK_a.
- Microbiological degradability gives information of the degradation rate. In literature there is information about degradation rates from laboratory conditions as well as from field conditions. Depending on the conditions the reported values can differ. The degradability is often reported as half time $t_{1/2}$, which is the time for halving of the initial concentration.

Besides these material properties the water transport in the landfill and the residence time in the landfill are of importance for the prediction.

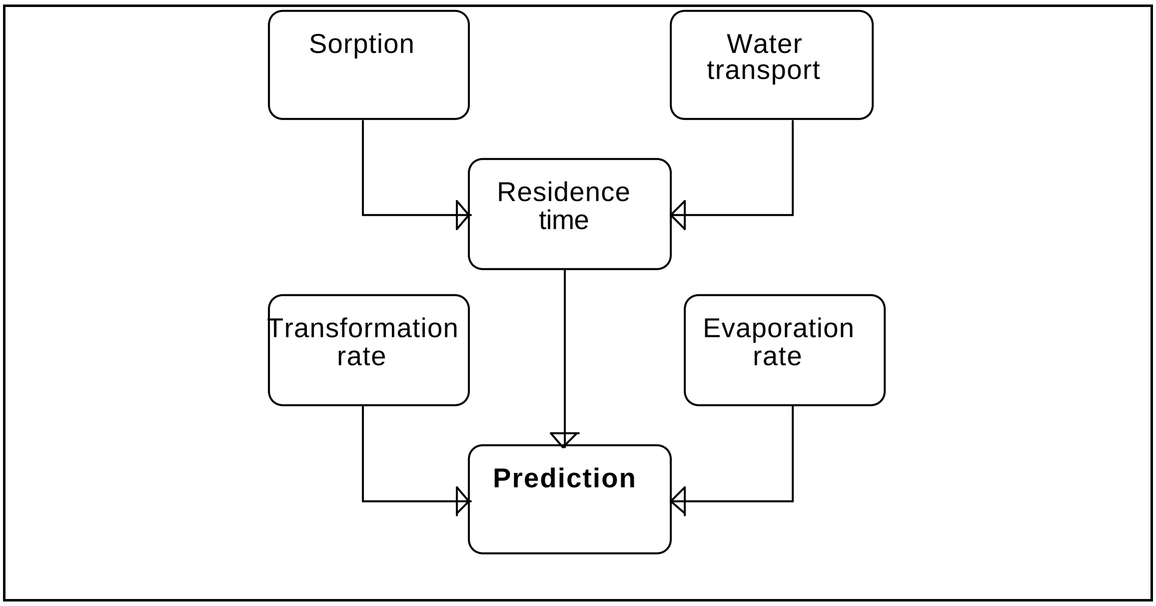

Figure B.1. The conceptual fate model for specific organic compounds (Öman, 1995)

Table B.1. Example of fate of some specific organic compounds (based on data presented by Öman et al, 1997; and Öman and Wennberg, 1997)

Compound	Molecular structure	log K_{OW}	pK_a	H *Pa m^3/ mol*	Anaerobic biological transformation		Processes				Emission in field test	
					General description	half time $t_{1/2}$	Sorbed	Evaporated	Degraded	Retained in waste	Emitted in gas	Emitted in leachate
Dichloromethane	CH_2Cl_2	1,2	-	260	Good to medium	<0,5 day	+	++	+++	-	+++	-
Tetrachloromethane	CCl_4	2,7	-	2000	Medium degrad.	3 - 14 days	++	+++	++	-	++	-
1,2-Dichloroethane	$CH_2Cl - CH_2Cl$	1,5	-	110	Degrad.	98 days-4,5 years	+	++	++	-	+++	-
Tetrachloroethene	$CCl_2=CCl_2$	2,6	-	2300	Medium degrad.	<19 - 218 days	++	+++	+	++	+++	-
1,4-Dichlorobenzene	$C_6H_4Cl_2$	3,4	-	160	Pers.	>1000 days	+++	++	-	++	(++)	-
2,4-Dichlorophenol	$C_6H_3Cl_2OH$	3,2	7,9	semi-vol.	Degrad.	<9 days	+++	+	++	-	(+)	-
Bensoenitril	C_6H_5-CN	1,6	-	semi-vol.	Degrad.		+	+	+++	-	(+)	+
Di ethyl phtalate	C_6H_4-(COO-$CH_2CH_2)_2$	3,4	-	0,011	Degrad.	40-100 days	+++	-	+++	+	(-)	++
2,3,4,5,6-Penta-fluorobenz-hydrol	CF_5-CH (C_6H_5)OH	3,1	-	non-vol..	Pers.		+++	-	-	++	(-)	+++
2,2,3,3,4,4,4-Hepta-fluoro-1-butanol	OH-CH_2CF_2-CF_2-CF_3	3,5	-	non.-vol.	Pers.							
MCPP, 2-(4-chloro-2-methyl-phenoxy) proionic acid	CH (COOH) (CH_3)-O-$C_6H_3Cl(CH_3)$	1,3	3,1	0,1	Pers.		+	+	-	+	(+)	++++
Lindane, γ-hexa-chlorocyclo-hexane	$C_6H_6Cl_6$	3,7	-	0,32	Not pers.	100-720 days	+++	+	+	++	(+)	-

Remarks:

() Parenthesis represents estimated values, as the actual emission data was not analysed (Öman et al 1997)

\+ Significant process or significant emission, graded + to ++++.

