Excerpt "landfill models" from the ecoinvent 2000 report on landfills

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The following text describes the model choices made for the ecoinvent 2000 landfill models. It is an excerpt of part III of the ecoinvent 2000 report No. 13 on landfills cited below. The full report is available to ecoinvent 2000 suscribers.

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Full report:

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5 Systems Characterisation

5.1 Introduction

Until the middle of the 20th century European urban solid wastes were collected and usually discharged to uncontrolled and unordered waste dumps. Concerns regarding water pollution, odorous air emissions, occurrence of fire and smoke, landscape protection, occurrence of vermin lead to laws prescribing ordered landfills in many countries.

Space restrictions but also pollution concerns and lacking public acceptance of new landfills made it difficult in the last twenty years to put up new landfill sites. Some countries, e.g. the Netherlands and Switzerland, have therefore mandated that no burnable wastes shall be landfilled directly, but only by way of incineration for waste volume reduction[4].

Switzerland knows three types of landfills. These landfills are regulated by the technical ordinance on waste (TVA 2000).

- Sanitary landfills (municipal waste landfills, organic landfill, Swiss German: 'Reaktordeponie')

- Residual material landfills (Swiss German: 'Reststoffdeponie')

- Inert material landfills ('cleanfill', Swiss German: 'Inertstoffdeponie')

Residual material landfills can be stand-alone sites or can be realised as compartments of sanitary landfills. Similarly there exists a *fourth kind* of 'landfill' that is only realised as a compartment of a sanitary landfill. These are the so called 'slag compartments' that only contain slag (bottom ash) from municipal solid waste incineration (CH German Schlackekompartiment).

While sanitary landfills can receive all kinds of biologically reactive wastes, the other types of landfills receive inorganic wastes with a varying extent of harmful contaminants. This material must be deposited in a physically stable form. Sites for all landfill types must avoid sites of high risk of landslide or flooding. Actual or potential groundwater utilisation has priority against landfill projects. All landfilled waste must be inventoried by the landfill operator. Controlled landfills in Switzerland usually feature a base and boundary sealing[5]. They feature a slanted base so gravitational water removal is secured. Sealed landfills also need to have a water collection system to secure pollutant monitoring and possible water treatment. After closure the landfilled must be monitored for several years and the surface must be restored either as agricultural or natural area (TVA 2000).

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[4] Germany plans to introduce such legislation for 2005.

[5] For Swiss inert material landfills a seal and leachate collection system can be excluded only if they are located outside of ground water zones for drinking water.

Fig. 5.1 **A new and empty landfill site with visible bottom seal sheet (left) and waste placement and compaction in a sanitary landfill with special loader (right)**

5.2 Sanitary landfills

Sanitary landfills are the most demanding type of landfill. They are designed to receive all kinds of waste: untreated municipal or household waste, building wastes, wastewater treatment sludge. If local capacities lack, they can also receive inert wastes or, pending special approval, hazardous wastes. Not allowed are liquid, infectious or radioactive wastes or explosives. No limiting requirements for the landfilled waste regarding composition or extraction tests results are defined. The landfill design must include base and boundary sealing, a water collection system and also a gas collection system due to the biologically reactive nature of the waste. Landfill gas is usually incinerated or converted to electricity. Restoration of the landfill site and a post-closure monitoring period of at least 15 years is mandatory. After a five-year transition period, landfilling of burnable wastes was prohibited in Switzerland in 2000[6].

For sanitary landfills several stages of development can be discerned.

1. Initial phase: The waste settles and is moistened up. Hydrolysis of hydrocarbons starts.

2. Oxygen and Nitrogen reducing phase: The remaining oxygen is used up in oxidising reactions. Nitrate and ammonia act as oxidising agents and are reduced to elemental nitrogen (N_2). Redox potential E_h is falling rapidly. Duration: first two weeks after waste placement.

3. Acidic anaerobic phase: Free oxygen is depleted (anaerobic conditions). Decomposition of hydrocarbons generates organic acids lice acetate and propionate causing the pH to drop temporary to 4 or 5. Metals are mobilised. Duration: First two months after waste placement.

4. Methane generating anaerobic phase. Further decomposition of hydrocarbons to CO_2 and methane (CH_4) as landfill gas. Carbonic acid (H_2CO_3) buffers the pH at 6 to 7. Continuos low redox potential. Temperature rises up to 120°C. The methane phase has an onset time of about 2 years and can last for 30 years.

5. Continuous leaching. While the landfill pH is buffered by acid neutralising materials, the landfill chemistry is in a quasi-steady state. Constant but comparatively low concentrations in leachate lead to continuous emission of pollutants from the landfill. The duration of this phase depends on the acid neutralising capacity of the landfill material and can last thousands of years.

6. Possible pH decrease. If the acid neutralising capacities of the acid buffer systems are used up by externally imported, or internally produced acids or buffer wash-out the pH of the landfill can fall sharply and formerly stable phases can be remobilised. Pollutant concentrations can rise significantly again.

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[6] However, in 2000 about 390'000 tons or 20 w-% of the waste landfilled in sanitary landfills was still burnable waste, but with a decreasing trend (BUWAL 2001e).

7. Humification and Reoxidation If the landfill has become unreactive, humification processes transform the landfill to a podzol (soil type). Intruding rain water oxidises the environment and redox potential E_h rises again. Precipitated metal sulfides can be oxidised, which lowers pH and increases metal mobility.

5.3 Inorganic landfills (slag, residual and inert material)

5.3.1 General description

'Inorganic landfills' is an ad-hoc term for landfills with mostly inorganic, low-carbon wastes, like incineration residues or inorganic building materials. Slag compartments, residual material landfills and inert material landfills can be regarded as inorganic landfills. In Switzerland, the total organic carbon concentration in the landfilled waste is legally regulated to be below 5 w-% in such landfills.

Fig. 5.2   **The Swiss residual material landfill Oulens VS. The blocks of solidified incinerator filter ashes are visible**

Compared to the intruding rain water, landfill bodies represent a high concentration deposit of several pollutants. The resulting concentration gradient between water and waste is a driving force for the continuous dissipation of pollutants into the leachate. The speed of dissipation depends on the dissolution and desorption characteristics of the involved materials and the geochemical stability of the mineral phases. Some incineration residues are thermodynamically unstable due to high temperature during incineration. This too, is a driving force of continuous change in the landfill. In time, landfill bodies undergo several mineralogical transformations. Over long timespans of hundreds or thousands of years, weathering processes can fundamentally change the mineralogical makeup of the landfilled material.

Following phases in an inorganic landfill can be discerned after waste placement:

1. Initial phase: The waste settles and is moistened up. Readily soluble salts like chlorides, sulfates, sodium and potassium are dissolved in the pore water and are mobile to be washed out with the leachate flow. The waste undergoes fast transformations especially in the humid environment of a landfill. Transformations consist of e.g. the corrosion and oxidation of metallic phases in fresh slags[7],

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[7] Zeltner (1992) finds that over 80% of the metallic iron in landfilled MSWI slag is oxidised to several types of iron oxides within 5 to 10 years.

hydration of metal oxides like CaO or MgO, and carbonation of hydroxides with CO_2 from infiltration water to carbonates[8]. Also glassy phases are immediately starting to hydrolize (Frühwirth et al. 2000:149). All these reactions are exothermic and can lead to significant temperature increases in the landfill[9]; see Fig. 5.4. Even in 10 year old slag landfills temperatures of 80°C could be observed (EKESA 1992). Reactions between amorphous glass phases and alkaline leachate creates secondary mineral phases (CASH phases = calcium aluminium silica hydroxides). Dissolution of calcium hydroxide creates a high pH (Frühwirth et al. 2000:149). Over time the pH, that can initially be above 12, drops continuously. Below a pH of 10, microbial activity can decompose part of the remaining organic carbon traces (Lechner 2001). Until the pH of 8.3 of the next phase is reached, up to 50 years can go by (Kraxner et al. 2001:18).
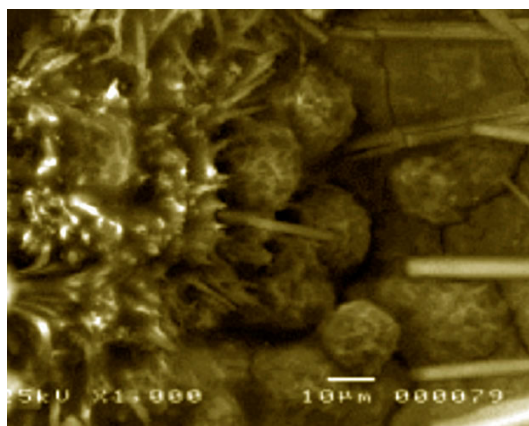

Fig. 5.3 **Example of weathering products in MSW bottom ash. Calcium silicate hydrate CSH on the left and calcium aluminium silica hydroxides CASH on the right (Kraxner et al. 2001:72).**

2. Carbonate buffer phase: The pH of the landfill pore water is buffered by the presence of carbonates to a value of around 8.3. As the pH is a master variable for landfill development, the landfill is in a quasi-steady-state during this phase. Depending on the acid neutralising capacity ANC, the calcium content, the quantities and pH of the intruding rain water, this phase will last for some 1000 to some 100'000 years. During that phase, natural weathering processes can transform minerals into thermodynamically more stable forms. A sequence of secondary mineral neoformations and transformations oxide→hydroxide→carbonate→silicate→... takes place (Sabbas et al. 2001). Carbonation of hydroxides, mentioned in phase 1, continues in this phase. The vitrified phases in slag can be transformed to more accessible clay phases by long-term weathering (Zevenbergen et al. 1995, Zevenbergen 1998). The liberated heavy metals from such phases might be bound again in secondary calcium aluminium silicates and -hydroxides (CASH phases) (Huber et al. 1996:5).

3. Possible pH decrease. If the acid neutralising capacity of the carbonate buffer system is depleted, the pH of the landfill can fall sharply below 7. Silicates, aluminium- and iron (hydr)oxides could buffer the system and affect the mobility of heavy metals (Sabbas et al. 2001). Formerly stable phases – e.g. CASH phases – can be remobilised. Pollutant concentrations can rise significantly again. A pH drop to a value of 4 must be considered as a worst case scenario (Huber et al. 1996:5, Johnson et al. 1995).

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[8] Similar to the hydration and hardening in cement: $CaO + H_2O = Ca(OH)_2$; $+ CO_2 = CaCO_3 \cdot H_2O$.

[9] The high temperature can damage landfill seals or lead to incrustations in leachate collection pipes by increased evaporation. These reactions can be enforced before landfilling, e.g. by slag quenching with water after incineration or by extended surface storage prior to landfilling. Even after water quenching, the temperature can reach 60°-70°C in surface stored slag heaps (Huber et al. 1996:4)
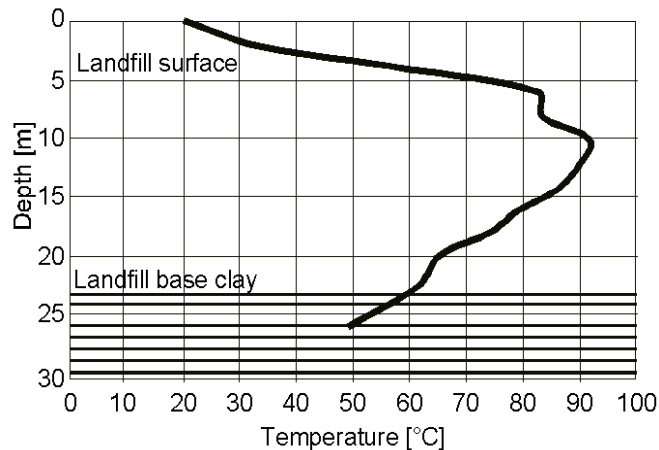
Fig. 5.4   Temperature measurement along a drilled hole in a slag landfill (Turk 1995)

The duration of carbonation ($MeO + CO_2 \rightarrow MeCO_3$) depends on the metal oxides present, the available carbon dioxide and the landfill depth. Frühwirth et al. (2000:182) calculates for a 15 m deep slag landfill a carbonation duration of 30 years, while the carbonate buffer phase lasts 5000 years.

Huber et al. (1996:91ff.) show that for MSWI slag the dominant loss of acid neutralising capacity occurs not through in-flow of protons in acid rain or acid producing internal reactions, but from the wash-out of buffering carbonate minerals.

Hellweg (2000:79) makes thermodynamic calculations of the reactions in a slag landfill body and finds the carbonate buffer is washed out after 56'000 years, using a atmospheric CO_2 partial pressure of $10^{-3.5}$ atm (316 ppm). In natural soil the CO_2 partial pressure is usually higher (10^{-2} atm, 10'000 ppm) due to decomposition in the detritus layer. Rain water intruding in landfills can be exposed to higher CO_2 partial pressures, when flowing through the vegetated landfill cover. Sensitivity calculations with a CO_2 partial pressure of 10^{-2} atm induced a threefold increase of carbonate washout and the carbonate buffer phase ends after 17'000 years. Also heavy metals like Cd, Zn, Pb, Cu were washed out 3 to 5 times faster during the carbonate buffer phase due to the lower pH.

5.3.2   Slag compartments

As explained above, slag compartments are not stand-alone sites but are separated parts of sanitary landfills. A separation that prohibits contact with other municipal waste is mandatory. Compartments are therefore not separate from sanitary landfills in a logistic sense, but they are separate in a physical and chemical sense. Landfill design requirements are the same as for sanitary landfills. Slag from MSWIs shall not have more than 3 w-% organic carbon (TOC). It is legally possible to recycle slag in road or dam constructions (TVA 2000, Art. 13). However, in the current Swiss practice all MSWI slag is landfilled (BUWAL 2001f).

5.3.3   Residual material landfills

Residual material landfills can receive wastes that are low in organic carbon (< 5 w-%) and are not biologically or chemically reactive in water. Less than 5 w-% of the pulverised waste shall be soluble when mixed with 10 parts water. Additionally, the waste must pass two extraction tests regarding

several parameters[10]. Residual materials landfilled are mainly filter ashes from municipal waste incineration and complying industrial wastes. The MSWI filter ashes are solidified with cement to comply with TVA extraction tests for residual material. A post-closure monitoring period of at least 10 years is mandatory.

