

Substance Management of Anthropogenic Residues



Substance Management of Anthropogenic Residues

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Wissen und Erkennen sind die Freude und die Berechtigung der Menschheit.

Knowledge and recognition are the joy and raison d'être of humanity.

Alexander von Humboldt

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Abstract

Landfill Mining (LFM) is an approach to managing anthropogenic residues of past decades in accordance with current technological, economic, political, societal and environmental conditions and regulatory frameworks. LFM is defined as resource recovery from closed or active landfills by means of excavation, processing and recycling of waste. Apart from the main objective to recover resources, environmental risks – such as groundwater pollution and uncontrolled landfill gas emissions – can also be eliminated. Previous research has focused on the resource potential of landfills and the related climate impact due to the incineration of refuse-derived fuel (RDF) and to avoiding landfill gas emissions. However, LFM involves many stakeholders and depends on a large number of factors along the process chain. In addition, landfills – worldwide – consist mainly of soil-like materials ("soils"), which are of little or no market value.

The objective of this dissertation is to evaluate LFM from prospection to processing and recycling using data from eight mined landfills, and to place an emphasis on soils. Material flow analysis (MFA) and substance flow analysis (SFA) were considered appropriate methods to assess the resource potential of LFM and the ecological performance of its processes.

In a first step, prospection sampling was evaluated in terms of contamination prediction quality by comparing samples from preliminary investigations with those from excavations. This evaluation also involved comparing two investigation methods - core drilling and grab crane. In addition, using statistical methods enabled (a) the identification of contaminant patterns within and between landfills, (b) the determination of potential indicator elements, and (c) the evaluation of legal limit values with regard to manage substance flows. Secondly, assessing the contaminant reduction effectiveness of processing equipment consisted of comparing contaminant concentrations in fine-, middle- and coarse-grained soils from four different processing trains. After that, material flows, energy consumption and related emissions of all LFM operations were analysed in a regional context. Finally, a modified PEST analysis (political, economic, socio-cultural and technological) enabled the identification of factors affecting material flows and influencing operation processes.

In terms of prospection, sampling using a grab crane and core drilling showed sufficiently accurate prediction results for most heavy metals, cyanides (CN), polycyclic aromatic hydrocarbons (PAHs), sulphate, barium, benzo[a]pyrene (BaP), pH and electrical conductivity (EC). Samples from grab-cranes proved better at predicting contamination concentrations than those from core drilling, even for smaller investigations of ten samples. Substance dispersions did not affect the reliability of prediction. Analysing contaminant patterns showed correlations between several heavy metals, sulphate and EC, as well as ammonium nitrogen and biodegradability. Sulphate, pH and total organic carbon (TOC) were the most efficient indicator elements. Legal limit values have turned out to be efficient to manage substance flows with regard to chloride, sulphate, dissolved organic carbon (DOC), cadmium, lead and zinc. In contrast, flows of polychlorinated biphenyl (PCB), PAHs, BaP, fluoride, mercury and biodegradability tended to be unaffected by legal limit values.

With regard to processing, concentrations of heavy metals, PAHs and TOC could be sufficiently reduced in the coarse-grained soils, while the concentration differences of leaching test parameters, such as pH, EC, barium and DOC proved to be heterogeneous and less pronounced (except for sulphate). In contrast to the common substance accumulation in the fines, fluoride and chloride tended to accumulate in coarse-grained soils. Comparing screens with a mesh size of 35 mm, 50 mm and 70-80 mm indicated that 50 mm openings performed more effectively with respect to contaminant redistribution and the proportion of material flows (fine- and coarsegrained soils). However, the optimum mesh size might be between 35 mm and 50 mm.

Analysing material flows and sub-processes in a regional context revealed that transportation required the largest share of energy and produced most (58%) of the emissions, followed by processing (27%). Transportation distances of soils turned out to be greater (84 km) than assumed in previous studies (5 to 50 km). The PEST analysis – with the categories technology, economy, institutions / laws, or-ganisation and landfill properties – revealed the complexity of LFM projects due to their individual character, the broad range of stakeholders and numerous interfaces. Consequently, flexibility, pragmatism and coordination of stakeholders turned out to be key factors.

Keywords:

Landfill mining, regional material flow analysis, substance flow analysis, carbon foot print, influencing factors, logistics of soils, transportation, processing technology, sampling, indicator element, contamination prediction, limit value, remediation, construction and demolition waste

Zusammenfassung

Landfill mining (LFM) ist eine Methode deponierte Abfälle unter Berücksichtigung gegenwärtig technologischer, wirtschaftlicher, politischer, gesellschaftlicher, ökologischer und gesetzlicher Rahmenbedingungen zu verwerten. Primäres Ziel ist die Rückgewinnung von Wertstoffen, außerdem kann durch den Rückbau sanierungsbedürftiger Deponien das Grundwasser vor Verunreinigungen geschützt und Deponiegasemissionen vermieden werden. Bisherige Forschungsarbeiten konzentrierten sich auf die Untersuchung des Wertstoffpotentials und Klimaauswirkungen die einerseits durch die thermische Verwertung rückgewonnener Wertstoffe (Kunststoffe, Holz) und andererseits durch die Vermeidung von Deponiegasemissionen bedingt sind. Jedoch stellte sich heraus, dass das Wertstoffpotenzial gering ist und Deponien zu einem großen Anteil aus bodenähnliche Bestandteilen ("Boden") bestehen dessen Entsorgung in der Regel mit Kosten verbunden ist. Außerdem zeigte sich in der Praxis, dass eine Bewertung allein auf Grundlage des Wertstoffpotenzials nicht ausreicht, da die Durchführung von LFM-Projekten von zahlreichen Faktoren entlang der Prozesskette (Erkundung, Aufbereitung und Verwertung) beeinflusst wird.

Ziel dieser Dissertation ist die Bewertung von LFM von der Wiege bis zur Bahre wobei ein besonderes Augenmerk den Böden gilt. Für die Bewertung des Wertund Schadstoffpotenzials sowie der ökologischen Auswirkungen von LFM-Prozessen wurden Materialfluss- und Stoffstromanalysen durchgeführt. Die Bewertung der Prozesse Erkundung, Aufbereitung und Verwertung basiert auf Daten von bis zu acht rückgebauten Deponien.

Die Zuverlässigkeit der Erkundungsmethoden (Rammkernsondierung und Baggerschurf) wurde anhand eines Vergleichs von Bodenproben aus den Voruntersuchungen und Haufwerksbeprobungen (Deklarationsanalytik aufbereiteter Abfälle) bewertet. Außerdem wurden mittels statistischer Verfahren für den Bodenanteil von Deponien a) Schadstoffregelmäßigkeiten identifiziert, b) Indikatorsubstanzen zur Schadstoffprognose abgeleitet und c) gesetzliche Grenzwerte hinsichtlich der Wirksamkeit zur Lenkung von Schadstoffströmen beurteilt. Um die Effektivität der Aufbereitung zu bewerten wurden die Schadstoffkonzentrationen der Fein-, Mittelund Grobkornfraktionen verglichen. Die anschließende Erstellung einer regionalen Materialflussanalyse (Abfälle, Baumaterialien und Energieträger) ermöglichte den Einfluss der regionalen Infrastruktur und den Energiebedarf sowie Emissionsausstoß der einzelnen Prozesse zu bestimmen. Abschließend wurde eine modifizierte PEST-Analyse (soziokulturell, technologisch, ökonomisch, politisch) durchgeführt um die Einflussfaktoren der Materialflüsse und Prozesse zu identifizieren.

Die Vergleich der Erkundungsmethoden (Rammkernsondierung und Baggerschurf) zeigte, dass beide Methoden eine zuverlässige Prognose für Schwermetalle, Cyanide, polyzyklische aromatische Kohlenwasserstoffe (PAK), Sulfate, Barium, Benzo[a]pyren, pH-Wert und elektrische Leitfähigkeit ermöglichten. Die Probenahme mittels Baggerschürfe erzielte präzisere Ergebnisse auch für kleine Stichprobengrößen (n=10). Die Streuung von Schadstoffen wirkte sich nicht auf die Prognosequalität aus.

Die Analyse zu Schadstoffregelmäßigkeiten ergab starke Korrelation zwischen Schwermetallen, Sulfat und elektrischer Leitfähigkeit sowie Ammoniumstickstoff und Atmungsaktivität (AT4). Als geeignete Indikatorsubstanzen konnten Sulfat, pH-Wert und gesamter organischer Kohlenstoff (TOC) identifiziert werden. Der Vergleich von Schadstoffkonzentrationen in Böden mit gesetzlichen Grenzwerten zeigte eine effiziente Steuerung von Schwermetallen, Chlorid, Sulfat und gelösten organischen Kohlenstoffströmen (DOC), nicht jedoch für polychlorierte Biphenyle (PCB), PAKs, Benzo[a]pyren, AT4, Fluorid und Quecksilber.

Die Aufbereitung in Bodenbehandlungsanlagen zeigte eine deutliche Reduzierung von Schwermetallen, PAKs und TOC in der Grobfraktion. Konzentrationsunterschiede von Substanzen und Parametern in Eluatanalysen (pH, elektrische Leitfähigkeit, Barium, DOC) waren, mit Ausnahme von Sulfat, insgesamt geringer und unregelmäßiger. Außerdem tendierte Fluorid und Chlorid in der Grobfraktion zu akkumulieren. Der Vergleich zwischen 35 mm, 50 mm und 70-80 mm Sieböffnungen ergab, dass 50 mm Öffnungen am effizientesten Schadstoffgehalte in der Grobfraktion reduzierten und gleichzeitig ein ausgewogenes Mengenverhältnis zwischen Fein- und Grobfraktion entstehen lassen. Jedoch weisen die Untersuchungen auf einen optimalen Sieböffnungsdurchmesser zwischen 35 mm und 50 mm hin.

Die Materialflussanalyse zeigte, dass Transporte den größten Energieverbrauch und Emissionsausstoß (58%) aufwiesen, gefolgt von der Abfallaufbereitung (27%). Die Transportdistanzen für Boden waren deutlich länger (84 km) als in bishreigen Untersuchungen (5 bis 50 km) angenommen. Die PEST-Analyse bestand aus der Bildung der Kategorien Deponieeigenschaften, Technologie, Wirtschaft, Institutionen/Recht und Organisation. Eine Vielzahl identifizierter Einflussfaktoren ließ einen hohen Grad an Individualität von LFM-Projekten und eine große Anzahl an Beteiligten mit zahlreichen Kommunikationsschnittstellen erkennen. Durch die daraus resultierende Komplexität zählten Flexibilität, Pragmatismus und die Koordination von Beteiligten zu den Schlüsselfaktoren.

Schlagwörter:

Landfill Mining, Deponierückbau, regionale Materialflussanalyse, Stoffflussanalyse, CO₂-Bilanz, Einflussfaktoren, Aushublogisitk, Transport, Aufbereitungstechnik, Probenahme, Indikatorelement, Schadstofferkundung, Grenzwert, Altlastensanierung, Bodenaushub, Bau- und Abbruchabfälle

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1 Introduction

1.1 Background and problem definition

Landfills are considered as contaminated sites posing a threat to human health and the environment. Landfill mining (LFM) is an approach to managing the anthropogenic residues of past decades in accordance with the current technological, economic, political, societal and environmental conditions of a region, and its regulatory frameworks. The main objective of LFM is recovery of recyclables, while individual projects also targeted the reclamation of land, extension of landfill lifetimes and prevention of environmental hazards (e.g. groundwater contamination, landfillgas emissions, landfill collapse; Krook et al. (2012)). However, recovery of recyclables also involves risks, as demonstrated by the reuse of contaminated lime from a Brazilian landfill which resulted in high dioxin concentrations in German dairy products (Torres et al., 2013; Weber et al., 2011).

LFM activities probably coincided with the creation of landfills. Recovery of recyclables has been recorded for the List landfill in Hanover/Germany in the 1930s, where the informal waste collectors needed a licence to collect materials from the landfill (Saniter and Köhn, 2001). Savage et al. (1993) reported recovery of soils for the purpose of fertilizer in orchards from a landfill in Tel Aviv in 1953. LFM activities significantly increased in the 1990s, especially in the USA (U.S.EPA, 1993; NYSERDA, 1998b; U.S.EPA, 1997). The main objectives – reclamation of landfill space and remediation of landfills – have also gained importance in Germany at that time (Göschl, 1995).

Previous research in LFM focused on characterization of materials, technology for excavation and processing, potential benefits, environmental impacts and safety issues (Krook et al., 2012). Geysen et al. (2012) introduced the concept of Enhanced Landfill Mining (ELFM) in 2010, that aimed at valorising the waste either as material or as energy. In this framework, the German collaborative research project "TönsLM" investigated ELFM as an alternative to the conventional landfill closure and aftercare (Krüger et al., 2016). The most recent international research has mainly examined the climate impact (Laner et al., 2016; Danthurebandara et al., 2015b; Frändegaard et al., 2013b), processing technology (Wanka et al., 2017; Danthurebandara et al., 2015c; Zaini et al., 2017), waste characterization (Hogland et al., 2018; Kaczala et al., 2017a) and socio-economic issues (Johansson et al., 2017a; Winterstetter et al., 2018; Bhatnagar et al., 2017; Fellner et al., 2018), while the current collaborative research project NEW-MINE (https://new-mine.eu/) is investigating processing technology, thermochemical conversion, upcycling opportunities and multi-criteria assessment to evaluate ELFM.

As yet, the European Commission has neither a strategy for dealing with landfills in future nor cost estimates of the total landfill-remediation costs (Vautmans, 2018). The European Enhanced Landfill Mining Consortium (https://eurelco.org/) and a few members of the European parliament are pushing for the integration of the ELFM concept into the amendments to Directive 1999/31/EC on the landfill of waste (Commission, 1999; Vautmans, 2018). This debate resulted in the development of the new term "Dynamic Landfill Management" (DLM) where ELFM is considered as an option for some landfills.

The question remains how society should deal with dumped materials from the past. LFM is considered an option to address this issue. However, LFM depends on a large number of factors along the process chain and involves many stakeholders, resulting in difficulties to evaluate opportunities and threats of LFM (Hermann et al., 2015). Every process – from prospection to processing and recycling – involves uncertainties where the previous process might significantly affect the following process all along the work-flow.

The composition of landfills – usually identified by preliminary investigations – is decisive for the planning of processing and for the determination of recycling opportunities. Consequently, the precision of preliminary investigations is of major importance, since errors will propagate and affect the complete process chain. The ISO18400-104 (2018) and ISO14688-1 (2018) standards define the approach to sample soils. However, previous LFM investigations have not applied these standards and a review of studies revealed great differences in sampling methods and sample preparation. These investigations are usually based on very few samples from test pits and sampling was usually carried out using a grab crane or core drills. For instance, the studies of Masi et al. (2014) and Rong et al. (2017) are based on one composite laboratory sample, while the studies by Mönkäre et al. (2016), Quaghebeur et al. (2013) and Zhou et al. (2015) involved six, ten and 22 samples, respectively. The distribution of contaminants in large composite samples tended to be more homogeneous, while sampling without composites resulted in stronger deviations. The main differences of previous studies were (a) pattern of sampling locations, (b) number of samples, and (c) preparation of samples. Composite samples are less suitable for evaluating the waste layers in terms of composition, age and extent (Patil, 2013). Substances showing strong variations might substantially affect the reliability of preliminary investigations and the interpretation of their results requires experience and knowledge. The precision of prediction usually depends on sample number and variation of data values, though a large sample number increases the costs. Apart from difficulties in comparing the different sampling approaches, the fundamental issues – to what extent preliminary investigations represent landfill compositions and which substance patterns might exist – have so far not been investigated.

Waste composition analyses are the most studied research subject and showed that landfills – worldwide – consisted mainly of soil-like materials (from now on referred to as "soils") and combustibles (Krook et al., 2012). However, the investigation on soils from landfills have not received much research (Krook et al., 2012; Kaczala et al., 2017a). With regard to substances in soils, previous research usually provided average contaminant concentrations and their standard deviations, while substance patterns and their prediction have rarely been researched (Brandstätter et al., 2014; Kaczala et al., 2017a). In addition, these studies are usually based on individual landfills neglecting to analyse substance patterns on a regional or international scale. Brunner and Rechberger (2017) concluded that the prediction of contamination, based on indicator elements, necessitates experience and knowledge. Consequently, to improve knowledge on contaminants requires research on substances with regard to patterns and relationships.

Processing is the second most studied main topic of LFM research (Krook et al., 2012). Processing enables the separation of waste resulting in individual materials for recycling, recovery or disposal. Previous research topices included recovery techniques, such as steam and plasma gasification (Zaini et al., 2017; Bosmans et al., 2013), processing trains consisting of screens, eddy current separators, crushers, air knives and manual sorting (U.S.EPA, 1993; Stessel and Murphy, 1999; Maul and Pretz, 2015), or soil washing technology (Wanka et al., 2017). Processing trains have been investigated in terms of the processing rate, stream purity, product quality, size distribution of materials and screen combinations, but not with regard to the generation of low contaminated soils for reuse.

Since landfills consist mainly of soils, their processing results strongly affect reuse opportunities and disposal costs (Krook et al., 2012). Jani et al. (2016) concluded that large soil quantities are crucial for economic assessment, since soils are of little financial value, or even involve costs for processing and disposal. Previous studies on soils based on dry screening tests were usually carried out on a laboratory scale (Masi et al., 2014; Rong et al., 2017; Jain et al., 2005; Di Maria et al., 2013; Quaghebeur et al., 2013). These investigations employed screens with openings up to 15 mm - this size might be hardly ever used in full-scale projects. Consequently, their conclusion that contaminant concentrations are lower in soils less than 10 mm might be without significance in practice, where coarse-grained soils seemed to be less contaminated due to the physical fact of less surface (Schachermayer et al., 1998). Consequently, the question arises to what extent might dry screening redistribute contaminants in soils in full-scale projects.

Once waste has been separated, its reuse, recycling, recovery or disposal depends on many factors apart from material characteristics (Hermann et al., 2016). For instance, Goeschl and Rudland (2007) reported disposal of plastics from the mined Sharjah landfill, due to long transportation distances to a waste-to-energy (WtE) plant. Van Passel et al. (2013) concluded that reuse options of materials and markets for recycled products are of major importance. LFM evaluations were usually based on theoretical models involving assumptions (Danthurebandara et al., 2015b; Frändegaard et al., 2013a; Jain et al., 2014; Laner et al., 2016). The quality of these models depended on data from practical experience and results varied strongly due to the different model choices and selected input parameters. Consequently, the uncertainty of these models is difficult to estimate. For instance, Hermann et al. (2016) determined 27 input variables, while Frändegaard et al. (2013a) used ten factors for his model and Laner et al. (2016) eight. Danthurebandara et al. (2015b) did not take transportation into account, Van Passel et al. (2013) and Laner et al. (2016) calculated low emissions from transportation, whereas Frändegaard et al. (2013a) identified transportation as the second-largest factor for added green-house gases. With regard to LFM operations (i.e. excavation, processing, rehabilitation and transportation), so far only Jain et al. (2014) have researched the environmental impact; however, this study was based on data from a single landfill and neglected significant quantities of soil. Consequently, the question arises how to evaluate LFM taking all materials, processes, the regional setting and business environments into account.

1.2 Objective and research questions

This dissertation investigates the management of materials – particularly soils – from landfills taking into account technological, economic, societal, political, legal and ecological factors. The investigation covers the LFM process chain from prospection, to processing and recycling based on analyses of materials, substances and flows from eight mined landfills. The objective of this research is to evaluate the current practices of LFM and to provide a better basis to amend circular economy policies with regard to management of old landfills. This research evaluates:

- The reliability of preliminary investigations analysing
 - to what extent substances and parameters can be predicted
 - how perform the two investigation methods using a grab crane and core drillings
- Substance concentrations in soils examining
 - which substance patterns exist
 - which substances may serve as indicator element
 - to what extent legal limit values can be an instrument to manage substance flows
- The effectiveness of processing investigating
 - whether and to what extent substances can be redistributed in soils
 - which screen mesh opening size most effective redistributes substances
- Regional material flows researching
 - which import, export and stocks are generated
 - how much energy consumption and emissions arise from operations
 - which factors influence LFM.

1.3 Thesis structure and approach

The present dissertation comprises four research articles evaluating the substance management of LFM along the process chain. The publication "Contaminants in landfill soils – reliability of prefeasibility studies" (Hölzle, 2017) evaluates the reliability of preliminary investigations, while the publication "Contaminant patterns in soils from landfill mining" (Hölzle, 2019b) analyses contaminant patterns and identifies indicator elements. Following on the publication "Dry screening – assessing the effectiveness of contaminant reduction in recovered landfill soils" (Hölzle, 2018) compares different processing technology in terms of substance redistribution in soils. The last publication "Analysing material flows of landfill mining in a regional context" (Hölzle, 2019a) investigates material and energy flows of LFM as well as factors influencing the business environment.

1.3.1 Materials and methods

The eight investigated landfills with municipal solid waste (MSW) and construction and demolition waste (CDW) were completely excavated and remediated in the German Federal State of Bavaria (Table 1.1). These landfills were used between the 1950s and 1980s and lacked bottom and surface sealings. The protection of drinking water abstraction required complete excavation. The landfills each had a waste quantity of up to 30,957 tonnes and surface area between 1,450 m² and 6,130 m². In total 121,133 tonnes of waste were excavated, treated and as far as possible recycled in line with the waste management hierarchy of prevent, reuse, recycle, recover and dispose (KrWG, 2012). Operations consisted of the following phases: pre-evaluation, evaluation, excavation, processing, recycling and rehabilitation.

Pre-evaluation and evaluation were carried out in accordance with the federal soil protection act (BBodSchG, 1998) and processing in line with the law on circular economy (KrWG, 2012). With regard to recycling, the technical guidelines for recycling soils ("RC guidelines", LAGA 2003), the German landfill ordinance ("LF ordinance", DepV 2009) and the waste wood ordinance (AltholzV, 2002) were most relevant.

Processing was carried out on-site if the groundwater situation was less sensitive and the size of the area allowed mobile processing equipment to be set up; otherwise, the waste was processed off-site at specialised plants. Processing involved the employment of mechanical screens, gravity separators, cross-belt magnets, crushers and air classifiers as well as manual sorting.

The landfills consisted of soils, CDW, plastics, wood, tyres, metals and hazardous waste. Soils and CDW were analysed for contaminant concentrations, since recycling and thermal recovery of scrap, wood, tyres and plastics did not require substance analysis. Analyses of inert waste mainly included heavy metals, organic compounds, ionic compounds (e.g. sulphate, ammonium nitrogen, fluorine) and further parameters such as EC, pH and biodegradability. The technical guidelines define - depending on the contaminant concentration - the potential reuse options for soil, for example in parking lots, noise barriers, sub-bases of roads, and backfilling of quarries and gravel pits (LAGA, 2003). More highly contaminated soils and CDW can be reused as substitute construction material at landfills, but if they exceed certain limit values must be disposed of at appropriate landfills (DepV, 2009). This dissertation is based on accounting, laboratory reports, consignment notes of transportation, daily construction records, reports of preliminary investigations, remediation assessments, project completions, field visits, documents and

Landfill	Area [m ²]	Excavated waste [t]	Disposal period
Ansbach	$4,\!185$	20,425	1960-1989
Lindau	$1,\!450$	4,197	1964-1972
Main-Spessart	$6,\!130$	25,828	1958-1975
Miltenberg A	5,820	$30,\!957$	1960-1977
Miltenberg B	3,900	4,027	1972-1977
Oberallgäu	$2,\!800$	9,311	1950-1975
Straubing	3,000	7,048	1950-1972
Traunstein	2,800	19,340	1964-1975

Table 1.1: Overview of the eight excavated landfills (Hölzle, 2019a).

communications of public and private stakeholders, stakeholder interviews, and data from the ProBas database (http://www.probas.umweltbundesamt.de) as well as ecoinvent database version 3.3 (Wernet et al., 2016). Soil samples were analysed at certified laboratories using standardized determination methods, generally ISO standards. Analyses of elements, substances, chemical compounds and parameters (referred to as substances in this thesis) were carried out for total concentrations and leaching tests (eluate analysis). Total concentration analyses included 21 substances and leaching tests 22. However, for certain statistical calculations not all substances were taken into account, particularly when values proved to be very low or below the detection limit (e.g. rare-earth metals, extractable organic halogens (EOX), phenols). In total the investigation included 301 samples from test pits and from excavation piles.

1.3.2 Method – prospection

Preliminary investigations enabled the identification of contaminant concentrations and their spatial distribution for environmental and economic evaluations. Two investigation methods – grab crane and core drilling – were assessed by comparing their samples with excavation samples. Evaluating the reliability of preliminary investigations involved a two-step approach of statistical calculations; firstly identifying deviations between preliminary investigation and excavation, and secondly verifying the significance of results (Figure 1.1). The statistical calculations comprised the calculation of (a) the weighted geometric means (GMs) and their coefficient of variations (CVs), (b) tests of normality and associated logarithmic transformations, (c) Mann-Whitney U test (MWW), and (d) Spearman's rho correlation coefficient.

