LCA Methodology

Product Specific Emissions from Municipal Solid Waste Landfills

Part I: Landfill Model

Part II: Presentation and Verification of the Computer Tool LCA-Land (Int. J. LCA 3 (4) 1998)

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Abstract

For the inventory analysis of environmental impacts associated with products in Life Cycle Assessment (LCA) there is a great need for estimates of emissions from waste products disposed at municipal solid waste landfills (product specific emissions). Since product specific emissions can not be calculated or measured directly at the landfills, they must be estimated by modeling of landfill processes. This paper presents a landfill model based on a large number of assumptions and approximations concerning landfill properties, waste product properties and characteristics of various kinds of environmental protection systems (e.g. landfill gas combustion units and leachate treatment units). The model is useful for estimation of emissions from waste products disposed in landfills and it has been made operational in the computer tool LCA-LAND presented in a following paper. In the model, waste products are subdivided into five groups of components: general organic matter (e.g. paper), specific organic compounds (e.g. organic solvents), inert components (e.g. PVC), metals (e.g. cadmium), and inorganic non-metals (e.g. chlorine,) which are considered individually. The assumptions and approximations used in the model are to the extent possible scientifically based, but where scientific information has been missing, qualified estimates have been made to fulfill the aim of a complete tool for estimation of emissions. Due to several rough simplifications and missing links in our present understanding of landfills, the uncertainty associated with the model is relatively high.

Keywords: Gas combustion, landfills; inorganic non-metals, emissions, landfills; landfill model, municipal solid waste, product specific emissions; landfills, product specific emissions; teachate treatment, landfills; metals, emissions, landfills; organic compounds, emissions, landfills; polymers, emissions, landfills; waste products, emission, landfills

Part I: Landfill Model

1 Introduction

The disposal processes occurring throughout the product life cycle should be included and environmental exchanges with the surroundings estimated for a complete Life Cycle

Inventory of a product. However, significant difficulties exist in estimation of product specific emissions from waste. Firstly, because many products of today are produced and distributed internationally, and waste (which may be produced in any life cycle stage) is frequently disposed and treated in many different countries. Secondly, because emissions from waste are determined by the actual waste treatment processes which vary significantly. Thirdly, because knowledge about the environmental impacts from waste treatment is so far primarily gathered to represent the facility or process as such, and are therefore difficult to relate to the specific substances that are processed.

Although reuse and incineration of waste gain importance in many countries in these years, municipal solid waste landfills are still important recipients for solid waste in large parts of Europe (HJELMAR et al., 1994) and methods to estimate the product specific emissions from landfills, i.e. the emissions caused by processing of the waste products are required.

Product specific emissions can not be measured directly at the landfills because (i) the waste products may be distributed at several landfills, (ii) the emissions from the studied substances are mixed with emissions from other substances present in the landfill (iii) the emissions will occur some time in the future during the coming decades or centuries. Therefore, estimates of product specific emissions must be based on modeling of the key processes in the landfills as well as in the environmental protection systems (gas combustion plants and leachate treatment plants) associated with the landfills.

The purpose of this paper is to present a generic landfill model useful for estimation of product specific emissions from municipal solid waste landfills. The model is based on a number of assumptions and approximations about landfills and landfill processes and it has been made operational in a computer tool called LCA-LAND presented and verified by NIELSEN et al. (1998). The aim has been to include all the most important landfill processes in the model, based on the highest existing level of knowledge about the subject. However, several important aspects have got no or only minor attention in the literature and qualified guesses are used to fill these gaps in our knowledge until more information is available.

Since the object of the study is very complex and inhomogeneous, several assumptions and average considerations have been made both concerning landfilling technology and basic physical, chemical and biological processes in the landfill simply to make generic modeling of the processes possible. However, it is the belief of the authors that these generalizations and occasional simplifications are justified by the need of an operational tool for estimation of product specific landfill emissions. Furthermore, compared to other approaches published so far (FINNVEDEN and HUePES, 1995; FINNVEDEN et al., 1995; FINNVEDEN, 1996), the model presented here is in many regards sophisticated, taking many previously overlooked processes and aspects into account.

2 System Definitions and Nomenclature

(1) Object of the study

Any waste product disposed at landfills as waste after any stage of the life-cycle of a product, including the actual product when disposed after use. Waste products are subdivided into components which can be regarded as behaving individually in the landfill. The terms "waste product" and "components" are hereafter used to characterize the entire waste product respectively the relevant elements of the waste product.

(2) Types of landfills

Since emissions from landfilled waste products are governed by the actual physical, chemical and biological conditions in the landfill and these conditions are determined by the general composition of the present waste, it should be emphasized that the focus of this study is on municipal solid waste landfills only. Other types of deposits (e.g. chemical waste deposits, electronic waste deposits or demolition waste deposits) are different with respect to physical, chemical and biological conditions and should be object of specific investigations. Energy and resource consumption (e.g. associated with transportation) at the landfills are neglected and only direct emissions from the landfills are considered.