5.3.4 Inert material landfills

Inert material landfills can receive unproblematic inorganic wastes. These wastes need to consist to at least 95 w-% of rock-like or mineral material like silicates, carbonates or aluminates. The waste composition must not exceed limits set for lead, cadmium, copper, nickel, mercury, and zinc. Less than 0.5 w-% of the pulverised waste shall be soluble when mixed with 10 parts water. The waste must pass two extraction tests regarding essentially the same parameters as for residual material landfills, but the limits are generally set 10 times lower. Inert wastes landfilled are clean or tolerable excavation material, road foundations, road tarmac (excluding old, tar-containing asphalt) and building wastes like concrete, tiles, bricks, glass, gypsum, asbestos-reinforced cement (BUWAL 2000). A post-closure monitoring period of at least 5 years is mandatory.

5.4 Underground deposits

Underground deposits are ordered waste storage spaces that are usually created in old potash or rock salt mines. No underground deposits exist in Switzerland, but some Swiss waste is exported to the German salt mines Herfa-Neurode and Heilbronn. Underground deposits are monitored and separated from the conventional salt mine part. Wastes are stored in steel drums, large steel containers or big bags.

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[10] The limit values to respect are set for aluminium, arsenic, barium, lead, cadmium, chromium III and VI, cobalt, copper, nickel, mercury, zinc, tin, ammonia, cyanides, fluorides, nitrites, sulfites, sulfide, phosphates, dissolvable organic carbon DOC, biochemical oxygen demand BOD_5, hydrocarbons, lipophilic non-volatile chlorinated hydrocarbons, chlorinated solvents and pH value.

Fig. 5.5   Separation of one completed compartment by a brick wall in the underground deposit Herfa-Neurode. At the ceiling the rock salt vein is visible[11]

In Germany four underground deposits for non-radioactive wastes exist[12]; all in old potash or rock salt mines. In 1999 these deposits received 226'000 tons of hazardous wastes like incineration ashes from coal power plants and MSWIs, wastes from metal-producing and chemical industry, contaminated soils and dismantling wastes, electronic components like transformers and condensers (Borsch 2001). In Herfa-Neurode approximately 80'000 tons of hazardous waste are deposited annually in a depth of 700–800 meters and over an underground area of 20 km^2 (Brendel 2000).

Since 1999, a new deposit also exists in Wittelsheim (France-Alsace) operated by StocaMine with a licensed capacity of 320'000 tons of waste. A new salt mine deposit by Minosus Ltd. in Winsford, Cheshire, United Kingdom, seeks official licensing since 1999, but meets continued opposition by adjacent residents.

In Germany there are also 20 coal mines where the exploited veins are filled up with material to mechanically stabilise the shafts (German 'Berg(e)versatz', also called 'mine-valorisation'). The material used is excavation material but since 1991 also hazardous wastes like filter ashes is mixed in. No monitoring or ordered storage of the deposited materials is required by law. Since the material is used to stabilise the mine, this type of disposal can be declared as 'recycling' (Jansen 2000). This controversial practice is not under consideration here[13].

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[11]   Picture from the operator of Herfa-Neurode 'Kali + Salz GmbH' http://www.k-plus-s.com/medien/images/herfa-neurode.jpg . More pictures of Herfa-Neurode can be seen at http://www.ks-entsorgung.com or in the preview section of http://www.photoweb.de.

[12]   1. Herfa-Neurode in Hessen (operating since 1972), 2. Heilbronn in Baden-Württemberg (since 1987), 3. Zielitz in Sachsen-Anhalt (since 1995), 4. Borth in Nordrhein-Westfalen (since 1997). A fifth deposit Riedel, near Hannover in Niedersachsen in the process of licensing (Borsch 2001).

[13]   Germany has accepted an ordinance that demands the same management standards for coal mine stabilisation with hazardous waste as for salt mine underground deposits. The ordinance is effective as of October 2002 (BMU 2002).

Only orderly disposal in underground salt mines – where some Swiss hazardous wastes are deposited – is inventoried here. Orderly disposal includes the record-keeping of waste samples, the full documentation of waste composition and location, which allows retrieval. Indeed, some wastes like disodium cyanamide, calcium formiate and arsenic trioxide have already been retrieved from Herfa-Neurode and used as secondary raw materials in the chemical industry (Brendel 2000). 7000 transformers containing PCB are stored separately, as it is expected that they can be recycled for their copper content in the future (WEKA 1993).

5.5   Emissions from landfills

Emissions to air and water from landfills can be separated into indirect emissions and direct emissions. Indirect emissions occur from the production of used materials and fuels. Direct emissions can be divided into emissions from landfill operations like transports, waste spreading and landfill shaping, and emissions that originate from the waste itself.

Emissions from the waste are usually the most important burden within the LCIA result of a landfill. Depending on the waste composition, waste reactivity and degradability the waste material is transferred to the landfill gas and the water outflow from the landfill (leachate). For reactive wastes in a sanitary landfill gas production can occur up to several decades. Leachate is produced continuously in landfills with permeable top-cover. In landfills with impermeable top-cover ('dry tombs') leachate is produced (again) after the top seal has been rendered unfit through ageing[14]. In any case leachate is usually produced continuously over an essentially unlimited timeframe.

Leachate can contain significant amounts of pollutants washed from the waste material in landfills. The temporal dynamics of these emissions are difficult to predict over such long timespans, but are governed such parameters as pH-value[15] and redox potential (E_h) in the landfill body as well as the chemical composition, chemical speciation and mineralogy of the waste. Usually landfills develop a quasi-stable emission level after an initial settling phase. During that settling phase emissions can be high and need to be monitored.

The pH value development of a landfill depends on the acid neutralising capacity ANC of the landfilled waste, which is diminished by

-   rain water inflow containing acidifying pollutants (NO_x, SO_2, HF etc.) or carbonic acid (HCO_3^-) from atmospheric CO_2,

-   the acid produced during the degrading or weathering of the waste and

-   the washing out of buffering materials.

Once the acid neutralising capacity is used up, which can be after thousands of years after landfill closure, the pH drops sharply and the landfill turns acidic. This change in acidity leads to an increase in solubility of formerly stable phases containing heavy metals and consequently to large emissions in the leachate. This scenario is likely to occur sooner in sanitary landfills containing biologically

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[14]   Landfills that receive a top seal after closure ('dry tombs') are initially closed-systems, but will become open systems after the failure of these technical barriers and penetration of rain water (Lee & Lee 1999). Technical barriers are bound to become faulty after decades or a few hundreds of years (Lee & Lee 1999, Covelli & Baumann 2000:6). So technical barriers only achieve a delay of emissions, in the case of dry tombs with a high risk of sudden and grave deterioration after centuries of eventlessness, when the dry environment is activated by water intrusion. Dry tombs are not common practice in Switzerland.

[15]   Lysimeter experiments with MSWI slags showed that pH is the most important influence on the leachate concentration of Pb, Cd, Zn, Ni, V, Co, Mn, Al, Ca, Mg and can cause changes of a factor of over 100, while sulfate, B , Si and easily soluble salts show no pH dependency. Other tested influences were liquid/solid-ratio, grain size, ageing, chemistry changes through drying (Kraxner et al. 2001:100).

reactive wastes, and later in the inorganic landfills (slag compartments, residual material landfills, and inert material landfills).

In a less dramatic scenario the acid neutralising capacity can remain positive and no dramatic increases in emissions occur over time. Nevertheless, normal weathering and gradual changes in the redox chemistry of the landfill (aerobic regime) can turn stable mineral phases into more soluble phases and subsequent wash-out (Huber et al. 1996:90). Results from leaching or availability test are used in environmental studies to gain information on the leaching behaviour of waste materials. Such tests typically do not last longer than hours or days. In such a short time it is not possible to simulate the weathering of the constituting minerals that occurs over long timespans (see e.g. section 'Heterogeneity in material' on page 32). Current leaching tests can therefore merely give an indication of the leachability of the *current* waste material or for a short term phase, but not of the *complete* long-term leachability of a material altered in time[16]. Also legislative leaching tests for landfill in Switzerland and the EU are performed on fresh, not weathered samples and are therefore are merely a test for the short-term behaviour of those wastes (Kraxner et al. 2001:17)[17].

The continuos throughput of leachate water secures the open-system character of a landfill after closure. In that manner, possibly small concentrations of pollutants can be gradually washed out from the landfill during very long timespans, usually following an initial settling phase. In the following section, I shall discuss the problems and solutions of integrating such long-term effects and emissions in life cycle inventories.

5.5.1   Future emissions in LCA

Previous studies on landfills indicated that the pollutant potential remaining in a landfill after one hundred years after waste placement is significant (e.g. Zimmermann et al. 1996, Sundqvist et al. 1997, Hellweg 2000, Doka et al. 2002). Landfill experts generally agree that regarding long-term behaviour landfills cannot be regarded as stable systems, even if catastrophic events like landslides or floods are excluded (Leuenberger 1999, Sabbas et al.1998, Huber et al. 1996, Lechner 2001). The designated barrier systems – 'inertisation', solidification, sealing sheets etc. – deteriorate in time and have a limited functional lifetime. There is a relevant and plausible potential that the remaining pollutant load in a landfill will be *completely* released, *if long enough timespans are considered*.

In other words, only a minor part of the harmful substances contained in waste have been released to the environment after one hundred years. From a LCA point of view, landfills postpone emissions from todays wastes into the future. This remaining future pollution potential must not be ignored, since life cycle assessment is concerned with creating a complete synopsis of environmental burdens attributable to a process, *wherever* or *whenever* they occur.

Inclusion of long time horizons in life cycle inventories and in LCA is problematic and needs special understanding. First, a choice of the time frame of the assessment has to be made. Second, the future emissions must be predicted using a landfill model, because no measurements can be made in the far future. The choice of the temporal system boundary is a relevant key question, because it influences to

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[16]   Such tests were used to estimate long-term emissions in former waste LCI studies (Zimmermann et al. 1996, Hellweg 2000). This method is not continued in this study. An alternative idea was to use sequential extraction data which allows to discern different phases in waste materials, e.g. easily dissolvable, carbonate phases., metals bound to amorphous FeO_x/MnO_x, metals bound to crystalline FeO_x, organic fraction and metal sulfides, metals in crystal lattices/silicates (see Huber et al. 1996, Frühwirth et al. 2000). This idea was abandoned for the same reason that sequential extraction is unable to simulate future material changes due to weathering, which turns all mineral phases into leachable phases (Personal communication with A.C. Johnson, EAWAG Dübendorf, September 12, 2002).

[17]   Kraxner et al (2001) validate several leaching methods for waste materials, which also include methods with *enforced artificial weathering*.

a large extent the type of landfill model. In the next section I will look at the problem of temporal system boundaries.

The default procedure for future emissions in LCA

Within the framework of LCA it has been common practice to neglect space or time issues. For example, ISO 14042 states in chapter 8: *"LCIA typically excludes spatial, temporal, threshold and dose-response information, and combines emissions or activities over space and/or time."* (ISO 2000). This implies that the mentioned types of information are also disregarded in the LCI stage of LCA, i.e. emissions are inventoried *regardless of their time of release*. All emissions are treated as if they would be released at the reference point of time[18] and within one reference geographic region[19]. LCIA practice currently lacks the tools to model damages to *future* environments from future emissions. Hence, as the default procedure in LCA, future emissions are included in the assessment without any kind of weighting and are treated just like short-term emissions[20].

Alternative procedures: temporal discounting of future emissions

Besides this default procedure of treating future emissions just like present emissions, alternative procedures of dealing with future emissions are imaginable. It is possible to differentiate and weigh future emissions differently in the assessment than present emissions. Weighting future emissions *lower* than present emissions is called positive temporal discounting. Weighting future emissions *higher* than present emissions is called negative temporal discounting. For both types of discounting and the default procedure (no discounting) several arguments can be put forward. These arguments are discussed at large in (Hellweg 2000, Hellweg et al. 2002, Doka et al. 2002). Some of the most memorable arguments for either procedure are given in the following paragraphs.

Some arguments for positive temporal discounting: In economics, discounting future monetary benefits is done, e.g., because an individual might have a pure time preference. He/she rather has the profits now, than later, possibly by mere impatience. Also economists usually presume a growing economy with continuously increasing productivity of capital. This makes current benefits more valuable than the same nominal amount of benefits in the future. Another reason for positive temporal discounting can be that the further we go into the future the more the individual might be unable to take advantage of these profits because there is an increasing probability that the individual has died[21] (Hofstetter 1998). By symmetry, future *monetary costs* can also be subject to discounting. For example, costs that arise in the future are less serious than the same costs today. By this reasoning it can be argued, that future *environmental burdens* are less bothersome than present burdens.

Some arguments for negative temporal discounting: One could argue that future generations might be compensated financially for the environmental impact. This compensation should be high enough to satisfy those damaged. The necessary money could be invested on the capital market. However, considering that environmental goods and resources might become very scarce while monetary wealth might increase in the future, this compensation cost could be very high, even approaching infinity. In

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[18] The reference time point is usually the present with the current environmental state.

[19] Currently, the reference geographic region depends on the LCIA valuation method and can be e.g., Switzerland, Netherlands, or Western Europe.

[20] However, also encountered are hard *temporal cut-offs*, i.e. all emissions after a certain time span, e.g., 150 years, are completely neglected, which is a form of temporal discounting. But, within the considered period usually no temporal discounting is performed.

[21] This argument can also be stated in the forms that every generation has its own problems, and our generation should only be bothered with its own problems.

other words, people who do not lack money might demand a very high compensation for an additional risk to their health.

The annual pollutant loads in a future environment are bound to be higher than currently. This leads to higher burdens per kilogram pollutant than today, i.e. increased LCIA classification factors for future emissions (Hellweg 2000:141ff.).

Another reason for positive discounting can be that future releases often represent an uncertain and/or poorly manageable risk. A *risk avoiding* personality (e.g. the Egalitarian archetype) will find a future and uncertain release less desirable than a present and certain release, and therefore weigh present emissions higher than future emissions.