\- Not identified emission or not a significant emission

Estimating emissions

Available information is not sufficient to calculate emission factors for specific organic compounds in general, but some indications can be given. The Table B.1 gives some indications of the fate of some compounds. A preliminary model to roughly estimate emission factors are given as follows:

- Compounds with Henry's constant $H > 300\ P{\cdot}m^3/mole$ will likely be emitted as gas in the landfill gas.
- Compounds with the octanol-water distribution factor $log\ K_{OW} < 3{,}4$ will likely be emitted in the leachate.
- Slowly or easily biodegradable compounds can be assumed to be microbially degraded, according to Equation 5.1. Since the surveyable time periods comprise a longer period, it is likely that several of the more or less persistent compounds will be more or less degraded. However, there is a risk that toxic intermediates are formed that can be emitted by the leachate.
- Other compounds will need a deepened study, where the rates of different reaction ways are compared with each other.

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Appendix C. Additives in paper and plastic

Additives in paper

Additives in paper have been discussed in Sundqvist et al (1997). Additives are made to paper to achieve many different properties. Filling materials are used to increase opacity and smoothness and to save fibres. Chemicals can be added to adjust pH and to increase the wet strength and stiffness. A surface layer, coating, is often added to avoid dust formation and to improve the brightness and printing properties. Since different types of pulp, with varying properties and content of lignin, are often mixed in the same product, there are hundreds of different compositions of paper. Here some main groups of paper are listed, and “normal” compositions are given as a guideline, see Table C.1. The figures are for Swedish production, but the total amounts of paper additives used in Finland give the same picture (Nurmi 1993).

Table C.1. Mean composition of some kinds of paper, % of total weight.

	Fine paper, writing or printing paper of high quality	Coated fine paper, a coating slip has been applied to both sides	Newsprint, daily newsprint paper and similar printed matter	Magazine paper, weekly journals, magazines, catalogues, etc.	Sack kraft paper, for paper sacks	Liquid carton board, for packaging of liquids	Cardboard, paperboard and other similar
Pulps							
Bleached chemical pulp: kraft or sulphite pulp, bleached with hydrogen peroxide, chlorine dioxide or ozone	75	57		29		44	55
Unbleached chemical pulp: kraft or sulphite pulp					99		
Mechanical pulp: TMP or groundwood pulp			99	38			
Chemithermo-mechancial pulp, CTMP						30	37

Continued on next page

Table C.1. Cont..

	Fine paper, writing or printing paper of high quality	Coated fine paper, a coating slip has been applied to both sides	Newsprint, daily newsprint paper and similar printed matter	Magazine paper, weekly journals, magazines, catalogues, etc.	Sack kraft paper, for paper sacks	Liquid carton board, for packaging of liquids	Cardboard, paperboard and other similar
Chemical compounds							
Cellulose, hemicell.	75	57	73	57	85	66	82
Lignin			26	10	14	8	10
Lime	19	21		5		2.6	3.2
China clay		14		24		2.6	3.2
Bentonite	0.5	0.4				0.4	0.5
Alum					0.4	0.3	0.4
Cationic starch	5	4	0.5	0.5		0.7	0.9
AKD or ASA (alkyl compounds forming esters with cellulose)	0.2	0.1				0.2	0.2
Polyacrylamide	0.1	0.1	0.1	0.1			
Optical whiteners	0.5	0.5					
Styrene-Butadiene-rubber		2		3		0.3	0.4
CMC (carboxymethyl cellulose)		0.2		0.3		0.03	0.04
Polyacrylic acid		0.1		0.1		0.01	0.01
Polyethylene			0.5			20	
Polyethylene imine			0.2	0.2			
Resin glue					0.2	0.2	0.2
PhF-resin (phenol-formaldehyde)					1		
MF-resin		0.2				0.01	0.01
Organically bound Chlorine	0.006	0.005		0.002		0.003	0.004

In the landfill the additives will take part in the other reactions:

- Lime ($CaCO_3$) will act as a buffering agent in the landfill and possibly speed up the conversion from the acidogenic to the methanogenic phase. Some CO_2 will be released as gas, but most of the carbon will be found as carbonate in the leachate.
- China clay and bentonite will be stable for a very long time. It may have some buffering and sorbing effects, but it will mainly be filling and decrease the water permeability
- Alum is used to make rosin size. The sulphate ion will not stay in the paper, just the aluminium rosin size.
- Iron oxide pigments and carbon powder from ink will slowly leave the landfill in the leachate, iron to some extent as fine particulate sulphide.
- Some of the organic additives will be degraded:
  - Relatively fast: cationic starch, AKD and ASA, CMC, linseed oil alkyd, Zn/Ca-rosinate
  - Slowly or very slowly: styrene-butadiene rubber, polyacrylic acid, Al-rosin size, phenol-formaldehyde resin
- The following compounds will probably not be totally degraded in the surveyable time period. Nitrogen in the form of NH_3 in the gas and NH_4^+ in the leachate will leave the landfill. The nitrogen content is given below.