The identification of deviations between preliminary investigation samples and

excavation samples consisted of a substance concentration comparison using the GM. The subsequent calculation of the CV allowed on the one hand to identify dispersion patterns, and on the other hand to evaluate the impact of substance dispersions on prediction quality. To determine if substance concentrations of preliminary investigation samples were similar (H_0) to those of excavation samples, the MWW was used since previous tests of normality showed that measurements were not distributed normally and logarithmic transformations had an insufficient effect. The relationships between the (a) CV and geometric mean differences, (b) CV of preliminary investigations and excavation, and (c) CV and MWW asymptotic significance were cross-checked using Spearman's rho correlation coefficient.



Figure 1.1: Statistical tests to compare preliminary investigation samples with excavation samples.

1.3.3 Method – contaminant patterns

Contaminant patterns were analysed with regard to the following four aspects: (a) the CV of the mean of each landfill was calculated to determine concentration variations within and between landfills, (b) the Spearman rank correlation coefficient (ρ) was used to qualify the correlations between substances, (c) the frequency of legal limit value exceedances allowed to identify problematic contaminants, and (d) cooccurrences of substances exceeding legal limit values showed relationships between contaminants (Figure 1.2). The results of these statistical analyses formed the basis to determine indicator elements. Indicator elements allow representation of a group of substances with specific properties such as frequently limit value exceedances, strong correlations or uncorrelatedness.

In contrast to quantifying material flow analysis (MFA), substance flow analysis (SFA) focuses on individual substances resulting from processes. Carrying out an SFA enabled the evaluation of legal limit values to manage substance flows in terms of recycling, recovery and disposal. Classified soils were compared to identify if substance concentrations in highly contaminated soils were in general higher or were limited to individual substances.



Figure 1.2: Statistical tests to identify substance patterns and to determine indicator elements.

1.3.4 Method – processing

Four different processing trains were compared with regard to contaminant redistribution in soils, since contaminants are assumed to accumulate in fines. Evaluating the efficiency of processing trains involved a two-step approach to statistical calculations: (a) quantifying substance concentration differences between fines and coarse-grained soils, and (b) verifying the significance of differences (Figure 1.3). The GM was used to quantify and compare substance concentrations of soils of different grain sizes. To verify the significance of substance concentration differences, the MWW was used since substance measurements proved to be positively skewed. The MWW determines if the substance concentrations of fines, medium-grained and coarse-grained materials are similar (H_0) or differ significantly, and consequently if substances can be accumulated in fines. In addition, the relation between GM differences and MWW significances was cross-checked using Spearman's rho correlation coefficient.



Figure 1.3: Statistical tests to compare substance concentrations of fines and coarsegrained soils.

1.3.5 Method – recycling

An MFA was carried out to identify waste streams, as well as the energy demand and related emissions of processes (e.g. excavation, processing, transportation). MFA is a method to analyse flows (input and output), stocks and processes of a system, taking into account time and space (Brunner and Rechberger, 2017). The objective of an MFA is to identify resource potentials and risks for human health and the environment by evaluating transformation, flows and storage of materials within a defined system (Stanisavljevic and Brunner, 2014). Materials consist of goods (plastics, tyres, construction materials), while processes comprise transformation of materials (processing, transportation) and flows describe the ratio of quantity per time (Figure 1.4). The mass balance principle defines that the input quantities into a system are equal to the outputs, taking into account changes in stocks.

To identify factors influencing stakeholder decisions and affecting operations and material flows, the business environment of LFM was analysed using an adapted PEST analysis (political, economic, socio-cultural and technological; Fahey and Randall (2001)). This analysis enabled the structured grouping of influencing factors into the classes economy, technology, organisation, institutions/laws, and landfill properties. The latter class comprised internal factors such as waste composition,



landfill size, topographical aspects etc.

Figure 1.4: Input and output flows of materials, energy and emissions.

2 Prospection - evaluating investigation methods

Contaminants in landfill soils – Reliability of prefeasibility studies

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Abstract

Recent LFM studies have researched the potential for resource recovery using samples from core drilling or grab cranes. However, most studies used small sample numbers, which may not represent the heterogeneous landfill composition. As a consequence, there exists a high risk of an incorrect economic and/or ecological evaluation. The main objective of this work is to investigate the possibilities and limitations of preliminary investigations concerning the crucial soil composition. The preliminary samples of landfill investigations were compared to the excavation samples from three completely excavated landfills in Germany. In addition, the research compared the reliability of prediction of the two investigation methods, core drilling and grab crane. Sampling using a grab crane led to better results, even for smaller investigations of 10 samples. Analyses of both methods showed sufficiently accurate results to make predictions (standard error 5%, level of confidence 95%) for most heavy metals, cyanide and polycyclic aromatic hydrocarbons (PAHs) in the dry substance and for sulphate, barium, benzo[a]pyrene (BaP), pH and the electrical conductivity (EC) in leachate analyses of soil type waste. While chrome and nickel showed less accurate results, the concentrations of hydrocarbons, TOC, DOC, PCB and fluoride (leachate) were not predicle even for sample numbers of up to 59. Overestimations of pollutant concentrations were more frequently apparent in drilling, and underestimations when using a grab crane. The dispersion of the element and elemental composition had no direct impact on the reliability of prediction. Thus, an individual consideration of the particular element or elemental composition for dry substance and leachate analyses is recommended to adapt the sample strategy

and calculate an optimum sample number.

2.1 Introduction

Landfill mining is considered a forward-looking concept for resource recovery and remediation. Increasing prices of raw materials inspired a discussion of landfill mining for resource recovery and strategies avoiding environmental pollution. Landfill mining is a part of technospheric mining and focuses on the reclamation of recyclable waste from active or inactive landfills (Johansson et al., 2013). The materials investigated include plastics, paper, wood, compost, rubble, metals and rare earth elements (Danthurebandara et al., 2015b; FWPRDC, 2005; Gutiérrez-Gutiérrez et al., 2015; Kurian et al., 2007; Passamani et al., 2016; Quaghebeur et al., 2013). In addition to continuous research on economic topics (Johansson et al., 2017b; Kieckhäfer et al., 2017), the focus of recent LFM studies has shifted to ecological (Frändegaard et al., 2013a; Gusca et al., 2015; Van Passel et al., 2013) and socio-economic issues (Damigos et al., 2016; Gaglias et al., 2016; Marella and Raga, 2014). The concept of ELFM valorises energy and materials using innovative transformation technologies, taking into consideration social and ecological criteria (Danthurebandara et al., 2015b; Jones et al., 2013). Due to the huge volumes of soil involved, the feasibility of landfill mining largely depends on its characteristics, pollutants and possible reuse options (Krook et al., 2012; Jain et al., 2013; Masi et al., 2014; Van Vossen, 2013; Zhou et al., 2015). The soils are of little financial value, or even involve costs for processing and disposal, but due to the large volume they are of major importance for economic and environmental assessment (Gusca et al., 2015). Landfill mining studies and evaluations are usually based on a very few samples from test pits carried out by drilling or grab cranes. The reliability and/or the statistical power of sampling depend primarily on sample number; consequently, there is some uncertainty about the significance of these study results. Seismic techniques, such as crosshole and multichannel analysis of surface waves, are useful methods, particularly for large landfills, to determine the composition, heterogeneity, the lateral and vertical extent, and moisture content in different depths (Abreu et al., 2016; Boudreault et al., 2016). Geophysical techniques may also be applied to identify leachate flumes, and to determine their shape and the contaminant migration route in order to appropriately locate wells for groundwater monitoring (Casado et al., 2015; Lopes et al., 2012). The sampling methods of recent pilot studies vary in sample type, number, quantity, stratification, sampling pattern, manner of creation of composite samples, equipment to obtain samples, etc. The sample numbers range from one composite sample (Masi et al., 2014; Rong et al., 2017) and six (Mönkäre et al., 2016) to ten point samples (Quaghebeur et al., 2013). The creation of large composite samples leads, in Masi et al. (2014); Rong et al. (2017), to more homogeneous results, in contrast to larger variations by sampling without composites in Zhou et al. (2015); Quaghebeur et al. (2013). Thus, large composite samples might not represent the heterogeneity of landfills nor allow for the evaluation of the composition of various layers, according to age, origin or other characteristics (Patil, 2013). The representativeness of random sampling depends on the sample number and spread, though a large sample number entails high costs. The reliability of preliminary investigations and assessments based on a small selection of samples is doubtful with regard to scattering elements and elemental compositions (denoted herein as "elements"). However, the common calculation formula for sample numbers could be larger than necessary, as a sample from a landfill is to some extent a cluster sample. In this respect, a smaller sample number might be sufficient, and therefore the risk of inaccurate assessment as well as the costs of laboratory analyses will be lower. Since the quality of landfill soils is crucial for off-site reuse and the large soil quantities may impact the economic feasibility of landfill mining, this investigation focuses on the contamination of soils. To determine the reliability of prefeasibility studies, the laboratory analyses of preliminary investigations were compared to excavation results. Therefore, objects of investigation in this work are:

- 1. the differences in reliability of the drilling and grab crane investigation methods
- 2. the possibilities and limitations of preliminary investigations concerning soil compositions
- 3. the identification of the behaviour of elements
- 4. the derivation of guide values for sample numbers

The results provide guiding values for estimating the sample number for particular elements and develop targeted sampling strategies. Thus, remediation specialists will have a better foundation to assess the resource recovery potential. In addition, sampling and analytical costs can be reduced whilst at the same time avoiding negative effects when making management decisions based on statistical samples. This paper is divided into two sections "material and method" and "results and discussion".

2.2 Materials and methods

This section consists of the site description, study design, laboratory analysis and statistical calculations.

2.2.1 Site description

Three landfills with MSW and CDW were completely excavated in the Federal State of Bavaria in Germany. These landfills had a surface area between 2,800 and $5,820 \text{ m}^2$ and waste quantity between 7,048 and 30,957 tons, and were used between the 1950s and 1970s (Table 2.1). Since 1972, the German waste law has required sanitary landfills. As the older landfills had neither bottom nor surface sealing, the protection of drinking water abstraction required complete excavation. Due to the EU waste management hierarchy of prevent, reuse, recycle, recover and dispose, all materials were separated and recycled to the extent possible (European commission, 2008).

2.2.2 Study design

The preliminary investigations involved test pits using grab cranes (2 m by 2m pit size) or, in the case of Traunstein, core drilling using a hydraulic hammer (drilled shaft of 80 mm). The cover thickness was 0.7 meters and the deposit depth 1.7 meters at the SR landfill, 0.7 meters and 6 meters at Traunstein, and 1.2 meters and 7 meters at Miltenberg, respectively. The density of pit per km^2 ranged from 3,000 at the SR landfill to 4,643 at Traunstein, compared to 700 pits per km^2 in Masi et al. (2014), 467 in Rong et al. (2017), 80 in Zhou et al. (2015) or 4.7 in Quaghebeur et al. (2013). The test pits of the grab cranes followed a regular grid, whereas drilling followed no clear pattern allowing more flexibility with regard to the identification of the horizontal and vertical landfill boundaries (Figure 2.1).During preliminary investigations, as far as possible one sample was taken of every layer or at minimum every meter, in line with the sampling standard DIN4023 (2006). Moreover, geomagnetic measures were employed to confirm the presence of barrels, seismic reflection to determine the lateral and vertical extent as well as the geological structure, and geoelectrical measures to identify leachate migration. The landfills were excavated with regard to the homogeneous composition of layers to the degree possible. The separation (off-site) of the waste into plastics and textiles, tyres, metals, glass, wood, asphalt, mineral residues (soil type waste) and hazardous materials was executed using mobile and stationary screens, as well as manual sorting. From every pile of mineral residues ten composite samples – each consisting of four samples - were taken, in line with the LAGA (2002). The piles were made up of soils with grain sizes similar to sand, gravel and cobble and were up to 600 cubic meters in size. Afterwards, the ten composite samples were mixed into one laboratory sample of 10 litres. From a total of 186 laboratory samples, the preliminary investigation included 97 samples of mineral residues and the excavation 89 samples.



Figure 2.1: Investigation test pits of Traunstein landfill (left) and Miltenberg landfill (right).

2.2.3 Laboratory analysis

Certified laboratories analysed the samples for heavy metals, and organic and physical parameters for the dry substance and leachate. The analysis using standardized determination methods – generally ISO standards – included in total nineteen parameters for the dry substance and twenty-two for leachate (Table 2.2). Samples of dry substance analyses passed through a 40 mm sieve and the overs were crushed before being added to the unders. The leachate analyses samples passed through a 10 mm sieve. Then a batch test in line with the standard EN 12457-4 was conducted.

2.2.4 Statistical calculations

The weighted geometric mean of each parameter was calculated separately for preliminary investigation and excavation samples, to permit the comparison of the methods and ensure the reliability of the investigation methods. Compared to the arithmetic mean, the geometric mean is less susceptible to outliers. The difference in the geometric means between preliminary investigation and excavation is presented in percent. The CV shows the dispersion and is also expressed in percent for better comparison of the parameters measured in different units (e.g. mg/kg, μ g /l). The preliminary visual check (histogram) and Shapiro-Wilk test revealed a positively skewed distribution of all parameters, with the exception of pH. Log10 and natural logarithmic transformations had no sufficient effect for most parameters. The non-parametric distribution required the Mann-Whitney U test (asymptotic significance p < 0.05, 2-tailed) which determines whether the preliminary investigation and excavation sample series come from the same landfill (H_0) , and consequently if the preliminary investigation is representative of the excavation pollutant concentrations. The MWW requires at least four samples of both groups and a minimum of 20 samples in total. Due to the requirements of a certain number and small differences in sample numbers, not all parameters could be tested. The analyses of 14 parameters from drilling were usually comprised of 59 preliminary investigation samples and 35 excavation samples. For 19 parameters the grab crane sampling included 20 ± 8 preliminary and 29 excavation samples. For the small SR landfill, the geometric means, differences in the geometric means and the CV were not considered. The MWW test was executed to check the reliability of the small sample numbers. The sampling inference precision is closely related to the sample number. The requested confidence level and accepted error determine the sample number and the resulting costs. The following formula was used to calculate the required sample number (n):

$$n = \frac{t^2 * CV^2}{SE^2}$$

where t is the value for a two sided confidence level; CV the coefficient of variation in percent; SE the standard error in percent. Additionally, the lower efficiency of nonparametric tests compared to the t-test requires adding a safety margin of up to 15%(Lehmann and D'Abrera, 2006). To avoid bias, the different landfill sizes and number of analyses were disregarded. The size of layers and piles were taken into account by weighting. The Kruskal-Wallis-test verified if the three landfills were independent. The asymptotic significance of all parameters was less than <0.000, except for benzo[a] pyrene (p 0.264). The significance values of the Kruskal-Wallis-test were verified for each chemical parameter using the Dunn-Bonferroni post-hoc-test by comparing pairs of groups (based on ranking). The adjusted significance (p) for all parameters was <0.000, but failed for arsenic (p 1.0). The correlation between the CV, differences in the geometric means and the asymptotic significance was crosschecked using – as the data showed a non-parametric distribution – Spearman's rho correlation coefficient (significance 2-tailed) and a scatter plot. For concentrations of chemical parameters below the limit of detection (LOD), the LOD divided by the square root of two was used in the statistical calculations. This replacement turned out to have the smallest relative difference compared to a replacement by zero, half of the LOD or the LOD itself (Croghan and Egeghy, 2003; Verbovšek, 2011).

2.3 Results and discussion

This section consists of the statistical analyses regarding the comparison of the geometric means of preliminary investigations and excavations and of the sampling methods drilling and grab crane. Afterwards, the reliability of the results was evaluated using the MWW test.

2.3.1 Differences between preliminary investigations and excavations

Both methods showed low differences of up to 25% between the means for the preliminary investigation and for the excavation samples for mercury, cadmium, thallium (<LOD), zinc (leachate), PAHs and benzo[a]pyrene, and large differences for hydrocarbons and PCB (Fig. 2.2). The large differences in PCB and hydrocarbons were caused by extreme outliers; the concentration of hydrocarbons might be decreased during excavation and screening due to volatility. The metals in leachate analyses below the LOD, such as arsenic, lead, cadmium and chrome remained the same in the preliminary and excavation analyses. Grab crane sampling showed the same pattern for the leachate analyses of nickel, molybdenum, mercury, selenium and thallium. Despite the larger sample number, marked differences were more fre-



Figure 2.2: Differences in the geometric means between preliminary investigation samples and excavation samples (in %): drilling (black), grab crane (white).

quently apparent in drilling. Drilling indicated moderate differences, in terms of

metals mainly overestimations ("+"), between 25 and 50% for lead, copper, nickel, zinc and CN, and larger differences for chrome (+52%) and arsenic (+81%). Chrome, lead and zinc revealed extreme outliers, although no large differences existed in their mean values. The overestimations may be caused by homogenization during excavation, transport, piling and taking samples from piles. Using a grab crane resulted mostly in small to moderate differences in terms of metals, particularly underestimations ("-"). Arsenic, barium (-10%, leachate) lead, cadmium, chrome, copper, nickel, mercury and zinc showed low differences (<25%), with the exception of nickel (-64%, leachate). For non-metals, using a grab crane led either to large overestimations or to small or moderate underestimations. Differences of up to 25% were detected for pH, electrical conductivity and CN and moderate to large differences for sulphate, fluorine (leachate), chlorine, DOC, PCB, total organic carbon TOC and hydrocarbons (Figures 2.2 and 2.3). The overestimation of dissolved organic carbon DOC and TOC concentrations might be related to the aeration during excavation and screening, although the carbon content is thought to derive from plastics. The underestimation of chlorine and sulphate concentrations might be connected with the dispersion of localised sources during excavation and screening as well as precipitation during excavation and transport. The concentrations of volatile halogenated hydrocarbons (VHH), EOX, benzene, toluene, ethylbenzene, and xylenes (BTEX), phenol and cyanide (leachate) remained on average below the LOD. In terms of non-



Figure 2.3: Differences in the geometric means (in %) of grab crane analyses.

metals, the differences were frequently large for both methods, although no tendency for over or underestimations existed. However, the drilling samples were only analysed for a few elements, on which basis no preference either for drilling or the grab crane could be recommended. The mean of heavy metal concentrations (Table 2.3) in soil type waste found in the dry substance were in line with Jain et al. (2005),
slightly higher than reported by Zhou et al. (2015), and slightly lower than reported by Quaghebeur et al. (2013). The values of copper, lead and arsenic were similar to those of Rong et al. (2017). The concentrations of metals in leachate analyses, such as arsenic, lead, cadmium, chrome, copper and zinc were typically negligible or below the LOD; similar results were recorded for antimony, molybdenum, mercury, nickel, selenium and thallium in grab crane analyses. The low solubility of heavy metals mirrored the results of the leaching experiments of Xiaoli et al. (2007), although her values were considerably higher. This difference might be related to the soluble portion of heavy metals leached after 40 years, and/or of low solubility, due to the anaerobic conditions and – particularly for high pH-values – the bonding of heavy metals to Fe-oxides and for some elements to organic matter (Øygard et al., 2008). Analyses of grab crane samples showed for polychlorinated biphenyl PCB a mean value of 0.04 mg/kg, benzo[a]pyrene: 0.1mg/kg, fluorine: 0.4 mg/l, PAHs: 1.0 mg/kg, TOC: 2.8%, DOC: 11.3 mg/l, hydrocarbons: 92.3 mg/kg and sulphate: 98.4 mg/l (Table 2.3). In this study the mean values did not differ substantially between the landfills at Miltenberg and Traunstein. For grab crane samples, the analyses of elemental compositions, such as naphthalene (drilling also), VHH, EOX, BTEX, phenols and cyanides (leachate), remained below the LOD. In particular, the mobility of VHH, BTEX and phenols might have led to their low concentrations in the mineral residues. In terms of metals, the CV was generally high with the exception of arsenic and nickel. In contrast lead, cadmium, copper, chrome, mercury and zinc had large CVs between 72% and 242% (Fig. 2.4). For copper a CV of 76% was recorded in leachate analyses of grab crane samples, and for barium a CV of 27%, respectively. Using a grab crane tended to have lower CVs which might be caused by the slight homogenization when taking samples. The dispersion of arsenic, chrome, copper and mercury in the dry substance compared well to the values found in Quaghebeur et al. (2013) and Zhou et al. (2015). In addition, the values for lead and zinc were similar to Zhou et al. (2015). The dispersion of copper, nickel and zinc coincided with the values reported by Masi et al. (2014). A high CV of between 96% and 324.5% was detected for PCB, hydrocarbons, benzo[a]pyrene and PAHs in the dry substance. For grab crane samples, a high CV was recorded for sulphate (127%), moderate CVs for DOC (49%), TOC (60%) and electrical conductivity (74%), and low CVs for pH (4%) and fluorine (14%). A high CV might indicate a more heterogeneous contamination. These localised sources, are more difficult to detect by drilling using a regular grid. The high degree of dispersion and difficulty in predicting PCB, might be caused, in addition to dispersion during excavation and processing, by the rounding effects of the rough measuring unit (mg/kg), accompanied by values below 0.1. A high degree of dispersion did not



Figure 2.4: Coefficient of variation of preliminary investigations (in %): drilling (black), grab crane (white).

coincide with a large difference in the means, except for PCB. In particular, for grab crane samples a high CV and at the same time low differences in the mean existed for most heavy metals. The CV of cadmium was 242% and the difference in the means 2.1%; for copper the values were 103% and 5.6%, respectively. No correlation was found between the CV and the differences in the mean (Spearman's ρ -0.37, p (uncorr.) 0.19), neither for drilling nor for grab crane sampling (ρ -0.19, p 0.45). The high degree of dispersion and difficulty in predicting PCB might be caused by the rounding effects of the rough measuring unit (mg/kg), accompanied by values below 0.1. Moreover, PCB seems to be found infrequently and dispersed during excavation. On average the concentrations remained below the German limit value RC1 (LAGA, 2003) and/or D1 (DepV, 2009) limit value (Table 2.3). Mineral residues of class RC1 can be reused at areas with good hydrogeological conditions without security measures (e.g. cover liner) for earthworks, such as road sub-bases, landfill top covers or noise barrier earth berms. However, several stockpiles exceeded the RC2 limit value of the technical guidelines for recycling soils (RC2) limit value and therefore had to be disposed of due to elevated maximum concentrations of copper (690 mg/kg), lead (3,000 mg/kg), zinc (2,800 mg/kg), sulphate (650 mg/l) and TOC (3.8 %). Less frequent excessive concentrations included PCB (5.54 mg/kg)and PAHs (46 mg/kg). Sources of elevated concentrations of heavy metals may be batteries, paints, timber preservatives and metal sheets, of sulphate CDW, of TOC grinded plastic particles, of PCB sealing compounds and burned PCB containing plastics and of PAHs tar paper, waste oils, ashes and asphalt. Despite the smaller sample number and frequent overestimations, the grab crane sampling produced the better results with regard to the similarity of the means from preliminary investigations and excavations. Although a high CV was found, the differences between preliminary investigation and excavation remained small to moderate. Drilling usually revealed higher CVs and larger differences in the means. These high CVs might be caused by the shape of the test pit, as circles have smaller perimeter per area than squares. Therefore, the sample is more homogeneous and the sampling variance higher. The risk of overestimation, in particular with drilling, can be reduced by preparing composite samples from transect sampling, and/or by cautious consideration of outliers. However, composite sampling impedes the determination of the properties of the various waste layers in terms of age, origin and composition. Zhou et al. (2015) recommend reducing the measurement uncertainty of PCB and PAHs sampling by drilling with high resolution spacing of 17 meters. Alternatively, geophysical methods in combination with geostatistical procedures for 3-D analysis enable optimising the location and the number of sampling stations (Boudreault et al., 2016). Small diameter drilling might be recommended to identify hot spots in a more cost-effective manner, although large objects such as asbestos cement roofing might be missed. Using a grab crane the variance can be reduced by rectangular test pits instead of quadratic ones, because rectangles have a greater perimeter to area ratio and are less susceptible to heterogeneity. Finally, in this study the correlation between the CV of the preliminary investigation and excavation was 0.70 (Spearman's ρ , p (uncorr.) 0.007) for drilling and ρ 0.65 (p 0.003) for the grab crane. This correlation suggests a similarity between the preliminary investigation and the excavation samples and therefore a reliable prediction regardless of the CV. Accordingly, the CV appears to be less important for calculating the sample number.

2.3.2 Limitations of preliminary investigations

High dispersion requires a large sample number to reduce uncertainty. Calculating the sample number on the basis of the CV (Table 2.3) resulted in several hundred or thousand samples and up to 16,848 for PAHs, except for fluorine which required 38 samples and pH two). Adding the 15% safety margin for non-parametric tests to the calculated sample number would result in 19,375 samples for PAHs. Other sample number calculation formulas, such as the one from Mason (U.S.EPA, 1992), are quite similar as are the calculated sample numbers. However, due to the fact that every sample is a composite, even smaller sample numbers might yield sufficient precision for an inference. Usually, the drilling sample number was 59 in the preliminary investigation and 34 for the excavation, and using a grab crane 20 ± 8 and 29, respectively. The MWW test verified that the concentration patterns of preliminary and excavation samples are almost the same; therefore, the preliminary investigation is representative. Both methods showed results which are sufficient for prediction with respect to cadmium, copper, lead, mercury, zinc, PAHs and benzo[a]pyrene in the dry substance (Fig. 2.5). In addition, drilling achieved the significance level of 5% for CN ($p \ 0.12$) and copper ($p \ 0.81$, leachate), and the grab crane for leachate analyses of sulphate, pH, barium, zinc and electrical conductivity (Fig. 2.6). For these parameters the results of the preliminary investigation compared well to the excavation; thus, the sample number was sufficient. Neither method achieved the



Figure 2.5: Significance (p) of the MWW test for drilling (black) and grab crane (white).