Some arguments for no temporal discounting: Within the framework of sustainability, we should not consider damages to future generations less important than damages to the present generation. Doing so would undermine the notion of sustainability. Also applicable is the 'polluter pays principle' as decreed by Swiss environmental law (BUWAL 1986): We have the use of the utility, we cause the damage, we should be held responsible for the *full* extent of the caused pollution. Obviously, this notion is also the core concept of life cycle assessment.

Discussion of temporal discounting

Whether discounting of long-term emissions is appropriate, depends on the goal of an LCA as well as the subjective preferences expressed in the impact assessment LCIA. In general, LCA is a tool to develop less environmentally burdening products and services or optimise ecological efficiency. The motivation for that is the insight of producers or consumers that present consumption levels in developed countries are unsustainable: Consumption at present level with current impacts could not be sustained for an indefinite time. The notion of sustainability includes the concept that the present generation should not fulfil its needs while jeopardising the means for the future generation to fulfil their needs. LCA is a tool for the environmental part of this sustainability discussion by pointing out less burdening options. **The mere shifting of burdens into the future shall not be regarded as a sustainable solution of environmental problems[22].** All of the possible arguments put forward to argue in favour of positive temporal discounting are in obvious disregard of this understanding of LCA. A pure time preference ('impatience') leading to positive temporal discounting implies that future people do not have equal rights as present people (Finnveden 1997). Furthermore, it *"is in the egoistic interests of present persons, those responsible for creating the waste, not to bear the consequences of their actions and instead to force those consequences on others who do not deserve them. Such an egoistic position is not ethically defensible"* (Shrader-Frechette 2000). LCA aims at displaying potential damages to *create a motivation* to reduce the risk of those damages actually happening. So, LCA needs to *show* those damage potentials and not conceal them by anticipating that

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[22]   Let's consider two alternative product options: Product A produces 90 burden points in the short-term, and an additional 10 burden points in the long-term future. Product B produces 50 burden points in the short-term, but an additional 450 burden points in the long-term. If we adopt a *short-term perspective*, product B with 50 points seems preferable over A with 90 points. Product B successfully transfers the major part of its attributable burden into the future. In a long-term perspective there is no reason to discount future emissions, if these emissions are really attributable to a product option, and product B with 500 points is found to be more burdening than product A with 100 points. For waste processes the release of waste components is clearly attributable to the waste product and there is no reason in LCA to discount those emissions from a disposal process. A short-term perspective and discounting of long-term emissions favours processes and products which shift burdens and blame to future generations. This cannot be regarded as a sustainable conduct.

others will have to deal with the problems[23]. Otherwise, incentives to decide in favour of less burdening technologies/products would disappear (Steen 1999).

It can be concluded that the LCA of landfill processes should generally also include long-term emissions, since the exclusion of those emissions is usually not defensible within the ecological sustainability discussion and the overall understanding of LCA as a sustainability tool. Due to the possibly controversial perception of long-term emissions it is a good idea to inventory long-term emissions separately. This allows for sensitivity analysis of LCA results with and without long-term emissions.

Timeframes in this study

Within the framework of this inventory study, long-term emissions are modelled and included in the inventory of landfilled wastes. To have a rudimentary distinction of short-term versus long-term emissions, emissions that occur *after 100 years* after deposition are inventoried in a special emission subcategory (*'water emissions, long-term'*), while emissions that occur before 100 years are inventoried in the same subcategory as present emissions.

> **Emissions from landfills are split up into emissions that occur in the first 100 years after waste deposition and emissions that occur afterwards.**

The point of 100 years is an arbitrary choice. It includes the complete or larger part of the surveyed periods[24] of landfills. It coincides with the choice made in studies of the Department of Systems Ecology at Stockholm University (e.g. Finnveden 1998, Sundqvist et al. 1997). The *uniform* separating time mark of 100 years is a deviation of the choice made in former LCI studies of ETH Zürich (Zimmermann et al. 1996, Hellweg 2000, Doka et al. 2002) where the *variable* surveyed periods of the different landfills were used to separate short-term emissions from long-term emissions (i.e. 40, 75 and 150 years, respectively. See footnote 24).

Cut-off at background concentrations?

It could be argued that pollutant concentrations in leachate become very small over time and have little ecological relevance. Following this argument, leachate emissions which, for example, fall below some natural background concentration value should not be inventoried, since they are assumed to have no ecological relevance. Following objections to this view can be put forward.

- The notion that concentrations observable in nature are benign and can be used as a cut-off criteria of relevance should be challenged as a whole. If, in a hypothetical situation, humans would pollute all rain water only *exactly up to* the background concentration, the pollutant load in water would double, since the anthropogenic pollutants would *add* to the actual natural background. It is not

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[23] It can be imagined that a human society in the far future will have the means to clean up the environment at virtually no cost because of large advances in technology, e.g. with nano-robots. Todays long-term burdens are then of no or little consequence, because they can be easily avoided by intervention and remediation. This can be regarded as science-fiction, because such technology does not exist today and the capabilities of future societies cannot be guaranteed. On the other hand long-term leaching is the normal development of landfills, which is not doubted by any landfill expert, and was observed e.g. in 2000 year old roman landfills (Lee & Lee 2000). There is little justification in disregarding a highly likely process on the grounds of a pure speculation.

[24] Surveyed periods are the lengths of time a landfill's emissions are actively monitored by the local government. Surveyed periods are *at least* 10 or 15 years (TVA 2000) and are estimated to be 40, 75 and 150 years for residual material landfills, slag compartments and sanitary landfills, respectively (Zimmermann et al. 1996:B.13).

guaranteed that this doubling of load would still be benign, but the understanding presented above would perceive no harm and no signal in an according LCA would be generated.

- In LCA, generally no concentration or dose-response information is heeded, i.e. flows are inventoried in the form "0.3 kg Cd per functional unit" and not as "0.3 kg Cd (in a concentration of 0.5 mg/l) per functional unit" (ISO 2000, cf. also section 'The default procedure for future emissions in LCA' on page 15). This makes sense in LCA, as the *sum total* of *all* the contributions of a pollutant flux (within a region and during a certain time) determines ecosystem and human health states[25]. This annual total of emissions determines the ecological impact, *irrespective if this total is made up from a few large or many infinitesimally small sources*[26]. Critical loads can be reached by many diffuse, low-concentration sources, too. In LCIA the total contributions to a burdened region are valuated (actual load or working point). Hence, there is no justification to exclude emissions just because they are small.

- The notion that small enough concentrations are not damaging (Paracelsus principle) is factually true on an individual organism level. But including it in an LCA would signify that diluting emissions with a large enough amount of media (air, water, soil) would solve environmental problems. This would send the entirely wrong – even illegal – message to decision makers. As mentioned above, LCA has a larger ecosystem view and not only an individual organism or local view.

- It has been pointed out by (Sundqvist 1998) that it may be irrelevant to use today's acceptable levels as a cut-off, since the environment may be more or less sensitive in the future due to a changing background contamination.

- Even if the argument would be accepted, and concentrations below an acceptable or natural background level would be ignored in the inventory, little would change in the outcome. An exponential decline model could be used to predict the concentration development of the landfill leachate (see section 'Short-term emissions' on page 21). The levels in unpolluted surface waters can be used as the natural background concentrations[27]. The time t when a certain leachate concentration c_x is reached, can be calculated according to Eq. 5.1 below (definition of variables see section 'Short-term emissions' on page 21). Time spans until geogenic background concentrations are reached in slag compartment or residual material landfill leachate, are in the order of thousands of years for highly soluble elements like C, S, N, but hundred thousands to *tens of millions* of years for heavy metals like Zn, Pb, Cd, Cu. If a timeframe were chosen, that would end, when *all* elements have reached their natural background concentration, *more than* 94% of the theoretically mobilisable fraction of an element would have been washed out already (smallest share is for Pb). In other words, heeding the 'natural background cut-off' argument – and ignoring its incompatibility with LCA methodology – would decrease the long-term emission potential *at best* by only 6%. With uncertainty factors in the heavy metal content of landfilled waste which can easily be above 300% (GSD[2]), these 6% would influence the overall result only negligibly.

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[25] This is the consequence of the **I=PAT** formula by (Holdren & Ehrlich 1974), i.e. '**I**mpact on the environment' [pollutant/year in a region] equals the multipication of '**P**opulation' [persons/region] times '**A**ffluence' [consumed products/person*year] times '**T**echnology' [pollutants/product].

[26] On a *local* scale and for assessment of *acute* damages, high density of large polluters can be relevant. Current LCIA methods presently look at chronic effects occurring at current background levels (dose-response working point) and are calibrated on national or continental levels. Also LCA is not a precision tool to accurately model local effects, but a coarse tool to create a *synoptic life-cycle view* establishing the *relative importance* of many different environmental effects or effect potentials.

[27] 52 mg Ca/l, 14.75 mg S/l, 7.2 mg Cl/l, 1.8 mg C/l, 0.03 mg Zn/l, 0.01 mg Cu/l, 0.0016 mg Cr/l, 0.0015 mg Pb/l, 0.0001 mg Hg (unreferenced geogenic background concentrations in Zimmermann et al. B.17).

$$\text{Eq. 5.1} \quad t = \ln\left(\frac{c_x}{c_o}\right)\Big/ -k$$

$$-k = \ln\left(1 - \frac{STTK}{TK_\infty}\right)\Big/ 100a = \left(V \cdot c_o / m \cdot TK_\infty\right)$$

For all these reasons (mainly B) a limited timeframe with a 'natural background cut-off' is not used in this study. The long-term phase is in principle an open-end timeline.

5.5.2   Modelling of landfill emissions

Introduction

As concluded above the inventory of landfill processes includes emissions occurring during a potentially unlimited time span after the deposition of the waste. As pointed out before, these future emissions must be modelled based on assumptions, since no measurements are available for the far future emissions of landfills.

Similar to the direct emissions of MSWI (see part II of this report), the emissions from landfilled waste are understood as *waste-specific* emissions, i.e. the emissions are derived from the chemical composition of a specific waste and transfer coefficients for chemical elements. These transfer coefficients determine for each landfill which percentage of a certain chemical element is released to the environment. Landfill transfer coefficients essentially make up the landfill model used here. The need to inventorise *waste-specific* emissions is a major difference to other existing landfill models, that usually model the *average* behaviour of the landfill, i.e. the emissions of the average mixture of landfilled waste.

$$\text{Eq. 5.2} \quad Emission_{media,phase,i}\ [kg\ i / kg\ waste] = TK_{media,phase,i}\ [-] \cdot waste\ composition_i\ [kg\ i / kg\ waste]$$

In the landfill model, emissions of a certain chemical element i (e.g. zinc, copper, cadmium) to a certain environmental media (usually pore water, but also to air for sanitary landfills) during a certain landfill phase (here only the short-term phase < 100 years and long-term phase > 100 years are distinguished) are determined. The emissions are calculated from the transfer coefficient for that element i, for the appropriate media (water or gas) and landfill phase (short-term or long-term[28]). The transfer coefficient is multiplied by the content of the element i in the assessed waste. In that way the various transfer coefficients represent the characteristics of the assessed landfill and the waste composition guarantees that the result is waste-specific. It is currently not possible to have waste-specific transfer coefficients, except for sanitary landfills the short-term and long-term transfer coefficients are adapted to the degradability of the assessed waste.

To summarise, the general method in this report to create waste-specific landfill models is to first calculate *average* transfer coefficients from *average* operation and *average* waste compositions. In the case of inorganic landfills these average transfer coefficients are multiplied directly with the specific waste composition under consideration to calculate waste-specific emissions, cf. Fig. 5.6 top half. In the case of sanitary landfills, *waste-specific* (not average) transfer coefficients are calculated from *average* landfill data and waste-specific degradability parameter D, cf. Fig. 5.6 bottom half. All calculations apply to several chemical elements, represented as matrix vectors in Fig. 5.6.

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[28] By convention, the term 'long-term transfer coefficients' refers to the *total* release of elements during the short-term *and* the long-term period (total availability or total long-term emissions). The term 'short-term transfer coefficients' refers only to the emissions during 0–100a (short-term emissions). The emissions >100a need to be calculated by difference (total minus short-term).
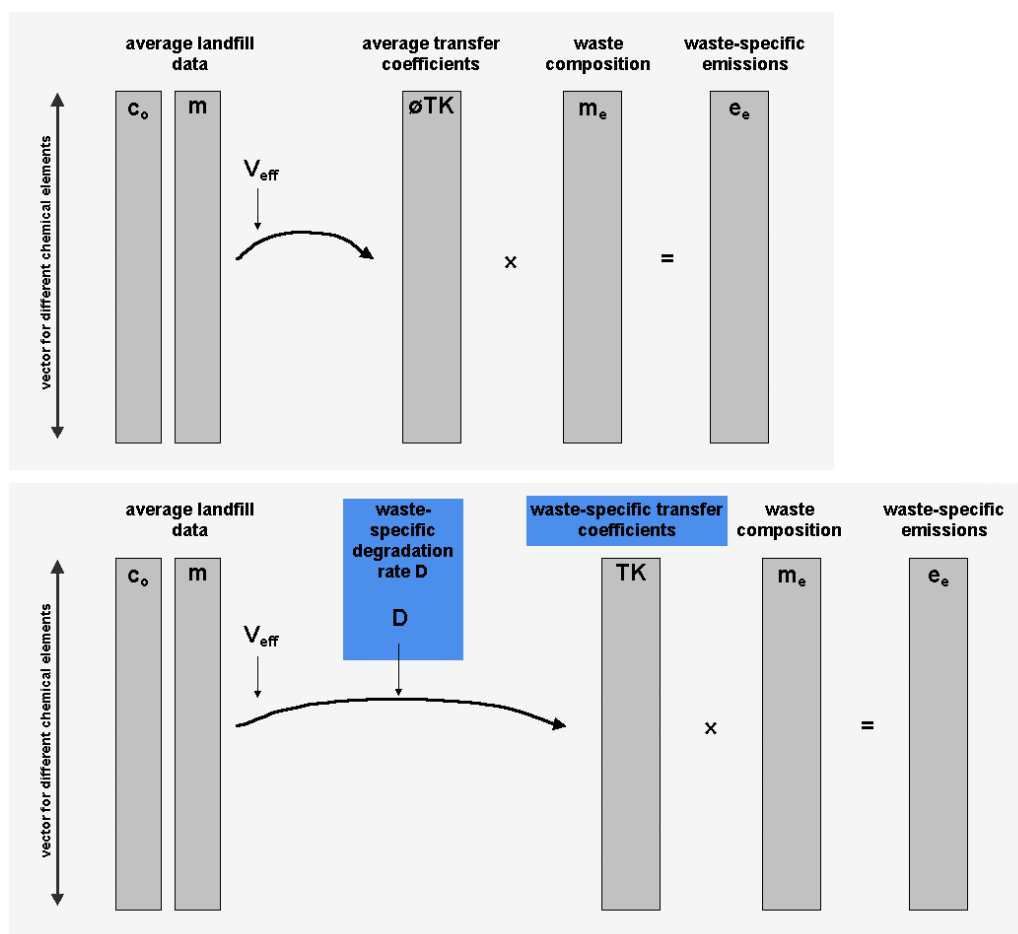
Fig. 5.6 **General calculation scheme for landfill emission models in this report, for inorganic landfills (top half) and sanitary landfills (bottom half)**

Lifetime of barriers and groundwater threat

The landfill is designed to collect leachate and discharge it to a sewer or to a surface water body. It can be assumed that for the short-term leachate <100 a these systems remain intact − at least for the majority of the time. But base seals and collection systems of landfills have a limited lifetime. This includes the geological barrier (clay layer). Considering long timespans, base seals will leak, collection systems will fail and leachate will enter the ground below the landfill. Baccini et al. (1992) consider a lifetime of 50 years for base seals. Frühwirth et al. (2000:128) discuss the special circumstances and ageing processes of geological barriers in a landfill and consider a lifetime of a clay barrier[29] of 100 years to be realistic.