(3) Geographic area

Landfill emissions are significantly determined by climatic conditions. In order to limit impacts of variations of climatic conditions in different countries, the geographic area of this study is limited to European countries with average precipitation and other countries with similar climates. The model can be extended to cover more countries with other climates in the future.

(4) External pollution control systems

Many landfills are equipped with leachate treatment systems and/or gas combustion systems in order to reduce pollution and utilize landfill gas for energy production. Processes in leachate treatment plants and gas combustion plants directly related to the waste product are taken into account in modeling of emissions to the environment, but energy and resource consumption at these installations are neglected.

(5) Time perspective

The general purpose of LCA is to assess the environmental impacts from products during their whole life-cycle from extraction of raw materials to the final disposal. However, waste products disposed in landfills may continue to cause emissions to the environment for hundreds of years, and modeling of emissions must therefore be based on predictions. Since the present municipal solid waste landfill is a rather recent creation (emerging around 1970) no experience exists regarding the long-term behavior of the disposed waste. We find that emissions from landfills can be predicted appropriately (based on the existing knowledge and acceptable extrapolations) for the first 100 years, while the fate of the compounds is more or less unpredictable hereafter. Therefore, predictions of emissions to the environment from landfilled products only include the first hundred years in this study.

(6) Environmental recipients

The landfills are considered to be a part of the technical system during the first 100 years of their existence, hereafter they are considered to be a part of the environment. Hence, in addition to emissions to atmosphere, water and soil during the first 100 years of the landfills existence, remains in the underground after hundred years are listed in the inventory.

(7) Physical borders

Determination of product specific emissions to the environment from products disposed at landfills requires clear definitions of the borders between landfill and environment. The bottom border of the landfill is localized between the original geological material (e.g. soil or rock) and the landfilled waste or landfill bottom barrier. The top border is localized between the atmosphere and the waste or landfill top-cover.

(8) Societal interaction

Due to societal interactions such as remediation or building activity, the landfill conditions may become significantly altered during the studied 100 year period. However, the importance of such actions and their potential consequences for the landfill conditions are absolutely unknown, and we assume that all landfills are left unaffected during the first 100 years of their existence.

(9) Impact assessment

Emissions to water, atmosphere and soil are treated as emissions from the other parts of the product system, i.e. their potential contributions to various environmental impact categories (acidification, stratospheric ozone depletion, global warming etc.) are calculated and added to contributions from the rest of the product system (WENZEL et al., 1997). Waste substances which remain in the landfill throughout the 100 year period are totally or almost stable and immobile and their further fate is almost unpredictable. Since the long term stable components do not cause any known environmental impacts, they should be disregarded in the impact assessment step and only appear in the inventory as discussed above.

(10) Underground use

Although landfilling of waste occupies space in the underground (during a short period for degradable and mobile components and a long period for heavily degradable and non-mobile components) and potentially limits the usefulness hereof, this is not accounted for in the model. Future studies should address how underground use associated with disposal of waste should be treated in LCA.

The above system definitions are summarized in short by NIELSEN et al. (1998).

3 General Landfill Model

The amount and composition of emissions from waste products disposed at landfills are determined by (i) the physical/ chemical and biological conditions in the landfill, (ii) the composition of the waste product and (iii) the physical/ chemical properties of the waste product components. Modeling of emissions from waste product disposed at landfills must therefore be based on a fundamental understanding of the most important processes taking place in the landfills as well as the associated leachate treatment systems and gas combustion systems.

Since modeling of emissions from a landfill to the environment in itself is very complex and this study aim at covering a whole range of different landfills in different countries, a simplified model of transportation routes for components into the environment is required to make the modeling possible. Figure 1 shows the emission routes from landfills either directly into the environment (atmosphere or water) or via gas combustion plants or leachate treatment plants. All types of aquatic recipients (e.g. groundwater, seawater, lakes) have been pooled in one group called water, since differences in sensitivity of the various recipients to the components are considered an issue of the subsequent impact assessment phase of the LCA.

Since environmental protection systems at landfills are controlling the amount and type of emissions from the disposed waste products, there is an obvious need to classify landfills into a limited number of groups according to their protection systems, to facilitate the modeling. In the following, three main categories of environmental protection systems at landfills are considered:

- 1. top-cover
- 2. leachate collection and treatment units
- 3. landfill gas collection and combustion units

Since the considered environmental protection units represent a whole range of different methods and technologies at different landfills, some general assumptions are made for each category:

(1) Top-covers range from simple layers of soil to advanced configurations of gas and water drainage layers, separation layers and lining membranes (Cossu, 1994). Infiltration of water from precipitation into the waste is an important parameter in modelling of emissions from landfills, which is largely controlled by the presence and type of the cover. In order to take differences in infiltration into account, we differentiate between two types of top covers.