5% significance level for PCB and nickel. And drilling failed for arsenic, chrome, zinc and hydrocarbons ($p \ 0.004$) in the dry substance, while the grab crane failed for TOC, DOC and fluorine (Figs. 2.5 and 2.6). However, for DOC, TOC, fluorine and barium the smaller sample number (12) in the preliminary investigations using a grab crane should be taken into account, as the disproportion in sample numbers



(12:29) might have affected the results of the MWW test. Although PAHs theo-

Figure 2.6: Significance (p) of further leachate analyses (except TOC) of grab crane samples.

retically required the largest sample number (16,848 samples), 28 samples from the grab crane achieved a considerable significance level of 0.81 and 59 samples from drilling 0.63. The same pattern applied for cadmium, copper, lead, mercury, zinc, sulphate and conductivity. Hence, for these parameters such large sample numbers may not be necessary. No correlation resulted in a cross check between the CV and the significance of the MWW test (drilling: Spearman $\rho 0.43$, p (uncorr.) 0.12; grab crane: ρ 0.27, p 0.26). For example, cadmium, copper, lead and PAHs in the dry substance showed a high CV, as well as good significance levels. The correlation of the differences in the means and the significance of the MWW test was moderate for grab crane sampling (ρ -0.67, p 0.002), but negligible for drilling (ρ -0.15, p 0.6). On the whole, heavy metals, PAHs and parameters of leachate analyses usually revealed acceptable results with regard to making predictions. Compared to the grab crane drilling samples were less reliable as predictors of the excavation results. The similarity between grab crane and pile sampling might have led to the more significant results. The relation between the CV, the differences in the means and the significance of the MWW test suggests three patterns. The first is characterized by a high CV, small differences in the means and a high MWW test significance. Fig. 2.7 shows for example the similarity of PAHs values from the preliminary investigation and the excavation, despite the high CV. The first pattern also includes most heavy metals in the dry substance, for instance cadmium, copper, lead, mercury and zinc. These elements seem to be quite predictable, thus a moderate sample number might be sufficient and a safety margin unnecessary. Nevertheless, due to their high CV, composite sampling is not recommended to identify hot spots. The second pattern was characterized by a low CV, small differences in the means and a high significance. For example, the pH values were quite similar in the preliminary investigation and the excavation and did not disperse. Hence, no safety margin seems to be necessary and even small sample numbers or composite sampling might be sufficient for prediction. This pattern tends to include parameters of leachate analyses, such as pH, conductivity and barium. However, if these parameters show high dispersion, a safety margin is recommended. The third pattern reveals a moderate to high CV, a moderate to high difference in the means and a low significance level. Fig. 2.7 shows the large differences accompanied by a high CV for PCB values and nickel between the preliminary investigation and the excavation samples. The same applies to C10-C40, DOC and TOC; consequently, a safety margin is generally required and composite sampling not recommended.

2.3.3 Possibilities and limitations of a small sample number

To test the effectiveness of a smaller sample number, the MWW test was conducted using the data from the smaller SR landfill. Usually, at the SR landfill the sample number for preliminary investigation was 10 and for excavation 11, and at the Miltenberg landfill 20 ± 8 and 29, respectively. The analyses of eleven chemical parameters included ten samples from preliminary investigations and eleven from excavation. For nearly all parameters the results were comparable to the extensive grab crane investigation at Miltenberg, with regard to the differences in the means and the significance of the MWW test (Fig. 2.8). The landfills did not differ significantly for PAHs, cadmium, copper, lead, mercury, zinc (dry substance and leachate) and electrical conductivity. Nevertheless, neither achieved the 5% significance level for DOC; the SR landfill also failed the 5% level for pH, due to frequent higher values in excavation samples. In contrast, the SR landfill achieved the significance level for TOC; this would support the suspicion that disproportions in sample numbers (Miltenberg 12:29) affects the MWW test results. Despite the small sample number and high dispersion for some parameters, reliable results were achieved in particular for heavy metals in the dry substance. Thus, a small sample number using a grab crane was sufficient for some parameters, probably due to compositing. Larger sample numbers did not necessarily lead to better results. The minor importance of the CV and sample number, as well as the strong impact of the sampling method, should be generally valid. However, the unique character of each landfill affects the concentration patterns of elements.



Figure 2.7: Comparison of preliminary investigation (black) and excavation (red) values of PAHs, pH (leachate), PCB and nickel (grab crane).



Figure 2.8: Significance (p) of grab crane sampling with different sample numbers.

2.4 Conclusions

On the whole, many heavy metals and PAHs in the dry substance revealed good results to form the basis of predictions. In contrast, PCB, TOC, DOC and C10-C40 have to be considered more carefully. The grab crane samples were more reliable as predictors of the excavation results than the drilling samples. Consequently, small diameter drilling might be recommended to identify hot spots in a more cost-effective manner. In general, the sample number affected the reliability of prediction for the specific element differently. The CV was of minor importance for the prediction and hence, the statistical calculation formula leads to unnecessarily high sample numbers. Even a small sample number using a grab crane was sufficient for some parameters, probably due to compositing. Since the particular elements, the analysis method and the sample taking method seems to have greater impact on inference precision than the sample number, the sampling strategy can be adapted and the costs reduced. To optimise the reliability of prediction, further research should focus on the shape of the test pit. Better results for drilling samples might be achieved by creating composite samples from transect sampling and using a grab crane by rectangular test pits instead of quadratic ones. Due to the greater perimeter to area ratio the results might be less susceptible to heterogeneity. Further research should investigate the effectiveness of combining geotechnical and geophysical techniques to optimise selective excavation with regard to localised sources of contamination. Also elemental compositions that are difficult to predict, such as PCB, DOC, TOC and C10-C40, should be examined to gain insight into their behaviour.

Disposal	period	1950-1972	1964 - 1975	1960-1977
Excavation	samples	11	45	33
Investigation	samples	6	09	28
Pit density	$\mathrm{per} \ \mathrm{km}^2$	3,000	4,643	3,608
Number of	pits	9	13	21
Surface [m ²]		3,000	2,800	5,820
Excavated	material [t]	7,048	18,662	30,957
Landfill		SR landfill	Traunstein	Miltenberg

excavated landfills.
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period of th
nd disposal
i, surface, a
: Amount
Table 2.1

Table 2.2: Parameters of laboratory analyses, standard of determination methods, and units.

Parameter	Determination	Unit
	\mathbf{method}	
Solid		
As, Cd, Cr, Cu, Ni, Pb, Zn	ISO 11885	mg/kg
BTEX (benzene, toluene,	ISO 22155	mg/kg
ethylbenzene, and xylenes)		
Benzo[a]pyrene ($C_{20}H_{12}$)	ISO 18287	mg/kg
CN	ISO 11262	mg/kg
EOX (extractable organic halogens)	DIN 38414-17	mg/kg
Нg	ISO 16772	mg/kg
Hydrocarbons (C10-C40)	ISO 16703	mg/kg
VHH (volatile halogenated	ISO 22155	mg/kg
hydrocarbons)		
Naphthalene $(C_{10}H_8)$	ISO 18287	mg/kg
PAHs (EPA)	ISO 18287	mg/kg
PCB (polychlorinated biphenyl)	ISO 10382	mg/kg
Tl	ISO 17294-2	mg/kg
TOC (total organic carbon)	ISO 10694	% dry substance
Leachate (l.)		
As, Ba, Cd, Cr, Cu, Mo, Ni, Pb,	ISO 11885	$\mu { m g}/{ m l}$
Sb, Se, Tl, Zn		
Нg	ISO 17852	$\mu { m g}/{ m l}$
Cl	ISO 10304-1	mg/l
F	DIN 38 405-D4	mg/l
Cyanide (total, free)	ISO 14403	$\mu { m g}/{ m l}$
EC (electrical conductivity)	EN 27888	$\mu{ m S/cm}$
pH-value	DIN 38404-5	-
DOC (dissolved organic carbon)	EN 1484	mg/l
Phenols	ISO 9562	$\mu { m g/l}$
SO ₄	ISO 10304-1	mg/l

Element	Drilling mean	Grab crane	${\color{black} {\rm Limit} \ {\rm value}^b}$				
		mean					
As	13.4	9.1	50				
Ba $(l.^a)$	-	44.7	5				
Pb	125.0	194.5	300				
Cd	1.2	1.8	3				
Cr	39.1	39.3	200				
Cu	120.3	120.3	200				
Cu (l. ^a)	LOD	4.6	150				
Ni	34.4	24.7	200				
Hg	0.2	0.1	3				
Zn	408.2	650.3	500				
Zn (l. ^a)	7.7	13.2	300				
PAHs	3.1	1.0	15				
C10-C40	82.8	92.3	500				
$C_{20}H_{12}$	0.3	0.1	1				
TOC (%)	-	2.8	1				
PCB	0.02	0.04	0.5				
pH $(l.^a)$	-	7.9	6-12				
EC	-	455.8	1000				
SO_4 (l. ^{<i>a</i>})	-	98.4	100				
$F(l.^a)$	-	0.4	5				
CN total	0.3	LOD	30				
DOC $(l.^a)$	-	11.3	50				

Table 2.3: Geometric means of preliminary investigations and limit values.

 $^a leachate$ analysis, $^b limit$ value RC1 and D1 for Ba, DOC, TOC, F

3 Contaminant patterns in soils from landfill mining

Contaminant patterns in soils from landfill mining

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Abstract

LFM is used to close the material loops by recovering recyclables from landfills. Previous research has focused on material composition reporting that, worldwide, landfills mainly consist of soil-like materials ("soils") and combustibles. Although soils have been investigated in a few studies, the results are limited to the presentation of substance concentrations without further analysing the substance patterns (e.g. correlation between substances). This research identifies similarities in substance concentrations within and between landfills, analysing approximately 300 soil samples from eight excavated landfills. Statistical tests enabled the determination of substance variations and correlations. Substance concentration correlations were found between several heavy metals (in particular zinc), sulphate and electrical conductivity, as well as ammonium nitrogen and biodegradability. With regard to contamination prediction, sulphate, pH and TOC proved to be the most efficient indicator elements. Legal limit values have proven to be efficient to manage substance flows in terms of chloride, sulphate, cadmium, lead and zinc, but were ineffective with respect to biodegradability, PCB, BaP and CNs.

3.1 Introduction

The concept of LFM is designed to close the material loops towards a circular economy, recovering landfill waste. At the same time, the excavation of unsafe landfills enables the prevention of environmental hazards, such as groundwater contamination and the release of green house gases. Previous research has focused on

waste composition, often emphasising the potential for resource recovery (Krook et al., 2012). However, LFM generally involves dealing with large quantities of soillike materials (referred to as "soils" in this paper) of little or no market value, and reusing soils strongly depends on contaminant concentrations (Jani et al., 2016). Previous investigations of soils often compared their characteristics in different age waste layers and research was limited to individual landfills (Burlakovs et al., 2016; Kaartinen et al., 2013). Contamination prediction and substance patterns – such as relationships among substances – have not received much research (Brandstätter et al., 2014; Kaczala et al., 2017a). Although municipal landfills, worldwide, consist mainly of soils (Krook et al., 2012; Parrodi et al., 2018), soils have so far not been compared on a regional or international level with regard to similarities in substance concentrations. The present investigation identifies substance concentration patterns within and between landfills employing statistical methods based on soil samples from eight completely excavated landfills. Substance concentrations of classified soils were compared with legal limit values to evaluate the effectiveness of regulations to manage substance flows, since high concentrations of only one or few substances are usually decisive for classification. This study seeks to:

- analyse substance patterns within and between landfills
- identify indicator elements for contamination prediction
- evaluate legal limit values with regard to manage substance flows

3.2 Materials and methods

Eight landfills in Germany, used between the 1950s and 1980s to dispose of MSW and CDW, were completely excavated (see Appendix A for landfill overview). The landfills showed similarities in terms of waste composition, size and age; they were located up to 440 km away from each other. Measurements prior to excavation showed that the methanogenic phase of the landfills was completed and the biodegradable portion of the waste mostly depleted. The protection of drinking water abstraction and, in one case instability, required their complete excavation. Mechanical screens (vibrating grizzlies, star, trommel, flip-flop and vibrating screens), magnetic separators, crushers, air classifiers and conveyor belts for manual sorting were used for the separation of waste and soils. Processed waste from landfills consisted of soils, CDW, plastics, wood, tyres, metals and hazardous waste. The potential reuse of soils depended on the contaminant concentrations, physical characteristics of the soils and the regional demand for them. Regulations define requirements for the reuse, recovery and disposal of soils and CDW with regard to purpose and location, such as (a) landscaping, (b) road construction and earthworks, (c) landfills, (d) backfilling quarries and pits, (e) underground mines and (f) building construction. CDW has thus far not been used in building construction, due to high quality requirements, low quantities available and possession of mineral raw material deposits by the concrete industry. More than ten different regulations specify the reuse, recovery and disposal of soils; however in this study, the technical guidelines for recycling soils ("RC guidelines") and the German landfill ordinance ("LF ordinance") were mostly applied for the classification of soils (DepV, 2009; StMUV, 2011). Classification of soils by RC guidelines is mainly based on total concentrations of heavy metals and chemical compounds, while the LF ordinance requires more leaching tests, in particular of heavy metals (see appendix B for substances and limit values). The RC guidelines define mineral materials from class Z0 to Z1.2 (referred to as RC1 in this paper) as appropriate for reuse without cover (e.g. parking lots, noise barriers, backfilling of quarries and gravel pits), and soils of class Z2 ("RC2") for reuse with an impermeable cover (e.g. sub-bases of roads). At landfills soils can be reused as construction material, such as base liners, covers, roads or disposed of. The LF ordinance defines four surface landfill classes: from D0-limit value of the German landfill ordinance (DepV, 2009) (D0) for low contaminated to D3 for heavily contaminated waste. In total 301 samples were taken from test pits and from excavation piles in line with the German sampling standard (DIN4023, 2006; LAGA, 2002). The analyses were carried out at certified laboratories using standardized determination methods, generally ISO standards in accordance with the LF ordinance and RC guidelines (Table 3.1). Analyses of substances comprised heavy metals, organic and inorganic compounds, as well as physical parameters (e.g. pH, EC) including seventeen parameters in soils (total concentration) and nine in leaching tests. In total many more substances, such as heavy metals in leaching tests, rare-earth metals, organic compounds and phenols were analysed. However, the values usually proved to be very low or below the detection limit, and therefore were not taken into account in this study. In the laboratories soil samples for total concentrations analyses passed through a 40 mm sieve and the overs were crushed using a jaw crusher before being added to the unders (DIN19747, 2009). The leaching test samples passed through a 10 mm sieve before being batch tested in line with EN 12457-4 (2002). Substance patterns were researched with regard to: a) concentration variations within and between landfills, b) correlations between substances, and c) frequency of legal limit value exceedances (Fig. 3.1). Using the CV

Parameter	Determination	Unit				
	\mathbf{method}					
Total concentration						
As, Cd, Cr, Cu, Ni, Pb, Zn	ISO 11466 and	mg/kg				
	11885					
Benzene, toluene, ethylbenzene,	ISO 22155	mg/kg				
and xylenes (BTEX)						
Benzo[a]pyrene (BaP)	ISO 18287	mg/kg				
Cyanide (CN)	ISO 11262	mg/kg				
Hg	ISO 16772	mg/kg				
Hydrocarbons (C10-C40)	ISO 16703	mg/kg				
Polycyclic aromatic	ISO 18287	mg/kg				
hydrocarbons (PAHs)						
Polychlorinated biphenyl (PCB)	ISO 10382	mg/kg				
Total organic carbon (TOC)	ISO 10694	% dry substance				
pH-value	ISO 10390	-				
Biodegradability (Biodeg.)	AbfAblV/DIN	mg 0 ₂ /g dry				
	38414-8	substance				
Leaching tests (eluate analysis)						
Preparation of leaching batch	EN 12457-4					
test						
As, Ba, Cr, Ni, Zn	ISO 11885	$\mu { m g/l}$				
Cl^- , sulphate (SO ₄)	ISO 10304-1	mg/l				
F-	DIN 38 405-D4	mg/l				
Electrical conductivity (EC)	EN 27888	$\mu { m S/cm}$				
pH-value	DIN 38404-5	-				
Dissolved organic carbon (DOC)	EN 1484	mg/l				
Ammonium nitrogen (NH ₄ -N)	DIN 38406-5	mg/l				

Table 3.1: Parameters of laboratory analyses, determination methods and units.

enabled the comparison of substance dispersion even with different measuring units (e.g. mg/l, mg/kg, μ S/cm). The following formula was used to calculate the CV, which is expressed as a percentage:

$$CV = \frac{\sigma}{\mu} * 100$$

where σ is the standard deviation and μ the mean. A high CV indicates a heterogeneous dispersion of substances due to the infrequent disposal of particular objects, characteristics of objects (coarse-grained or bulky materials), previous mixing of waste, volatility of substances and methodological limitations of sampling, while common and daily disposed waste results in a low CV. The CV was calculated for the mean of each landfill to determine similarities within and between landfills, and of all landfills to identify general substance dispersions. Weighting took layer sizes from core drills of preliminary investigations and pile quantities into account. Each landfill was considered as an individual unit for statistical calculations. This one-toone weighting of landfills (landfill A : landfill B...: landfill H) prevented a bias due to differences in landfill size and number of analyses. Statistical calculations were carried out using PAleontological STatistics (Past3.0), GNU PSPP Statistical Analysis Software (release 0.8.1) and IBM SPSS Statistics (16.0). Brunner and Rechberger



Figure 3.1: Overview of statistical tests to identify substance patterns and of the approach to determine indicator elements.

(2017) suggest determining indicator elements (also referred to as "surrogate indicator") to generate maximum information with minimum effort. Indicator elements can be selected to represent a group of substances with specific properties by a small number of substances. However, this approach requires knowledge and experience to choose representative substances. A correlation analysis enabled the identification of correlating and non-correlating ("independent") substances simultaneously. A preliminary visual check (histogram) and Shapiro-Wilk test revealed a positively skewed distribution of all parameters (except pH), and logarithmic transformations had an insufficient effect. Due to the non-parametric distribution the Spearman rank correlation coefficient (ρ) was chosen, and $\rho > 0.7$ (bilateral correlation significance 0.01) defined as a strong relation ($\rho > 0.5$ moderate relation). Substance concentrations below the LOD were replaced using the division of the LOD by the square root of two (Croghan and Egeghy, 2003; Verbovšek, 2011). With regard to the frequency of legal limit value exceedances, substance concentrations were compared, as far as possible, with RC2 limit values of the RC guidelines, since up to this level soil recovery is frequently possible and inexpensive. For substances not listed in the RC guidelines, the DO limit value of the LF ordinance was used. The frequency (f) of limit value exceedances, which is expressed as a percentage, was calculated as follows:

$$f = \frac{n}{N} * 100$$

where n is number of measurements exceeding the limit value and N the total number of samples. Substances frequently exceeding legal limit values were analysed with regard to serve as an indicator element (i.e. representing other substances of interest for contamination prediction). A substance flow analysis was carried out to assess the effectiveness of legal limit values to manage substance flows in terms of recycling, recovery and disposal. The average concentration of each substance was calculated for – in line with legal limit values – classified soils.

3.3 Results and discussion

This section consists of waste composition analyses, substance concentrations in soils, substance variations within and between landfills, substance correlations, legal limit values exceedances and substance flows.

3.3.1 Waste composition analyses

The landfills consisted on average of 88% soils, 4.4% CDW (including 0.01% asphalt), 1.8% plastics, 0.3% scrap, 0.2% wood, 0.1% tyres and 5% topsoil of the cap (Table 3.2). Hazardous waste, such as batteries, asbestos and bitumen, comprised less than 0.1% of the total waste. The material composition of the landfills did not vary substantially for scrap, tyres, wood and hazardous waste, but did for plastics (0.1-5%) and CDW (0.5-17.8%). Apart from the original waste composition, waste type proportions depended on the employed equipment and processing efforts. Consequently, missing numbers for scrap, wood and types did not reflect an absence of those. The landfills investigated here showed a high proportion of soils similar to those of "set 3" in Laner et al. (2016), which reflects the composition of older landfills researched by Hogland et al. (2004) and Masi et al. (2014). Apart from structural requirements for soils and CDW, the technical guidelines define – depending on the contaminant concentration – mineral materials from class Z0 to Z2 (referred to as RC2 in this paper) as appropriate for reuse such as parking lots, noise barriers, sub-bases of roads, backfilling of quarries and gravel pits (LAGA, 2003). Soils and CDW of higher contamination can be reused as substitute construction material at landfills, but if they exceed certain limit values must be disposed of at appropriate landfills. With regard to reuse and disposal of inert waste at landfills, the German landfill ordinance defines four surface landfill classes: from D0 for low contaminated to D3 for heavily contaminated waste (DepV, 2009). The landfills investigated on average consisted of 30% RC material (21% RC1 and 9% RC2), 18% D0, 22% D1 and 18% D2-limit value of the German landfill ordinance (DepV, 2009) (D2). The proportion of classified soils differed substantially between the landfills, as a result of (a) applied regulations, (b) soil characteristics, (c) the employment of different processing equipment, and (d) different tendering procedures. Better processing results seemed to be achieved when the company carrying out the project became the owner of the excavated material and was not just a service provider (Hölzle, 2018).

Tyres	Wood	Scrap	Plastics	CDW	Topsoil	D2	D1	D0	RC2	RC1	Material	
0.1	0.2	0.3	1.8	4.4	5.0	18.0	21.5	18.4	8.8	21.5	Average	
0.1		0.3	4.0	9.1			30.6	1.8	27.1	27.0	Traunstein	Table
0.2	0.1	0.2	5.0	0.6	19.0	30.8	6.7		37.4		Straubing	e 3.2: Materi
0.1	0.4	0.3	1.8	0.5	9.3	70.6	10.8	4.3		2.0	Miltenberg A	al composition
0.1		0.1	0.1		4.4	39.5	41.9			14.1	Miltenberg B	of the landfills i
	0.2	0.4	0.1				27.6			71.7	Oberallgäu	and arithmetic
	0.3	0.5	2.6	6.9				89.7			Lindau	means.
			0.1		3.2		16.0	41.4	2.3	36.9	Main-Spessart	
	0.9	0.8	0.8	17.8	4.4	3.5	38.3	9.7	3.8	20.0	Ansbach	

le
3.2:
Material
composition
of the
landfills
and
$\operatorname{arithmetic}$
means.

3.3.2 Substance concentrations in soils

In terms of substance concentrations, zinc showed the highest average (median) of 350 mg/kg and a maximum value of 2,800 mg/kg, and PCB the lowest at 0.01 mg/kg and 0.5 mg/kg, respectively (Table 3.3). The median of the mixture of BTEX and arsenic (leaching test) remained below the limit of detection. With regard to leaching tests, the highest concentrations were recorded for sulphate (median 54 mg/l, max. 650 mg/l, and the lowest for fluoride (median 0.3 mg/l, max. 0.6 mg/l). The mean of most heavy metals (total concentrations) were in line with FDEP (2009), Hull et al. (2005) and Jain et al. (2005), but substantially lower than in the fines reported by Masi et al. (2014), Rong et al. (2017), Quaghebeur et al. (2013), Kaczala et al. (2017b) and Jani et al. (2016). The higher concentrations in these fines might be related to the small grain size (<10 mm) compared to the grain size of approx. 0-80 mm in the present study. Previous studies reported decreasing metal concentrations in fines smaller than 10 mm (Jain et al., 2005; Masi et al., 2014; Rong et al., 2017; Rousseaux et al., 1992) and also of coarse-grained soils and inert waste larger than 35 mm (Hölzle, 2018; Schachermayer et al., 1998). In contrast to the fines of the Metsäsairila landfill in Finland, cadmium, lead and mercury concentrations proved to be higher in the present study, while copper, chrome, TOC and DOC tended to be lower, and arsenic, nickel and zinc values were similar (Särkkä, Heikki et al., 2016). With regard to leaching tests, the pH-value, EC, sulphate, fluoride and chloride proved to be lower than found in Wanka et al. (2017) and Jani et al. (2016). The low values in leaching tests might be related to leaching processes in the landfill during the after-care phase starting in the 1970s.