Polyacrylamide	20 % N
Optical whiteners	19 % N
Polyethylene imine	33 % N
Melamine-formaldehyde resin	55 % N

- The most common optical whitener also contains 11 % S. The sulphur will mainly be transformed to sulphide. Some of the sulphide will leave as hydrogen sulphide in the gas or leave with the leachate, while the most will be bound to metals.
- Another group of compounds is high molecular mass hydrocarbons that will not be much affected in the surveyable time period. These are polyethylene, waxes mineral oil (high boiling point), etc.
- Organically bound chlorine in paper is found in many different compounds formed during bleaching with chlorine gas or chlorine dioxide. The amount given in the Table is from bleaching with chlorine dioxide. The figure will be higher if chlorine gas is used (this is not used in Sweden any more) and close to zero if no chlorine at all is used.
- The chlorine compounds that are formed with bleaching without chlorine gas have a low degree of chlorination and are relatively easily degradable. The rest products will be methane, carbon dioxide, water and mainly chloride ions.

Additives in plastics

Different additives are added to plastic materials in order to improve processability, properties and prevent degradation. The effectiveness of the additives depends not only on the chemical nature of the additive, but also on the ability of the additive to persist in the material throughout the intended life time of the product. If degradation-preventing additives are released from the plastic matrix an increased degradation of the remaining plastic matrix is to expect. Additives in plastics have been discussed in Sundqvist et al (1997).

The additives are incorporated into polymers at concentrations of 0,01-1,0 wt-%. They are generally grouped under a name that explains their action, e.g.

antioxidants
metal deactivators
light stabilisers
heat stabilisers
plasticisers
lubricants
colourings
flame retardants and smoke suppressants
antistatic agents
fluorescent whitening agents
biostabilisers.

The additives used have different chemical structures and molecular sizes. Most of them are considerably smaller than the macromolecules, but larger than traditional solvents.
Typical concentrations of the various additives vary typically between 0,03 % and 1,0 %, except for plasticisers in PVC where the amount of additive can reach as much as 25 - 30 %. Many packages, which are in contact with foods, use epoxidized fatty acid esters (e.g. epoxidized soybean oil) as plasticisers. The amount of plasticiser in PE, PS and PET is generally low, if any.

The loss of the additives from plastics in the municipal solid landfill will continue until equilibrium solubility is reached in the plastic material. The equilibrium solubility and the migration rate are dependent on the temperature. Since the oxygen and nitrate reducing stage is very short, sometimes only a few days, the temperature in the landfill will decrease to ambient rather quickly and the migration rate of the additive thus slow down. On the other hand, if water penetrates the plastics it will be possible for the additive to migrate and be released and transferred to the leachate. This continuous migration of additive from the plastic materials will prohibit the equilibrium solubility to be reached.
In the longer perspective (i.e. the hypothetical time-period), the polymer will be more and more degraded and also more porous. That means that the remaining amount of additive will come into direct contact with leachate water and moisture. The remaining amount of additive will then be emitted to the environment

It is very difficult to give exact numbers of the degradation rates after migration of additives. We have, however, studied the degradation of polyethylene and monitored the percentage degradation in a long-term perspective. Here it was demonstrated that the percentage degradation increased with a factor two in PE samples deprived of the anti-oxidant and subject to biodegradation for about two years (Albertsson and Banhidi, 1980). The loss of additives should probably affect the resulting degradation rate of plastics by two modes:

- The migration of stabilising additives will result in the remaining plastics being more susceptible to various degradation factors (oxidation, thermo-oxidation, photolysis, etc.). This increases the degradation rate on a purely chemical basis.
- Loss of additives such as the plasticizers from PVC will leave a very brittle and porous material with higher probability for swelling and fragmentation. These increase the degradation rate more indirectly on a more physical basis.

It can be anticipated that the additives will leave the plastics by different rates. The loss of the stabilisers should during the surveyable time-period have the largest influence on the resulting degradation rate. The loss rate is determined by the rate of volatilisation of material from the plastic. Material depleted from the surface is be replaced by diffusion from the bulk.

From the above, the following conclusions can be drawn. The additives used in PE, PS, PVC and PET are numerous and every manufacturer uses his own recipes. The two principal factors governing the loss of additive from the plastics are the additive loss from the plastic surface and migration rate through the bulk of the material. The effect of the loss of additives on the resulting degradation rate is of two types. A purely chemical effect is seen when the stabilisers leave the material with increased probability for degradation. In some instances it is likely to expect that all or nearly all of the stabilisers have already left the material during the time in which the plastics are in use. The other type of effect is experienced when PVC is deprived of its plasticiser. As one third of PVC might be plasticiser the remaining polymer matrix will be porous and brittle giving increased probability for fragmentation.

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