Do emissions into the ground[30] pose a risk? A threat to human health exists, if the pollutants reach the groundwater. But even if the pollutants remain in the ground below the landfill site, a ecosystem damage risk of plant take-up, e.g. by trees, remains. Stefanie Hellweg (2000:98ff.) calculates the retention time of pollution fronts until they reach the groundwater table heeding several soil parameters like infiltration rate, preferential flow paths, cation exchange capacity, acidity, clay

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[29] Clay layer of 50 cm thickness and water permeability (percolation coefficient) k_f of 1^{-10} to 1^{-11} m/s.

[30] 'Ground' signifies a horizon *15 to 25 meters deep* below the landfill, and not a (vegetated) *surface* soil, that is sometimes considered in LCA as emission media, e.g. for agricultural emissions.

content, iron oxide content, organic content and distance to groundwater table. About 20% to 40% of the metals reach the groundwater directly through flow in continuous macropores. Macropore flow occurs relatively quickly, it interacts hardly with the soil matrix and the transfer to groundwater levels takes only approximately 3 to 25 years. The remaining 60% to 80% of the leached metals are slowed through sorption and reach the groundwater table later (so called 'matrix flow'). Retention times for metals transported by matrix flow are around 400 to 3000 years for Cd^{2+}, Ni^{2+}, Zn^{2+}, Cu^{2+}, Cr^{3+}, and 5000 to 33'000 years for Pb^{2+} and Hg^{2+}. These values are calculated with default values for Switzerland proposed by (Hellweg 2000:107). For macropore and matrix flow combined, average retention times of 1000 years result (12'000 years for Pb^{2+} and Hg^{2+}). Over the time span considered for the landfill models (60'000a) the majority of the emitted pollutant load can be expected to reach the groundwater.

Erosion and climate change

In the long timespans considered, erosion is a probable influence on landfill development. For Swiss landscape and climate erosion rates of 5 to 100 cm in 1000 years are realistic (Baccini et al. 1992). However, the geotextiles usually applied on the landfill surface during closure have high resistance against erosion. The decomposed or remaining waste can have very variable erosion characteristics. With the erosion rates cited above, a landfill of 15 m thickness plus 4 m recultivation layer would be eroded within 19'000 to 380'000 years. In such a long timespan the climate and accordingly erosion strength can change.

Catastrophic events like flooding or landslides can short-cut erosion in the long run, although Swiss landfills must be built outside (todays) flooding zones. Changing climate can increase precipitation[31], water and wind erosion regimes. Cooler periods can promote glacier formation, which increases the likeliness of landscape remodelling and distribution of landfill contents. On the other hand, new permafrost soils can postpone landfill leaching.

These effects sustain the notion of a *worst case* that represents a complete mobilisation of all pollutants from the landfill. This worst case will be heeded in the uncertainty data of the long-term transfer coefficients.

Short-term emissions

For the time after the placement of the waste, information is available in the form of leachate and landfill gas measurements of present landfills and closed landfills. Also the gas production during the methane phase of a sanitary landfill is well understood and can be used to calculate emissions to air.

The exponential model

Short-term emissions transfer coefficients can be derived from measurements of the leachate in existing landfills, by assuming that the measured trends will continue into the future. In 1989 Hasan Belevi and Peter Baccini introduced a popular formula to predict long-term emissions from sanitary landfills after the initial active phase (Belevi & Baccini 1989). The behaviour of sanitary landfills was assessed by extrapolating the observations made in existing landfills. The time after the initial active phase (methane phase) is observed. The landfill body is assumed to be a simplified chemical reactor without concentrations gradients. The leachate concentrations can then be predicted based on the following parameters[32].

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[31] Like predicted from global warming model calculations.

[32] The parameters are adapted here to relate to 1 kg of waste and to the variables used in this report.

c_o Mean concentration of the observed element in the leachate during the *first year* after the active landfill phase [g/l].

V Mean annual leachate output from the landfill per kg waste. Assumed to be constant [l/a per kg waste].

m Total amount of the observed element in the landfill at the end of the initial active phase [g/kg waste].

TK_∞ Fraction of the observed element that can be mobilised at the end of the initial active phase [kg/kg].

All these parameters are constant or assumed to be constant after the initial active phase. From these constants the time-dependent concentration of the observed element in the leachate $c(t)$ is calculated according to the following formula:

Eq. 5.3
$$c(t) = c_o \cdot e^{-(V \cdot c_o / m \cdot TK_\infty) t}$$

where t is the time variable in years.

This model features an exponential decline of concentrations from the starting level c_o. An example from the original publication (Belevi & Baccini 1989) is shown in Fig. 5.7. Similar exponential decline formulas have been formulated e.g. by Rowe (1991) or Kruse (1994).
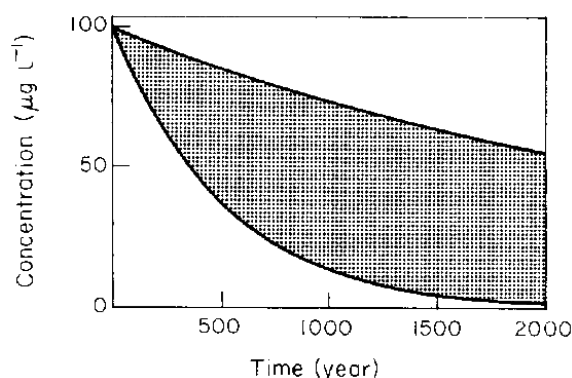

Fig. 5.7   Concentrations dynamics in leachate according to simplified exponential decline. The range is defined by variance in the content in the waste *m*. Original chart from (Belevi & Baccini 1989)

The term $m \cdot TK_\infty$ in the exponent of Eq. 5.3 signifies the total mobilisable amount of the element in the landfill per kg average waste at the end of the initial active phase[33]. This amount is the total initial emission potential and is represented by P_o.

Eq. 5.4 $P_o = m \cdot TK_\infty$ Total initial emission potential per kg average waste

If the fraction c_o/P_o is large, then a relatively large amount of the total initial emission potential has been washed out in the first year, and the element can be considered to be washed out fast. The concentration $c(t)$ shows a rapid decline for 'fast' elements in the sense above and for large volumes of

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[33]   In the original form, Belevi and Bacini (1989a) formulated the equation for *c(t)* for the whole waste mass of the landfill. Here the formula is normalised to refer to 1 kg of average waste. In the original form, the total initial emission potential $m \cdot TK_\infty$ is represented by the total mass of the landfilled waste *M* times the initial concentration of the element in the waste that can be mobilised m_o, which is the equivalent expression, but for the whole waste mass of the landfill and not just 1 kilogram.

leachate water flow *V*. Belevi and Baccini determined the mobilisable fraction of an element by performing a leaching experiment on pulverised waste samples with four consecutive leaching steps with distilled water for 0.5, 5, 50, and 288 hours respectively (L/S = 10). After that time all mobilisable matter was assumed to be extracted, which is an underestimation, *"since the experiment does not guarantee that further extractions give negligible contributions"*, as the authors note themselves (Belevi & Baccini 1989).

Mobilised and remaining mass in the exponential model

Since the model follows a simple first-order decay dynamic, the *current* leachate concentration *c(t)* is always proportional to the *remaining* mass of mobilisable matter. This remaining mass of mobilisable matter can be regarded as the remaining emission potential *P(t)*, i.e. the amount of potentially mobilisable pollutant that has not yet been emitted. Similarly, the initial leachate concentration c_o (i.e. t=0) is equally proportional to the total initial emission potential P_o. From this proportionality follows:

Eq. 5.5 $\quad \dfrac{c_o}{P_o} = \dfrac{c(t)}{P(t)}$ and from that $\quad P(t) = P_o \cdot \dfrac{c(t)}{c_o}$

The removed and emitted amount of mobilisable matter *E(t)* can be calculated as difference of P_o and *P(t)*:

Eq. 5.6 $\quad E(t) = P_o - P(t) = P_o \cdot \left(1 - \dfrac{c(t)}{c_o}\right) = m \cdot TK_\infty \cdot \left(1 - e^{-(V \cdot c_o / m \cdot TK_\infty)t}\right)$

The fraction of mobilised element *E(t)* in relation to the total initial amount *m* at any time *t* is equal to the transfer coefficient for that time *t*:

Eq. 5.7 $\quad TK(t) = \dfrac{E(t)}{m} = TK_\infty \cdot \left(1 - e^{-(V \cdot c_o / m \cdot TK_\infty)t}\right)$ $\quad$ Exponential transfer coefficient

With this formula the transfer coefficient for any point in time can be calculated. The transfer coefficient *TK(t)* approaches TK_∞ for large times *t*. The specific annual leachate volume *V* will later be replaced by the effective leachate volume V_{eff}, heeding preferential flow in leachate (see section 'Heterogenity in landfill hydrology' on page 30).

Exponential versus linear approach

It is interesting to note the relation of Eq. 5.7 for exponential decline with a linear approach, i.e. assuming that the observed leachate concentrations are *constant* over time and are always equal to c_o. The transfer coefficient $TK_{lin}(t)$ in such a linear model would be calculated from Eq. 5.8.

Eq. 5.8 $\quad TK_{lin}(t) = \dfrac{V \cdot c_o \cdot t}{m}$ $\quad$ *,with $TK_{lin}(t) \leq 100\%$* $\quad$ Linear transfer coefficient

In general, a term *(1-e^{-x})* equals approximately *x*, if *x* is much smaller than unity. This approximation can be applied to Eq. 5.7, with *x* being the term $V*c_o*t/(m*TK_\infty)$ in the exponent. If this term is much smaller than unity, it means that the element under consideration is not easily leached out (c_o is small compared to $m*TK_\infty$) and that after the time *t* still a majority of the leachable mass remains in the landfill. Hence, if $V*c_o*t/(m*TK_\infty)$ is much smaller than unity, Eq. 5.7 can be approximated with $TK_{exp}(t) = TK_\infty*V*c_o*t/(m*TK_\infty)$, which equals $V*c_o*t/m$, i.e. the equation for the linear model (Eq. 5.8). It can be concluded that for elements which are washed out over time spans considerably longer than 100 years, there is *no perceptible difference between a linear and an exponential approach* to determine the *short-term* TK. Elements which do not fulfil this condition are the easily soluble ones, like sodium, potassium or chlorine.

Supplementary formulas

If TK_∞ and $TK(T)$ for a specific time T are known, any $TK(t)$ can be calculated:

$$TK(t) = TK_\infty \cdot \left(1 - e^{-k \cdot t}\right)$$

Eq. 5.9

$$-k = \ln\left(1 - \frac{TK(T)}{TK\infty}\right) \Big/ T$$

The time t until a certain leachate concentration c_x is reached is calculated with following formulas.

$$t = \ln\left(\frac{c_x}{c_o}\right) \Big/ -k$$

Eq. 5.10

$$-k = \ln\left(1 - \frac{TK(T)}{TK\infty}\right) \Big/ T$$

Application of the model for short-term behaviour

Although the Belevi-Baccini-model (Eq. 5.3) was derived based on observations from sanitary landfills after the reactive methane phase, it can be applied to any landfill, assuming that it has reached a quasi-steady-state and leachate concentrations are ruled by first-order kinetics. Although inorganic landfills have initial settling reactions, this formula will be used *for the residual landfill and the slag compartment* to calculate transfer coefficients of soluble elements. For the sanitary landfill another approach will be used that considers the different waste-specific degradation rates to derive waste-specific transfer coefficients (see chapter 6.1 'Waste-specific emissions from sanitary landfills' on page 42). For the inert material landfill no direct leachate emissions will be used, cf. chapter 9.1 'Emissions from inert material landfills' on page 73.

For the residual landfill and the slag compartment the following procedure to calculate average short-term transfer coefficients is used:

For easy soluble elements an exponential decline of the leachate concentration is assumed. Easy soluble elements are the monovalent ions (Na^+, K^+, Cl^-, F^-, Br^-, I^-), oxianion-forming elements (WO_4^{2-}, HVO_4^{2-}, $Cr^{(VI)}O_4^{2-}$, MoO_4^{2-}, HBO_3^{2-}, $HSeO_4^-$, SbO_4^{3-}, $HAsO_4^{2-}$) and also nitrogen (as nitrate NO^{3-}).