3.3.3 Substance variations within and between landfills

Zinc and copper varied strongly (>90%), while pH values (total concentration and leaching test) almost did not vary at all (Fig. 3.2). High variations (>75%) were also found for ammonium nitrogen, PAHs, PCB, chrome, lead and CN. However, a small number (72) of ammonium nitrogen analyses may have probably resulted in a high CV, while rounding effects of the rough measuring unit and values below 0.1 might have led to an increased variation of PCB. Heavy metals tended to vary more (except for nickel), while the variation of substances of leaching tests generally were lower. The low variations in leaching tests might be related to frequent low substance concentrations. The heavy metal dispersions proved to be similar to those found by Brandstätter et al. (2014), Masi et al. (2014) and Zhou et al. (2015). In addition, ammonium nitrogen, sulphate, EC and pH (leaching test) dispersions were in line with Brandstätter et al. (2014). A high CV indicates the disposal of objects

Parameter	Unit	Median	P75	Max	Limit
As	mg/kg	8	10	22	150
As	mg/l	LOD	LOD	12	60
Ba	mg/kg	52	69	140	2000^{a}
Pb	mg/kg	130	210	890	1000
Cd	mg/kg	0.9	1.9	5	10
Cr	mg/kg	29	45	87	600
Cr	mg/l	LOD	LOD	90	150
Cu	mg/kg	62	130	828	600
Ni	mg/kg	29	34	110	600
Ni	mg/l	LOD	LOD	390	200
Hg	mg/kg	0.2	0.3	3	10
Zn	mg/kg	350	590	2800	1500
Zn	mg/l	21	43	490	600
PAHs	mg/kg	2.5	6	46	20
C10-C40	mg/kg	76	210	970	1000
BaP	mg/kg	0.2	0.5	3	-
BTEX	mg/kg	LOD	LOD	9	5
TOC	% dry substance	1.7	2.5	11	1^a
DOC	mg/l	4.5	6	38	50^a
PCB	mg/kg	0.01	0.05	0.5	1
CN total	mg/kg	0.1	0.7	4.7	100
Biodeg.	$mg 0_2/g$	0.2	0.4	1.2	5^a
pН	total	7.9	8.2	12.3	-
	concentration				
pН	leaching test	8.1	8.2	11.9	5.5 to 12
EC	$\mu S/cm$	271	415	1510	1500
Cl-	mg/l	1.4	2.5	60	30
SO_4	mg/l	54	130	650	150
F ⁻	mg/l	0.3	0.4	0.6	1^a
NH ₄ -N	mg/l	0.4	0.8	4.6	1^a

Table 3.3: Total averages (median), 75 percentile, maximum, limit values (RC2/D0) and number of analyses.

 $^{a}D0$ limit value otherwise RC2

which were not part of daily MSW and CDW streams. In addition, heterogeneous waste composition and hotspots might be a result of the disposal of coarse-grained or bulky materials, previous mixing of waste, volatility of substances and methodological limitations of sampling. Tar paper, waste oils and ashes (from households and landfill fires) might have led to increased PAHs values, and other sources of mercury might include electrical components and wood preservatives; sulphate from dry wall; PCB from sealing compounds and electronic devices (e.g. condensators, fluorescent light ballasts; hydrocarbons from workshop waste and machinery; zinc from metal sheets and colours; copper from electronics, metal sheets and wood preservatives; cadmium from colours and PVC; chrome from wood preservatives and colours; lead from batteries, anti-corrosive paint, roofing sheets and water pipes; and CN from galvanisation residues). Comparing substance concentration variations within and between landfills enabled the following substance classification: Class A) substances with variations between landfills of more than 50% and at the same time stronger variations within landfills than between landfills, class B) substances with variations between landfills of more than 50% and a CV between landfills resembling the average of the CVs within landfills, and class C) substances with similar variations between (<50 %) and low variations within landfills. Class A substances usually showed a high CV (up to 430%) within landfills, but average substance concentrations proved to vary less between landfills ($\sim 75\%$; see Fig. 3.3). The range of CVs within landfills was usually more than 100% (except for lead). Class A comprised PAHs, mercury, sulphate, PCB, hydrocarbons, cadmium, lead and ammonium nitrogen. Variations of lead and ammonium nitrogen remained low within landfills, while the concentrations of mercury, sulphate, hydrocarbons and cadmium tended to vary less between landfills. Substances of class B were characterized by strong variations between landfills, whereas the range of CVs within landfills proved to be low to moderate. The variation between landfills was approximately equal to the mean of the individual landfill variations. Class B consisted of copper, zinc, CN and to some extent (due to low CVs within landfills) of chrome and biodegradability. Class C included substances with similar variations between landfills (<50%) and low concentration variations within landfills, i.e. similar concentrations in all samples. Class C included BaP, chloride, TOC, EC, barium, DOC, nickel, arsenic, fluoride and pH (total concentration and leaching test). The CVs within landfills of BaP and EC proved to be higher than the CV between landfills, which - in contrast to class A substances – remained below 50%. Measurements of pH (total concentration and leaching test) revealed remarkable similarities within and between landfills. No



Figure 3.2: Substance dispersions expressed as the coefficients of variation (CV in %).

regularities were observed between the landfills with regard to dispersion, i.e. no landfill frequently showed low or high CVs. Cadmium, chrome, copper, mercury and nickel concentration variations between landfills showed similarities to Parrodi et al. (2018) review of fines from international landfills.

3.3.4 Substance correlations

Heavy metals correlated frequently with other substances. For instance, zinc showed moderate to strong correlations ($\rho > 0.5$) with nine from 23 substances, while copper and mercury correlated with eight substances, ammonia nitrogen with seven, lead, chrome and arsenic with six, and TOC with five, respectively (Table 3.4). The Spearman rank correlation test revealed a strong relation between zinc and cadmium $(\rho \ 0.84)$, copper $(\rho \ 0.84)$ and lead $(\rho \ 0.78)$, as well as between copper and cadmium $(\rho \ 0.77)$ and lead $(\rho \ 0.73)$. High correlations were also recorded for chrome and nickel ($\rho 0.85$) as well as for EC and sulphate ($\rho 0.87$). Biodegradability correlated with ammonium nitrogen (ρ 0.70), barium (ρ 0.69) and BaP (ρ 0.68). A strong correlation between BaP and PAHs (ρ 0.86) indicated BaP as a frequent member of PAHs. For heavy metals, moderate correlations were found between arsenic and cadmium, copper, lead, mercury and zinc ($\rho \sim 0.6$), but not for chrome and nickel. However, chrome and nickel tended to correlate with copper, mercury and zinc $(\rho \sim 0.6)$, though not with lead and cadmium. TOC correlated with several heavy metals, such as copper (ρ 0.63), lead (ρ 0.56), mercury (ρ 0.66) and zinc (ρ 0.69), but not with chrome, cadmium and nickel. Ammonium nitrogen tended to correlate with the pH value (ρ -0.65; leaching test), hydrocarbons (ρ 0.61), DOC (ρ 0.61), sulphate (ρ 0.58), EC (ρ 0.57) and BaP (ρ 0.57). The eluate analyses of Kaczala et al. (2017a) revealed – with regard to substances of this study – strong correlations between lead and zinc ($\rho 0.71$, present study $\rho 0.78$), TOC and zinc ($\rho 0.81$, present study ρ 0.69), as well as between TOC and DOC (ρ 0.65, present study ρ 0.02). In line with Brandstätter et al's (2014) regression modelling, EC and pH (leaching test) showed relationships with ammonium nitrogen and sulphate but not with chloride. Measurements of pH-value (leaching test) tended to be uncorrelated, since in 16 of 23 cases ρ was 0.15 or less. The same applied to hydrocarbons (15 cases), PAHs and chloride (14), DOC and biodegradability (13) and BaP (11). Consequently these substances might be considered as "independent" substances/parameters not representing other substances. Substances and parameters of leaching tests tended to correlate less, with the exception of ammonium nitrogen. In addition, heavy metals proved to be remarkably uncorrelated ($\rho < 0.15$) with PAHs, hydrocarbons and pH (leaching test). Plastics and wood were most probably origin of high TOC

Chapter 3. Contaminant patterns in soils



Figure 3.3: Substances classified by variations within and between landfills.

and DOC concentrations, since no correlations ($\rho \leq 0.18$) were found with PAHs and hydrocarbons.

3.3.5 Legal limit values exceedances

TOC measurements exceeded in 75.2% of cases the D0 limit value, whereas zinc (39.5%) and sulphate (34.3%) showed frequently RC2 limit value exceedances (Fig. 3.4). Moreover, ammonium nitrogen (19.5%), copper (14.6%) and lead (10.3%) concentrations proved to be frequently greater than the RC2 or D0 limit value, while the exceedances of pH (total concentration), BaP, PAHs, hydrocarbons and cadmium remained below 10%, and of BTEX, EC, PCB, mercury, fluoride and chloride below 5%. Arsenic, barium, chrome, nickel, DOC, CN total, biodegradation and pH (leaching test) always remained below the limit. Frequent limit value exceedances of zinc, copper, lead and cadmium were in line with Adelopo et al. (2018) survey of heavy metal pollution in landfill precursors. Heavy metals tended more frequently to exceed the limit values, while substances of leaching tests usually remained below the limit values (except for sulphate and ammonium nitrogen).

3.3.6 Substance correlations

In many cases two, sometimes up to four, substances exceeded the limit values. Fig.3.5 shows that sulphate and TOC exceedances co-occurred in 50.4% of all sulphate or TOC exceedances. Sulphate concentrations exceeded solely (i.e. no cooccurrence with TOC exceedances) the limit value in 2.5% of cases, whereas 47.1%TOC exceedances did not involve sulphate exceedances. These 50.4% co-occurrences represented 39% of total sample numbers, since TOC or sulphate did not exceed in every sample the limit value (see Fig. 3.5 percentage of total sample number in brackets). In terms of TOC, the differences were small between the co-occurrence percentage of exceedances and the co-occurrence percentage of the total sample number, due to numerous TOC exceedances. Less frequent limit value exceedances of zinc and EC resulted in a greater difference. Frequent co-occurrences were also recorded for TOC with zinc (42%), ammonium nitrogen (32%) and copper (17%). Zinc co-occurred with copper (41%), lead (29%) and cadmium (24%), while pH (total concentration) coincided with EC (22%). However, the co-occurrence percentage of total sample number remained low for zinc with copper (18%), lead (13%) and cadmium (10%), as well as for pH with EC (2%). Infrequent co-occurrences were observed for hydrocarbons with TOC (9%) and zinc (6%). Co-occurrences of zinc with other heavy metals coincided with its high correlations (see subsection 3.3.1). In spite of the frequent combination of TOC and sulphate limit value exceedances, a correlation between these substances could be observed. With regard to limit

pH*	40	14	.12	12	01	.03	.02	26	.13	.15	18	35	.08	04	20	14	65	.02	47	15	07	14	08	
pН	.04	.16	19	52	13	32	27	44	31	32	03	.16	40	18	.08	16	.35	.25	10	.51	05	29		
Ba*	.23	60.	-20	.08	.40	.34	.25	.04	.22	.29	.04	9.	.25	.47	.13	69.	.41	00.	.18	.25	.27			
Cl-*	.54	.08	.19	.04	.23	.08	.18	.16	.02	.01	- 60.	00.	.03	.07	.33	.03	.35	.05	.34	.06				
F-*	.02	-20	.10	.51	.26	.47	.39	.41	.48	.54	.13	00.	-64	.02	.13	.03	.29	.15 -	.10					
SO_4*	.87	.18	.18	.11	- 30	.21 -	.28 -	.18	.01	.18	.41	.32	00.	.32	11.	.06	<u>8</u>	.02						
PCB	.14	30	.33	.21	.35	.35	.33	.27	34	39	.25	.24	.31	.40	.20	- 80.	.21							
NH4-N*	.57	.33	37	.17	14	17	16	.18	15	22	.61	.57	20	21	.61	.70								
Biodeg.	19	.25	00.	13	.10	.05	.22	00.	.11	.06	.26	68.	.23	.32	.08									
DOC*	.28	.03	-04	.41	.06	.22	.07	.39	.25	.10	60.	.28	.12	.02										test
TOC	.04	.18	.47	.29	44.	.63	.56	.32	99.	69.	.06	.25	53											ching
CN	.02	60.	.28	.38	.33	44.	39	.33	55.	.49	.15	.13												*leac
BaP	.32	.86	.14	06	.15	.06	.17	.02	.07	.01	.42													
C10-40	.39	.34	14	12	.04	.01	.15	-,11	13	04														0.15
Zn	.13	.06	.62	.58	.84	.84	.78	.57	.61															d U
Hg	.06	.11.	.53	.55	.49	.64	.50	.55																0.14
Ni	.26	08	.45	.85	.46	.61	.40																	2
Pb	.22	.05	.57	.40	.73	.73																		×0.
Cu	.21	.02	.64	.59	.77																			7: p
Cd	.25	.06	.64	.41																				0.5
Cr	.15	15	.34																					0.7
As	.25	.15																						م ۱۸
PAHs	.15																							
	EC*	PAHs	As	Cr	Cd	Cu	Pb	Ni	Hg	Zn	C10-40	BaP	CN	TOC	DOC*	Biodeg.	NH4-N*	PCB	SO_4^*	*	C]-*	Ba*	Ho	

Table 3.4: Spearman rank correlation test (ρ , correlation significance <0.01, bilateral) of substances and parameters.



Figure 3.4: Frequency (in %) of RC2 limit value exceedances (D0 limit values were substituted for non-existent RC2 values).





Figure 3.5: Co-occurrences of substances with TOC, zinc and EC exceeding the legal limit values (*leaching test).

value exceedances, zinc indicated cadmium, copper and lead, and to lesser extent sulphate and TOC (Fig. 3.6). The indication rate of zinc was less than 40 % for PAHs, hydrocarbons and ammonium nitrogen. In contrast, sulphate limit value exceedances represented all hydrocarbon exceedances and most of ammonium nitrogen, cadmium and lead exceedances. Using sulphate as indicator element resulted in moderate $(\sim 55\%)$ indication of copper, zinc and TOC, while PAHs were satisfactorily (38%) indicated. However, the number of cadmium, PAHs, ammonium nitrogen and hydrocarbon exceedances ranged from 14 to 16, which might result in a more uncertain indication rate. TOC showed good results (>90%) for the indication of ammonium nitrogen, PAHs, lead, cadmium, copper, zinc and sulphate, whereas hydrocarbon exceedances were completely represented. The indication rate of PAHs, cadmium and hydrocarbons might differ due to a number of exceedances ranging form 12 to 14. It should be noted that, TOC exceeded limit values nearly twice as often as zinc or sulphate. Infrequent exceedances of mercury, BaP, PCB, BTEX, pH (total concentration), EC, fluoride and chloride made their indication difficult. due to a number of exceedances below ten. TOC tended to cover mercury, BaP, PCB and fluoride best, while sulphate indicated EC exceedances better. With regard to BTEX, pH (total concentration) and chloride, the same indication rate was observed for zinc, sulphate and TOC. However, pH (total concentration) exceedances (six samples) were not at all covered by these indicator elements. A combination of sulphate and TOC turned out to best indicate the analysed substances, resulting in an indication rate of more than 90% for PAHs, copper and lead, as well as of 100%for cadmium, zinc, hydrocarbons and ammonium nitrogen. In terms of PAHs, cadmium, hydrocarbons and ammonium nitrogen, number of exceedances ranging from 12 to 16 might significantly increase the uncertainty of prediction. Exceedances of pH (total concentration) remained undetected, and consequently, the selection of the pH value as indicator element would be a useful asset. Moreover, adding pH (total concentration) to the indicator element set would increase the indication rate of EC exceedances to 100%. Using zinc as indicator element would not improve prediction, since sulphate and TOC covered all with zinc related exceedances. Brandstätter et al. (2014) multivariate regression modelling showed sufficient prediction of twelve substances using loss on ignition (which is frequently used instead of TOC), EC, pH (leaching test) and chloride as predictor variables. EC and chloride rarely exceeded legal limit values in the present study – probably due to the age of the landfills – but might be a suitable indicator element. In line with Brandstätter et al. (2014) lack of accuracy for sulphate and zinc prediction, analyses of these substances might



be an asset for contamination prediction. Finally, zinc, chrome, sulphate, TOC and

Figure 3.6: Indication rate (in %) of limit values exceedances using zinc, sulphate and TOC as indicator elements.

ammonium nitrogen might be suitable indicator elements due to high correlations, co-occurrences and frequent legal limit value exceedances, whereas pH (total concentration), PAHs and hydrocarbons might be chosen as indicator elements due to missing relations to other substances ("independent indicator element"). A combination of sulphate, TOC and pH proved to indicate efficiently limit value exceedances of 14 substances. For less degraded waste, EC and chloride analyses might increase prediction quality in accordance to the observations of Brandstätter et al. (2014).

3.3.7 Substance flows

Since exceedances of one to three substances were decisive for soil classification, flows of other substances and their accumulation tendencies might remained unaffected. Substance flow analysis allowed to verify if substance concentrations were in general higher in contaminated soils and, consequently, limit values efficiently managed substance flows. Average substance concentrations were calculated for each soil class and expressed as a percentage. Concentrations of biodegradability, PCB, BaP, CNs, copper, PAHs, fluoride and hydrocarbons proved to be on average higher in RC1 and RC2 soils, while chloride, cadmium, zinc, sulphate and lead tended to accumulate in D1 and D2 soils (see Fig. 3.7). The latter also applied to some extent for DOC, arsenic and EC. Sulphate, cadmium and chloride remarkably accumulated in D2 soils, whereas CNs and hydrocarbons showed accumulations in D1 soils and mercury in D0 soils, respectively. TOC showed similar concentrations in all soil classes, probably due to insignificant differences between the limit values of different soil classes. Soils could be often classified as either RC2 or D0 due to similar limit values; however, analysis requirements for substances and parameters are to some extent different for these limit values. The RC guidelines limit values are mainly based on total concentration analyses, while the LF ordinance requires mostly eluate analyses. For instance, the RC guidelines do not include a biodegradability limit value, probably resulting in higher measurements in RC soils. All in all, the system of limit values guided only to some extent the substance flows, since the concentrations of individual – and sometimes infrequent – substances were decisive for classification. Consequently, high concentrations of a single parameter frequently led to higher classifications although concentrations of all other contaminants remained low. Limit values proved to efficiently manage substances showing frequent exceedances, such as cadmium, zinc, sulphate and lead, but not TOC. Limit values also did not affect flows of biodegradability, PCB, BaP and CNs, as well as of copper, PAHs and fluoride to some extent.

3.4 Conclusions

Previous research reported that, worldwide, municipal landfills consist mainly of soils (Krook et al., 2012; Parrodi et al., 2018). In the present study, substance patterns in soils from LFM were investigated with regard to dispersion within and between landfills, correlations between substances, frequency of limit value exceedances and substance flows. One objective was the identification of indicator elements for contamination prediction. Zinc showed the strongest correlations and frequent limit value exceedances; however, indication of other substance limit value exceedances



was moderate. Sulphate and TOC proved to be suitable as indicator elements, due to correlations, co-occurrences and frequent legal limit value exceedances. However, pH exceedances were not represented probably due to a lack of correlation. Consequently, the addition of pH as an "independent" indicator element is recommended, as well as EC and chloride might improve prediction quality in line with the observations of Brandstätter et al. (2014). Legal limit values proved to be efficient to manage flows of substances which frequently exceeded limit values, such as cadmium, zinc, sulphate and lead (except for TOC), as well as chloride and to some extent DOC. However, limit values were ineffective in terms of biodegradability, PCB, BaP and CNs. Since the landfills of the present study showed certain similarities (waste composition, size, age), further research emphasis should thus focus on factors resulting in similarities, such as landfill specific properties (type and combinations of disposed of materials, waste age, micro-climatic conditions, etc.) and regional settings (geogenic background values, industrial production, recycling systems, consumption patterns, economic development, climate). In addition, the suitability and reliability of sulphate and/or zinc as indicator elements requires further research.
4 Processing – assessing the effectiveness of dry screening

Dry screening – assessing the effectiveness of contaminant reduction in recovered landfill soils

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Abstract

Landfill mining LFM – an alternative to landfill closure – is designed to recover resources. As landfills consist mainly of soils, the soil quality and possible reuse options are crucial for the economical assessment. The objective of this study is to redistribute contaminants – primarily chemical – in excavated landfill soils for off-site reuse using exclusively mechanical processing equipment. The waste from two full-scale landfill remediation projects were processed in four different processing plants. The efficiency of contaminant redistribution in soils at each processing plant was investigated using statistical methods. At all four processing plants considerable lower concentrations of heavy metals, PAHs, TOC and sulphate were observed in the coarse-grained soils appropriate for off-site use. In contrast, the differences in leachate analyses (pH, EC, chloride, fluoride, barium, DOC) proved to be heterogeneous, less pronounced between fines and coarse-grained soils. Fluoride and chloride sometimes even showed higher concentrations in the coarse-grained. Screens with a mesh size of 50 mm performed more efficiently in terms of contaminant reduction and proportion of material flows (unders and overs) compared to openings of 35 mm or of 70 mm and larger. However, the results indicated an optimum between 35 mm and 50 mm. To ensure the reuse of soils, the determination of contaminants and the grain size distribution should be established in preliminary investigations which are an indispensable requirement for selecting optimal processing equipment and appropriate screen openings.

4.1 Introduction

LFM is considered as an option to remediate landfills and eliminate the environmental risks of groundwater contamination and emissions, as capping is not a permanent solution. At the same time, the recovery of material enables the closure of the circular economy. The feasibility of LFM depends primarily on the composition of landfills. Thus, in the last decades the characterization of deposited material has been extensively researched (Krook et al., 2012). Studies have focused either on the general composition of waste (Hogland et al., 2004; Kaartinen et al., 2013; Quaghebeur et al., 2013; Wolfsberger et al., 2015) or on the recovery of a specific material, i.e. metals for recycling (Gutiérrez-Gutiérrez et al., 2015; Wagner and Raymond, 2015), plastics for generating energy (NYSERDA, 1998b,a; Passamani et al., 2016) and soils to use as compost (Kurian et al., 2007; Masi et al., 2014; Mönkäre et al., 2016; Rong et al., 2017; Zhou et al., 2015) or as landfill cover (Jain et al., 2005; NYSERDA, 1998a,b). Since most landfills worldwide consist primarily of soil, the soil characteristics and pollutants are crucial for reuse and the economical feasibility of LFM (Jani et al., 2016; Krook et al., 2012). In previous studies chemical analyses of soils typically targeted heavy metals (Jain et al., 2005; Kaartinen et al., 2013; Wolfsberger et al., 2015), whereas in studies focusing on compost recovery, biological and chemical analyses – i.e. of nitrogen, phosphorus, potassium and the biochemical methane potential (Das et al., 2002; Masi et al., 2014; Mönkäre et al., 2016; Zhou et al., 2015) – and germination tests (Prechthai et al., 2008; Rong et al., 2017) have been carried out. However, the soils recovered have only (rarely) been used as on-site daily or landfill cover in a few full-scale projects (Jain et al., 2013; U.S.EPA, 1993). Some research has also focused on processing technology, such as the performance of different types of equipment in terms of the processing rate, stream purity, product quality, size distribution of materials, costs and the combination of different screens (U.S.EPA, 1993; Stessel and Murphy, 1999; Maul and Pretz, 2015). In contrast to the regular employment of screens in LFM case studies, soil processing technologies have tended to focus on soil washing rather than dry screening (Dermont et al., 2008; Voglar and Lestan, 2013). The use of soil washing technology leads to an accumulation of contaminants in the liquid, while using dry sieving equipment may not affect adhering impurities (Wanka et al., 2017). However, soil washing is more complicated and often involves the use of acids and/or chelating agents. The concentrations of heavy metals of different soil sizes were rarely analysed without being a specific research objective (Hogland et al., 2004; Jain et al., 2005; Masi et al., 2014; Rong et al., 2017). With the exception of using mechanical equipment in the study of Hogland et al. (2004), all studies employed laboratory scale manual sieving. Thus, the results might not be representative of full-scale projects, in particular those employing small mesh sizes, i.e. 0.425/6.3 mm (Jain et al., 2005), 4/10 mm (Masi et al., 2014) and 5/10 mm (Rong et al., 2017). Higher contaminant concentrations in fines were frequently observed, since a larger surface facilitates the adhesion of substances. However, the contamination differences for individual substances was not quantified in previous studies and the results not verified using statistical methods. The objective of this study is to produce a soil of higher quality meeting the legal requirements for off-site applications (i.e. noise barrier earth berms, sub-bases of roads, top cover of landfills or backfilling of quarries and gravel pits). The second goal is to maximize mass flows of recoverable soils. Only mechanical processing equipment and the existing processing plants are used in two full-scale projects subject to economic constraints. Therefore, this research seeks to analyse:

- the processing effectiveness of contaminant distribution in soils of different grain sizes
- the quantification of concentration differences of substances using statistical methods and
- the efficiency of different processing trains and screen mesh opening sizes

4.2 Materials and methods

This section consists of the waste processing plants (WPPs) description, laboratory analyses and statistical methods. Between the 1950s and 1970s the landfills at Traunstein (TS) and Miltenberg (MIL) in southern Germany were used to dispose of municipal solid waste (MSW), excavated soil and CDW. The TS landfill had a surface area of 2,800 m2 and a waste quantity of 18,662 tonnes, and the MIL landfill 5,820 m² and 30,957 tonnes, respectively. In the last five years these unlined landfills were completely excavated, to protect the nearby drinking water extraction. The German waste law (KrWG, 2012) requires the treatment of the excavated waste in line with the EU waste management hierarchy of prevent, reuse, recycle, recover and dispose (European commission, 2008). 2.1 Waste processing plants (WPP) At the MIL1 a grizzly screen (70 mm) was followed by a conveyor belt for the manual separation of the overs, such as plastics, textiles, scrap and wood (Fig. 4.11). The unders (0-70 mm) fell into an under-screen box (50 mm) and the medium sized material

(50-70 mm) passed under a cross-belt magnet to remove ferrous metals, while the soil type material (<50 mm) was directly transported and used off-site as landfill cover. The remaining waste stream (50-70 mm) from the under-screen box, consisting mainly of plastic films and clumpy soils, was split into two different processing trains for further treatment. Main objective was to separate the soil and adhering fines from plastic films, while processing at train two also targeted the production of a low contaminated coarse-grained soil. Using different types of 20 mm screens led to a further break up of the waste matrix (50-70 mm), and adhesive fines (<20 mm) dropped. Processing train one used a vibrating screen (20 mm), where the overs were shredded before passing through a star screen (20 mm). Bricks and plastics were removed manually from the overs, and the remaining soil was subsequently mixed up with the soils (<20 mm) from the star and vibrating screens. The vibrating screen broke up the waste matrix insufficiently and thus the quantities of fines (<20 mm) remained negligible. At processing train two, plastics were picked up from the remaining fraction (50-70 mm) on a conveyor belt followed by a trommel screen (20 mm). From the trommel overs plastics were once again collected. In contrast to processing line one, using a trommel screen proved to be more efficient in segregating fines from the mixture of soil and plastics resulting in a proportion of 50% fines (<20 mm). With regard to statistical calculations, these fines were considered to be fine-grained soils. The MIL 2 consisted of a vibrating grizzly screen (70 mm) and a cross-belt magnet located at the overs output stream (Fig. 4.2). Two workers collected plastics, wood, textiles and scrap from the unders and overs. Due to the high amount of waste excavated in a short period the processing at several WPPs was necessary. For the TS landfill 14,363 tonnes were transported to the WPP Traunstein 1 (TS1) and then, due to excessive workload, 4,299 tonnes to the WPP Traunstein 2 (TS2); for the MIL landfill 10,470 tonnes were transported to the WPP Miltenberg 1 (MIL1) and 20,487 tonnes to the WPP Miltenberg 2 (MIL2). The WPPs usually had mobile screens, except MIL1 which had some immobile equipment. The TS1 consisted of a vibrating grizzly screen with an under-screen box (35 mm) and a convevor belt for manual sorting of the overs (>80 mm), such as glass, debris, wood, plastics and textiles, tyres, debris and scrap (Fig. 4.3). The medium fraction (35-80 mm) was passed through a gravity separator to recover stones which were then mixed with the debris from manual sorting and crushed. The remaining medium fraction passed a 40 mm flip-flop-screen followed by a manual sorting of the coarse materials into plastics and wood. The crushed stones and debris were screened using a vibrating mash screen, which consisted of a top deck with 50 mm openings and a



Figure 4.1: Scheme of the waste processing plant Miltenberg 1 (MIL1).