- Clay or synthetic geotextile top-covers, where 5% of the precipitation is assumed to enter the waste (through holes and cracks).
- Soil top-covers, where 50% of the precipitation is assumed to enter the waste (BLAKEY, 1992).

Landfills without top-cover are classified in the same group as landfills with soil top-covers with respect to infiltration, since infiltration is assumed to be at the same level. Clay and synthetic geotextile top-covers are hereafter called "water-stopping top-covers".

(2) Leachate collection systems and leachate treatment units may be constructed in several different ways with varying efficiencies and long-term stability. The fraction of leachate

Fig. 1: Substance and energy transportation routes from waste products disposed in municipal solid waste landfills. Rounded boxes represent environmental recipients, rectangular boxes represent technical units. Full arrows indicate material transportation. Dotted arrows indicate energy transportation

collected for treatment at waste water treatment plants is an important factor in modeling of emissions to water. However, the subject is difficult to address because leachate collection systems are differently designed at different landfills and because the efficiency of the leachate collection may change as liners and membranes age (CHRISTENSEN et al., 1994b). In the reviewed literature no information about individual or average efficiency of leachate collection systems has been found. We assume that all leachate collection systems are 80% efficient and hence that 20% of leachate produced at landfills equipped with leachate collection systems is emitted to aquatic recipients through leaks in the membrane.

Leachate treatment units associated with landfills may vary significantly in construction, ranging from simple low cost plants adjacent to the landfill to municipal waste water treatment plants with several advanced cleaning steps. Since application of leachate treatment plants have not been investigated further in this study, it is assumed that all leachate treatment plants are municipal waste water treatment plants. Sludge from the leachate treatment plants may either be spread on farmland (as fertilizer), be combusted at incineration plants or be disposed at landfills. Although the further fate of sorbed compounds from the landfills depend on the specific sludge treatment, all sorbed compounds in the leachate treatment plant are assumed to be emitted to the soil environment.

(3) Landfill gas collection systems and combustion units may be constructed in several different ways with differing efficiencies and long-term stability. The fraction of landfill gas collected for combustion at landfills equipped with gas combustion systems is an important parameter in the inventory of landfill emissions of volatile components and in modeling of the associated energy production. However, the subject is difficult to address, partly due to lack of information in the reviewed literature, and partly due to the fact that landfill gas collection may be stopped for economical reasons in the future before all gas has been emitted. Without any deeper background, 50% of the total gas production is assumed to be collected for energy production and 50% is assumed to be emitted directly to the atmosphere at landfills equipped with gas collection systems.

The landfill gas is used for heat and/or electricity production (STEGMANN, 1989) and both types of energy should be considered in the inventory. However, to our experience, the generally available data about energy production at landfill gas combustion plants are not sufficiently detailed to permit specific calculations of heat- and electricity production. Therefore, energy production is calculated as heat, based on specific energies of combustion and an average combustion efficiency of 85% (O'CALLAGHAN, 1992).

The environmental protection systems are used in different combinations at different landfills depending on traditions and technological state in different countries. Leachate emissions and amounts of substances remaining in the landfill after 100 years is determined by the combination of topcover and leachate treatment while landfill gas emissions are assumed to be independent of the presence of landfill top-cover and leachate treatment units. Therefore, all landfills are organized in four groups (L1 to L4) which behave individually with respect to leachate emissions and amounts of substances remaining in the landfill, and two groups, G1 and G2 which behave differently with respect to landfill gas emissions (\rightarrow) *Table 1* and *Figure 2*). The landfill grouping with respect to leachate emissions (L1 to L4) and the landfill grouping with respect to landfill gas emissions (G1 and G2) are independent of each other and characterize the landfills in two different ways.

Fig. 2: Illustration of landfill properties for landfill types L1 to L4. Water-stopping top-cover include top covers made of synthetic geotextiles or clay. See also Table 1

Table 1: Landfill characteristics

a)cover made of clay or synthetic membrane material

b)LGC is the term used for G1-landfills by NIELSEN et al. (1998)

at a reasonable level of effort.

The fractions of waste disposed at landfill types L1 to L4 respectively G1 and G2 are used to describe landfill technology in individual countries and the result is a simple but valuable set of landfill profiles characteristic for each country. Data collection based on the above concept has been performed in The Netherlands, Germany and Denmark (NIEI,SEN et al., 1998). It was the experience from this study that the required data were available at a central level in

4 Division of Waste Products into Components

these countries and that the information could be collected

Waste products are composed of a number components, which may act differently in the landfill. Therefore, waste products must be divided into a number of components which should be considered separately in the modeling of emissions. The division of waste products into their components must be based on a physical/chemical understanding of the waste product and insight into landfill processes in general. In order to facilitate modeling, five different groups of components with individual behavior in the landfill have been identified:

- Specific organic compounds (SOCs)
- General organic matter (GOM)
- Inert components
- Metals
- Inorganic non-metals

For each component of the waste product, the weight and relevant physical and chemical properties should be used in modeling of emissions as described in more detail in sections 5 to 9. The five groups should in principle cover all waste products. However, in cases where compound specific data are absent (e.g. for the more rare metals) model estimations of emissions may have to be based on default values determined from properties of more frequently occurring components, or the modeling is impossible.