For all other elements (mostly cationic metals) a constant leachate concentration is assumed. This corresponds well with results from thermodynamic calculations of the leaching behaviour in long-term landfills models (Hellweg 2000). The concentrations of these elements in the leachate are governed by the conditions and processes in the landfill. If the conditions do not change, the concentrations will remain constant. Heterogeneity in landfill hydrology and waste distribution can lead to variations in leachate, but the average leachate concentration is established during the flow of the leachate through the landfill, with the leachate having average residence times of years.

The transfer coefficients are calculated from Eq. 5.7 and Eq. 5.8 on page 23. The necessary information for c_o and m is derived from literature values (see Tab. A.2, Tab. A.3, and Tab. A.4 on page 110ff. in the appendix). Data from various landfill sites is collected, implying that a collection of values from various sites gives mean values representative of a average generic and typical landfill site. The reason for this is that no complete data set for all necessary elements from one single landfill site is available.

A total mobilisable fraction TK_∞ of 100% is used in Eq. 5.7, with the exception of chromium. In slag, 75% of chromium is present as chromite (Cr^{III} in $FeCr_2O_4$), which is very stable and not available even under most aggressive conditions (Huber et al. 1996:47). For the TK_∞ of chromium a value of 25% is used. In Eq. 5.8 the values are corrected to ≤100%. For most elements a complete mobilisation is assumed. In reality, this mobilisation potential is initially not necessarily 100%, but will be achieved over time through weathering. The exact weathering dynamics are not modelled here, so in the model

the TK_∞ is constant over time[34]. Where no information on c_o and m is available, transfer coefficients are estimated from other sources, or as proxies from other elements based on chemical similarities.

Long-term emissions

This section relates only to the residual material landfill and slag compartments, i.e. inorganic landfills. The calculation of sanitary landfills (organic landfills) are specified in chapter 6.1 'Waste-specific emissions from sanitary landfills' on page 42.

As mentioned in footnote 28 on page 19 the long-term transfer coefficient refers to the total fraction of an element in waste, that can be mobilised and emitted in the long-term perspective. The master variables that ultimately control the pollutant potential are the pH of the pore water (leachate) and the mineralogy of the landfilled material. The pH determines which chemical species are mobile. The pH and redox potential are usually closely related (Huber et al. 1996:52). Mineralogy of the waste determines if phases are stable under the encountered conditions. Both variables are intertwined and can change in time. Ongoing geochemical weathering processes change the mineralogy of the waste. The development of the pH value depends on the acid neutralising capacity of the landfilled material, which in turn is defined by the minerals present.

Meaning of uncertainty ranges for long-term transfer coefficients

The prediction of landfill development over very long time spans is vague at best and depends on many uncertain parameters like development of geochemical weathering, climate conditions, vegetation. Accordingly the transfer coefficients which describe the landfill development can cover a large range representing the differences between a best case and a worst case.

The goal of this study is to estimate *waste-specific* emissions. Theoretically, a specific waste can influence the behaviour of its constituting elements in the landfill. For example lead in a glass matrix that is transferred to MSWI bottom ash will likely have a different range for its long-term transfer coefficient than finely dispersed lead from the incineration residue of a plastic. MSWI bottom ash is a heterogeneous material and the term 'slag' is somewhat misleading as only about 40 w-% consists of molten phases (Lichtensteiger 1995). About 50 w-% if the 'slag' contains material, which pass through the incineration grate without changes in mineralogy (ceramics, the core of larger metal parts, rocks from minerals like quarz, feldspar, calcite) (Lichtensteiger 2002). However, for inorganic landfills it is not yet possible to derive waste-specific transfer coefficients, as the mineralogical fate of constituents of all specific waste fractions is not known. Within this study all inorganic landfills have therefore *average generic* and not waste-specific transfer coefficients. In this model, all elements in all types of wastes feature an *average behaviour* as conceived for the elements in the average landfilled waste.

For the inorganic landfill model it is necessary to obtain a range for the long-term transfer coefficients. This range shall express the likeliness that an element is leached out from the landfill. The lower 2.5% confidence value or minimal value shall represent the fraction of the element that is emitted *at least*, i.e. even under most optimistic circumstances this fraction is most certainly emitted. The upper 97.5% confidence value or maximal value shall represent the fraction of the element that is emitted only under the most unfavourable conditions and thus represents a worst case. The mean value shall

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[34] Exemplary, numerical simulations with a mobilisation potential which *changes in time* were performed. For demonstration purposes it was assumed that the mobilisable fraction is in the beginning only 20% of the total amount (without any measured evidence for that figure) and increases over time with a square-root dynamic, i.e. proportional to $\sqrt{time}$. This dynamic is likely to occur for progressive weathering. Compared to the used model with *constant and complete mobilisation potential throughout* (TK inf. = 100%) the resulting TK were 0% to 20% smaller, depending on point in time (i.e. between identical and 80% of the values from the used model). This incompleteness of the used model must be accepted due to lack of field data and indeed seems acceptable, compared to the large uncertainties already observed in landfill measurements of current composition and content, which form the basis of TK calculations in this study.

represent an intermediate case, ideally representing the conditions that are to be expected in an average or likely case. As long-term landfill development can currently not be observed or measured, the definition of the intermediate and best-case development case must of course remain speculative. For the worst-case, however, there is a upper limit: 100% discharge; i.e. the landfill cannot emit more pollutants than were present in the landfilled waste.

Application of the model for long-term behaviour

The following procedure is applied for the long-term development of residual material landfills and slag compartments. The model choices for the calculation of the *short-term* transfer coefficients of different elements are maintained. The leachate dynamics (exponential decline or constant) are *continued into the long-term future*. At some time, the acid neutralising capacity ANC of the landfilled material will be used up and the carbonate phase ends. The loss of ANC is mainly caused by washout of calcium and magnesium. If the carbonate phase ends, the pH drops and metal cation concentrations in leachate increase, cf. arrow 1 in Fig. 5.8. The concentrations of metal oxianions *decreases* as they are less soluble at low pH, cf. arrow 3 in Fig. 5.8. The washout of other easy soluble monovalent ions (Na^+, K^+, Cl^-, F^-, Br^-, I^-) and nitrate NO_3^- is unaffected by pH[35]. The factor of concentration increase for metal cations at higher pH values is measured for some elements in (Johnson et al. 1996). The factor x_e is 158 for Cd, 100 for Cu, 251 for Pb, and 158 for Zn. For other elements an average value of 167 is used[36]. Oxianions are less soluble at low pH, i.e. the factor x_e is <1 and assumed to be 0.01. For silicon x_e is also 0.01 as it is only soluble as hydroxide at high pH, cf. arrow 5 in Fig. 5.8.

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[35] See also footnote 15 on page 13.

[36] These increases in concentration of two orders of magnitude are probably conservative. Kraxner et al. (2001:161) find increases of *three orders of magnitude* when moving from pH 8 to pH 4 for lead and zinc, and a bit lower for copper, manganese and iron. For some bivalent metals Me^{2+} solubility is controlled by hydroxide phases ($Me^{2+} + H_2O = 2\ H^+ + Me(OH)_2$). Based on data from (Chandler et al 1997) Kraxner et al. (2001:162) derive a simplified rule for such metals by which each decrease in pH per unit increases metal concentration (as mg/l) by two orders of magnitude. A an increase of 3 in pH therefore increase metal concentrations by *six orders of magnitude*. This latter rule however disregards formation of complexes and ion pairs.
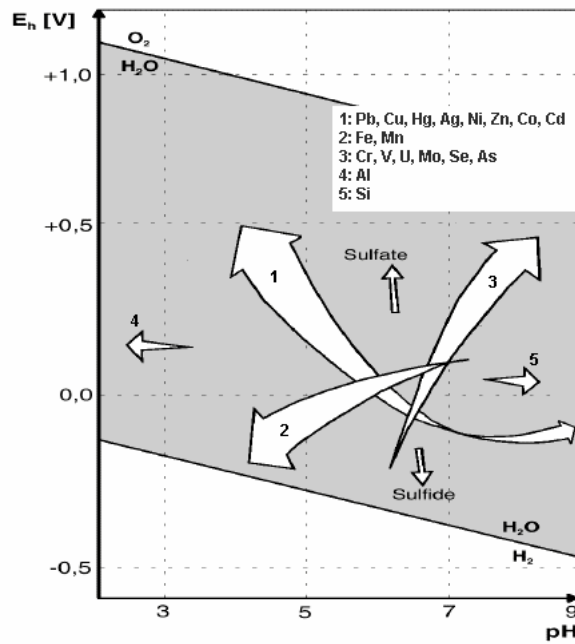
Fig. 5.8   Diagram of pH versus redox potential E_h and relative metal mobility. Arrows indicate directions of *increasing relative mobility* for the corresponding metals. Adapted from (Förstner et al. 1989)

The washout at low pH will fundamentally continue unlimited. There is fundamentally no end to washout other than the complete removal of material from the landfill site. All known effects can only delay complete washout (Sabbas et al. 1998). This is the reason to set the *worst case* or maximal value for long-term transfer coefficients to TK_∞.

To calculate a *mean* transfer coefficient value, only the modelled emissions up until the next expected midland-covering glacial period are heeded (see Fig. 5.9). Glaciers reshape and remodel the landscape and break up the biosphere. When glaciers cover the Swiss midland, landfills will be eroded too and their contents redistributed in the landscape[37]. The question when the next glacial period in Switzerland can be anticipated is discussed in the section 'The next glacial period in Switzerland' on page 29 and is answered there with 'approximately 60'000 years from now'. In case the carbonate phase lasts until the next glacial period occurs, the mean value will be calculated from the emissions that occur up to the next glacial period.

A *best case* or minimal value for the transfer coefficients is derived from the following arguments. It is established that landfills emit pollutants and will continue to do so. During the carbonate phase a certain pollutant fraction is leached out, but after the carbonate phase a substantial pollutant potential still remains. After the carbonate phase, the remaining pollutants are washed out with increased rates due to a decrease in pH. The best case assumes that this pH drop does not occur, and total long-term emissions are *at best* as large as the emissions during the carbonate phase alone (see Fig. 5.9). In case the carbonate phase continues until the next glacial period occurs, the minimal value will be set equal to the mean value[38]. The three values for best, average and worst case represent a triangular distribution.

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[37] The landfill then ceases to exist, but its redistributed contents represent an environmental impact after the retreat of the glaciers.

[38] Due to late changes described in section 'Uncertainty of transfer coefficients and emissions' on page 36 this best case or minimal value will not be used in the model. Instead a lognormal distribution will be estimated from the medium and worst case value.
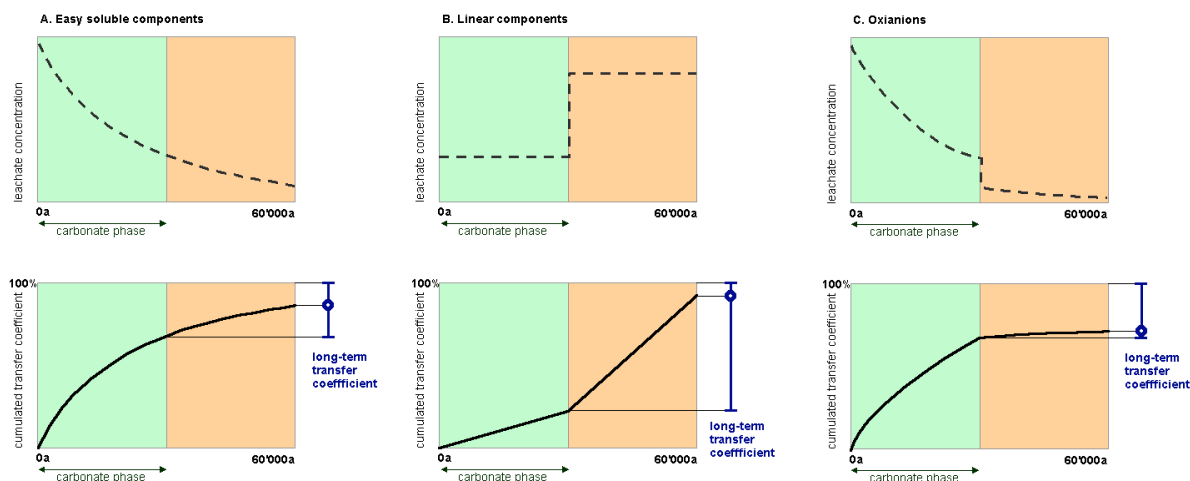
Fig. 5.9 **Calculations scheme for long-term transfer coefficients for different elements: A.) Exponential decline for soluble elements with no change after the carbonate phase (left), B.) Constant concentrations for metal cations with an increase after the carbonate phase (middle), C.) Exponential decline of concentrations for metal oxianions with a decrease after the carbonate phase (right). The range indicated at the right side of each panel represents the derived uncertainty range for transfer coefficients**

With the choices made above, the three values for best, average and worst case actually represent a timeline in the modelled landfill development. The best case marks the end of the carbonate phase, the mean value marks the time of the next anticipated midland-covering glacial period in Switzerland (the end of the landfill), and the worst case represents an infinite time[39]. These marks in time complement the information of the short-term emissions which occur during the first 100 years after deposition. However it would be wrong to understand for example a best case value for long-term *emissions* as a value which merely discounts emissions after the carbonate phase. The best case emission value is not solely determined by the minimal long-term transfer coefficient alone, but also by the *best case waste composition* (i.e. lower boundary pollutant concentration in the waste). The best case is a value derived for optimistic scenarios for *all* determining factors.

For some elements the values of c_o or m are not known from literature. But the values for the long-term transfer coefficients can be calculated from the short-term transfer coefficients (STTK) derived above. For exponentially decreasing leachate concentrations the formula is:

$$TK(t) = TK_\infty \cdot \left(1 - e^{-k \cdot t}\right)$$

Eq. 5.11
$$-k = \ln\left(1 - \frac{STTK}{TK_\infty}\right) \Big/ 100a$$

where STTK is the short-term transfer coefficient = *TK(t=100a)*

For constant leachate concentrations the formula is:

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[39] Such long timeframes are already invoked in LCA for example in the LCIA stage. Modelled environments for calculating characterisation factors of the LCIA method CML'01 have in principle infinite timeframes (Guinée et al. 2001). In actuality they cover very long timeframes (e.g. millions of years for soluble pollutants like fluoride in marine water).