Figure 4.2: Scheme of the waste processing plant Miltenberg 2 (MIL2).

bottom deck with 25 mm openings. The overs (>50 mm) passed under a cross-belt magnet to separate iron from the residual plastics. Finally, processing generated three different soil grain sizes: fines (0-35/50 mm), medium-grained (35/50-80 mm) and coarse-grained soils (>80 mm). According to ISO14688-1 (2018), the fines range from sands to gravels, the mediums from coarse gravel to cobble, while the coarse-grained soils are cobble. The TS2 consisted of a trommel screen with a mash size



Figure 4.3: Scheme of the waste processing plant Traunstein 1 (TS1).

of 80 mm (Fig. 4.4). Afterwards the trommel unders passed through a star screen (35 mm) and the trommel overs through a gravity separator. Both equipments were followed by an air knife to separate the gravel from a solid recovered fuel like material (SRF), mainly plastics and textiles. In addition, wood and scrap was separated from the coarse material (>80 mm). The processing resulted in a fine-grained soil (0-35 mm), a medium-grained soil (35-80 mm) and coarse cobbles (>80 mm).

4.2.1 Laboratory analyses

In total the research included 63 laboratory samples; the processed soil piles at the Miltenberg WPPs were comprised of 31 samples and at the Traunstein WPPs 32. From every pile up to 600 cubic meters, ten composite samples – each consisting of four samples – were taken, quartered and mixed into one laboratory sample of 10 litres in line with the LAGA (2002) regulation. Thus, every laboratory sample was composed of 40 individual samples. Certified laboratories characterized the concentrations of elements and elemental compositions (now referred to only as "elements" in the samples and measured as well physical parameters (Table 4.1). The dry substance samples passed through a 40 mm sieve and the overs were crushed



Figure 4.4: Scheme of the waste processing plant Traunstein 2 (TS2).

before being added to the unders. The preparation of leachate analysis samples involved sieving (10 mm) before being batch tested in line with EN 12457-4 (2002). The analyses using standardized determination methods – generally ISO standards – included in total fifteen parameters for the dry substance and seven for leachate.

4.2.2 Statistical calculations

The weighted geometric mean (now referred to only as "mean") of the elements reflected the concentrations most suitable, as it was less susceptible to outliers and the positive (right) skewed distributions. The mean was calculated separately for the fine, medium- and coarse-grained soil samples, taking into account the pile quantities (i.e. weighted geometric mean). The differences in the means are expressed as a percentage to better compare the parameters measured in different units (e.g. mg/kg, μ g/l). To verify the significance of the differences between fines and coarsegrained soils, statistical tests require certain quantities of samples. Therefore, the medium-grained soils (35/50 - 80 mm) of the TS WPPs were considered to be coarsegrained (>35/50 mm). With the exception of pH, all parameters showed a positive skewness (histogram and Shapiro-Wilk test) and logarithmic transformations had an insufficient effect. Therefore, the non-parametric distribution required a rank test – such as the MWW – to verify the significance of the results. This test determines if the contamination of the fine and coarse-grained soils is similar (H_0) , or if the soils differ significantly and consequently that the pollutants can be reduced in the coarsegrained soils. To meet the requirements for a minimum of 20 samples and significance (asymptotic - p < 0.05, 2-tailed), the MWW test was calculated for each landfill but not for the WPPs. In addition to verify the results, the Spearman's ρ correlation coefficient which was applied by cross-checking the correlation between the results Table 4.1: Parameters of laboratory analyses, standards of determination methods, and units.

Parameter	Determination	Unit		
	\mathbf{method}			
Dry substance				
As, Cd, Cr, Cu, Ni, Pb, Zn	ISO 11885	mg/kg		
CN	ISO 11262	mg/kg		
Нg	ISO 16772	mg/kg		
C10-C40	ISO 16703	mg/kg		
C_{10} H ₈ (naphthalene)	ISO 18287	mg/kg		
PAHs(EPA)	ISO 18287	mg/kg		
PCB	ISO 10382	mg/kg		
TOC	ISO 10694	% dry substance		
Biodegradability	AbfAblV/DIN	${\rm mg} \; 0_2/{\rm g} \; {\rm dry}$		
	38414-8	$\operatorname{substance}$		
Leachate analysis (l.)				
Preparation of leaching batch	EN 12457-4	-		
test				
Cl^-, SO_4 (sulphate)	ISO 10304-1	mg/l		
F ⁻	DIN 38 405-D4	mg/l		
EC	EN 27888	$\mu { m S/cm}$		
pH-value	DIN 38404-5	-		
DOC	EN 1484	mg/l		
Ва	ISO 11885	mg/l		
EOX	DIN 38414-17	mg/kg		

of the MWW test (asymptotic significance) and the differences in the means. For concentrations of elements below the LOD, the LOD divided by the square root of two was used instead of a replacement by zero, half of the LOD or the LOD itself. This replacement turned out to to have the smallest relative difference (Croghan and Egeghy, 2003; Verbovšek, 2011).

4.3 **Results and discussion**

This section consists of the following subsections: composition of the processed waste and soils , contaminant concentrations of the fines and coarse-grained soils, contaminant concentrations of the fines, medium-grained and coarse-grained soils and the significance of the contaminant concentration differences using the MWW test .

4.3.1 Composition of the processed waste and soils

Both landfills consisted mainly of a mixture of soils, decomposed matter and CDW, plastics and textiles ("plastics"), and to a lesser extent of up to 0.5% asphalt, wood, scrap, glass and tyres (Fig. 4.5). The wood was incinerated in waste wood energy plants and the plastics in various incineration plants, the tyres in cement plants, while the scrap was recycled.

4.3.1.1 Soil classes and quantities

The classification of soils depends on their level of contamination and if the physical properties make it suitable for construction purposes. The reuse of soils is regulated in the LAGA ordinance (LAGA, 2003; StMUV, 2011), which contains seven classes ranging from not contaminated soils (Z0) to heavily polluted soils (Z5). Soils up to class Z1.2 (from now on referred to as "RC1" in this paper) can be reused in construction without cover and of class Z2 ("RC2") with cover (e.g. paved road). The disposal of soils is regulated in the landfill ordinance (BMU, 2009), including five landfill classes from DK 0 to DK IV (from now on referred to as "D0 to D4-limit value of the German landfill ordinance (DepV, 2009) (D4)" in this paper). Low contaminated soils, unsuitable for construction, can be inexpensively disposed of at D0 landfills, which possesses only a geological barrier, or used as cover at lined landfills. Usually in this study, soil of class RC1 was reused for backfilling of quarries and gravel pits, and of class RC2 and D0 for the top cover of lined landfills. The more contaminated soils (D1-D3) were disposed of at lined landfills (off-site). From a total of 30,249 tons of soils from the Miltenberg landfill, 32% were processed at MIL1 and 68% at MIL2 (Fig. 4.6). The soils examined here consisted mainly of fines, which were defined, depending on the specific process train, by passage through a 50 mm



Figure 4.5: Waste composition of the Miltenberg (left) and Traunstein (right) landfills.

screen at MIL1 and in the case of MIL2 even a 70 mm screen. Consequently, MIL2 revealed large quantities of fines (82%) compared to MIL1 (62\%). At MIL1, 77\% of the processed soils were composed of class D2 (56% fines; 21% coarse-grained), 12% of D1 (6% fines; 6% coarse-grained), and 11% of RC1 (coarse-grained). At MIL2, all the fines were of class D2 (82%), while the coarse-grained soils consisted of D0 (11%), D1 (4%) and D2 (2%). Thus, the coarse-grained soils showed lower contamination. However, the quantities of contaminated fines at MIL1 remained high, and at MIL2 the contaminant concentrations in the coarse-grained soils were only slightly lower. The more extensive processing at MIL1 produced 11% of the RC1 material. From a total of 15,591 tons of soils from the Traunstein landfill, 75%were processed at TS1 and 25% at TS2 (Fig. 4.7). At TS1, the extensive processing, using various types of equipment and a crusher, as well as larger screen openings (up to 50 mm) yielded a larger proportion of fines (TS1: 73% vs TS2: 50%). There were 19% medium-grained soils at TS1 and 31% at TS2, while the coarse-grained soils were 10% and 19%, respectively. At TS1, the largest proportion was made up of RC1 material (23% fines, 13% medium-grained, 10% coarse-grained) and of RC2 material (23% fines, 5% medium-grained). The remaining fines consisted of D0 (2%), D1 (21%) and D3 (3%). Thus, the medium-grained and coarse-grained soils could be completely used off-site for construction, and the fines – despite their large quantity – mostly used. The amount of material for disposal remained low (26%), however 3% was of class D3. At TS2, the largest proportion consisted of D1 material (41% fines, 31% medium-grained, 10% coarse-grained), while both RC1 (coarse-grained) and RC2 material (fines) contained 9% each. Hence, the processing at TS2 showed negligible results with regard to processing soils for reuse.



Figure 4.6: Mass balances of soils at the MIL WPPs by grain sizes and contamination classes.



Figure 4.7: Mass balances of soils at the TS WPPs by grain sizes and contamination classes.

4.3.1.2 Contaminant concentrations of soils

At the MIL landfill the heavy metal values tended to be higher than at the TS landfill (Table 4.2). Zinc showed the highest total average of 522 mg/kg at MIL and 332.7 mg/kg at TS and mercury the lowest at 0.14 mg/kg and 0.36 mg/kg, respectively. With regard to the elemental compositions and leachate analyses, the TS landfill indicated substantially higher values of PAHs, hydrocarbons, cyanide and PCB, whereas the MIL landfill showed higher concentrations of chloride and sulphate. PCB showed the lowest total average of 0.01 mg/kg at MIL and 0.09 mg/kg at TS, while hydrocarbons were the highest at 63.1 mg/kg and 172.3 mg/kg, respectively. The concentrations in the fines of lead, copper, zinc, PAHs and TOC frequently exceeded the RC2 and/or D0 limit values, as did in some cases sulphate, DOC, PCB and pH. The mean of heavy metal concentrations found in the dry substance were in line with FDEP (2009) and Hull et al. (2005), slightly higher than reported by U.S.EPA (1993) and by Zhou et al. (2015), and slightly lower than reported by Masi et al. (2014) and by Quaghebeur et al. (2013). Comparing these results to previous studies, the pH values were similar to those of Rong et al. (2017), Jani et al. (2016) and Zhou et al. (2015) and slightly higher than reported by Hogland et al. (2004), Prechthai et al. (2008) and U.S.EPA (1993). In contrast, the EC proved to be lower than found in Hogland et al. (2004), Wanka et al. (2017) and Zhou et al. (2015), similarly lower results were reported for chloride (Hogland et al., 2004) and TOC (NYSERDA, 1998b; Quaghebeur et al., 2013). The low conductivity and chloride values might be related to leaching processes occurring over the course of 40 years due to the absent surface sealing, while the lower TOC values might be caused by the intensive processing and manual removal of wood and plastics.

4.3.2 Contaminant concentrations of fines and coarse-grained soils

To enable the comparison between all WPPs, the medium-grained soils (35/50 mm - 80 mm) of the TS WPPs were considered to be coarse-grained (>35/50 mm). In terms of metals, the WPPs generally showed a similar pattern, with considerable differences in metal concentrations between fines and coarse-grained soils (Fig. 4.8). The greatest differences were reported for lead, cadmium, copper, mercury, and zinc (>35%). In addition, substantial differences in arsenic, chrome and nickel were recorded at TS1 (>50%). The differences in barium (leachate) varied substantially, though for no discernible reason processing at TS1 led to higher concentrations in the coarse-grained soils. In all, TS1 performed best, while TS2 and MIL1 had the weakest results. Generally, the concentration differences of lead (74%) were higher and of chrome (32%) lower than reported in Hogland et al. (2004, Pb: 58%, Cr:)

Parameter	TS average	MIL average	Limit value
	(max)	(max)	
As (mg/kg)	8.0 (21.0)	10.2 (22.0)	150
Ba (mg/l)	62.6 (220)	43.0 (93.0)	2000^{a}
Pb (mg/kg)	96.6 (350)	146 (890)	1000
Cd (mg/kg)	1.1 (2.8)	1.3 (4.8)	10
Cr (mg/kg)	29.1(50.0)	46.6 (73.0)	600
Cu (mg/kg)	93.1 (690)	119 (660)	600
Ni (mg/kg)	26.9 (91.0)	36.2 (110)	600
Hg (mg/kg)	0.36(1.1)	0.14(0.80)	10
Zn (mg/kg)	333 (850)	522 (1200)	1500
PAHs (mg/kg)	3.1 (19.1)	1.2(45.9)	20
C10-C40 (mg/kg)	172(550)	63.1 (210)	1000
C_{10} H ₈ (mg/kg)	$0.07 \ (0.31)$	$0.02 \ (0.17)$	1.0
Cyanide (mg/kg)	0.8(4.7)	0.1(1.1)	100
TOC [%]	1.7(4.0)	1.3(2.8)	1.0^{a}
Biodegrad. (mg $0_2/g$)	0.3(1.2)	$0.2 \ (0.5)$	5.0^{a}
PCB (mg/kg)	$0.09 \ (0.50)$	$0.01 \ (0.11)$	1,0
pH (leachate)	8.6 (7.9-11.9)	7.9 (7.7-11.0)	5.5-12
EC ($\mu m S/cm$)	267(1490)	291 (1130)	1500
$Cl^- (mg/l)$	1.7(4.3)	3.3 (19.5)	30
$SO_4 (mg/l)$	49.4 (330)	123 (650)	150
F^{-} (mg/l)	0.2(0.4)	0.3 (0.5)	1.0^{a}
DOC (mg/l)	5.2 (15.0)	4.9 (24.0)	50 ^a

Table 4.2: Total averages and maxima for the two landfills, and limit values (RC2).

^alimit value D0



Figure 4.8: Metal distributions (in %) in fines (black) and coarse-grained soils (grey).

65%) and Prechthai et al. (2008, Pb: 60%, Cr: 54%). In those studies in contrast, cadmium, copper, nickel and zinc accumulated in the coarse waste. However, their analyses were of mined waste and not soils. The accumulations in the fine material of all analysed heavy metals were in line with Rousseaux et al. (1992) organic matter of fresh waste, Schachermayer et al. (1998) construction and demolition waste, and - with the exception of chrome - Das et al. (2002) compost. In the first study the fine organic matter had a size less than 35 mm, and the last compared compost of particle sizes 19.1 mm and 9.5 mm. Studies which focused on sieved soils using openings of less than 10 mm suggest that the trend of heavy metal accumulation in fine-grained soils reverses for smaller particle sizes. Masi et al. (2014) reported that soils of a particle size of up to 4 mm revealed lower concentrations than the sieve-overs (<10 mm). Similar trends were recorded in pilot studies by Rong et al. (2017) comparing fine material <10 mm and <5 mm, and Jain et al. (2005) < 0.425 mm and < 6 mm. With regard to fresh waste, Rousseau et al. (1992) found lower concentrations in fines of particle sizes of up to 2 mm than in those of up to 5 mm, and Di Maria et al. (2013) detected the highest concentrations, using leachate analyses, in the material of 0.212-0.5 mm. It should be noted that, in full scale projects the efficient employment of sieves with openings of 5 mm and below depends largely on the soil characteristics. In terms of organic compounds (including cyanides), Figure 4.9 shows at all WPPs considerable concentration differences for PAHs and TOC (>50%) between the fines and coarse-grained soils, as well as moderate differences for biodegradability and naphthalene (>20%). In addition, the TS WPPs demonstrated considerable variation in the concentration of hydrocarbons, PCB and cyanide (>40%). While at the MIL WPPs, in the coarse-grained soils higher concentrations of hydrocarbons were recorded. However, the absolute hydrocarbon concentrations (63 mg/kg) remained considerably lower, compared to the TS landfill (172 mg/kg). With regard to hydrocarbons and cyanide, the TS WPPs showed a different pattern in comparison to those at MIL, which might be more related to the material properties than to the processing technologies. Thus, the effectiveness of mechanical screening might be uncertain for these substances. In terms of leachate analyses, the differences of measured values between the fines and coarse-grained soils proved to be heterogeneous and less pronounced, with the exception of sulphate (Fig. 4.10). The differences of pH, chloride, fluoride, DOC and EC (except at MIL1) remained low or proved to be heterogeneous, while chloride and fluoride tended to accumulate in the coarse-grained soils. Wanka et al. (2017) also reported higher chloride concentrations in the coarse-grained soils (10mm –



Figure 4.9: Elemental composition distributions (in %) in fines (black) and coarsegrained soils (grey).

60mm), but did not analyse fluoride. Sources of fluoride might be wood treated with salt-based water soluble preservatives, while higher chloride concentrations in the coarse-grained might be a result of magnesite screed pieces. After World War II cement was rationed, and magnesite screed was a frequently used construction material. Thus, fluoride and chloride might be leached from the fines, but remained more in the coarse-grained pieces. Although at MIL1 EC varied considerably between the fines and coarse-grained soils, the measured values stayed low ($\leq 525 \ \mu S/cm$). At all WPPs the fines tended to have lower pH-values. The inconsistent patterns and lower concentration differences in leachate analyses might be also related to the fact that the concentrations of chloride (<5 mg/l), fluoride (<0.4 mg/l) and DOC (<10 mg/l) were generally low, and the soluble part of the elements, particularly in the fines, was leached over a period of 40 years. The better results at TS1 might be caused by the sophisticated processing train consisting of a combination of screen types. A vibrating grizzly screen at the beginning of the process train efficiently broke up the waste matrix and humid soils. Moreover, the absence of rotation made the vibrating grizzly less susceptible to break downs (caused by tights and audio/video tapes) and jamming (by textiles and plastics). Moreover, at TS1 various different sieves and equipment, such as flip-flop-screens, gravity separators and crushers, were employed specifically to better break up the compacted waste and soil lumps. With regard to the processing rate, vibrating grizzly screens performed more efficiently than star screens. To break up the waste matrix, slow speed shredders could be an alternative. It should be mentioned, that the processing train at TS1 was only realised through special governmental subsidies and the personal commitment of the managing director. Kieckhäfer et al. (2017) also found for excavated waste that the on-site employment of some mobile equipment turned out to have the highest economical potential compared to sophisticated processing efforts.

4.3.3 Contaminant concentrations in the fines, medium- and coarsegrained soils

Previously, processed soils were only classified into fines and coarse-grained soils, whereas the medium-grained soils at the TS WPPs were considered coarse-grained. In the following the separation into fines (f), medium (m) and coarse-grained (c) soils/cobbles, and the resulting substance distributions will be analysed. At TS1 the processing resulted in 71.7% fines (0-35/50 mm), 18.7% medium-grained soils (35/50-80 mm) and 9.6% cobbles (>80 mm), while at TS2 the proportions were 50% (0-35 mm), 30.6% (35-80 mm) and 19.4% (>80 mm), respectively (Figure 4.7). The employment of a crusher at TS1 resulted in the smaller proportion of cobbles. TS1



Figure 4.10: Distribution of measured values (%) in leachate analyses (fines: black; coarse-grained soils: grey).

showed considerable (36-85%) concentration differences in the metals, except for barium, between the fines and medium-grained soils, whereas at TS2 the differences were less pronounced (17-42%) and were recorded between the medium- and coarsegrained soils. (Fig. 4.11). This effect might be related, despite the more intensive processing at TS1, to the fact that a proportion of the fines (0-35/50mm) at TS1 was classified as medium-grained soil (35-80 mm) at TS2. Thus, screen openings of 50 mm might more effectively redistribute metals than openings of 35 mm, and yield a low contaminated coarse-grained soil for off-site reuse. The concentration differences of chrome, copper and nickel were usually lower, and at TS2 the highest copper values were recorded in the medium-grained soil. Differences in barium concentrations (leachate) remained rather low, in particular at TS1. Both WPPs



Figure 4.11: Metal distributions (in %) in fines (black), medium-grained (grey) and coarse-grained soils (white).

showed considerable concentration differences for PAHs, cyanides, TOC and PCB (>30%), moderate ones (>20%) for hydrocarbons and biodegradability, and those of naphthalene were negligible (Fig. 4.12). While at TS1 the concentration in the fines differed greatly from the medium-grained soils, at TS2 the concentration differences

were not so high and were usually recorded between the medium- and coarse-grained soils (except PCB). In terms of leachate analyses, the previous concentration pat-



Figure 4.12: Concentrations of substances (in %) in the fines (black), mediumgrained (grey) and coarse-grained soils (white).

terns were only recorded for sulphate (Fig. 4.13). EC, fluoride and DOC remained almost unchanged, while the concentrations of chloride and the pH-value tended to be higher in the coarse-grained soils. No specific reason could be determined for the peak of DOC in the medium-grained soils at TS2. Previous studies-examining different waste sizes – have not focused on soils from landfills, but on fresh waste, construction and demolition waste and more. The trend of heavy metals, PAHs, TOC to accumulate in the fine materials were in line with the results found in Schachermayer et al. (1998) CDW. The accumulation of lead and chrome in the fine materials were comparable to the values of mined waste found in Prechthai et al. (2008). In contrast, in Prechthai et al.'s (2008) investigation nickel, zinc and cop-



Figure 4.13: Concentrations (in %) in leachate analyses of fines (black), mediumgrained (grey) and coarse-grained soils (white).

per accumulated in the medium sized waste (25-50 mm). Rousseaux et al. (1992) also found higher concentrations of heavy metals in the fresh waste fine material (25-35 mm), while lead, nickel and zinc increased once again in waste larger than 50 mm.