The emissions from waste products disposed at landfills are often determined by the physical and/or chemical organization of the actual components. For instance, organic chemicals that are surrounded by plastic (e.g. phthalates in PVC) will generally be released much slower from the landfill than similar chemicals disposed directly at the landfill. This should be taken into account when the fate of the components are considered. Assessment of the importance of the actual organization of the components within the waste product must be based on physical/chemical understanding of the specific waste product and insight into landfill processes in general. No generally useful rules or recommendations can so far be provided to support this assessment.

The model assumptions and approximations that are presented and discussed in sections 5 to 9, are based on the general premise that the components of the waste product are released to the landfill environment within the first few years after disposal.

5 Specific Organic Compounds (SOCs)

All organic chemicals (including organic solvents) belong to the group of SOCs. The group includes compounds such as benzene, phenol, trichloroethene, formaldehyde and phthalates. In addition to the conditions in the landfill, the fate of SOCs is determined by the specific properties of each compound. SOCs may evaporate from the landfill, leach out of the landfill and/or become partly or fully degraded. SOCs are normally disposed at municipal landfills in small amounts and the interest in this group is primarily due to their toxicity. The total number of substances belonging to this group is in the order of 100,000 (EEC, 1990) and estimations of emissions must be based on model simulation because only a minor part of these have been investigated directly in the landfill environment.

5.1 In the landfill

During recent years several models have been developed for estimation of the fate of SOCs in landfills (e.g. LIN and HILDEMANN, 1995; KJELDSEN and CHRISTENSEN, 1997). The model presented by KJELDSEN and CHRISTENSEN (1997) is founded on basic physical, chemical and biological principles and is useful for general modeling of SOC-emissions by leachate and landfill gas as well as loss by degradation, and was preferred for this study. Originally, the model was prepared for short term estimations of emissions of SOCs under steady state conditions. For this study, the model has been modified, an now integrates emissions during the hundred year period under changing landfill conditions $(\rightarrow$ *section S.1.2).* The modified model is hereafter called the SOCmodel.

5.1.1 Compound specific parameters

Since the behavior of SOCs in the landfills is determined by their individual nature, a number of compound specific input parameters are required for prediction of their fate by the SOC-model: molar weight, diffusion coefficient i air, vapor pressure, Henry's law coefficient (k_H) , octanol-water partition coefficient (log K_{ow}) and first order degradation rate constant. All physical/chemical parameters have general validity, while the degradation rate constant is determined by the actual conditions in the landfill.

Measured physical/chemical data are often available in the literature (e.g. MONTGOMERY, 1996; MACKAY et al., *1992-* 1995; Hazardous Substance Data Bank, 1995) for commonly occurring compounds while information about less frequent compounds may be missing. Estimates of physical-chemical properties based on empiric molecular connectivity or quantitative structure-activity relationships, QSAR (e.g. MEYLAN and HOWARD, 1995) may be useful where experimental data are lacking.

Degradation rates in the landfill environment should be considered separately in the waste and in the top-cover. In the waste, redox conditions change from aerobic in the very beginning of the landfill's existence to anaerobic during a long period until all organic material has been degraded. After this, the landfill slowly turns aerobic again (FINNVEDEN et al., 1995). Degradation potentials and degradation rates are widely determined by the actual redox conditions and hence degradation may change significantly as time goes. However, for the modeling, the initial aerobic period is ignored (because it is short) and it is assumed, that the landfill is anaerobic throughout the 100 year period, and hence only anaerobic degradation processes need to be considered. The degradation of SOCs in the landfill is difficult to assess, mainly due to lack of experimental data (DEIPSER and STEGMANN, 1994; KROMANN and CHRISTENSEN, 1997) but also due to large variations in degradation potentials and degradation rates caused by variations in e.g. microbial composition, temperature and humidity both among different landfills and within each landfill. In cases where degradation rate data do not exist a conservative assumption (no degradation) can be useful, or degradation data obtained from anaerobic degradation studies of landfill leachate plumes (CHRISTENSEN et al., 1994a; NIEi.SEN al., 1995a,b) can be adapted. Data obtained in other environments than landfills should only be used with great care, and it should be noted that information about aerobic degradability (e.g. in waste water treatment plants) can not he extrapolated to predict the behavior of SOCs under anaerobic conditions in the landfill.