Eq. 5.12 $\quad TK(t) = \dfrac{STTK}{100a} \cdot t$ $\qquad$ corrected to $TK(t) \leq TK_\infty$

where STTK is the short-term transfer coefficient = $TK(t=100a)$

These formulas can be used to calculate the best case (minimal) transfer coefficient values, when t is set as the time the carbonate phase ends ($= t_e$). In case the carbonate phase would end after the next glacial period ($t_e > t_g$), these formulas are also used to calculate the mean value.

If the carbonate phase ends before the next glacial period ($t_e < t_g$), the leachate concentrations are altered by a factor x_e. The mean value of the transfer coefficients is then calculated heeding these post-carbonate concentrations. The time when the carbonate phase ends is t_e; the time when the next glacial period occurs is t_g.

Eq. 5.13
$$TK(t_g) = TK(t_e) + \left(TK_\infty - TK(t_e)\right) \cdot \left(1 - e^{-k \cdot x_e \cdot (t_g - t_e)}\right)$$
$$-k = \ln\left(1 - \frac{STTK}{TK_\infty}\right)\bigg/ 100a$$

For linear washout the formula is:

Eq. 5.14 $\quad TK(t_g) = TK(t_e) + \left(\dfrac{TK(t_e)}{t_e}\right) \cdot x_e \cdot (t_g - t_e)$ $\qquad$ corrected to $TK(t_g) \leq TK_\infty$

The end of the carbonate phase

To calculate the minimal values of the long-term transfer coefficients, the time t_e when the carbonate phase ends needs to be known. Carbonate producing metals are calcium, magnesium, strontium, barium and others, but usually calcium and magnesium carbonates contribute over 99.5% to the carbonate buffer. To determine the time when the acid neutralising capacity is used up, the complete washout of calcium and magnesium is modelled. With a constant washout during the carbonate phase, the time when an element is completely leached out can be determined by Eq. 5.15.

Eq. 5.15 $\quad t_e = m_{Ca,Mg}\bigg/ c_{o,Ca,Mg} \cdot V$

When calcium and magnesium are washed out, the carbonate phase ends and the landfill enters a new stage with lower pH. Usually calcium is the limiting element, i.e. calcium takes more time than magnesium to be washed out.

The next glacial period in Switzerland

To calculate the mean values of the long-term transfer coefficients, the time t_g, when the next Swiss midland-covering glacial period begins, needs to be known. In the last 2 Million years, 15 glacier advances were recorded in the Swiss alps (Schlüchter 2002). Approximately every 110'000 to 130'000a a glacial period occurs. Not all glacier advances did necessarily cover the Swiss midlands with ice. These ice covers erode soil and ground and reshape the landscape. Essentially the ecosphere is reshaped. But also in an altered ecosphere and the resulting biosphere, release of pollutants – e.g. heavy metals from glacier rubble – will have an ecological impact. This supports an upper, worst case long-term transfer coefficient of 100%. Following timeline was compiled from (Schlüchter 2002).

- 180'000 to 120'000a BP (before present): the Riss glacial period: The Swiss midlands were largely ice-free.

- 115'000 to 10'000a BP: the Würm glacial period, where

- 100'000a BP the first Würm glacier advance occurred.

- 60'000 to 28'000a BP: the Swiss midlands were ice-free, with a steppe/tundra climate and a forest limit at 1800 meters above sea level.

- 32'000 to 10'000a BP: the second Würm glacier advance occurred. Forest limit dropped to 800 meters above sea level.

- After 10'000a BP the Swiss midlands became ice-free again until today.

Hence, in the last 200'000 years the Swiss midlands were iced over only twice, and on average, every 67'000 years. In the future this period could presumably be extended by present anthropogenic global warming. A period of 70'000 years between midland-covering glacial periods is estimated here. 10'000 years since the last glacial period (second Würm advancement) have already passed. For the present study, a duration t_g of 60'000 years from now until the next midland-covering glacial period is estimated.

Heterogenity in landfill hydrology

Leachate does not flow homogeneously through a landfill. Regions with low water permeability can form which hinders leachate flow. On the other hand, preferential flow paths lets leachate flow relatively quickly through the landfill. This can lead to leachate which has hardly had exchange with the landfill body and is therefore not effective as a reactand media (i.e. media for washout of material). This slows down the landfill development as compared to a homogenous model with homogenous average infiltration. This postpones the end of the carbonate buffer phase further into the future. In the following paragraphs calculations with sample figures are performed.

Preferential flow in the leachate has a share of 10% to 50% in the total water discharge in a slag landfill (Hellweg 2000:77, based on a personal communication with Annette Johnson, EAWAG). The geometric mean is 22%.

If I is the rain water infiltration rate (in mm/a per m^2 area), then this volume must be (in the long-term average) released as leachate for reasons of mass balance[40]. The water flows through the water pores of the landfill. The water pore volume $V\%$ of landfills is estimated to be 31%[41]. Assuming all water pores are open and connected the average cross section area of water pathways is 0.31 m^2 per m^2 landfill area. Since the cross-section area of the flow-paths is smaller than the landfill area receiving the water, the leachate flow within the landfill must be *faster* than the infiltration speed at the surface suggests. The average leachate speed within the landfill is $I / V\%$. From this speed, the average residence time of the water in the landfill can be calculated. If the landfill has the height h in meters, then the *average* residence time of leachate water is T_a.

$$\text{Eq. 5.16} \quad T_a = \frac{h[m] \cdot 1000 \cdot V\%}{I[mm / m^2 \cdot a]}$$

T_a is approximately 0.6 years *per meter of landfill height*, and approximately 9.3 years for a landfill of 15 m height and infiltration of 500 mm/m^2a. Again, in the long-term average, after the landfill has been saturated, the average residence time *must* be achieved, otherwise the landfill would absorb or generate water. Therefore, if a part of the leachate is preferential with a residence time much *shorter* than average, then the *rest* of the water must flow more *slowly* to compensate the effect of the

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[40] Neglecting any uptake or liberation of waste in the waste and lateral flow to or from the landfill body through seal leaks.

[41] Calculated from a water content of 20% (Johnsson et al. 1998:370) and an average density of 1500 kg/m^3.

preferential flow and to maintain the overall average residence time of leachate, respectively mass balance (cf. Fig. 5.10). If $w\%$ is the preferential part of the collected leachate and T_p is the residence time of preferential leachate, then the residence time T_s of the non-preferentially flowing water is expressed with:

$$\text{Eq. 5.17} \quad T_s = \frac{T_a - w\% \cdot T_p}{1 - w\%}$$

With a residence time T_p of 9 weeks (0.17a) and a share of preferential leachate $w\%$ of 22%, the residence time of non-preferentially flowing water is approximately 11.7 years or 1.3 times larger than the average residence time of 9.3 years.
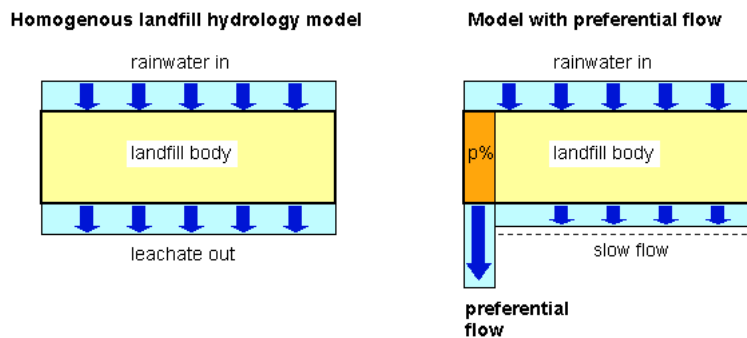

Fig. 5.10   Scheme of landfill hydrology with homogenous flow (left) and with preferential flow included (right). Not to scale

If 22% of the collected leachate (output) are obtained through preferential flow, it does not necessarily mean that 22% of the *landfilled contents* are exposed to preferential flow. A lot of 'fast', preferential water can pass through the landfill and still be in contact with just a marginal part of it (cf. Fig. 5.10). Residence times can in general be calculated by the formula 'stock [kg] divided by rate [kg/a]'. If we know the 'stock' of preferential water, i.e. the percentage of the total water *within* a landfill that is flowing preferentially, we can estimate which part of the landfill is actually in contact with preferentially flowing water. Let $p\%$ be the (unknown) part of the landfilled material that is exposed to preferential flow. Per 1 m² of landfill area there is a total mass of *1m²*h* δ* of landfilled material with density δ, with h being the landfill height. The average water content $v\%$[42] in this waste column is 20%. So, per 1 m² of landfill area there is a total water mass of *v%*1m²*h*δ*, which we will call M_w[43]. Accordingly, the 'stock of preferential water', i.e. the amount of water in the part of the landfill that is affected by preferential flow, is $M_w*p\%$. The output rate of preferential water is part of the leachate output, that flowed preferentially, i.e. $I*w\%$. The residence time for preferentially flowing water can therefore be written as:

$$\text{Eq. 5.18} \quad T_p = \frac{stock}{rate} = \frac{M_w \cdot p\%}{I \cdot w\%}$$

From that, p% can be isolated:

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[42]   Not to be confounded with the water pore volume V% defined above.

[43]   It is assumed that water distribution within the landfill is homogenous.

$$\text{Eq. 5.19} \quad p\% = \frac{T_p \cdot w\% \cdot I}{M_w} = \frac{T_p \cdot w\% \cdot I}{v\% \cdot 1m^2 \cdot h \cdot \delta \cdot}$$

Using the values derived above, a share of <1% of the landfilled material is actually in contact with preferentially flowing water[44]. In other words, most of the landfill is in contact with non-preferentially flowing water.

So the effect of including preferential flow in the landfill model landfill is twofold. First, *less* water than in the homogenous model is in contact with the majority of the landfill body (>99%), since 22% are preferential water with little contact to the landfill. Second, the remaining, non-preferential water body is *slower* than suggested by a homogenous model. Almost all of the landfill body is in contact with this 'slow' water. Both effects delay the landfill development.

The equations to calculate the transfer coefficients (Eq. 5.7 and Eq. 5.8 on page 23) are defined for the annual leachate volume V [l/a*kg waste]. This annual leachate volume needs to be replaced by the annual *effective* leachate volume V_{eff}. The effective leachate volume is the water that interacts with the landfill and promotes weathering reactions, i.e. the non-preferential 'slow' flow. The annual mass of leachate water flowing non-preferentially is I*(1-w%). The part of the landfill exposed to 'slow' flow is *(1-p%)*. Per m[2] of landfill area, the mass of waste exposed to slow flow is *(1-p%)*1m²*h* δ*. The effective annual leachate volume is:

$$\text{Eq. 5.20} \quad V_{eff} = \frac{I \cdot (1 - w\%)}{(1 - p\%) \cdot h \cdot \delta} = \frac{1 - w\%}{\left(\dfrac{h \cdot \delta}{I} - \dfrac{T_p \cdot w\%}{v\%} \right)}$$

with

I	Rain infiltration rate (~500 mm/m²a)
w%	share of preferential flow in leachate output (22%)
p%	share of the landfill exposed to preferential flow (<1%), cf. Eq. 5.15
h	landfill height (15 m slag, 10 m residual)
δ	waste density (kg/m³)
v%	water content in waste (20 w-%)
T_p	Residence time of preferentially flown water (0.17a)

With the above parameters, the effective leachate volume V_{eff} is 0.017 l/kg waste per year. This is volume is 20% smaller than in the homogenous modelling, and the carbonate phase therefore lasts 1.3 times longer than in the homogenous case. For each landfill type different V_{eff} are calculated in the corresponding chapters.

It is assumed that the complete landfill body is exposed to 'slow' flow, i.e. the accelerated weathering in the small share of landfill body exposed to preferential flow (p%) is neglected.

Heterogeneity in material

In the homogenous landfill model, it is assumed, that the landfill body is a homogenous mixture of water and solid material. In reality, the material has varying grain sizes, varying porosity and varying mineralogical composition. For large particles, the corrosion or weathering speed might become a rate determining step of washout, if these surface processes are slow enough. Corrosion of metals was observed to be quite fast (10 years for metallic iron). For glass particles, corrosion is thought to be

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[44] If an inhomogeneous water distribution is assumed and v% is increased, p% decreases.

slow[45]. Accordingly, heavy metals embedded in glass matrices will only be released slowly. If the dimensions of solid, unfractured domains of glass are large enough, heavy metals could be bound in such material for long times, even if favourable conditions of their washout (low pH after carbonate buffer phase) would establish in the landfill. In MSWI bottom ash (slag), approximately 40 w-% is molten, glass-like material (Lichtensteiger 1995). The size distribution from sieving in Fig. 5.11 suggests that there are over 30 w-% in slag that have a size over 1 cm. Some of this coarse material is bulk metal, not glass. Lichtensteiger (1995:2) points out that the molten phases in slag, while having particle sizes of millimetres up to centimetres, usually feature small scale associations of different phases at or below the millimetre scale. Speiser (2001:3) describes MSWI slags as "porous matrix with mixed-in phases on a nanometer to micrometer scale". Speiser (2001:5) gives a typical pore volume of 50% for molten, glassy phases in MSWI slag. There is however no information on the degree of fracture of the coarse glass material.
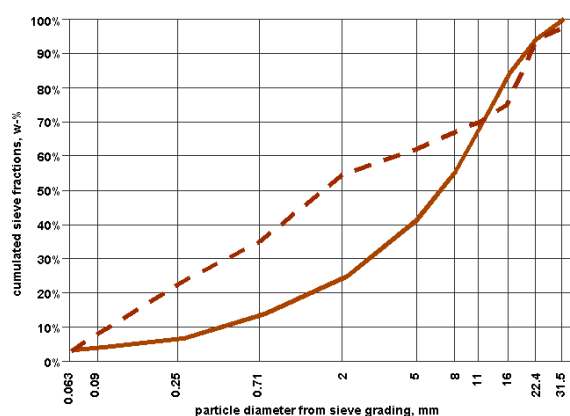

Fig. 5.11   Grading curve of MSWI bottom ash from size analysis with sieves. Adapted from Frühwirth et al. 2000:148 (hatched line) and Reichelt & Pfrang 1998:3(solid line)

It is important to note the difference between particle sizes derived from sieves and the unfractured bulk material dimensions, that are effective in corrosion processes. Fractures and open pores in the material can decrease the domain size, which is effective and applicable in corrosion estimates. Compared to homogenous, unfractured, unporous material, corrosion can be completed much earlier, if the material is fractured or porous. Since MSWI slag is usually quenched in water after the incinerator grate, i.e. transferred form 1000°C to <100°C, fractures are likely to occur.