4.3.4 Significance of the contaminant concentration differences using the MWW test

The sampling of the processed soils at the MIL landfill consisted of 24 samples of the fines and seven of the coarse-grained soil, at the TS landfill 22 and 10, respectively. The MWW test verified that the concentration patterns of the fines and coarse-grained soil samples differed significantly (p < 0.05, 2-tailed), and whether elements usually decreased in the coarse-grained soils. The processed soils of both landfills showed significant (i.e. p < 0.05) concentration differences for arsenic, lead, cadmium, chrome, copper, mercury, zinc, PAHs, TOC and sulphate, but not for chloride, fluoride and DOC (Fig. 4.13). In addition, MIL revealed significant differences between fine and coarse-grained soils of barium, naphthalene and EC. At TS significant differences were observed for nickel, hydrocarbons, cyanide, PCB and pH. With the exception of sulphate, parameters of leachate analyses proved to reflect less significant differences, particularly for fluoride and DOC. The less significant results of the MIL landfill might be caused in part by the slightly higher disproportion of sample numbers (MIL 24:7, TS 22:10). Generally a high difference in concentration proved to be significant (p < 0.05), except for the pH-value which revealed low differences and a high significance. This pattern was induced by the high homogeneity of concentrations in the fines (Fig. 4.14). In addition the relation between the concentration differences and the significance (MWW test) was verified using a correlation test (Spearman ρ). This test resulted in Spearman ρ -0.81 (p 0.00) at MIL and ρ -0.69 (p 0.00) at TS, and thus confirmed that the greater the differences in the means were, the more significant the reduction of elements in the coarse-grained soil proved to be. Figure 4.15 shows exemplary four typical distribution patterns in the fines and the coarse-grained soils of the TS landfill. The pattern in the top left figure reflects the high and significant concentration differences in PAHs, although the concentration distribution was heterogeneous. Similar patterns were observed for most heavy metals, sulphate, TOC and others. The top right chart presents the low but – due to the homogeneous distribution – significant concentration differences in the pH measurements. In the chart on the bottom left no significant differences (e.g. EC) were noted, for the most part, although the coarse-grained soil induced several high values. Despite the EC, DOC and fluoride behaved similarly. In the bottom



Figure 4.14: Significance of the differences between the fine and coarse-grained soils (TS black, MIL grey).

right figure: the concentrations of chloride revealed a moderate accumulation in the coarse-grained soil; however, the significance of the difference is low $(p \ 0.07)$. Similar results were obtained at MIL for nickel and at TS for naphthalene.

4.4 Conclusions

Landfills consist mainly of soil-like materials, thus the contaminant concentrations of soils are crucial for their reuse. In this study soils from two completely excavated landfills were processed at four different processing plants using mechanical equipment, in particular for dry screening. Processing targeted the reduction of contaminants in the coarse-grained soil, resulting in a soil suitable for off-site reuse complying with regulatory limits. Screen openings of 50 mm performed more efficiently than openings of 35 mm, or of 70 mm and larger, with regard to contaminant redistribution and proportion of material flows (unders and overs). Screen openings of 35 mm produced coarse-grained soils with higher contaminant concentrations compared to soils produced with a 50 mm screen. Whereas the simple separation using 70 mm screen openings led to high quantities of moderately contaminated fine-grained soils (<70 mm). However, the optimum screen opening size seemed to be between 35 mm and 50 mm. Employing a vibrating grizzly screen at the beginning of the process train proved to be more efficient in breaking up the waste matrix, and the absence of rotation led to fewer breakdowns and less clogging due to tights, audio/video tapes and other plastics. Alternatively, a star screen might be useful but the processing rate would be lower. Employing different screen types resulted in greater contaminant reductions in coarse-grained soils; however, the optimal equipment depends largely on the particular waste characteristics. Less humid, clumping or uncompacted materials allow the direct use of trommel and vibrating screens. Higher substance concentrations in fines were observed in previous investigations, and were verified in this study for many substances using statistical methods. Nonetheless, the concentrations of a few elements, particularly in leachate analyses, were noted to be higher in the coarse-grained soils (>35/50 mm). Heavy metals, PAHs and TOC were significantly (asymptotic - p < 0.05, 2-tailed) decreased by 35% to 91% in the coarse-grained soil, and moderate reductions were noted for sulphate and biodegradability (> 27%). Measurements of leachate analyses usually proved to be heterogeneous and less pronounced between fines and coarse-grained soils. The values of DOC, EC and pH in leachate analyses did not follow a regular pattern, while chloride and fluoride tended to accumulate in the coarse-grained soil. Nevertheless, the absolute values remained low. The reason for this pattern could not be identified but might be related to the leaching of the soluble part, particu-



Figure 4.15: Distribution of PAHs (top), EC (middle) and chloride (bottom) in fine (white) and coarse-grained soil (black).

larly in the fines, over a period of 40 years. Although the two landfills were located 450 km away from each other, the waste seemed to be quite similar due to their comparable age and origin (rural area). Moreover, both landfills were characterised by humid conditions, the lack of compaction of the waste during placement and the lack of engineered covers, which should taken into account when comparing results. Mechanical screens separate materials by secondary properties, such as size, density and others. Thus, the efficiency of contaminant accumulation depends strongly on the homogeneity of material categories, despite the fact that contaminants accumulate in the fines due to a higher surface area. Preliminary analyses are of major importance for selecting appropriate processing equipment, and soil washing equipment might be the better choice when leachate parameter values exceed national regulatory limits. However, complex processing equipment involves higher costs and might lead to a lower cost-effectiveness (Kieckhäfer et al., 2017). During the project a socio-economic issue also emerged: different tendering procedures seemed to influence the processing quality; screening results tended to be of a higher quality when the company carrying out the work became the owner of the excavated material and was not just a waste processing service provider. Further research should focus on a wider range of screen opening sizes, the combination of different processing technologies and a wider variety of waste types. Previous investigations reported notable contaminant reductions in soil particle sizes smaller than 10 mm (Jain et al., 2005; Masi et al., 2014; Rong et al., 2017; Rousseaux et al., 1992). Screen openings of up to 10 mm can be used for dry and sandy material, while the processing of clumping and coarse-grained material – as in this study – might not be efficient when using openings of this size. The production of low contaminated fines (<10 mm) might provide a further reuse opportunity or at least reduce disposal costs. However, the feasibility of screening soils using screen openings of less than 10 mm has to be examined prior to application in full-scale projects. Although screen openings of 50 mm performed best, the comparison to 35 mm openings indicated an optimum between the two sizes. To more precisely identify the patterns of contaminant redistribution and proportions of material flows (unders and overs), systematic testing of closely graduated screen openings (e.g. 5 mm grades) and a wide range from 5 mm to 70 mm is advised. Further research should also focus on the combination of processing technologies, using different types of screens, air knives and wet mechanical treatment equipment. Furthermore the modification of equipment with regard to excavated waste might improve the contaminant redistribution. In a subsequent project, one processing company reported significant improvements using a

recently developed windsifter – combining pressure and suction – to remove small objects comprised of wood, plastics, cardboard and leather. Despite the contaminant reduction, the processing rate and the percentage of low vs high contaminated soils are important. Performance improvements should be assessed with regard to cost-effectiveness, since soils are of low market value and expensive to transport. Further research emphasis should compare wastes with different properties (origin, age, compaction etc.), particularly in terms of processability and substance concentration irregularities (e.g. chloride, fluoride).

5 Recycling - regional material flows and influencing factors

Analysing material flows of landfill mining in a regional context

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Abstract

Landfill mining LFM is considered an option to recover recyclables and at the same time to prevent environmental hazards as well as an option to reclaim land. With regard to the environmental sustainability of LFM, previous research has focused on life-cycle assessment and the climate impact of energy generation using refusederived fuel from mined landfills. These studies were commonly based on hypothetical models paying little attention to emissions resulting from operations (e.g. excavation, processing, transportation). Since LFM involves a broad range of stakeholders, the objective of the present study is to investigate LFM empirically in a regional context using data from eight mined landfills in Germany. A material flow analysis, which included a calculation of energy consumption and related emissions from operations, was carried out and factors affecting material flows were determined based on environmental scanning. LFM operations required on average 103 megajoules diesel $\simeq 2.4$ kg) and 1.9 megajoules electricity per tonne excavated waste, producing 12 kg of CO₂-equivalents (Global Warming Potential: 100 years) (CO₂-eq.). Transportation proved to be the sub-process with the largest energy consumption by far, producing 58% of total emissions, followed by processing (27%). The average transportation distance was 122 km; however - in contrast to previous studies transportation distances for recovered soils (84 km) and asphalt (175 km) were considerably longer. Decisive for energy consumption were: a) the option of excavating waste lifts one at a time, b) an on-site processing option, c) processability of waste, and d) an on-site reuse option and/or nearby recovery facilities for soils. Flexibility, pragmatism and coordination of stakeholders proved to be key factors due to the complex and individual character of LFM projects as well as numerous interfaces.

5.1 Introduction

The concept of LFM has evolved to ELFM which aims to close the material loops to foster a circular materials economy, using landfilled waste by employing innovative transformation technologies (Jones et al., 2013). The research on LFM in the last two decades has focused on characterizing deposited material and to a lesser extent on technology needed for excavation and materials processing (Krook et al., 2012). Landfills were investigated in terms of material composition (Kaartinen et al., 2013; Quaghebeur et al., 2013) or characteristics of specific materials, such as plastics (Passamani et al., 2016; Zhou et al., 2014) or fines (Burlakovs et al., 2018; Rong et al., 2017). Research on technology has either focused on sorting equipment (Garcia Lopez et al., 2018; Wanka et al., 2017) or recovery techniques (Bosmans et al., 2013). In addition to their composition, the processability and reuse options of materials as well as markets for recycled products are crucial for profitable LFM (Van Passel et al., 2013). Johansson et al. (2017a) concluded that evaluating marketability requires an institutional, technical and organizational approach, while Hermann et al. (2016) observed a broad range of stakeholders involving many environmental and socio-economic factors. Recent studies have also identified project related issues, such as ecosystem services revitalization (Burlakovs et al., 2017) and evaluation perspectives (Winterstetter et al., 2018). Thus far, Danthurebandara et al. (2015b), Frändegaard et al. (2013a), Jain et al. (2014) and Laner et al. (2016) have developed models for life cycle assessment to evaluate the environmental impact and benefit of recycling and recovering materials (e.g. metals, plastics, paper) from LFM. These studies focused on the ecological benefits resulting from emission savings due to metal recovery and substitution of primary fuels using RDF. Laner et al. (2016) observed that ecological benefits are mainly determined by the waste composition, the efficiency of WtE-plants, the background energy system and landfill gas management. Energy savings from metal recovery might, worldwide, be very similar, due to insignificant variations of metal proportions in landfills and comparable scrap markets. The environmental impacts of LFM itself, i.e. excavation, processing, transportation and rehabilitation, have so far only been investigated at one landfill (Jain et al., 2014). However, this study did not take a significant quantity of soil into account, and the results are limited to a single case study. Krook et al. (2012) observed that landfills consist mainly of soil-like materials (referred to as "soils" in this paper) with limited reuse options due to the requirements of soil protection acts. Reuse and recovery of soils require prior processing, which can be carried out either on-site or off-site at processing plants. Processing usually results in a portion of low contaminated soils ready for reuse/recovery and a portion of contaminated soils that has to be disposed of or might be used to a limited extent as construction material at landfills (Hölzle, 2018). Transportation activities will increase if contaminated soils may not be dumped on-site or have to be treated at more sophisticated processing plants. In addition to the ecological impact, costs will arise since soils are of low or no economic value and expensive to transport. Transportation distances to recycling facilities are, apart from their economic importance, crucial for environmental assessment and hitherto have not been researched (Frändegaard et al., 2013a). For instance, Danthurebandara et al. (2015b) did not incorporate transportation in their model, Van Passel et al. (2013) and Laner et al. (2016) reported negligible emissions from transportation, while Frändegaard et al. (2013a) concluded that transportation is the second-largest factor for added green house gases. As a result of long transportation distances to a WtE-plant, plastics from the Sharjah remediation project had to be disposed of, while the reuse of the generally large soil quantities was crucial for the economic assessment (Goeschl and Rudland, 2007; Jani et al., 2016; Krook et al., 2012). Thus, in addition to their material composition, landfills have to be assessed with regard to their spatial context taking into account the regional infrastructure (e.g. WtE-plants, processing plants, recycling facilities) – and their business environments. The reliability of a model depends on the choice of selected parameters and the quality of input data, which is usually based on practical experience (Frändegaard et al., 2013b). The assumptions and model choices that have formed the basis of the above mentioned hypothetical case studies resulted in a level of uncertainty, that is difficult to quantify (Laner et al., 2016). For instance, (Hermann et al., 2014) determined a comprehensive set of factors influencing LFM which reflected the complexity of evaluation. It should be noted, though, that this study followed a theoretical approach, whereas empirical studies have been a common source to identify factors influencing waste management (Simões and Marques, 2012). is considered an appropriate instrument to assess LFM holistically, taking into account waste composition, flows, regional infrastructure and markets for recyclables. In contrast to previous models, this study is based on an inductive bottom-up approach and compares results of eight mined landfills to identify LFM aspects in practice. The research focuses on the material and energy flows of the LFM operations, i.e. site preparation, excavation, processing, transportation, rehabilitation and disposal, as well as on factors affecting material flows. The objective is to offer a greater insight into the current practice of landfill mining and to get an detailed overview on what factors influence LFM operations, in order to provide a better basis for the preparation of legal, institutional and organizational frameworks. This study seeks to:

- qualify and quantify materials and their flows in a regional context
- determine the energy consumption and related emissions arising from LFM operations
- identify factors affecting material flows and influencing LFM operations
- place a particular emphasis on soil recovery and transportation

5.2 Materials and methods

5.2.1 Site description

Eight landfills, used between the 1950s and 1980s to dispose of MSW and CDW, were completely excavated in the German Federal State of Bavaria. The investigated landfills each had a surface area of up to 6,130 m² and waste quantity of up to 30,957 tonnes (see Appendix C). In total 121,133 tonnes of waste, with a volume of 74,519 m³, were excavated. As these landfills had neither bottom nor surface sealing, to protect drinking water abstraction and, in the case of Oberall-gäu, instability their complete excavation was required. In line with the EU waste management hierarchy of prevent, reuse, recycle, recover and dispose, the German waste management directive requires separating and recycling the excavated waste to the extent possible (European commission, 2008; KrWG, 2012). Processing was carried out on- and/or off site and by means of mechanical screens, crushers, gravity separators, air classifiers, cross-belt magnets, and manual sorting.

5.2.2 Material flow analysis (MFA)

Metabolism, i.e. physiology, is defined as the transfer, storage and transformation of materials within a defined system and the exchange of materials with the environment (Baccini and Brunner, 1991). An MFA assesses the flows (input and output), stocks and processes of a system, taking into account time and space. Materials include both goods, e.g. plastics, tyres, wood, and substances, such as chemical elements and elemental compositions (Brunner and Rechberger, 2017). MFA is normally used to quantitatively analyse waste flows, while a SFA focuses on the transformation of wastes (Stanisavljevic and Brunner, 2014). The present case study is based on accounting, more than 2,000 consignment notes of transport, daily construction records, remediation reports project completion reports, documents of stakeholders (engineering consultants, hauliers, waste companies, environmental agency), stakeholder interviews, field visits and data from the ecoinvent database version 3.3 (Wernet et al., 2016). Processing of mined waste revealed soils, CDW, plastics and textiles ("plastics"), wood, tyres, metals and hazardous waste. Apart from structural requirements for soils and CDW, the technical guidelines define - depending on the contaminant concentration - mineral materials from class Z0 to Z2 (referred to as RC2 in this paper) as appropriate for reuse such as parking lots, noise barriers, sub-bases of roads, backfilling of quarries and gravel pits (LAGA, 2003). Soils and CDW of higher contamination can be reused as substitute construction material at landfills, but if they exceed certain limit values must be disposed of at appropriate landfills. With regard to reuse and disposal of inert waste at landfills, the German landfill ordinance defines four surface landfill classes: from D0 for low contaminated to D3 for heavily contaminated waste (DepV, 2009). The material flow of hazardous waste – consisting mainly of batteries and asbestos - was not further analysed due to the very small quantities.

5.2.2.1 Sub-processes

In this case study, the process of LFM consisted of six sub-processes: site preparation, excavation (including presorting, treatment of leachate and surface water, and pumping of groundwater), transportation, processing of excavated waste (on-site or off-site), disposal and rehabilitation or restoration (referred to as "rehabilitation" in this paper; Fig. 5.1). Site preparation involved expanding or resurfacing of access roads, the occasional felling of trees, removing topsoils, installing working facilities (for office containers, waste containers, engines) and, where necessary, installation of groundwater pumps and water treatment equipment. In a few cases the access road had to be resurfaced after the project. For the excavation tracked excavators were generally used and sometimes wheeled or tracked loaders as well. At all landfills, containers were designated for the collection (presorting) of particular bulky objects (scrap, tyres, wood, asphalt), barrels and bulk bags for hazardous materials. Waste separation was carried out either by excavation of waste lifts one at a time, or by on-site and/or off-site processing. At the Ansbach and Straubing landfills, there was sufficient space and the groundwater situation was less sensitive enabling on-site processing to some extent. Soils rarely had to be transported to an interim storage site. The off-site process train was composed of a vibrating grizzly, trommel screen, cross-belt magnet, air knife and conveyor belt for manual separation,

for all of which the energy consumption and emissions had to be calculated. On the other hand, the on-site process train consisted of a trommel screen and a star screen. At on- and off-site processing, the energy calculations also included the use of a wheeled loader. To some extent, waste layers could be removed separately and directly transported away. All sub-processes involved transportation to deliver or remove waste and materials. Disposal included energy consumption and related emissions for construction and operation of residual material landfills.

5.2.2.2 Flows

The mass flow rate was defined as metric tonnes per day (t/d), since the limit was the daily capacity of processing plants. All other sub-processes were usually more flexible and less sensitive to quantity changes. Flows consisted of excavated waste, construction material (e.g. for access roads), fuel, electricity, emissions (off-gas) and water from drainage which resulted from excavation operations (see Fig. 5.2). Water quantities were not part of the mass balance; however, these were calculated since pumping of groundwater and treatment of surface water had a considerable affect on energy demand during excavation. Oxygen consumption for diesel combustion and energy generation was not taken into account, while methane emissions were not an issue, after 40 years of decomposition of organic matter. Solid materials, water and emissions (CO_2 -eq., GWP 100 years; referred to as " CO_2 -eq." in this paper) were calculated in tonnes, while electricity and diesel consumption in gigajoules (GJ). The import consisted of (a) asphalt and gravel for the construction of working areas and access roads, (b) gravel and top soil for rehabilitation, (c) diesel for excavation machines, trucks and processing equipment, and (d) to a lesser degree electricity for water pumps, water treatment plants, magnetic belts and conveyor belts for manual separation. The storage (stock) comprised gravel and top soil for rehabilitation, gravel and asphalt for road construction and disposed soil as well as asphalt from excavated waste. The export included processed waste, water from drainage, construction material from site preparation and emissions from diesel engines and electricity generation. Small material flows (plastics, wood, scrap and tyres) were bundled and allocated to the largest flow (on-site processing, off-site processing or excavation by waste lifts one at a time). For instance due to their size, tyres were mainly separated at the landfill and were thus, allocated to the "excavation by waste lifts" flow. The quantities of excavated materials and their transportation were recorded in detail, while the quantities of water, access road construction material, as well as energy consumption for site preparation, excavation and rehabilitation were only available for a few landfills. To overcome this lack of data, existing data



Figure 5.1: Sub-processes of LFM: transportation, site preparation, rehabilitation, excavation, processing and disposal.



Figure 5.2: Input and output flows of materials, energy and emissions, as well as excluded energy benefits (right).

was converted with regard to landfill size and site condition, and incorporated into the case studies lacking complete data sets. The remaining information and figures for some standardized sub-processes, e.g. energy demand and emissions for transportation, excavation, track construction, disposal and rehabilitation, were taken from the ecoinvent database (see Appendix D).

5.2.2.3 Spatial and temporal system boundary

The system boundary started at production of construction material for constructing access roads and working areas as well as rehabilitation and ended at the gate of plants (e.g. WtE-plant) where processed materials were used. When materials were disposed of, the disposal activity was taken into consideration, which means the material flow calculations ended for reused soils at the gate of a pit or landfill, for CDW at the gate of the reseller, for asphalt at the landfill, recycling plant or the road construction site, for plastics, tyres and wood at the gate of the incineration plant and for scrap at the gate of the smelting plant. Thus, energy savings and emissions from recycling, thermal recovery and landfill gas utilization were not part of the system. Water and heat consumption, e.g. for the operation of landfills, the construction of trucks, etc., were also not taken into account. Transportation encompassed the entire transport life cycle, including the construction, operation, maintenance and end of life of vehicle as well as road infrastructures. Transportation of construction machines and personnel was not considered.

5.2.3 Analysis of influence factors

The environment of LFM was scanned on an empirical basis for factors influencing operations and affecting material flows. The identification of influence factors involved scanning of regulations, guidelines, tenders, reports (e.g. preliminary in-
vestigation, remediation assessment, rehabilitation planning, project completion), correspondence between stakeholders, further documents of engineering consultants, hauliers, waste companies, contracting authorities and the environmental agency, as well as interviewing stakeholders. PEST analysis (political, economic, socio-cultural and technological) enables the creation of an overview on what macro-environmental factors determine business (Fahey and Randall, 2001). Identified influence factors were grouped based on an adapted PEST analysis using the classes economy, technology, organisation, and institutions/laws. In addition, landfill properties constituted a fifth class, comprising internal factors such as waste composition, size, topography etc. The functioning of influence factors, that means their relationships, causality, interaction and strength of influence is not part of this study.

5.3 Results and discussion

5.3.1 Material flow analyses

The excavated landfills consisted mainly of soils (87%) and CDW (5%), and to lesser extent of plastics, scrap, wood, tyres and topsoil of the cap. The soils were made up of 33% RC2 material, 21% D0, 19% D1 and 15% D2 (Table 5.1). The landfills showed a high proportion of soils similar to older landfills researched by Hogland et al. (2004) and Masi et al. (2014). In total 143,596 tonnes of solid material were moved, not including pumped groundwater and collected surface water resulting from excavation. The daily import was 77 t/d, while the export consisted

Waste type	%
RC2	32.6
D0	21.0
D1	18.6
D2	15.0
Topsoil	5.1
CDW	5.0
Plastics	2.1
Scrap	0.4
Wood	0.3
Tyres	0.1

Table 5.1: Average composition of the investigated landfills.

of 441 t/d waste and materials, not including 73 t/d of ground and surface water

resulting from excavation operations (Fig. 5.3). Consequently, the stock decreased by a net 364 t/d including 86 t/d new stocks. The import consisted of 22 t/d gravel and 2 t/d asphalt for site preparation (track, access road and working area construction), as well as 28 t/d topsoil and 25 t/d gravel for rehabilitation. New stocks (86 t/d) resulted from rehabilitation (74 t/d), disposal (9 t/d) and - since tracks and access roads were usually not dismantled - site preparation (3 t/d). Topsoil not containing waste was reused for rehabilitation, but due to limited space in two cases the topsoil had to be transported off-site and stored temporarily (7 t/d). The standard deviation (SD) of the imported materials, stored materials and water were usually high, since their type and quantities differed from project to project. Import quantities and storage of gravel, asphalt and topsoil were highly dependent on the construction of access roads and the subsequent use of the landfill. The output in total was composed of separated waste (440 t/d), gravel (0.5 t/d) and asphalt (0.2 t/d from the site preparation, while 9 t/d of separated waste were disposed of and remained within the system boundaries. In addition, excavation operations resulted on average in an output of 1 t/d treated surface water and 72 t/d pumped ground water. Groundwater was pumped in only one case, while surface water was collected and treated in two cases. In six of eight projects, the waste was transported to processing plants (280 t/d), while 101 t/d of waste were processed on-site. However, 44 t/d of on-site processed waste required further treatment at processing plants. In a few cases homogeneous waste lifts (68 t/d) could be excavated separately and were transported away without further processing. In total, the separated waste consisted of 409 t/d soils, 17 t/d CDW, 11 t/d plastics, 1.7 t/d scrap, 1.3 t/d wood, 0.4 t/d tyres and 0.2 t/d asphalt. With regard to soils, processing resulted altogether in 145 t/d RC2, 91 t/d D2, 88 t/d D0 and 85 t/d D1 (not including 8 t of disposed of D1). Processing at stationary plants resulted mostly in soils of class D0 (79 t/d), RC2 (78 t/d), D2 (69 t/d) and D1 (68 t/d). On-site processing produced soils of class D1 (22 t/d), D2 (22 t/d), RC2 (6 t/d) and D0 (6 t/d). Excavation by waste lifts enabled the separation of soils of class RC2 (61 t/d), D1 (3 t/d) and D0 (3 t/d). Processing at stationary plants turned out to be more efficient than on-site, resulting in more low contaminated soils of class D0 and RC2, while on-site processing usually produced medium contaminated soils of class D1 and D2. Excavation by waste lifts was carried out in particular to separate low contaminated soils (RC2) for backfilling at nearby pits. Similar scrap proportions existed at all projects; thus, the uncertainty remained low (SD: 64%). The same applied to plastics, wood and tyres, resulting in an uncertainty of 110%. Dos Muchangos et al.