In the top-cover, redox conditions are determined by two competing factors: (i) oxygen intrusion from the atmosphere and (ii) oxygen consumption from oxidation of methane produced in the waste during degradation of general organic matter $(\rightarrow section 6)$. The result is a redox gradient in the top cover (JONES and NEDWELL, 1990) which changes as the landfill ages and methane production varies and hence significant differences exist in degradation potential for SOCs in the bottom of the of the cover and in the top of the cover. Degradation of SOC in landfill covers has only received minor attention in the literature (e.g. KJELDSEN et al., 1996) and the few degradation rates which have been determined in experimental studies should be used with care due to large variations.

Since methane is one of the most important SOCs emitted from landfills $(\rightarrow section 6)$, special attention is given to this compound. Methane is subject to aerobic degradation processes during transportation through the top cover as observed by WHALEN et al. (1990) and JONES and NEDWELL (1993). Based on experimental studies, WHALEN et al. (1990) proposed that 50% of methane produced at landfills in the United States is degraded in landfill top covers. However, due to lack of supportive studies and significant variations in methane degradation rates (four orders of magnitude in one landfill, JONES and NEDWELL, 1993), we find the cautious assumption that no methane is degraded in top covers most appropriate until more information is available.

5.1.2 Landfill parameters

In addition to the specific properties of SOCs, also a number of general landfill parameters are required for modeling of emissions of SOCs. Although landfills are very heterogeneous with respect to construction and physical conditions and furthermore many conditions change as the landfills age, we attempt the description of an average landfill by the following values:

- bulk density of the waste, $p(b,w) = 0.6$ tons of dry waste \cdot (m³ waste)⁻¹
- volumetric water content in waste, $\varepsilon(w,w) = 0.4$ m³ water \cdot (m³ waste)⁻¹
- volumetric air content in waste, ε (a,w) = 0.1 m³ air · (m³ waste) $^{-1}$
- fraction of organic carbon in waste, $f_{\text{av}} = 0.1$ kg organic $carbon \cdot (kg$ dry waste) $^{-1}$
- height of waste $L(w) = 10$ m.
- volumetric air content of cover, ε (a,c) 0.11 m³ air \cdot (m³ cover) -I
- total porosity of cover, $\varepsilon(c) = 0.45$ m³ \cdot (m³ cover)⁻¹
- tortuosity, $\tau(c) = 0.02863$ [-]
- thickness of cover, $L(c) = 1$

All parameters are further described in KJELDSEN and CHRISTENSEN (1997). Estimates of average height of waste thickness of landfill covers are based on DOEDENS and CORD-LANDWEHR (1989) respectively Cossu (1994).

Gas generation caused by the degradation of organic matter in the landfill is an important factor for convective transportation of volatile SOCs in the landfill gas. Therefore, assumptions about gas generation during the hundred year period are required. However, gas generation changes significantly as the landfills age and organic matter disposed in the landfill is degraded. Hence, assumptions about gas generation are made as function of time. Although many factors influence the methane production in landfills (e.g. waste composition, temperature, and humidity (CHRISTENSEN and KJELDSEN, 1989), we made following simple assumptions based on NILSSON et al. (1995) and BELEVI and BACCINI (1989):

First year: 5 m³ gas \cdot (m³ waste)⁻¹ · year⁻¹ Year 2 to 20: gas generation decreases linearly to 0 m^3 gas \cdot $(m³$ waste)⁻¹ · year⁻¹ Year 21 to 100: 0 m³ gas \cdot (m³ waste)¹ · year¹

The flux of water into the landfill which is caused by rainfall is also an important parameter in modeling of emissions of SOCs dissolved in leachate. In major parts of European land areas mean annual precipitation ranges between 400 and 1500 mm year¹ (The Times Atlas, 1992) and an average precipitation value of 1000 mm year¹ has been assumed for average European countries. The fraction of precipitation entering the waste was discussed in section 3.

Synthetic top covers installed at some landfills significantly control the SOC diffusion from the landfill into the atmosphere. Several water-stopping top-covers are based on synthetic geotextiles and an approximate diffusion coefficient of 10^{-12} m²s⁻¹ (based on PARK and NIBRAS, 1993) is assumed as standard for all SOCs disposed at landfills equipped with water-stopping top-cover.

5.2 In the gas combustion plant

The fate of SOCs in the gas combustion plant depends on the nature of the compound. Some compounds are totally mineralized, some are passing unaffected through the plant and some are partially combusted. Therefore, in addition to specific combustion energies (e.g. CRC, 1984), the fraction of combusted SOC as well as specific amounts of combustion products emitted to the atmosphere are required as input for modeling of the emissions.

5.3 In the leachate treatment plant

The fate of SOCs in the leachate treatment plant is governed by the actual conditions in the plant as well as the nature of the specific compounds. The subject is difficult to address because processing conditions in various leachate treatment plants are heterogeneous. However, the technical guidance document (European Commission, 1996) on risk assessment of chemicals within the European Union propose a generic model which estimates the fraction of degraded, evaporated, sorbed and discharged SOCs. The model has been described by e.g. STRUIJS et al. (1991) and a modified version of the model suggested by MIKKELSEN et al. (1996) is used in this study.