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[45]   Personal communication with A.C. Johnson, EAWAG Dübendorf, September 12, 2002.
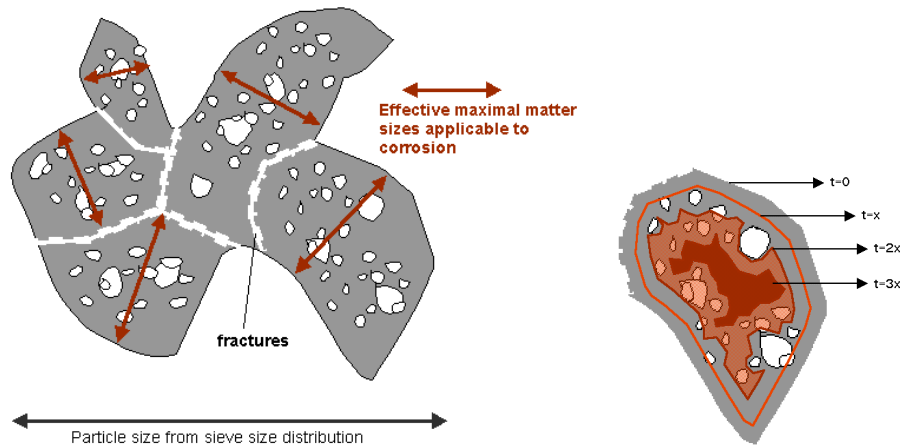
Fig. 5.12 **Schematic of a glass particle showing the difference between the particle size from sieved size distribution and domain sizes applicable to corrosion (left). On the right the qualitative temporal dynamics of particle corrosion and the effects of closed pores are shown**

Speiser (2001:12) points out that the corrosion of glass phases in MSWI slag landfills is much faster, than in natural glassy minerals, such as volcanic basalt. Conversions of glass phases via hydration to gels and subsequently conversion to clay minerals has been observed in slags only 12 years old. Heavy metals, formerly enclosed in the glass matrix, were converted to fine suspensions of calcium salts and carbonates (Speiser 2001:12, Huber et al. 1996:90) . Similar processes in natural glasses take thousands of years. MSWI slag, which was stored open-air for 3 months, already shows signs of glass corrosion and formation of new calcium-silicate-hydrates visible on a micrometer scale (Speiser 2001:143).

Gross et al. (1999) measured corrosion rates of commercial $CaO-(Mg,Fe)O-Al_2O_3-SiO_2$ glass samples. Disks of glass were exposed to 120 days of distilled water with 0.154 mol NaCl at 37°C and buffered to varying pH values. These conditions are similar to those in a landfill. Landfill leachate has elevated temperatures of approximately 25°C and contains 0.16 mol NaCl. The corrosion rates found[46] were between 0.19 mg/cm^2 (at pH 7.4) and 310 mg/cm^2 (at pH 3). At pH 9 a rate of 0.33 mg/cm^2 was found. During the carbonate phase of an inorganic landfill the leachate pH is approximately 8.3. Under these conditions, a glass corrosion rate in the range of 0.19 to 0.33 mg/cm^2 could be expected. With a glass density of 2200 kg/m^3, a corrosion speed of 2.6 to 4.5 micrometers per year can be calculated. With corrosion speeds of this magnitude, a *large, unfractured, homogenous glass domain* with a diameter of 1 cm would be corroded within 1000-2000 years. Gross et al. (1999) report increasing corrosion speed with decreasing pH. This would signify that during landfill development the corrosion of glasses would increase as the pH drops.

Theoretical, thermodynamic calculations on corrosion speed of glass phases in MSWI bottom ash performed by Yan (1998:45) shows ranges between 12.3 and 0.08 micrometers per year, compatible with the ranges above. These are reported as initial rates that slow down over time.

These reported corrosion speeds are also compatible with observations about alterations in the surface domains of glassy blast furnace slags (Olbrich & Frischat 2001). The surface altered to a depth of 150 nanometers *within only 7 days* in an alkaline solution of 0.1 M KOH. This solution is considered representative for early pore water solutions in this material. Even the concentration of network elements (Al, Si), which constitute the glass matrix, is altered. This extrapolates to an alteration rate of

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[46] The probe contained SiO_2 42.4%, Al_2O_3 19.9%, TiO_2 2.08%, FeO 5.92%, CaO 18.7%, MgO 6.5%, Na2O 0.61%, K_2O 2.91%, P_2O_5 0.5%, MnO 0.92%.

7 micrometers per year. This figure is not a corrosion or dissolution rate, and it is also daunting to extrapolate a 7-day result to years, because alteration rates might slow down over time. Presuming that a matrix alteration precedes corrosion, this rate is however compatible with annual corrosion rates in the micrometer range reported by Gross et al. (1999) cited above. These results also underline the dynamic and non-inert properties of glasses in the surface domain. Olbrich & Frischat (2001) observe surface transition from mellilite glass to pyroxene and further to forsterite glass *within one hour* in a solution of 0.1 M KOH. Alterations in the surface roughness are also visible on a micrometer scale. All these observations occur at a high pH of approx. 13. As noted before, the corrosion speed increases with decreasing pH (Gross et al. 1999).

An uncertain point is, which percentage of pollutants are contained in the glassy phases of MSWI slag. Corrosion of glass might be slow, but if heavy metals are predominantly contained in other more accessible phases or near the surface of glass particles, the delaying effect of glass corrosion won't be noticeable.

It can be concluded that the occurrence of glass phases in waste pose *some* barrier function that delays emissions into the future. Ultimately, also glasses will be corroded and their constituting materials will be further weathered. With the limited available information, the delay can not be quantified but seems to be in the range of tens to hundreds of years, depending on the size of unfractured, homogenous glass domains. This range of time seems small compared to the total long-term time period of 60'000 years modelled in this study[47].

The calculations of transfer coefficients in the present landfill model are based on the relation between total content of elements in the waste (*m*) and the *measured* concentrations of these elements in the leachate (c_o). Cf. Eq. 5.7 and Eq. 5.8 on page 23. Glassy phases, that would initially prevent elements from leaching out, are heeded in this *measured* leachate data. It was decided above not to correct for the effects of glassy phases. This negligence actually leads to *smaller* inventoried emissions, because any initial abating effect of glasses occurring in the first hundred years or so, is extrapolated *without correction* into the long-term future. In this respect, the whole landfill inventory profits from any abating effect of initially present glasses.

Categorisation of landfill emissions

In chapter 'Lifetime of barriers' on page 20 it was concluded that it is just a question of time until pollutants, that leaked into the ground below the landfill, reach the groundwater. In this study all long-term emissions >100 a are integrated over time (see section 'The default procedure for future emissions in LCA' on page 15). Consequently, retention times are not detailed in this study. It is assumed that after 100 years, the collection system fails, the base seal leaks and leachate is entering the ground below the landfill. Hence, the *long-term emissions are inventoried as emissions to groundwater* (exchange subcategory 'Water, ground-, long-term'). The exchange subcategory 'soil' would not be appropriate, as this normally refers to emissions to the soil *surface* with e.g. wind erosion which does not occur in deep soil.

The short-term emissions of inorganic landfills are expected to be collected and discharged into a surface water body. The exchange subcategory 'Water, river' is chosen for short-term emissions. The short-term emissions of sanitary landfills are collected, discharged into a sewer and treated in a municipal wastewater treatment plant.

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[47] Assuming a homogenous distribution of elements in the slag material, assuming that 40% of slag is glassy, and 30% of the glassy phase has (overestimated) domain sizes of over 1 cm with corrosion times of over 1000 years, the mistake in emissions over the course of 60'000 years from neglecting these glassy phases is less than 1%.

Uncertainty of transfer coefficients and emissions

Transfer coefficients of landfills bear significant uncertainties, mainly because of the long time spans involved. Transfer coefficients represent the fate of a chemical element in a landfill. It is important to distinguish this *uncertainty in fate* from the *uncertainty in the waste composition*[48]. The uncertainty of the waste composition is already heeded in the waste input. In defining a uncertainty value for transfer coefficients, one has to ask, how uncertain the landfill behaviour is even for a perfectly defined waste.

Similar to the transfer coefficients for incineration (see part II), it can be argued that a major pathway (i.e. a large transfer coefficient) has a low uncertainty, as this pathway seems to be popular and 'well travelled'. For example chlorine is well soluble in leachate and is likely to be washed out rapidly. The transfer coefficient is large and the heterogeneity in the landfill can have but a small influence on the overall chlorine emissions. On the other hand the outcome of a minor pathway can be influenced by many stochastic events of which there are many in the heterogeneous milieu of the landfill body.

The uncertainty for short-term transfer coefficients (ST) is assumed to follow a lognormal distribution. It is assumed that for minor pathways (i.e. for small values of the mean m_{ST}) the GSD_{ST}^2 is approximately 10; i.e. the confidence range for a ST value is between $m_{ST}/10$ and $m_{ST}*10$. The GSD for large m_{ST} decreases and approaches 1 (100%) for $m_{ST} = 1$. To derive GSD_{ST} values from m_{ST} values the following formula is used (cf. uncertainty calculations for TK in part II on incineration):

$$\text{Eq. } 5.21 \quad GSD_{ST} = N \cdot \ln(m_{ST}) + c \quad \text{with N = -0.18, } m_{ST} \text{ as [kg/kg] and c =1}$$

The value of N is chosen so that the GSD^2 value for the smallest m_{ST} values are approximately 10. The smallest m_{STTK} are in the range 0.0006% to 0.001%. The lognormally distributed short-term transfer coefficients are multiplied with the lognormally distributed waste composition to obtain the *short-term emissions*.

The uncertainty of the long-term transfer coefficient (LT) are given as a triplet of minimal, average, and maximal values (lower = l_{LT}, mean = m_{LT}, upper = u_{LT}). Those values were derived with the procedure described in section 'Long-term emissions' on page 25. These values represent a triangular distribution. It was first intended to use this distribution directly for transfer coefficients and accordingly to enter long-term emissions as a triangular distribution. Discrepancies in the implementation of triangular distributions in the ecoinvent 2000 database made it impossible to use this approach without violating the mass balance for the medium case[49]. Instead a lognormal approach is used also for long-term transfer coefficients. It is assumed that the medium and maximum case value

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[48] The uncertainty range shown in leachate development in Fig. 5.7 from (Belevi & Baccini 1989) on page 22 is entirely derived from the variability in the waste composition.

[49] In the landfill model the average or medium case value of a triangular distribution is actually a Maximum Likelihood value MLV. It was originally intended to use this Maximum Likelihood Value as the mean (average) value for emissions in the inventory. However, for the ecoinvent 2000 database it is mandatory to enter the *arithmetic mean* of the triangular distribution (Frischknecht et al. 2003a). The arithmetic mean is expressed by (Min+MLV+Max)/3, i.e. the unweighted mean of the minimal, the maximal and the MLV (geometrically, the mean expresses the *centre of gravity* of the triangle, while the MLV is the *top corner*). In the originally intended model calculation, the MLV would have fulfilled mass balances, i.e. the use of MLV as the average value would assure that the emitted mass is not larger than the mean input mass. The *arithmetic mean* however can become larger than the MLV, if the distribution is left-skewed. Since for the maximum emission value the upper limit waste composition is used (and not the average) the *arithmetic mean* does not fulfil mass balances. With the *arithmetic mean* as the average case for emissions (as prescribed for triangular distributions in ecoinvent 2000) the mass balances cannot be fulfilled. This shortcoming has led to abandoning triangular distributions in this report altogether and some late changes.

represent the mean and upper value of a lognormal distribution. The lognormal distribution has then a GSD^2 of $(maximal/mean)^{50}$.

The long-term coefficients expressed in Tab. A.7 on page 117f. express the *total* transfer for the short-term and the long-term emissions *together*. To avoid double counting the short-term emissions must be subtracted from the total long-term emissions. This subtraction is performed already on the level of transfer coefficients, i.e. transfer coefficients for the emissions >100a alone are calculated. These transfer coefficients are labelled ΔLT, i.e. the difference between the total long-term and the short-term transfer coefficients. Following procedure is applied.

The mean ΔLT is derived by subtracting the mean short-term transfer coefficient from the mean long–term transfer coefficient.

$$\text{Eq. 5.22} \quad m_{\Delta LT} = m_{LT} - m_{ST}$$

The maximal ΔLT is derived by subtracting the lower short-term transfer coefficient value from the maximal long–term transfer coefficient value.

$$\text{Eq. 5.23} \quad u_{\Delta LT} = u_{LT} - l_{ST} = u_{LT} - \left(\frac{m_{ST}}{GSD_{ST}^2} \right)$$

As explained above, the distribution of the ΔLT TK is assumed to be lognormal. The GSD of this lognormal distribution is derived from the mean and upper value:

$$\text{Eq. 5.24} \quad GSD_{\Delta LT} = \sqrt{\frac{u_{\Delta LT}}{m_{\Delta LT}}} = \sqrt{\frac{u_{LT} - \left(\frac{m_{ST}}{GSD_{ST}^2} \right)}{m_{LT} - m_{ST}}}$$

The long-term emissions after 100a are then calculated by multiplication with the landfilled waste composition. Both factors are lognormally distributed and the GSD of the resulting emission is calculated by using the uncertainty propagation formula for multiplication.