(2017) recorded uncertainties in MFA of municipal solid waste ranging from 29% to 96%. Uncertainty of soil quantities ranged from 17% to 267%. RC2 and D1 soils showed the lowest uncertainties; however, different soil quantities from on-site and off-site processing as well as from excavation by waste lifts considerably affected the uncertainty level. Since fewer projects involved on-site processing, the uncertainty of soils there tended to be higher (SD: 136% to 267%). The same applied to the uncertainty of ground water and the export of gravel and asphalts. On average, the daily waste flow was approx. 449 t, but up to 601 t when processing was carried out off-site using two processing plants and, at the minimum, 320 t for on-site processing. Moreover, increasing storage of on-site processed soils - due to the time necessary for carrying out chemical analyses - became a problem over time. The stationary processing plants were designed for the separation of CDW and their usual capacity was higher. The separation of the excavated waste required more time and effort than CDW from current waste streams. The capacity of one plant alone was not sufficient to process the waste stream of the landfills exceeding 10,000 tonnes, due to limited space for storage of excavated and processed waste. Since excavation can be carried out faster than processing, using off-site processing plants provides an advantage in timing and even in quality. In a few cases processed waste had to be forwarded to a second processing plant to separate particular contaminants, whereas on-site processing allowed sending specific wastes directly to an appropriate processing plant. In line with the waste hierarchy, 5% of materials (topsoil) were reused, 0.3% recycled, 91.1% recovered, 1.9% thermally recovered and 1.7% disposed of (Table 5.2). The recovery of soils depended primarily on the degree of contamination and local recovery options. Processing focused primarily on removing plastics and wood, due to the 5% TOC limit, and to a lesser degree on scrap. Nevertheless, frequently individual contaminants led to an inferior classification. Consequently, transportation to a soil treatment plant turned out to be more economical, making it possible to avoid the high disposal costs of D2 and D3 landfills. However, transportation costs and thereby emissions increased. Processing resulted in a large proportion of RC2 soils, which could be used for backfilling of quarries and gravel pits. More contaminated, often fine-grained (<50mm), soils were used as construction material at landfills of class D1 and D2. CDW was crushed before being reused for instance in road sub-bases. Asphalt was initially added to the production of new asphalt. Due to bituminous (pitch) content, in later projects it was reused for sub-bases of roads, disposed of at landfills or recovered at underground waste storage facilities. Plastics were usually separated at a processing plant and to a lesser extent directly at the



Figure 5.3: Mass-per-time-flow diagram for solid materials and water during the period of 1 day.

landfill. Plastics could not be recycled due to adhesive particles, and were thus thermally recovered in incineration plants. Less frequently, plastics were manually sorted at specialised processing plants, to avoid the high MSW incineration costs (approx. 140 EUR/t), and those free of adhesive particles were sent to RDF power plants. types were thermally recovered at cement plants due to adhesive particles. Wood was thermally recovered at waste wood incineration plants or coincinerated at a lignite power plant. The waste wood regulation specifies the recovery of waste wood and defines five classes with regard to the degree of contamination (AltholzV, 2002). Since the quantities of wood recovered were low, the type heterogeneous, and thermal treatment plants existed either for untreated or highly contaminated wood, the wood was incinerated in plants for highly contaminated wood without prior analyses. The scrap was sold to local merchants; however, the revenues were negligible. Metal flows and transportation were difficult to analyse since merchants collect scrap from different sources and classify it before selling it to wholesalers or smelting plants. Consequently the metal flow was characterized by more and changing actors. However, the scrap flow could be researched in detail for the Straubing landfill. The scrap merchant there sold iron to two steel mills and non-ferrous metals to regional wholesalers. The aluminium wholesaler shredded and resold it, probably to Austria.

Process	Proportion	Material type
Reuse	$5.0 \ \%$	Topsoil
Recycling	0.3%	Metals (0.3%), asphalt (< 0.1%)
Material	91.0 %	Soil (86%), CDW (5%), asphalt
recovery		$({<}0.1\%$)
Energy recovery	1.9%	Plastics (1.5%) , wood (0.3%) , tyres
		(0.1%)
Disposal	1.7%	Soil (1.7%) , asphalt $(<0.1\%)$

Table 5.2: Waste types and quantities with regard to the waste hierarchy.

5.3.2 Calculation of energy consumption and emissions

In terms of energy consumption, the daily import was on average 46 GJ diesel ($\simeq 1.1$ tonne) and 0.8 GJ electricity, producing approximately 5.2 t/d of CO₂-eq., which means on average 103 MJ diesel ($\simeq 2.4$ kg), 1.9 MJ electricity and 12 kg of CO₂-eq. per tonne excavated waste (Fig. 5.4). In contrast, Vitale et al. (2017)

reported 200 MJ/t fuel and 41.4 MJ/t electricity consumption for demolition waste from residential buildings. Laner et al. (2016) calculated up to 640 kg CO₂-eq. per tonne mined waste including emissions from thermal recovery, but also showed savings of up to -1550 kg of CO₂-eq./t depending on the energy system (heat and electricity) and its share of renewables. With regard to energy generation at regional WtE-plants, thermal recovery of one tonne of processed waste would result in production of approximately 175 MJ, consisting of 152 MJ from incineration of plastics, 13 MJ of tyres and 10 MJ of wood (see Appendix E). Energy savings from avoided primary steel production due to ferrous scrap recycling would be 56 MJ. Danthurebandara et al. (2015a) similarly determined higher benefits from RDF usage than from metal recovery. However, their calculation of electricity generation resulted in 2.556 MJ per tonne of waste, while Frändegaard et al. (2013b) estimated a district heating generation of 13,500 MJ/t. These high values might be related to a larger proportion of plastics and a higher assumed calorific value, as well as to a higher efficiency of WtE-plants in the latter study. Jain et al. (2014) observed larger CO_2 savings from metal recovery than from incineration of RDF avoiding coal usage for electricity generation. In addition, Laner et al. (2016) and Danthurebandara et al. (2015a) identified that avoided landfill gas emissions are of major importance for ecological assessments, in particular at early stages of waste degradation. Transportation proved to be the main energy consuming sub-process (28) GJ/d), followed by processing (14.6 GJ/d), excavation (2.1 GJ/d), rehabilitation (1.1 GJ/d), site preparation (0.9 GJ/d) and disposal (0.3 GJ/d). Thus, 58% of emissions resulted from transportation and 27% from processing. Danthurebandara et al. (2015a) also found that separation and transportation dominated most environmental impact categories when substituting coal using RDF, while the impact of excavation proved to be negligible and site preparation was not taken into account. In contrast, Jain et al. (2014) reported that mining operations (i.e. excavation and processing) resulted in six times more emissions than transportation, but significant quantities of soils were not part of the calculations. Laner et al. (2016) concluded that emissions from transportation are irrelevant taking the background energy system, composition of excavated waste and WtE technology into account. However, transportation distances for soils were assumed to be significantly smaller than in the present study. With regard to electricity, the main consumption took place during off-site processing (0.4 GJ/d), followed by site preparation and rehabilitation (each 0.1 GJ/d), as well as excavation and disposal (each <0.1 GJ/d). Transportation to processing plants required most of the diesel (13 GJ/d) and generated the

highest emissions (1.6 t/d \simeq 31 % of total emissions), while transportation to recycling/recovery facilities necessitated 7.1 GJ/d for off-site processed waste, 5.6 GJ/d for on-site processed waste, and - due to nearby backfilling options for soils - 0.8 GJ/d for excavated waste by lifts. Diesel consumption for transportation of rehabilitation materials was 1.3 GJ/d and of materials for site preparation 0.2 GJ/d. In relative terms, transportation of waste to processing plants required 46 MJ/t, while subsequent transportation of separated materials from the processing plant necessitated 22 MJ/t, on-site processed materials 55 MJ/t, waste excavated by lifts 13 MJ/t, rehabilitation materials 25 MJ/t, materials for site preparation 9 MJ/t and removing materials from site preparation 2 MJ/t. Thus, processing at stationary plants increased energy consumption of transportation by 23% compared to on-site processing. However, transportation from processing plants to recycling/recovery facilities could not be completely recorded and the diesel consumption might be slightly higher. Apart from the transportation to and from processing plants (SD: 73% and 46%), all other transportation showed a high uncertainty (SD: 110% to 200%) with regard to diesel consumption. The SDs of energy consumption are equal to those of emissions and are therefore not presented in Figure 5.4. Greatly varying transportation distances resulted in a high uncertainty, since plastics, wood and asphalt had to be transported far. In addition, in a few cases transportation for site preparation and rehabilitation was necessary which only further increased uncertainty. Finally, fuel consumption for transportation might be considerably reduced by reusing soils on-site or nearby. Similarly, Frändegaard et al. (2013a) found higher green house gas emissions for transportation from the landfill to a processing plant than from processing plants to recovery or disposal facilities. On-site processing performed better in terms of emissions for transportation distances greater than 300 km; however, the percentage of plastics significantly affected the net emissions. In contrast, Gusca et al. (2015) recorded a reduction of environmental impacts by 28%for on-site processing compared to an off-site processing plant 12.6 km away. In the model of Danthurebandara et al. (2015a) transportation and separation both topped the environmental impact categories; however, a major benefit of using refuse derived fuel from LFM was the reduction of transportation of coal. The study by Jain et al. (2014) showed a significant higher GWP for excavation and processing ("material recovery scenario") than for transportation. It should be noted, though, that this study did not take soils of daily, intermediate and final cover into account. Processing proved to be the sub-process with the second largest energy consumption (14.6 GJ/d), comprising 11.5 GJ/d for off-site processing and 3.1 GJ/d for on-site processing. Using a wheeled loader required most of the energy for processing (\sim 40%), followed by the employment of different mechanic screens, and at processing plants air knives, conveyor belts and cross-belt magnets. The uncertainty of energy demand was directly related to the waste quantity uncertainty. Emissions from processing could generally be reduced using electrical equipment and renewable energy sources. The processing train consisted of less equipment in comparison to that used by Goeschl and Rudland (2007) or Wanka et al. (2017). Consequently, energy consumption and emissions might actually be higher. For instance, the sorting technology used by Laner et al. (2016) required 35 to 83% less diesel, but nearly 70 times more electricity. Excavation required 2.1 GJ/d diesel for the excavator, while the electricity consumption of groundwater pumps and surface water treatment at individual projects was 0.03 GJ/d. Uncertainty remained low (SD: 25%) since this sub-process proved to be comparable for all projects. In contrast, Trani et al. (2016) recorded 26% less fuel consumption for excavation at earthworks. Rehabilitation required in total 1.0 GJ/d comprising 0.5 GJ/d diesel to return the land to its previous state as well as 0.5 GJ/d for the gravel production. Using primary gravel at individual projects resulted in a higher uncertainty (SD: 136%), in particular for electricity consumption (SD: 300%). Electricity consumption can be reduced significantly by using recycled materials instead of primary gravel, while diesel consumption largely depended on transportation of gravel and topsoil. Ecological land restoration proved to be considerably less energy-consuming than rehabilitation. Site preparation comprised in total 0.9 GJ/d. The construction of roads and working facilities, in a few cases consisting of asphalt, consumed the most energy (0.8 GJ/d), while removal of topsoil required 0.1 GJ/d. Uncertainty turned out to be high (230%) due to the utilization of asphalt at individual projects. Electricity consumption can be reduced significantly using recycled materials instead of primary gravel, while diesel consumption largely depended on the construction of asphalt access roads. Disposal of small quantities of asphalt and soil required 0.2 GJ/d. Although energy consumption was low, uncertainty proved to be high (SD: 350%) since asphalt and soil were disposed of at only one project each. Finally, the uncertainty was most affected by the number of projects concerned and with regard to the transportation of processed waste by the network density of recycling and recovery facilities.

5.3.3 Transportation analyses

Since the absolute diesel consumption of different transportation tasks depended on the transportation distances and material quantities, a further analysis of the distances for each material type was carried out. The average transportation dis-



Figure 5.4: Energy consumption and emissions in CO_2 -eq. of sub-processes during the time period of 1 day.

tance was 122 km for the output (143 km for off-site processed waste), 20 km for the input and 2.6 km for the stored material. Transportation consisted of 828,781 km (equal to 20.7 times the circumference of the earth). Transportation to processing plants varied between 21 km and 263 km (on average 104 km), lower than assumed by Danthurebandara et al. (2015a, 150 km) and Frändegaard et al. (2013a, 300 ± 150 km), but higher than in the case study of Gusca et al. (2015, 12.6 km). Transportation distances ranged from 41 km for CDW, to more than 84 km for soils, 100 km for tyres, 133 km for wood, 134 km for plastics, 175 km for asphalt to 268 km for scrap (Fig. 5.5). The transportation distances for CDW remained low, since plants processing CDW are more common, sometimes resulting in transportation distances of only 20 km. However, transportation calculations usually stopped at the gate (reseller), due to later reuse. The transportation distances for CDW were in line with Doka (2007, 32.7 km), Trani et al. (2016, 25 km), Laner et al. (2016, 20-50 km), Vitale et al. (2017, 30 km), and considerably lower than assumed by Frändegaard et al. $(2013a, 400\pm200 \text{ km})$. In terms of soils, frequent exceedances of zinc, sulphate and TOC in soils required more efficient off-site processing, and resulted in considerably greater transportation distances of soils (84 km) than assumed in previous studies (Frändegaard et al., 2013a; Laner et al., 2016). With regard to plastics, the average distance to a waste incineration plant was 69 km - since there is a tight network of MSW incineration plants –, and 221 km to an RDF power plant, due to required additional sorting at specialized plants. Thus, avoiding relatively high costs of MSW incineration plants resulted in a tripling of transportation. Assumptions for transportation of RDF in previous studies varied significantly ranging from 50 to 100 km in Laner et al. (2016), to more than 150 km (Danthurebandara et al., 2015a), 400 ± 200 km (Frändegaard et al., 2013a) and 600 km (Jain et al., 2014). The high values of 400 km and 600 km might be related to the socio-geographical conditions in Sweden and the weaker network of waste incineration plants in the USA. However, if plastics could be recycled due to material purity and/or advanced technology, transportation might increase significantly. Compared to plastics, tyres could be separated at the landfill resulting in less transportation. Nevertheless, the distance might be greater since cement plants are scarce and the recovery of tyres was only recorded in two projects. Restrictions regarding bituminous contaminants resulted in long transportation distances for asphalt (on average 175 km). Transportation was 19 km when excavated asphalt was recycled to produce new asphalt, 324 km when simply disposed of and 335 km when reused as a sub-base for roads. In subsequent projects, asphalt was transported – using inland waterway transportation – up to 800 km for backfilling mines. The average transportation distance of scrap to a smelter was 268 km, including 68 km from the landfill to a merchant. In terms of iron, the subsequent transportation from the merchant was 147 km or 252 km to steel mills and in the case of aluminium 58 km to the wholesaler and 99 km to the smelter. The copper flows could not be reproduced; however the distance from the first merchant to the closest and most common used smelters ranged between 222 km and 277 km. In contrast, Frändegaard et al. (2013a) calculated 800 ± 400 km for ferrous and non-ferrous metals, while Laner et al. (2016) assumed 250 km for densely populated areas and 500 km for regions with lower population density. Lightweight materials, such as plastics, tyres and woods, might be compressed or shredded on-site to generate efficient payloads. Road transportation proved to be the most economical and flexible mode of transport. Khooban (2011) concluded that road transport is characterized by low investment, operation and maintenance costs, but observed that for distances greater than 350 km rail-road transportation becomes more economical than road transport, due to lower fuel costs. Transportation distances of materials for rehabilitation (29 km) and site preparation (15 km)remained low, due to their wide availability and negligible price differences between the suppliers. With regard to off-site processed soils, the average transportation



Figure 5.5: Average transport distances for different waste types, construction materials and to facilities.

distance of 84 km included 36 km transportation from a processing plant to a pit

or a landfill, while Doka (2007) estimated for CDW 15 km from a processing plant to an inert material landfill. Soils of class RC2 were transported on average 53 km, and of D0 127 km, D1 99 km, D2 88 km, respectively (Fig. 5.6). In contrast, Laner et al. (2016) assumed a distance of 10 to 20 km to an landfill for the recovery of residues, while Frändegaard et al. (2013a) transportation distance of fines at 10 ± 5 km involving on-site separation and reuse. Removing waste lifts separately during excavation allowed the creation of homogeneous piles, and these - when not containing MSW - could be reused off-site without prior processing. Soils of class RC2 were transported on average 28 km for backfilling quarries and pits. The landfill network is less tight, therefore transportation of D0 (78 km) and D1 (75 km) soils increased due to their limited use as construction material at certain landfills. Further processing of D1 soils was often not economical, due to small asphalt pieces and other particles. In addition, interim storage was needed in one case, due to daily acceptance limits at a landfill, thus increasing the transportation distance by 13%. The excavation of waste lifts one at a time, was particularly interesting for projects close to quarries and pits; otherwise, on-site processing enabled transportation distances to be reduced. In the last project, a combination of excavation by waste lifts and on-site processing enabled a reduction in transportation; however, 17 % of the soils still had to be forwarded to a processing plant. On-site processing resulted in transportation distances of 64 km (RC2), 93 km (D0), 91 km (D1) and 102 km (D2), while off-site processing increased transport to 90 km (RC2), 157 km (D0), 115 km (D1) and 74 km (D2). In the case of D0, additional processing at specialized plants led to contaminant reductions while increasing transportation distances; long transportation distances for on-site processed D2 soils resulted from the low number of D2 landfills. Apart from regulations, the decision between further processing - requiring transportation over longer distances - and higher landfill costs was economically-driven. The emissions of heavy metals during transportation were low compared to the reduction of these metals in soils at processing plants. While 100 km transportation produced 3.3 mg/t, processing resulted in 299 g/t less zinc in coarse-grained soils (>35 mm); a similar relationship existed for lead emissions from transportation (0.1 mg/t) vs reduction of lead in soils (135 g/t), chrome 0.06 mg/t vs 15 g/t, copper 0.04 mg/t vs 63 g/t, nickel 0.02 mg/t vs 16 g/t, cadmium 0.02 mg/t vs 1 g/t, mercury 0.01 mg/t vs 0.3 g/t, arsenic 0.0002 mg/t vs 5 g/t and PAHs 0.2 mg/t vs 3 g/t, respectively (Hölzle, 2018; Wernet et al., 2016).



Figure 5.6: Transportation distances of different soil classes by type of recovery.

5.3.4 Factors influencing material flows

Scanning the environment revealed the complexity and variety of factors influencing LFM in practice. Taking into account the concept of PEST analysis, identified factors were classified into the categories: economy, technology, organisation and institutions/laws (Fig. 5.7). An additional class constituted the landfill properties (materials and site characteristics), including internal factors such as composition, waste characteristics (i.e. chemical, biological and physical), material processability, and site properties (size, access, on-site processing option, topography, neighbourhood, option for on-site reuse of materials, environmental risks such as groundwater contamination). Economy (i.e. finance and market) might be classified into production factors (operation scale; interest rate; credit terms; costs of labour, waste recovery, processing, transportation, backfilling and disposal; availability of waste facilities and subcontractors; business strategies and connections), performance factors (prices, potential use of materials and diversity of recyclables) and marketrelated factors (development, trends, competitors, demand and acceptance for recyclables, costs of substituted raw materials, competition from recyclables of other waste streams). Enterprises made prudent investments in processing equipment to reduce the risk of low returns due to the lack of follow-up projects. Nunes et al. (2009) similarly determined a higher risk of failure for large-scale recycling centres of CDW. Mining of larger landfills requires an investment in appropriate processing facilities along with the continuity and volume of material flows. However, economic feasibility depends on market development driven by incentives and regulations. Technology-related factors primarily involved the effectiveness and efficiency of processing equipment in terms of throughput, separation of materials and substances (i.e. impurities and contaminants), susceptibility to clogging, required maintenance, energy consumption, mobility, versatility and adaptability to different waste compositions. The technological performance of WtE-plants and existing systems for heat and power generation are also of importance with regard to climate impact (Danthurebandara et al., 2015b; Laner et al., 2016), which was not, however, part of this study. With regard to organisation, coordination of enterprises and authorities along the chain of sub-processes, as well as flexibility and pragmatism of stakeholders proved to be of major importance (since preliminary investigations did not represent the heterogeneous landfill composition and the exact disposal area boundary). Further social issues included education level, enterprise experience and company "internal" issues. Network density, diversity, adaptability and capacity of waste facilities (i.e. processing plants, landfills, backfilling pits, WtE-plants) were crucial properties of the regional infrastructure. In terms of capacities, the consideration of simultaneous waste streams from recycling and other LFM projects was essential. The distance to potential LFM sites might be decisive for processing plant investments, particularly with regard to long transport times due to dense traffic in large cities. For instance, Nunes et al. (2009) identified a lack of space in urban areas for the installation of CDW processing plants in Brazil. In contrast, Chinda (2017) observed that infrastructure, compared to regulatory issues and open-mindedness, had the least influence on reverse logistics implementation of CDW in Germany and Thailand. Other factors influencing organisation included loads for return trips and e-commerce for recyclables and recovered materials. Lockrey et al. (2016) reported that a lack of coordination amongst CDW actors resulted in a transportation bottleneck and that enterprises diversified their services to avoid empty backhauls. The LFM projects showed a high degree of individuality, many stakeholders with different objectives, numerous interfaces and interdependencies, and exogenous influences. Coordination of stakeholders seemed to bear a high optimisation potential including the need to introduce coordination instruments. Flexibility turned out to be a key factor, particularly with regard to handling difficult and abnormal tasks, adapting to changing conditions and meeting the requirements of contracting entities and supervisory authorities. Political factors ("institutions/laws") comprised mechanisms and instruments (e.g. tax-based financing for the remediation of contaminated sites, incentives, subsidies, taxes, commitment, rules and prohibitions), institutional and regulatory issues, such as institutional preparedness, expenditures for administrative procedures and legal issues (e.g. acceptance exceptions of landfills for certain substance limit values). One project benefited from special governmental subsidies resulting in investments in processing equipment. Nunes et al. (2009) proposed reducing taxes and providing loans at lower interest rates to promote CDW recycling,

while Lockrey et al. (2016) reported a lack of financial support to transform the CDW recycling industry. Capacities and acceptance, and not only of institutions, also proved to be an issue. For instance, in one project, uncontaminated soils were not accepted at a landfill due to glass shards, while at another project types and plastics were mistakenly disposed of. Similarly, Johansson et al. (2017a) research on the readiness of institutions dealing with LFM observed deficiencies with regard to skills and institutional affiliation. In addition different contracting and tendering procedures influenced the working processes and waste processing. The results tended to be of a lower quality when the company carrying out the work was a service provider and did not become the owner of the excavated waste. In contrast to the holistic assessment of Hermann et al. (2016) some economic factors (e.g. competition, business strategies and connections), organisational factors (internal company issues, contracting and tendering procedures) and institutional factors (education level, capacifies and readiness of institutions) were identified. In terms of CDW recycling, previous studies (Chinda, 2017; Lockrey et al., 2016; Nunes et al., 2009) primarily determined the need for a change in stakeholder perception, open-mindedness toward the use of recyclables and the introduction of classification, standardisation and control for recycling construction materials. The development of multi-criteria decision-making applications requires a comprehensive analysis and an evaluation of factors influencing LFM. Apart from site specific conditions, the regional situation should be taken into account with regard to the individual purpose, since material flows might vary considerably in different settings (e.g. developing countries). Enhancing the reuse of mined waste – in particular soils - requires research into a variety of topics, such as processing technology, diversification of reuse options, appropriate mechanisms and instruments to increase acceptance and demand (e.g. the leading role of the public sector in reusing soils in construction projects), as well as organisational issues (e-commerce, coordination and timing to match supply and demand, designation of strategic locations in urban areas for interim storage and transaction).

5.4 Conclusions

Analysing material flows of eight mined landfills in a regional context enabled the identification of energy consumption and related emissions in sub-processes (i.e. site preparation, excavation, processing, disposal, rehabilitation and transportation) and revealed the importance of the regional waste infrastructure (e.g. processing plants, WtE-plants, backfilling facilities, landfills, etc.). The present study focused on the energy consumption of LFM operations and did not take benefits from material re-

Technology	Orga	nisation
Versatility and adaptability to different wastes	Capability, flexibility and pragmatism of stak	eholders
Design (size, set-up and shutdown time)	Network density and capacity of waste infra	structure
Throughput of processing equipment	Diversity and adaptability of waste infra	structure
Efficiency of substance segregation		
Effectiveness of separation La	ndfill Company internal and strateg	ic issues
Energy consumption Waste compos	E-commerce for the second s	naterials
Mobility and weight Contaminants	and processability Loads for re	urn trips
Economy Options for on-site	nd transportation distance Instituti	ins/laws
Scale of operation Landuse at ar	id around landfill Administrativ	e efforts
Interest rate, credit terms Environ	nental risks Principles (waste hierar	chy etc.)
Business strategies and connections	Contracting and tendering pr	ocedures
Development, trends and competition	Instruments (incentives, subsidi	s, taxes)
Costs for backfilling, disposal, WtE-plants	Regulations (waste, soil, water, er	lissions)
Demand and price for recyclables and energy	i je samihan bus samtuses saitisened	, , , , , , , , , , , , , , , , , , ,
Costs of processes (processing, transportation, etc.)	Capacities, acceptance and reaumess of in	RUOINNI
Diversity and acceptance of material reuse/recovery	Legal aspects (e.g. acceptance exceptions of	andfills)

Figure 5.7: Classified factors influencing landfill mining.

covery and recycling into account. In addition to material and energy flow analyses, an adapted PEST analysis – using the categories economy, technology, organisation, institutions/laws and landfill properties - enabled the identification of factors influencing operations and affecting material flows. Transportation to processing plants - followed by processing - required by far the most energy and, consequently, produced the greater part of emissions $(CO_2-eq.)$. Frequent contaminant exceedances in soils required more efficient off-site processing, and resulted in greater transportation distances of soils (84 km) than assumed in previous studies. In addition, the capacities of existing processing plants proved to be insufficient, although landfills were still comparatively small ($\leq 30,907$ tonnes). The energy balance and related emissions might significantly change by extending the system boundary, resulting in benefits from energy recovery by incinerating plastics, wood, tyres and landfill gas as well as energy savings from metal recycling. Since transportation of soils resulted in the largest portion of emissions, optimizing LFM benefits strongly depends on: a) the option of excavating waste lifts one at a time, b) processability of waste taking results from preliminary investigations and the processing technology into account (Hölzle, 2018, 2017), c) on-site processing option, d) on-site reuse option and/or nearby recovery facilities for soils, e) reuse possibilities (material characteristics and legal restrictions), f) markets (competition with raw materials and recyclables from current waste flows). In terms of nearby reuse options and markets, the coordination and timing with construction projects - particularly of roads and other earthworks - is crucial to match supply and demand. The landfills showed similarities in terms of composition, size and age; however, they were mined by several enterprises in different ways enabling me to analyse more differentiated influence factors. The PEST analysis revealed numerous factors influencing LFM, notably flexibility, pragmatism and coordination of stakeholders. The empirically determined influencing factors might represent a valuable asset for further studies involving system dynamics, stakeholder analyses or complex models. Potential fields of research constitute the analysis of influencing factors with regard to their relationships, interactions (feedback loops) and strengths.