6 General Organic Matter (GOM)

General organic matter includes all sorts of organic materials of biological origin which are easily degradable under the strongly reduced redox conditions in the landfills. GOM includes materials such as wood, cotton, fat, sugar, meat and paper. The main components in GOM are C, H, O, N and S but also trace amounts of other elements (e.g. Fe, P, Mg, CI) are often among the constituents. The main concern associated with landfilling of GOM is the formation of methane, which due to its radiative absorbing capacity serves as at potent greenhouse gas when emitted to the atmosphere (THORNELOE, 1996). The focus of this section is therefore on emissions originating from the carbon in GOM. Significant amounts of other constituents, should be considered as separate components (e.g. metals or inorganic nonmetals, se following sections). Various types of GOM behave similarly with respect to emissions during the 100 year period in the landfill and are therefore treated identically in the following.

6.1 In the landfill

GOM is decomposed into three main products during methanogenic degradation in the landfill: methane $(CH₄)$, CO, and dissolved organic matter. The decomposition rate of GOM depends on the nature of the GOM (ARIGATA et al., 1995), but is generally assumed to be quite fast. According to first order degradation rate constants proposed by ARIGATA et al. (1995), 80% of the slowest degradable GOM are degraded within the 100-year period. Although it may not be absolutely correct, all GOM is assumed to be decomposed under anaerobic conditions (\rightarrow *section 5.1.1*) within the 100-year period. Methane is a volatile organic compound and its fate in the landfill is discussed in **section** 5. All CO_, is assumed to be emitted to the atmosphere. Since GOM originates from recent biological activity, the emitted CO₂ is not considered as a net contribution to the atmospheric content of CO₂, and it is therefore considered to be neutral with respect to global warming (HAOsCHILD and WENZEL, 1997).

Based on general information about the fate of carbon in landfills (BELEVl and BACCJNI, 1989) *99%* of the carbon content is assumed to be emitted as gas (CO, and methane), and 1% of the carbon content is assumed to be emitted as unspecific organic matter dissolved in the leachate. The amounts of CO, and methane produced pr. gram of GOM is specific for each type of GOM and can be determined by stoichiometric calculations as described by Cossu et al. (1996).

6.2 In the gas combustion plant

Methane is the only combustible atmospheric emission from GOM. Since methane is very flammable (Hazardous Substance Data Bank, 1995) all methane is assumed to be mineralized completely to CO, and H₂O with an energy yield of 55.7 KJ g⁻¹ (CRC, 1984).

6.3 In leachate treatment plant

Dissolved unspecific organic matter leaching from GOM disposed at landfills is considered to be easily biodegradable and non-volatile in the leachate treatment plant. Therefore, all organic matter derived from GOM is assumed to be completely mineralized to $CO₂$ and $H₂O$ in the leachate treatment plant. The fate of methane in the leachate treatment plant is estimated by model simulation as discussed in **section** 5.3.

7 Inert Components

The group of inert components includes all substances which are persistent in the landfill, and do not evaporate to the atmosphere or dissolve in the leachate but remain intact in the landfill during the hundred year period. The group of inert components includes for instance heavily degradable polymers (e.g. polyvinyl chloride, PVC and polyethylene, PET (FINNVEDEN et al., 1995; Møller et al., 1996) and geological materials (e.g. clay, sand and stones). The inert components are not toxic, and the main problem associated with their existence in the landfill is due the fact that they occupy space. Since the inert components are not subject to transportation processes discussions of their fate in the gas combustion plant and leachate treatment plant are not relevant.

8 Metals

The group of metals includes all metallic elements (e.g. aluminum, iron, cadmium, copper, lead, nickel and zinc). The main concern associated with emissions of metals is due to the potential toxicity of certain species.

8.1 In the landfill

The transportation of metals from the landfill into the environment is determined by sorption, precipitation and complex formation processes, and the relative importance of each processes is specific for each metal (CHRISTENSEN et al., 1994a). The transportation mechanisms of metals in landfills are very complex and no mechanistic models, useful for estimation of emissions have been found during our literature review. Therefore, estimates of metal emissions from landfills are based on emission factors (total emissions during approximately one century) deduced from measured metal concentrations in leachate (FINNVEDEN, 1996). Although large uncertainty is associated with the emission factors, they provide a useful indication of the fact that metals are emitted extremely slowly and that the main fraction remains in the landfill after the 100 year period.

The emission factors presented by FINNVEDEN (1996) are based on studies of landfills with a significant leachate generation. Therefore, emissions of metals from landfills equipped with water-stopping top-cover are adjusted by the ratio of the leachate generation at landfills with water-stopping top-cover to the leachate generation at landfills without water-stopping top-cover $(\rightarrow \text{section 3})$ in this study.