This procedure neglects the information provided by the minimal TK value in Tab. A.7 on page 117f.. The minimal TK value applied in the database is determined by the mean and the GSD value. I.e. the used minimal LTTK does *not* refer to the emissions, that occur up to the end of the carbonate phase, as discussed in 'Long-term emissions' on page 25. The deviations of this minimal value (ΔLT) to the minimal data expressed in Tab. A.7 on page 117f. (given from ST and LT) are of minor importance.

As explained in sections 'Short-term emissions' and 'Long-term emissions', the transfer coefficients are largely calculated from literature data on the average waste composition *m* and the average initial leachate concentrations c_o from slag compartments and residual material landfills. The literature data is shown in the appendix (Tab. A.2, Tab. A.3, and Tab. A.4 on page 110ff. in the appendix). For the 'common' heavy metals like Cd, Cr, Cu, Hg, Ni, Pb, and Zn there is usually a lot of data. For some more 'exotic' elements there is little data available. Given the large possible variations in waste composition and leachate concentrations, the uncertainty is high for poorly referenced elements, i.e.

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[50] The transfer coefficients will in reality not follow a lognormal distribution. A transfer coefficient cannot go beyond the range of 0–100%. The lognormal distribution is always left-skewed, has a long tail to the right and has a non-zero probability for values larger than 100%. For transfer coefficients distribution that is right-skewed for mean values close to 100% would be more suitable (e.g. the beta distribution. Not available in ecoinvent 2000). Also suitable is a discrete range between 0 and 100%, i.e. zero probability outside 0% ≤ x ≤ 100%. In the model, however, distributions will be narrow for most elements (i.e. the mean is close to the upper boundary value) and the probability for values larger than 100% is very small.

there is not enough data to allow averaging over several landfill sites. Still, this data is used – even if derived from few data points – as this represents the measured state of knowledge. The resulting uncertainty from inferior averaging is assumed to be of minor importance compared to the uncertainty ranges derived above.

5.6 System boundaries

The goal of this study is to inventory disposal of different waste materials in landfills and subsurface deposits, so that this data can be used in LCAs of waste-producing activities. The slag compartment and residual material landfill are also used for the inventory of waste incineration (see part II).

Fig. 5.13 shows the process chain for landfilling of municipal waste. First the waste needs to be transported from the location of the waste-producing activity to the landfill. *Within the scope of ecoinvent 2000 this transport must be inventoried by the waste-producing activity and not the waste disposal process.* To avoid double counting the waste inventories devised in this report, do not contain the transport of the waste[51]. In the sanitary landfill, leachate is collected the first 100 years and treated in a municipal wastewater treatment plant. The resulting wastewater treatment sludge is incinerated in a municipal incineration plant. The incineration residues are landfilled in slag compartments and residual material landfills.
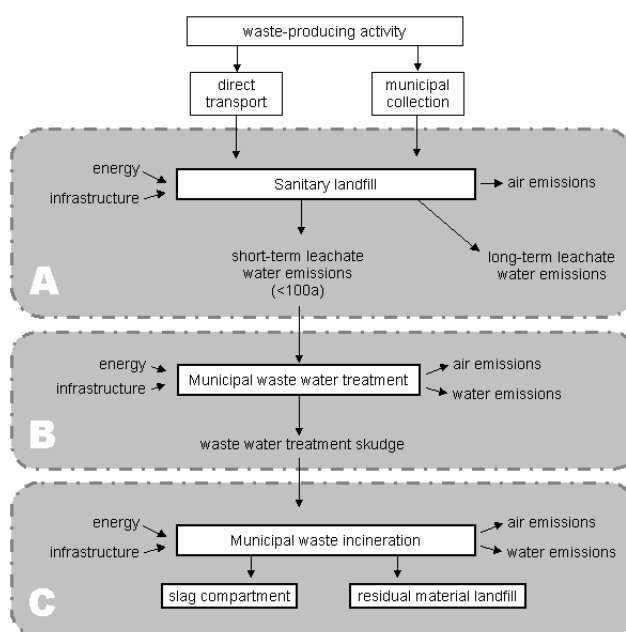

Fig. 5.13   Process chain for landfilling of municipal waste

The landfilling of waste leads to direct air and water emissions as well as land use burdens. Indirect burdens originate from the consumption of energy and infrastructure materials. The data modules devised in this study contain all the burdens from the landfill (A), and from the wastewater treatment (B) and from incineration and slag/residual landfills (C). The burdens associated with wastewater treatment are described in part IV of this report. The waste-specific emission data module also calls up two data modules for landfill infrastructure and for process-specific burdens, see Fig. 5.14. Both these modules are independent of waste composition.

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[51] This is in contrast to the waste inventories performed before. In (Frischknecht et al. 1996, Hellweg 2000, Doka 2002) the waste inventories already contained the transport to the landfill.
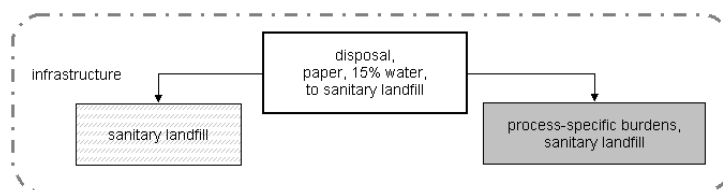
Fig. 5.14   System boundary and data modules for the inventory of landfilling of municipal waste

5.7   Necessary waste-specific data

To inventorise waste-specific burdens, the most important information within the landfill models is the elemental composition of the waste. An extensive vector of elemental concentrations in the waste can be specified by the user in the Excel calculation workbook on incineration '13_MSWI.xls' located on the on the CD-ROM. This data is given in kilogram element per kilogram wet waste. All composition data – also upper and lower heating values – refers to the *wet composition* of the waste. The corresponding water content can be specified for information purposes, although it has little consequence later in the calculations, but might be of significance if the heating values are extrapolated using the formulas of Michel (1938). For energy balance calculations, the upper and lower heating value must be specified in megajoules per kilogram wet waste. The lower heating value should be consistent with the given water content.

For disposal in sanitary landfills, the user must additionally specify the degradation rate during the first 100 years (See chapter 6.1.1 'Waste-specific degradability in sanitary landfills' on page 42).

Within the ecoinvent 2000 project, CO_2 and CH_4 air emissions are separated into emissions originating from biomass (paper, cardboard, vegetable waste, wastewater sludges) and from fossil material (plastics, mineral oil, solvents). The user must specify, what percentage of the given carbon content is biogenic.

For disposal in residual material landfills, the user can specify, if the waste is solidified with cement before deposition. The same solidification procedure as for MSWI residual material is assumed (50% residue, 30% cement, 20% water. See part II on incineration).

5.8   Waste heat from landfills

Decomposition of municipal waste produces a lot of heat. The decomposed material liberates waste heat from the microbial breaking up of chemical bonds. Some of the waste's energy is still contained as chemical energy in the landfill gas as methane. Some of the landfill gas is incinerated and the methane energy is liberated. In sanitary landfills the full upper heating value of the waste *without the chemical energy in non-incinerated methane* is inventoried as waste heat to air and water.

Also in inorganic landfills temperature gradients are observable due to e.g. carbonation of metal oxides and -hydroxides in ashes (cf. Fig. 5.4 on page 10). These energies dissipate and represent real waste heat emissions of such landfills. However, the energy needed for the formation of such energy containing ash minerals originates from the incineration of waste in the waste incinerator. The full waste heat amount of the waste is already inventoried in the incineration process, i.e. storage of chemical energy in incineration residues was not heeded in the incineration in accordance with the ecoinvent 2000 methodology (Frischknecht et al. 2003a). To avoid double counting of waste heat, no waste heat emissions from incineration slag and residue landfills are inventoried. The case of heat from corrosion processes of metals in waste is discussed in part I on incineration for steel and aluminium metal waste to municipal waste incineration.

5.9   Landfill infrastructure

In landfills some construction materials are used for ground work, base seals, flank seals and leachate collection systems that can be regarded as the 'infrastructure' of a landfill, while the landfilled waste is the 'product' of the landfill. In most industrial processes the infrastructure or production means are used to perform a desired task on the product. The product enters, proceeds through and leaves the infrastructure. The infrastructure is removed or replaced when aged. By contrast, the landfill infrastructure remains on site after landfill closure and is *not removed*. In a manner of speaking the infrastructure becomes part of the landfill. Also the landfill 'product' – the waste – remains in the 'factory' – the landfill – and is not removed. The landfill infrastructure is more like a permanent 'packaging' or 'property control device' for the waste, and much more closely combined with the 'product' – the waste – than production infrastructure is with its product.

In the ecoinvent 2000 project, infrastructure must be inventoried in separate data modules. This allows for LCI calculations with or without infrastructure. The landfill infrastructure is inventoried accordingly in a separate data module. Users who wish to use LCI data without infrastructure should check the compatibility of their rationale of doing so with the fact that the 'stationary' infrastructure of landfills is not included in such data.

5.10   Land occupation and transformation of landfills

Land occupation is inventoried separately from land transformation in ecoinvent 2000. Usually in industrial processes, land occupation is associated with infrastructure, e.g. areas occupied by factories. In landfills, the land occupation occurs not by mere infrastructure, but by the 'product', i.e. the waste itself during deposition. Land occupation and transformations for landfills are therefore not inventoried within the landfill *infrastructure* module (see chapter 5.9 'Landfill infrastructure'), but along with the *process expenditures*[52]. In this respect, land use of landfills resembles agriculture or open pit mining of coal, where the product itself leads to land use and not the production means.

5.10.1 Ecoscarcity eco-factors for occupied landfill volume

For land transformations and occupations associated with landfills the surface type with the CORINE code 132 ('dump site') is suitable. In ecoinvent 2000 this code is differentiated into several types for several near-surface landfill types (codes 132b-132e in Tab. 5.1). This is not to suggest, that the ecological quality of these landfill types are significantly different[53]. The reason for this differentiation is that in the LCIA method of ecological scarcity[54] a midpoint eco-factor per kilogram landfilled waste exists (BUWAL 1998). Ecoscarcity does not differentiate the different surface landfill types, so the *same* eco-factor is applied to each kilogram of landfilled waste[55]. This eco-factor is 500 eco-points/kg. In the LCIA calculations of the ecoinvent database, direct valuation of technosphere processes is not possible. It is therefore not possible, nor pragmatic the valuate each waste module 'Waste XY in landfill type Z' with this eco-factor directly[56]. To be able to fully assign the Ecoscarcity method in ecoinvent 2000, an approach via the landfill land area was chosen. The landfill land area is inventoried with every landfill waste module. The necessary land area for the landfilling of one kilogram waste

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[52] One exception is the land use for the access road, which is considered to be an infrastructure part.

[53] Though a sanitary landfill with vermin and food wastes will have a different internal biodiversity and also a different impact on the biodiversity of the surrounding land than a inert material landfill. These effects are not quantified in this report.

[54] Synonyms: Ecoscarcity, BUWAL method, Swiss Ecopoints, German: Umwelt-Belastungs-Punkte, UBP'97.

[55] There are however different ecofactors for wastes to underground deposits (salt mines) and for radioactive wastes.

[56] This would be not pragmatic because each time a *new* landfill waste module were created, the LCIA calculation matrix for Eco-scarcity would have to be expanded to include that module.

can be calculated from the landfill depth and the waste density. This area is inventoried in the database as a transformation to and from a landfill area (in m^2) and as an occupation of landfill area for the duration of the landfill operation (in m^{2}a) for each kilogram of waste. Since the average depth and waste density is different for each landfill type, different areas per kilogram waste result. Since the concerned area is inventoried directly as a land transformation, it is possible to attach an adapted waste eco-factor in 'eco-points per m^2 landfill area' to the inventoried landfill area transformation[57]. The adapted eco-factor must be differentiated for the different landfill types (see Tab. 5.1). Using these modified eco-factors, each kilogram landfilled waste will be attributed a constant burden of 500 eco-points.

Tab. 5.1   Differentiated CORINE land types for landfills and ecofactors for landfill areas

CORINE code	Landfill type	Waste density kg/m^3	Landfill depth m	Kilogram waste per m^2 landfill area kg/m^2	Ecofactor per m^2 landfill area (from 500 eco-points/kg) eco-points/m^2
132	dump site (general)	–	–	20'000 [1]	**10'000'000**
132a	dump site, benthos [2]	–	–	0.92 – 3.85	**462 – 1925**
132b	dump site, sanitary landfill	1000	20	20'000	**10'000'000**
132c	dump site, slag compartment	1500	15	22'500	**11'250'000**
132d	dump site, residual material landfill	1600	10	16'000	**8'000'000**
132e	dump site, inert material landfill	1500	15	22'500	**11'250'000**

1   The landfill type 'dump site (general)' is not used in this report, but elsewhere in ecoinvent 2000 (e.g. uranium milling tailings, hard coal mining, silicone plant infrastructure). The value from sanitary landfill was adapted.

2   Used for offshore drilling wastes spread on the seafloor. With 260 m^2 covered area per drilling meter and 240 – 1000 kg waste per drilling meter.

5.11   Underground deposit volume occupation

In ecoinvent 2000, the volume occupied in underground deposits is inventoried as a consumption of a natural resource (Volume occupied, underground deposit). This exchange signifies the volume actually occupied by the waste and not the complete volume of the emptied salt vein[58]. This exchange can be used to apply the eco-factor for underground deposits from the LCIA method of ecological scarcity (BUWAL 1998). In this method each kilogram of waste in a underground deposit is valuated with 24'000 eco-points (irrespective of its composition). With an average density of 1600 kg/m^3 (estimate), an adapted eco-factor of *38.4 Million eco-points per m^3 occupied underground deposit volume* is calculated. This eco-factor is applied to the exchange 'Volume occupied, underground deposit, m3'.

No information on the complete volume of the inventoried underground deposit Herfa-Neurode is available. The complete deposit area is given as 20 km^2 (Brendel 2000). With an average ceiling height of 2.7 m a total volume of 54 million m^3 can be calculated.

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57   The area ecofactor is applied only to the 'transformation *to* dump site type Z' and not to 'transformation *from* dump site type Z'. Applying it to *both* would be double counting.

58   There is usually some empty space between the deposited waste and the cavern wall, cf. Fig. 5.5 on page 12.