

# Solid Waste Treatment Within the Framework of Life Cycle Assessment

## Metals in Municipal Solid Waste Landfills

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### Abstract

Due to a lack of available methods and data, the Inventory Analysis in many Life Cycle Assessments (LCA) often exclude important information concerning emissions from landfills. In light of this, a method for estimating emission factors for metals from municipal solid waste has been developed and is presented herewith. Emission factors, expressing the emitted fraction of the landfilled amount of the element during a surveyable time period (corresponding to several decades or a century), is suggested for several metals. It is suggested that these can be used in initial (screening) LCAs where the aim is to identify key-issues, i.e. important aspects of the system under study.

**Key words:** LCA/LCI, municipal solid waste; waste, LCA/LCI; municipal solid waste, metals; metals in municipal solid waste; landfills, municipal solid waste; waste, municipal solid, landfills; emissions from landfills; allocation, landfills; LCA, screening, emissions form landfills; LCA, case studies; case studies, LCA

### 1 Introduction

The Inventory Analysis of a Life Cycle Assessment (LCA) should include emissions from landfills [1–3]. However, this is often not the case [4], largely due to lack of methods and data [5]. There is thus a need for developing methods for estimating emissions from landfilling of solid waste that can be used in LCAs. There is also a need for generic emission factors which can be used in initial (screening) LCAs for identifying key-issues, i.e. important aspects of the system under study.

In a previous paper [6], a general approach for estimating potential emissions from landfilling of solid waste was discussed. As a continuation of this work, the present paper deals specifically with metals in municipal solid waste (MSW) landfills. For an LCA practitioner, emission factors,  $E_j$ , defined as

$$E_j = \frac{\text{Amount of emitted pollutant } j \text{ during a certain time period}}{\text{amount of landfilled pollutant } j} \quad (1)$$

would be useful. The amount emitted from a landfill per functional unit during a certain time period can then be calculated as the product of the emission factor and the amount landfilled per functional unit. (The functional unit describes the function and the amount of it that the system

under study is providing). The aim of this study has been to derive such emission factors and to discuss their applicability.

### 2 Method and Results

#### 2.1 General approach

Since the general approach for estimating emissions from landfills has been discussed in detail in [6], only a few key aspects will be mentioned here:

- The landfill is regarded as a part of the technical system. The emissions from it should thus be included in the inventory analysis.
- In connection with LCAs, it is usually not the total emissions from the landfill that is of interest, but instead the specific emissions that can be allocated to the product under study. The allocation of the emissions from the landfill to the incoming products should be based on chemical, physical and biological (in short 'natural') causation as far as possible.
- In order to be comparable with other data in the inventory analysis, the emissions from a landfill need to be integrated over a time period. Two time perspectives will be considered; the surveyable time perspective and the hypothetical, infinite time perspective. The surveyable time perspective is defined as the time it takes to reach a pseudo steady-state, after which the changes are slower than during the initial phases. In the pseudo steady-state, the chemistry is only slowly changing in response to external changes and concentrations may be controlled by equilibrium reactions. The surveyable time perspective should correspond to approximately one century but needs to be further defined for each specific type of waste. The hypothetical, infinite time perspective is defined by a complete degradation and spreading of all the landfilled material. The spreading may occur by leachate, gas, erosion or by transformation of the landfill into a part of the environment.

The surveyable time perspective for municipal solid waste landfills is suggested to be the period until the later part of the methane phase when the gas production is diminishing [4, 6]. This is a time period which is in the order of magnitude of several decades or perhaps a century [7], largely depending on the climate.

## 2.2 Metals in municipal solid waste

### 2.2.1 Introduction

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. 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. The process can thus be divided into two steps; the release and the further fate. Either of these steps may be the rate determining step. If the first step is rate-determining, the emission rate will depend on the matrix in which the metal is landfilled, if the second step is rate-determining, the emission rate is independent of the origin of the metal.

(The above is partly a simplification since metals need not be truly dissolved to be transported. They can also be transported as mobile particles. This, however, does not change the reasoning except in one case: If metals are released as colloidal particles and then transported by the same particles, without ever being truly dissolved in the leachate, then the transport rate may be the rate determining step and at the same time depend on the origin of the metal. This special case will, however, not be further discussed.)

It has been earlier suggested that mass balance data (e.g. from [8–10]) can be used for estimating emission rates [4]. Similar approaches have also been developed by others [11, 12]. However, such an approach is based on the assumption that the emission rate is independent of the origin of the metal, i.e. the release from the matrix is not a rate determining step. In the treatment below, the mass balance approach is examined, and then the assumption that the release rate is not rate determinant.