8.2 In the gas combustion plant

In the group of metals only Hg and Cd are assumed to evaporate from the landfill (FINNVEDEN, 1996) and only these two metals will reach the gas combustion plant. In the landfill

gas combustion plant 100% of both components are assumed to be emitted to the atmosphere. Hence, collection and combustion of landfill gas does not influence the atmospheric metal emissions.

8.3 In the leachate treatment plant

The behavior of metals in the leachate treatment plants is determined by the specific nature of the metals and the actual conditions in the plants. Since metals are not degraded in the treatment plant and does not evaporate into the atmosphere (LESTER, 1983), estimates of their fate can be limited to investigations of metal partitioning between the sludge phase and the water phase, and hence between soil and water as final recipients.

No models for estimation of metal partitioning between water and sludge have been found during our literature review and estimates are based on measured removal efficiencies at treatment plants (LESTER, 1983). Removal efficiencies may, however, vary significantly and average values from several investigations are recommended. Based on five investigations or more, LESTEa (1983) calculated following average removal efficiencies useful for estimation of emissions: copper: 66%, zinc: 69%, chromium: 66%, cadmium: 46%, lead: 64%, Nickel: 33%, mercury: 63%.

9 Inorganic Non-Metals

The group of inorganic non-metals includes compounds of all elements except organic carbon $(\rightarrow$ *sections* 5 and 6) and of course the metals. Due to significant differences in physical, chemical and biological properties, non-metals behave very differently in the landfill environment as well as in the gas combustion plant and leachate treatment plant. Hence, individual considerations must be made for each compound. The transportation of non-metals is very complex and no predictive models have been found for estimation of their fate. Thus, assumptions about their emissions are primarily based on measured emissions. Only the most frequent non-metals (chlorine, sulfur, and nitrogen) will be discussed here because information is missing or very scarce for the others.

9.1 In the landfill

Chlorine is generally assumed to be highly mobile in the landfill, and all chlorine is assumed to be emitted within the 100 year period (BELEVl and BACCINI, 1989). The main part (about 90%) of the total chlorine is emitted with the leachate, but also a fraction (about 10%) is emitted with the gas (BELEVI and BACCINI, 1989). In the reviewed literature, no information exist about the specific chemical structure of the emitted chlorine and it is assumed that 100% of chlorine is emitted with the leachate as CI to aquatic recipients. Since CI transportation is not retarded (CHRISTEN-SEN, 1992), it is assumed that all CI is emitted within the hundred year period, also at landfills equipped with waterstopping top-cover.

Sulfur is reduced to sulfide under the strongly reduced conditions in the landfill, and is partly emitted to the atmosphere (H, S) , partly emitted to water $(H, S$ and HS $)$ and partly precipitated (e.g. as metal sulfides). The emitted fraction of sulfur is generally assumed to be emitted within the first decade of the landfills existence (BELEVI and BACCINI, 1989), but much precipitated sulfide may remain in the landfill after this period. When the landfill turns aerobic (as assumed after the 100 year period, \rightarrow *section 5.1.1*) the precipitated sulfide may be oxidized and emitted slowly to aquatic recipients as sulfate (SO_4^2). Since SO_4^2 is assumed to be emitted after 100 years it is disregarded here, and only emissions of sulfide will be considered in the following.

According to transfer coefficients reported by BELEVI and BACCINI (1989), 3‰ of disposed sulfur is emitted with the gas to the atmosphere and 1%o is emitted with the leachate to water. Modeling of sulfur emission to aquatic recipients is quite complicated because speciation of sulfide is determined by pH, which changes significantly during the 100 year period (CHRISTENSEN and KJELDSEN, 1989) and varies within each landfill (SLEAT et al., 1989). The first dissociation constant of H₂S is pK = 7.04 (CRC, 1984) and pH in the leachate may range between 4.5 and 7.5 in the acid phase and between 7.5 and 9 in the methanogenic phase of the landfills existence (CHRlSTENSEN et al., 1994a). Hence, sulfur emissions to water may be dominated by H₂S in the acid phase and HS in the methanogenic phase. Due to lack of generally representative information about pH in the landfills and about release rates of sulfur from landfills, we assume that 50% of sulfur is emitted as H ₂S and 50% as H S². All sulfur emitted to the atmosphere is assumed to be emitted as H₂S, since no other inorganic compounds of sulfur are volatile. Since the fate routes of sulfur are very complex and assumptions about emissions are rater uncertain, no distinctions are made between landfills with and without water-stopping top-cover (clay/synthetic membrane)