### 2.2.2 The mass-balance approach

The emission factor as a function of time,  $E_j(t)$ , may be calculated by

$$E_j(t) = \frac{\int_0^t G_j(t) dt}{W X_j} \quad (2)$$

where  $G_j$  is the outflow of the element  $j$  from the landfill,  $W$  is the total amount of landfilled solid waste and  $X_j$  is the concentration of the element in the solid waste.

Based on field data from landfills with varying age, emission factors (called “transfer coefficients”) at a mean residence time of about 10 years were calculated by BACCINI et al. [8, 9]. This time period is rather short as representing the surveyable time period, but nevertheless the landfill seemed to be in the later part of the methane phase since the gas production was declining [8]. The emission factors were calculated for emissions by both gas and leachate. As far as metal emission is concerned, emission by gas is negligible in all cases except for Hg, and possibly Cd. These data are presented in Table 1 under column A.

If it can be assumed that the concentration in the leachate is constant during the integration time, equation 2 may be simplified to

$$E_j(t) = \frac{C_j A(t)}{X_j} \quad (3)$$

where  $C_j$  is the concentration in the leachate and  $A$  is the amount of leachate produced per amount MSW during the time  $t$ . If it is assumed that also the leachate production is constant, it may be calculated as the product of the amount of leachate per year and the number of years. EHRIG [10] assumed constant concentration in the leachate when calculating ‘transfer rates’. The latter may be recalculated to emission factors for the surveyable time period by multiplying with 30 years which is then assumed to be the time for significant gas production at these sites. The so calculated emission factors are presented in Table 1 under the column B.

A question which arises, however, is whether or not it is reasonable to assume constant concentrations during the surveyable time period. Since the conditions in the landfill are changing during the different degradation phases, it may be expected that the concentration in the leachate will also change. From a mass balance perspective the first aerobic phases, the initial phase and the oxygen and nitrate reducing phase, are generally too short to be of any significance. The acid anaerobic phase may, however, prevail for years, possibly decades, and are thus of relevance. Although many studies of leachate composition have been performed, relatively few have attempted to characterise the leachates as being acetogenic or methanogenic. Two studies are cited in Table 2, however, both being based on a number of samples from several landfills [13, 14]. It can be seen that for Fe, Mn and possibly Hg, the change is fairly large between the two phases. For Zn the change is approximately one order of magnitude. For the other metals, the change is smaller and the concentration in the two phases are within the same order of magnitude. It can thus be concluded that the constant concentration assumption for the surveyable time period is reasonable for some metals, but not all.

EGGELS and VAN DER VEN [12] also used a mass balance approach assuming constant concentrations during 30 years. Based on their data and equation 2, the emission factors presented under column C in Table 1 can be calculated.

The Tellus Institute [11] used a mass balance approach assuming constant concentrations during 55 years. The emission factors reported are very low, however, compared to other studies cited here, due to the calculated amount of leachate, 0,03 mm/yr which is low compared to, for example, the deposition of rain water. The leachate generation is typically 20 % of the annual precipitation [10]. If a leachate generation of 200 mm/yr is assumed, new emission factors can be calculated which are presented in Table 1 under column D. It can be noted, however, that in the study it was assumed that the landfill is active for 25 years followed by 30 years of post closure where the landfill is closed and capped

Table 1: Emission factors [kg emitted/kg landfilled] for the surveyable time period

Element	A (gas)	A (leach.)	B	C	D	E	F
Cd	$4 \cdot 10^{-6}$	$6 \cdot 10^{-5}$	$7 \cdot 10^{-4}$		$2 \cdot 10^{-4}$	$1 \cdot 10^{-4}$ – $7 \cdot 10^{-4}$	$5 \cdot 10^{-3}$
Cu	$2 \cdot 10^{-7}$	$2 \cdot 10^{-4}$	$1 \cdot 10^{-4}$	$8 \cdot 10^{-6}$		$1 \cdot 10^{-5}$ – $4 \cdot 10^{-5}$	$5 \cdot 10^{-4}$
Fe	$2 \cdot 10^{-7}$	$1 \cdot 10^{-4}$					
Hg	$4 \cdot 10^{-5}$	$6 \cdot 10^{-5}$	$6 \cdot 10^{-3}$			$2 \cdot 10^{-5}$	
Pb	$1 \cdot 10^{-7}$	$7 \cdot 10^{-5}$	$9 \cdot 10^{-5}$		$5 \cdot 10^{-5}$	$8 \cdot 10^{-6}$ – $2 \cdot 10^{-5}$	$1 \cdot 10^{-4}$
Zn	$4 \cdot 10^{-7}$	$2 \cdot 10^{-4}$	$4 \cdot 10^{-4}$			$5 \cdot 10^{-5}$ – $1 \cdot 10^{-4}$	$2 \cdot 10^{-3}$
Cr			$1 \cdot 10^{-3}$ – $2 \cdot 10^{-2}$	$3 \cdot 10^{-4}$	$3 \cdot 10^{-4}$	$4 \cdot 10^{-5}$	$7 \cdot 10^{-3}$
Ni			$5 \cdot 10^{-3}$			$3 \cdot 10^{-4}$	$1 \cdot 10^{-2}$
As				$2 \cdot 10^{-3}$	$1 \cdot 10^{-3}$		