Nitrogen species are reduced to NH_{4} ⁺ in the landfill. Dissociation of $NH₄$ ⁺ to form $NH₃$ (gas) is not very likely because $NH₄$ ⁺ has a pK₃ of 9.2 (Hazardous Substance Data Bank, 1995), and all nitrogen is therefore assumed to be emitted with the leachate to water (BELEVI and BACCINI, 1989). Leaching of NH₄ is retarded by ion-exchange processes in the landfill, but generally, the NH $₁$ ⁺ is assumed to be emitted rather quickly</sub> to the environment. According to transfer coefficients reported by BELEVI and BACCINI (1989) 6% of nitrogen should be emitted after a mean residence time of 9.4 year. Assuming that 6% of the remaining fraction of nitrogen is emitted every 9.4'th year throughout the 100 year period, it can be estimated that approximately 50% of the nitrogen is emitted during the first 100 years of the landfills existence. Since the estimations of $NH₄$ ⁺ emissions are based on studies of land-

fills with relatively high leachate production (leachate/precipitation ≈ 0.4 , BELEVI and BACCINI, 1989) and all NH₄+ is assumed to be transported with water, emissions from landfills equipped with water-stopping top-cover (clay/synthetic membrane) are adjusted to the lower leachate generation.

9.2 In the gas combustion plant

Chloride and NH_A ⁺ are not emitted with the landfill gas and only sulfur (H,S) will be discussed in this section. H,S is highly flammable (Hazardous Substance Data Bank, 1995) and all H_1 S is assumed to be completely converted to SO $_x$ </sub> in the gas combustion plant.

9.3 In the leachate treatment plant

Chloride is very soluble in water and is not susceptible to biological processes, sorption or precipitation. Thus, chloride passes the leachate treatment plant unchanged.

The fate of sulfur species (H₂S and HS⁻) and NH₄⁺ is among other things determined by pH and redox conditions in the leachate treatment plant. Sulfur may evaporate as H₂S, precipitate as metal sulfides or become discharged to aquatic recipients as HS/H₂S. During our literature review, no information was found about the importance of each transportation route and we realize that the subject is very difficult to address both by general mass balance studies (sulfide from the landfill is mixed up with sulfate/sulfide from other sources) and model simulations (sulfur is very sensitive to pH and pE changes which appear both at micro and macro scale in the waste water treatment plant). We assume that a of sulfur is emitted to water as sulfate, 1/3 is precipitated as sulfide and 1/3 is emitted to the atmosphere as H₂S.

Nitrogen in the form of $NH₄$ ⁺ may be oxidized to NO₃ in the aerobic parts of the waste water treatment plant and further reduced to N , in anaerobic denitrification steps. Thus, nitrogen may be emitted to water as NH_{4} ⁺ or NO_{3} or to the water/atmosphere as N_2 . The fate of nitrogen is determined by the efficiencies of aerobic steps and the presence and efficiencies of denitrification steps at the waste water treatment plants, and average estimations of emissions should be based on detailed information about the actual leachate treatment technologies applied in various countries. Collection of information about leachate cleaning technology is outside the scope of this study, and we assume that 80% of incoming $NH₄$ ⁺ is oxidized to NO₃. and 50% of $NO₃$ is reduced to $N₂$.

10 Discussion

In this paper a generic landfill model has been presented for estimation of emissions from waste products disposed as municipal solid waste for the LCA inventory. The mod-

Table 2: Uncertainty of estimates of emissions from various components in the landfill system as assessed by the authors

eling assumptions and approximations made in this study are, however, associated with uncertainty which lead to uncertainty of the following estimates of emissions. As already discussed in the introduction, it is not possible to measure emissions from the landfills, and hence not possible quantify the uncertainties of emissions. Therefore, the uncertainties associated with the modeling assumptions and approximations must be based on subjective estimates, which of course limit their value to more general conclusions. Table 2 shows the uncertainty of estimated emissions as assessed by the authors.

Table 2 shows that the uncertainty of estimates of emissions generally is considered to be high, but also that significant variation exist among different components in different parts of the landfill system.

The uncertainty of estimates of GUM emissions from landfills and gas combustion plants is low because estimates are based on stoichiometric calculations and quite certain assumptions that almost all GOM is gasified respectively combusted. The high uncertainty of SOC emissions from landfills (up to 100%) is partly caused by uncertainty of predictions of transportation and partly caused by the lack of information about degradation of SOCs in landfills. A completely degradable SOC may as a conservative assumption be considered as non-degradable resulting in an absolutely wrong estimate. This is, however, more related to lack of information about degradation of specific compounds than uncertainty associated with the model. FINN-VEDEN (1996) suggested that emission factors of metals in landfills are uncertain by one or two orders of magnitude which is quite high. However, the absolute emissions of metals are small in any case and the significance of the high uncertainty is limited.

Much uncertainty associated with the assumptions and approximations presented in this paper has emerged from insufficient information about specific processes and conditions. Thus, a substantial potential exist for diminishing the uncertainty of the assumptions and approximations and hereby improve the estimates of emissions from landfills. It **is** the hope of the authors that future research and development will lead to improvements of assumptions and approximations to limit their uncertainty and thereby improve the certainty of the final LCA inventory. Until then, the authors find the assumptions and approximations made in this paper to be the best available for modeling of landfill emissions in LCA, and stress the importance of taking the uncertainty into account when interpreting the results.

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