A: Data from Belevi and Baccini [9], corresponding to a mean residence time of 9.4 years.

B: Calculated from data in Ehrig [10], corresponding to 30 years.

C: Calculated from data in Eggels and van der Ven [12], corresponding to 30 years.

D: Calculated from data in the Tellus packaging study [11] and assumptions, see text.

E: Calculated from data in White et al. [15] corresponding to 30 years.

F: Calculated from data in Lagerkvist [16] and assumptions, see text.

to minimise inflow of water, but leachate may still be produced. Thus, the leachate generation rate will be different in the first 25 years rather than the following 30 years. However, the leachate generation rate reported in the study is averaged over the 55 year lifetime. Thus, if this average generation rate is compared to the leachate generation rate for an operating landfill, the average generation rate will appear low.

Based on data in WHITE et al. [15], for metal concentrations in leachate, amount of leachate produced during 30 years, and concentrations in MSW, a new set of emission factors can be calculated which are presented under column E in Table 1.

In another context, LAGERKVIST [16] calculated “half-lives” of some elements based on Swedish data. By assuming a surveyable time period of approximately 50 years, emission factors can be calculated which are presented under column F in Table 1.

By inspection of the data in Table 1, it can be noted that the differences between the calculated emission factors are in some cases fairly large. This is not unexpected, however, since both concentrations in leachate and in the solid waste, as well as the amount of leachate produced per ton of MSW, are highly uncertain data. By taking the median of the emission factors in Table 1, the “best available data” can be estimated and these are presented in Table 3 for both the surveyable and the hypothetical, infinite time period. The uncertainties and implications are further discussed below.

(It was noted above that the constant concentration approximation does not fully hold for Fe, Zn and Hg. The data in Table 3 for these elements are, however, the same as the results calculated from BELEVI and BACCINI [9], which are not based on the constant concentration approximation).

Table 2: Leachate concentrations [mg/l] during different phases

Parameter	EHRIG (1983)			ROBINSON and GRONOW (1993)	
	Acetogenic	Acid and met <sup>a</sup>	Methanogenic	Acetogenic	Methanogenic
Fe	920		20		
Mn	20		0.6		
Zn	5.6		0.6	6.8	0.8
As		0.13		0.010	0.009
Cd		0.005		0.01	<0.01
Co		0.05			
Cr		0.28		0.12	0.07
Cu		0.06		0.07	0.07
Ni		0.17		0.23	0.14
Pb		0.09		0.30	0.13
Hg				0.003	<0.0001

<sup>a</sup> No difference between phases could be observed between the acetogenic and methanogenic phases.

### 2.2.3 Release rates

It was noted above that the calculation of the emission factors is based on the assumption that the release rate 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. Some cases will be discussed below.

I. If the concentration of the element in the leachate is determined by equilibrium reactions, the release process is not

Table 3: "Best available data" for emission factors [kg emitted/kg landfilled]

Element	The surveyable time period	The hypothetical, infinite time period
As	$2 \cdot 10^{-3}$	1
Cd	$5 \cdot 10^{-4a}$	1
Cu	$7 \cdot 10^{-5}$	1
Cr	$7 \cdot 10^{-4}$	1
Fe	$1 \cdot 10^{-4}$	1
Hg	$1 \cdot 10^{-4b}$	1
Ni	$5 \cdot 10^{-3}$	1
Pb	$6 \cdot 10^{-5}$	1
Zn	$2 \cdot 10^{-4}$	1

<sup>a</sup> Approximately 10 % by gas, the rest by leachate.

<sup>b</sup> Approximately equal amounts by gas and by leachate.

the rate-determining step. This is the case even if it is not the release process that is the equilibrium reaction. In these cases, it is the equilibrium concentration and the leachate generation rate which determines the emission rate.

II. Several heavy metals, e.g. Cd and Pb, are used as additives in plastics. One of the factors influencing the release rate will then be the degradation of the plastic matrix. When the matrix degrades, the metals present will be in contact with the aqueous phase. If this contact also leads to dissolution into the leachate, the release rate will be at least as high as the degradation rate. A degradation of 1–5 % during the surveyable time period, which can be assumed for several plastics [6, 17], will then lead to a release rate of 0.01–0.05 [kg released/kg landfilled] which is higher than the estimated emission rates. It therefore seems reasonable to assume that for metals used as additives, the release rate will normally not be the rate-determining step.

III. For metallic materials, the release process is corrosion. The release rate thus equals the corrosion rate. Data for corrosion rates in landfills is, however, very scarce. It has been suggested that data for soil corrosion may be used as a rough approximation [17] where data for anaerobic soils may be especially relevant. Besides general corrosion, microbiologically induced corrosion (MIC) is also of interest when nearly neutral water (pH 4–9), at 10 ° to 50 °C is in continuous contact, especially under stagnant conditions, with different metals such as carbon steel, stainless steel, and alloys of aluminium and copper [18]. In anaerobic soils, as well as in landfills, sulphate-reducing bacteria (SRB) will generally be present. SRB may, while reducing sulphates to sulphides, catalyse the oxidation (corrosion) of metals. Soils are therefore generally considered to be more corrosive with reducing (anaerobic) redox potentials, which favour the presence of SRB [18]. It is therefore suggested that, in general, corrosion rates in soils are lower than corrosion rates in landfills. Typical data for soils may thus be used as a low estimate of corrosion rates in landfills.

The most extensive field tests of corrosion in soils are reported by ROMANOFF [19]. Some of the results are sum-

marised in Table 4 [20]. Results obtained in other studies are similar [20, 21] and these corrosion rates can be compared to the emission factors. However, since corrosion is a surface reaction, a geometry of the corroding product must be assumed. The diameter of a sphere can be calculated for which the corrosion rate (from Table 4) is equal to the emission rate (from Table 3). For structures larger than the calculated sphere, corrosion is the rate-determining step. For Fe, this diameter corresponds to approximately 15 m. For Cu and Pb similar results can be calculated. Since such large structures are not landfilled as MSW, it may be assumed that for these materials, the release process is not the rate-determining step.

Table 4: Corrosion of different materials for given exposure times in soil, [20] based on [19]

Material	Average Corrosion (mg/dm <sup>2</sup> day)	Number of soils
Open Hearth Iron (12 year exp)	4.5	44
Wrought iron (12 year exp)	4.7	44
Bessemer steel (12 year exp)	4.5	44
Copper (8 year exp)	0.7	29
Lead (12 year exp)	0.5	21
Zinc (12 year exp)	3.0	12

For low alloy steels, the corrosion rate is somewhat lower, but still in the same order of magnitude [19]. For high alloy steels (Cr more than 18 % and Ni more than 8 %) the weight losses were, however, significantly smaller, although pitting corrosion occurred in most cases [19]. This makes it difficult to estimate the corrosion rate in MSW landfills. The corrosion process may be the rate-determining step for high-alloy steels (also depending on the geometry of the landfilled product). If so, the emission factors calculated above may be over estimates for larger products made in high-alloy steels. Stainless steel is the dominant use of Cr and Ni in Sweden [22]. It would therefore seem reasonable to propose that stainless steel is a significant source of Cr and Ni in landfills, although this is not in accordance with other studies [23]. For Cr and Ni in non-stainless steel products (such as plastics and paper), the calculated emission factors (→ Table 1 and 3) may thus be underestimated.

### 3 Discussion and Conclusions

Based on the discussion above, it seems reasonable to assume that in general the release process will not be the rate-determining step, 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 estimated and can thus still be used for conservative calculations.

It was noted above that the 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. By consideration of the range in Table 1, and the uncertainties in the data used for the calculations, it is suggested 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. [3], 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 is regarded as a "black box": Nothing needs 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. For larger changes, the landfill processes, and therefore also the emission factors may change.

When calculating the emission factors, only the landfill process has been considered. Collection and treatment of leachate is therefore not included here. Assumptions on collection efficiencies vary in different studies, e.g. 98 % [12], or 70 % [15].

It is important to note that the emission factors for the surveyable time period ( $\rightarrow$  Tables 1 and 3) 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 after the methane-phase, metal concentrations in the leachate may rise again [16, 24]. This implies that the environmental impact 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.

Although the emission factors in Table 3 are uncertain, they may be useful for estimating emissions from landfills in initial ("screening") LCAs, where the aim is to identify key issues, i.e. important aspects of the system under study.

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