6 Summary and general discussion

The main objective of LFM is the recovering recyclables from landfills. The ecological and economic performance depends on numerous factors along the process chain. Investigating the most important processes prospection, processing and recycling enabled the evaluation of LFM in practice taking into account current technological, economic, political, societal and environmental conditions as well as regulatory frameworks. This dissertation is based on analyses of substances and materials from eight mined landfills.

The investigated landfills consisted of soils (88%), CDW (5%), plastics (1.8%), scrap (0.3%), wood (0.2%), tyres (0.1%) and topsoil of the cap (5%; Fig. 6.1). Hazardous waste (<0.1%) included mainly batteries and asbestos and was not further considered due to the very small quantities. The soils were made up of 23% RC1 soil, 10% RC2, 21% D0, 19% D1 and 15% D2. Compared to most previous studies, the landfills investigated here showed a high proportion of soils similar to those of "set 3" in Laner et al. (2016), which reflects the composition of older landfills researched by Hogland et al. (2004) and Masi et al. (2014). With regard to the waste



Figure 6.1: Median and range of waste type quantities.

hierarchy, 5% of materials (topsoil) were reused, 0.3% recycled (0.3% metals, <0.1%

asphalt), 91.1% recovered (86.1% soil, 5% CDW, < 0.1% asphalt), 2.1% thermally recovered (1.8% plastics, 0.2% wood, 0.1% tyres) and 1.5% disposed of (1.5% soil, <0.1% asphalt; Fig. 6.2). Reuse and recovery of soils depended on contaminant concentrations, physical properties (consistency, grain size) and regional recovery options. RC1 and RC2 soils were usually used for backfilling pits and D0 to D2 soils as construction material at landfills. Reuse options for CDW included the construction of road sub-bases and noise barrier earth berms. At the beginning asphalt was recycled to produce new asphalt, but was later disposed of at landfills or used for backfilling mines due to high PAHs concentrations. Wood was incinerated in waste wood energy plants or co-incinerated in lignite power plants, plastics were incinerated either at MSW incinerators or RDF power plants, and tyres in cement plants.



Figure 6.2: Waste quantities and types with regard to the waste hierarchy.

6.1 Prospection - evaluating investigation methods

The samples from preliminary investigations were compared with the samples from excavation of three completely excavated landfills. In addition, the investigation methods – core drilling and grab crane – were evaluated in terms of reliability of prediction.

Comparing the preliminary investigation samples from core drilling and grab crane showed small to moderate substance concentration differences in the GMs (<50%) between preliminary investigation and excavation samples for lead, cadmium, copper, nickel, mercury, zinc, CN, PAHs and BaP (Fig. 6.3). For both methods large differences were recorded for PCBs. Using a grab crane also resulted in differences up to 50% for arsenic, chrome, fluoride, sulphate, pH and EC. Core drilling tended to involve more frequent overestimations – probably due to the absence of homogenization processes – while using a grab crane more often resulted

in underestimations. Consequently, hotspots strongly affected the analyses of core drilling, since landfill compositions are assumed to be very heterogeneous as a result of varying material types in every truckload. Samples taken with a grab crane might mix waste, and therefore resemble stockpile samples which are strongly homogenized as a result of excavation, transportation, piling and composite sampling.

Metals (except arsenic and nickel), PAHs, BaP, hydrocarbons and PCBs dispersed strongly, and the CVs ranged from 72% to 326%. In contrast, the CVs of pH (4%) and fluoride (14%) remained small for grab crane samples. Heavy metal dispersions were similar to those of Quaghebeur et al. (2013); Zhou et al. (2015) and Masi et al. (2014). However, high CVs did not co-occur with large differences in the means between preliminary investigation samples and excavation samples. For instance, heavy metals dispersed strongly, though differences in the means were small; the CV of cadmium was 242% whilst the difference in the means remained at 2.1%.

A MWW test was carried out to determine the significance of results. Core drilling and grab crane sampling proved sufficiently predictive of cadmium, copper, lead, mercury, BaP and PAHs concentrations ($\rho > 0.05$). Moreover, core drilling showed sufficient results for CN and copper (leaching test), and grab crane sampling for leaching tests of barium, EC, pH, sulphate and zinc. Consequently, the sample numbers of preliminary investigations (core drilling: 59; grab crane: 20) were sufficient for these substances. However, both methods failed in terms of PCBs and nickel, while the number of core drilling samples was not sufficient to predict arsenic, chrome, zinc and hydrocarbons nor for grab crane samples of fluoride, DOC and TOC.

6.2 Contaminant patterns in soils from LFM

Analysing contaminant concentrations of eight mined landfills enabled (a) the identification of contaminant patterns within and between landfills, (b) determination of indicator substances for contamination prediction, and (c) evaluation of limit values with regard to the effectiveness of managing substance flows.

Lead, chrome, copper, zinc, ammonium nitrogen, PAHs, PCBs and CN varied strongly (CV >75%) within landfills, while pH values hardly varied (<10%). Heavy metal concentrations generally varied more, while the variation of substances in leaching tests generally was lower, probably due to low substance concentrations. Variations of heavy metals were in line with Brandstätter et al. (2014), Masi et al. (2014) and Zhou et al. (2015), in addition to these, ammonium nitrogen, sulphate, pH and EC proved to be similar to those reported by Brandstätter et al. (2014).



Figure 6.3: Geometric mean differences between preliminary investigation samples and excavation samples.

25-

-0 %

-25-

3

*/10

-75-

-20

Grab crane

 100^{-1}

125

75-

50-

A low CV indicates homogeneous substance dispersions due to common and daily disposed waste which often was mixed before. In contrast, infrequent disposal of coarse-grained or bulky materials results in a heterogeneous dispersion of substances and a high CV.

Comparing substance concentration variations in and among landfills revealed three patterns: (a) substances with strong variations in and among landfills, (b) substances with strong variations among landfills where the average of the individual landfill variations resembled the variation among landfills, and (c) substances with low variations in and among landfills.

Heavy metals – particularly zinc, copper and mercury – correlated ($\rho > 0.7$) frequently with other heavy metals and few substances (Fig. 6.4). In addition, frequent correlations ($\rho > 0.7$) were recorded for ammonia nitrogen (with PCB, pH, EC, hydrocarbons, BaP, DOC) and TOC (with heavy metals and CN). Individual strong correlations were observed between EC and sulphate ($\rho 0.87$), as well as BaP and PAHs ($\rho 0.86$). In contrast, pH-values did mostly not correlate ($\rho \le 0.15$), and hydrocarbons, PAHs, chloride, DOC, biodegradability and BaP tended to be uncorrelated. Kaczala et al. (2017a) reported similar strong correlations between lead and zinc ($\rho 0.71$, present study $\rho 0.78$), TOC and zinc ($\rho 0.81$, present study $\rho 0.69$), while the correlation between TOC and DOC proved to be remarkably stronger ($\rho 0.65$) than in the present study ($\rho 0.02$). Moderate to strong correlations ($\rho > 0.5$) between EC and ammonium nitrogen as well as sulphate were in line with Brandstätter et al.'s (2014) observations.



Figure 6.4: Frequency of correlations ($\rho > 0.5$) and uncorrelatedness ($\rho < 0.15$).

TOC (75% of samples), zinc (40%), sulphate (34%), ammonium nitrogen (20%), copper (15%) and lead (10%) exceeded most frequently the RC2 or D0 limit value, while arsenic, barium, chrome, nickel, DOC, CN, biodegradability and pH always

remained below the limit value. Adelopo et al.'s (2018) investigation of heavy metals in landfill precursors similarly showed frequent limit value exceedances of lead, copper and zinc.

With regard to co-occurrences, often two, sometimes up to four, substances in one sample exceeded the limit values. TOC exceedances proved to frequently co-occur with exceedances of sulphate (50%), zinc (42%), ammonium nitrogen (32%) and copper (17%), whereas zinc exceedances coincided with exceedances of copper (41%), lead (29%) and cadmium (24%).

A combination of TOC, sulphate and pH would best indicate contamination and other substance exceedances. TOC and sulphate exceedances represented 100% of ammonium nitrogen, hydrocarbons, cadmium and zinc exceedances, and more than 90% of lead, copper and PAHs exceedances. Adding pH as an indicator element turned out to be necessary, since pH measurements were highly uncorrelated and did not co-occur with other substance exceedances. Zinc indicated other heavy metals efficiently; however, TOC also covered these. It should be noted though that the high indication rate of TOC might be also related to its high limit value exceedances rate.

The indication of chloride, fluoride, mercury, EC, BaP, BTEX and PCB proved to be difficult due to infrequent limit value exceedances. Brandstätter et al. (2014) also concluded that loss on ignition (which is frequently used instead of TOC) and pH are efficient indicator elements, but suggested selecting EC and chloride as well.

Assessing the effectiveness of limit values to manage substance flows showed that arsenic, cadmium, lead, zinc, chloride, sulphate, EC and DOC tended to accumulate in contaminated soils (class D1 and D2). In contrast, average concentrations of fluoride, copper, CN, biodegradability, hydrocarbons, BaP, PAHs and PCB were higher in low contaminated soils of class RC1 and RC2. Consequently, the limit value system turned out to manage efficiently flows of few substances since one to two substances exceeding the limit value often resulted in a higher classification.

6.3 Processing – assessing the effectiveness of dry screening

Four mechanical processing trains (MIL1, MIL2, TS1, TS2) were compared to assess the processing efficiency of contaminant redistribution in soils of different grain sizes. Objective was the production of low contaminated soils for reuse.

Extensive processing using sophisticated processing equipment at processing plant TS1 resulted in a large quantity of fines (<35/50 mm) and significantly increased the amount of low contaminated soils. In contrast, processing efforts at MIL1 and in particular at MIL2 proved to be less intensive, and large screen openings (50/70

mm) produced remarkable quantities of contaminated fine to medium-grained soils.

Heavy metals accumulated in the fines at all four processing plants, showing remarkable redistributions (>35%) for cadmium, lead, copper, mercury and zinc (Fig. 6.5). Strong redistributions (>50%) were recorded for arsenic, chrome and nickel at TS1. Accumulation of heavy metals in the fines were in line with Rousseaux et al.'s (1992) organic matter of fresh waste and Schachermayer et al.'s (1998) CDW.

With regard to chemical compounds, strong (>50%) accumulations of PAHs and TOC in the fines were recorded at all processing plants, as well as moderate (>20%) ones of biodegradability and naphthalene. In addition, concentration differences of >40% were observed for CN, hydrocarbons and PCBs at TS1 and TS2.

Differences between fines and coarse-grained soils proved to be less noticeable and heterogeneous in leaching tests (except sulphate). The differences in chloride, DOC, EC, fluoride and pH usually remained below 30%, probably as a result of leaching processes having occurred in the after-care phase over a period of 40 years.

In contrast to the common pattern of an accumulation in the fines, fluoride accumulated in the coarse-grained soils. The same applied to chloride at the TS1 and TS2 processing plants in line with the findings of Wanka et al. (2017). Pieces of wood treated with preservatives might resulted in higher fluoride concentrations in the coarse-grained soils, while magnesite screed pieces might resulted in higher chloride concentrations. DOC, EC and fluoride remained nearly unchanged, whereas pH measurements tended to be more basic for coarse-grained soils.

The subsequent MWW test verified significant ($\rho < 0.05$, 2-tailed) concentration differences in arsenic, cadmium, chrome, copper, lead, mercury, PAHs, sulphate and TOC at TS and MIL, while the significance level was not achieved for chloride, DOC and fluoride. In addition, significant differences of cyanides, hydrocarbons, PCBs, pH and nickel were recorded for the processed soils of the TS landfill, and of barium, EC and naphthalene for the soils MIL landfill.

Soil washing technology might redistribute soluble contaminants more efficiently, but will – in line with Kieckhäfer et al.'s (2017) observations – involve higher costs. Apart from techniques, different tendering procedures affected the processing results; screening efforts and results decreased when the company carrying out the work was just a processing service provider and did not get the owner of the waste.

6.4 Recycling – regional material flows and influencing factors

The preparation of an MFA enabled the evaluation of LFM in practice taking into account technological, economic, societal, political, legal and ecological conditions.



Figure 6.5: Distributions in fines (black) and coarse-grained soils (red) at TS1 processing plant.

The analysis comprised material flows and stocks, as well as the energy demand and related emissions of operations.

The waste export was 441 tonnes/day and material import 77 t/d, resulting in a stock decrease of 364 t/d including 86 t/d new stocks (Fig. 6.6). The export consisted mainly of processed waste and small quantities of decommissioned construction material, while the import comprised materials for rehabilitation and site preparation. Stocks included materials for rehabilitation and site preparation as well as disposed of soils and asphalt. Soils (409 t/d) made up the greater part of the exports, followed by CDW (17 t/d), plastics (11 t/d), scrap (1.7 t/d), wood (1.3 t/d), tyres (0.4 t/d) and asphalt (0.2 t/d).

Uncertainty of scrap (SD: 64%), plastics, wood and tyres (each SD: 110%) remained low due to similar proportions at all landfills, while the SD of soils ranged from 17% to 267%. Dos Muchangos et al. (2017) reported an uncertainty of 29% to 96% for MSW streams in Maputo.

LFM operations required in total 46 GJ/d diesel and 0.8 GJ/d electricity, producing 5.2 t/d carbon dioxide (CO₂). In terms of one tonne waste, this means 103 MJ diesel ($\simeq 2.4$ kg), 1.9 MJ electricity and 12 t of CO₂. Benefits from thermal recovery of plastics, wood and tyres resulted in approx. 175 MJ per tonne waste, and energy savings from scrap recycling were 56 MJ/t.

Transportation (28 GJ/d, $\simeq 58\%$) needed the most energy, whereas processing (15 GJ/d), excavation (2 GJ/d), rehabilitation and site preparation (1 GJ/d each) as well as disposal (0.3 GJ/d) required less energy. Transportation of waste to processing plants proved to be the main energy consuming transportation process, followed by transportation to recovery facilities and delivery of construction materials. Diesel consumption and related emissions were 23% more for off-site than for on-site processing due to transportation. In contrast, Jain et al. (2014) observed that excavation and processing produced six times more emissions than transportation, while Laner et al. (2016) reported that emissions from transportation were negligible compared to those from the background energy system and WtE-plants.

Comparing the emissions of heavy metals to the air from transportation and heavy metal reduction in soils due to processing showed an insignificant impact of transportation. For instance, transporting one tonne of waste 100 km resulted in 3.3 mg zinc emissions to the air, while processing enabled a reduction of 299 g zinc in one tonne coarse-grained soils. However, the emissions of hydrocarbons to water and soil from the production of diesel required to transport waste 100 km might be equal to the reduction of hydrocarbons in processed soils (86g/t) when the origin of oil is Russia (86.4 g, EU average: 17.4 g; Jungbluth (2007)).

The average transportation distance for waste was 122 km, where transportation distances ranged from 41 km for CDW to 84 km for soils, 100 km for tyres, 133 km for wood, 134 km for plastics, 175 km for asphalt and 268 km for scrap. Consequently, transportation distances of soils were considerably greater than assumed in previous studies ($10\pm$ 5 km, Frändegaard et al. (2013a); 10-50 km, Laner et al. (2016)).

The adapted PEST analysis consisted of the classification of factors into the categories economy, technology, organisation and institutions/ laws. Analysing the economic environment showed that investments were limited to some processing equipment due to the lack of follow-up projects. In line with Nunes et al.'s (2009) findings of large-scale recycling centre failures, discontinuous waste streams from LFM increase the risk of low returns. The effectiveness and efficiency of processing equipment was the most dominant technological factor, while organizational issues included primarily flexibility of stakeholders and infrastructure issues (capacities of processing plants, and transportation distances to processing plants and disposal facilities). Political factors involved on the one hand government subsidies and on the other hand capacities of institutions. Lockrey et al. (2016) and Nunes et al. (2009) similarly reported the need for financial support – such as reduced taxes and provision of loans at lower interest rates – to promote CDW recycling. Finally, the waste composition and the option for on-site processing and reuse turned out to be most decisive with regard to landfill properties.



6.4. Recycling – regional material flows

Figure 6.6: Material flows, energy consumption and related emissions during the period of 1 day.

7 Conclusion and outlook

The evaluation of LFM along the process chain -i.e. from prospection, to processing and recycling - was carried out on the basis of materials and substance flows. The landfills of the present research showed similarities in terms of waste composition, size, age and origin of waste. Consequently, results might not be applicable to large landfills containing more recent waste and/or in other countries.

Comparing substance concentrations in preliminary investigation samples (prospection) with those of excavation samples showed that heavy metals and some chemical compounds could be sufficiently predicted. By and large, grab crane sampling reflected results from excavation sampling more accurately, probably due to similarities between sampling using a grab crane and sampling of stockpiles. The dispersion of substances did not affect the reliability of prediction; consequently, the sample number might be calculated for substances individually disregarding dispersion. With regard to prospection, potential fields of further research include:

- using rectangular grab crane test pits instead of quadratic ones since a greater perimeter to area ratio might reduce susceptibility to heterogeneity
- comparing and assessing the application of different sampling patterns in accordance with ISO18400-104 (2018) for landfills

Analysing substance concentration in soils from LFM showed that heavy metals correlated strongly and frequently among themselves and to lesser extent with TOC, while pH hardly correlated with other substances. Sulphate, TOC and pH indicated sufficiently limit value exceedances, but this combination might be extended by EC and chloride analyses in line with the findings of Brandstätter et al. (2014). However, waste composition and age should be taken into account for the selection of indicator elements. The limit value system guided to some extent substance flows since only one to two substances were usually decisive for classification. Further research should focus on extending the analyses of contaminant patterns to an international level and evaluating the suitability and reliability of sulphate and zinc as indicator elements.

Using mechanical screens efficiently redistributed heavy metals, PAHs, TOC and sulphate to the fine-grained soils, while fluoride and chloride tended to accumulate in the coarse-grained soils. Mesh openings between 35mm and 50mm resulted in an optimal proportion of material flows and contaminant redistribution. However, the optimum mesh size and equipment also depends on the waste composition and homogeneity, due to the fact that screens separate materials by secondary properties (e.g. size, density). Further research emphasis should thus focus on:

- investigating the efficiency of recently developed windsifters and ballistic separators in processing mined waste
- \bullet studying the feasibility of using mesh openings ${<}10$ mm to produce low contaminated fines

Analysing regional material flows and calculating the energy consumption of LFM processes showed that transportation – particularly to processing plants – required most of the energy. In contrast to previous LFM models, transportation distances for soils proved to be significantly longer. Off-site processing allowed waste to be processed more efficiently in terms of contaminant redistribution, timing and limited space; however, the combination of excavation by waste lifts with on-site processing proved to be most efficient. Crucial for the energy demand and related emissions of LFM were: (a) the option of excavating waste lifts one at time, (b) processability of waste, (c) an on-site processing option, and (d) distance to locations for reuse or recovery of soils. Since the present investigation did not take energy benefits from metal recycling and thermal recovery of RDF into account, their incorporation might significantly change the energy balance and emission quantities. To improve material flow management, further research should focus on options to enhance local soil reuse and recovery.

Using an adapted PEST analysis revealed that flexibility, pragmatism and coordination of stakeholders proved to be key factors, since LFM projects were characterized by difficult and abnormal tasks, changing conditions and particular requirements of contracting entities and authorities. In line with the findings of Chinda (2017); Lockrey et al. (2016); Nunes et al. (2009) and Johansson et al. (2017a), enhancing LFM requires a change in stakeholder perception and development of their capacities. With regard to system analysis and the business environment, further research should analyse the relationships, interactions (feedback loops) and strengths of influencing factors.

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Appendix A

Landfill	Area	Excavated	Year of	Years of	Project
	(m^2)	waste (t)	closure	operation	costs (€)
Ansbach	4,185	20,425	1989	39	$1,\!400,\!000$
Lindau	$1,\!45$	4,197	1972	8	729,544
Main-Spessart	$6,\!13$	25,828	1975	17	$1,\!222,\!484$
Miltenberg A	5,82	$30,\!957$	1977	17	$3,\!151,\!403$
Miltenberg B	3,9	4,027	1977	5	$428,\!89$
Oberallgäu	2,8	9,311	1975	25	980,111
Straubing	3	7,048	1972	22	$734,\!957$
Traunstein	2,8	19,34	1975	11	2,583,474

Overview of the landfills.

Appendix B

Limit values of the RC guidelines and LF ordinance.

		RC gi	uidelines	LF	r ordinar	nce
	Unit	RC1	RC2	D0	D1	D2
As	mg/kg	50	150			
As	mg/l	0,04	0,06	$0,\!05$	0,2	0,2
Ba	mg/l			2	5	10
Pb	mg/kg	300	1000			
Cd	mg/kg	3	10			
Cr	mg/kg	200	600			
Cu	mg/kg	200	600			
Ni	mg/kg	200	600			
Hg	mg/kg	3	10			
Zn	mg/kg	500	1500			
PAHs	mg/kg	15	20	30	500	1000
C10-C40	mg/kg	500	1000	500	4000	8000
BaP	mg/kg	1	1			
BTEX	mg/kg	1*	1*	6	30	60
TOC [%]	% dry substance			1	1	3
DOC	mg/l			50	50	80
PCB	mg/kg	0,5	1	1	5	10
CN	mg/kg	30	100			
Biodeg.	$\mathrm{mg}~\mathrm{0_2/g}$			5	5	5
pН	dry substance	5-9*				
pН	leaching test	6-12	5.5-12	5.5-13	5.5-13	5.5-13
EC	$\mu { m S/cm}$	1000	1500			
Cl-	mg/l	20	30	80	1500	1500
SO_4	mg/l	100	150	100	2000	2000
F ⁻	mg/l			1	5	15
NH ₄ -N	mg/l			1*	4*	200*

^{*}currently not applied

Appendix C

Landfill	Area [m ²]	Excavated waste [t]	Disposal period
Ansbach	4185	20425	1960-1989
Lindau	1450	4197	1964-1972
$\operatorname{Main-Spessart}$	6130	25828	1958-1975
Miltenberg A	5820	30957	1960-1977
Miltenberg B	3900	4027	1972-1977
Oberallgäu	2800	9311	1950-1975
Straubing	3000	7048	1950-1972
Traunstein	2800	19340	1964-1975

Overview of the eight excavated landfills.

Appendix D

Energy consumption, related emissions of sub-processes and potentials. Datasets are based on own case studies or are extracted from the Ecoinvent (version 3.3) database.

		6	0.7	m^2	Restoration consisting of 0.3 m topsoil	Rehabilitation
).2	0.	26	2.9	÷	Process-specific burdens	
		2	3.2	-	Residual material landfill construction	Disposal
		4	0.6	-	Star screen	
		17	1.6	t	Wheeled loader	
).2	0.		0.04	+	Conveyor belt	
1.7	0.		0.1	-	Air knife	
).1	0.		0.03	-	Cross-belt magnet	
		6	0.8	-	Trommel sieve	
		6	0.9	-	Vibrating grizzly	Processing
		2	0.2	tkm	24 t truck	
		1	0.1	tkm	40 t truck	Transportation
.4	0.		0.06	-	Groundwater punping	
4	7		0.6	-	Surface water treatment	Excavation
						excavation
		4	0.5	m^3	Excavation*	Site preparation $/$
						rehabilitation
14		13	3.8	. +	Gravel production	Site preparation $/$
2		34	9.1	m^2	Road renovation	
12		172	45.7	m^2	Road construction	Site preparation
(\mathbf{II})	(M	(MJ)	(kg)			
tricity	Elect	Diesel	CO_2 -eq.	\mathbf{Unit}	Type of equipment or activity	Sub-process

*Topsoil density: 1.3 t/m³; waste density: 1.2 t/m³

Appendix E

Energy	generation	from	thermal	recovery	and	energy	savings	from	metal	recovery.
00	0			•		00	0			•

Material	Energy	Calorific	Energy	Source
	conversion	value or	output/	
	efficiency	\mathbf{energy}	saving	
	[%]	saving	[MJ/t]	
		[MJ/t]		
Plastics	41.3 %	18	152	Agency (2001)
Wood	27,00~%	13.5	10	Fritsche (2005),
				industrial reference
Tyres	80,00~%	28	13	Industrial reference
Ferrous		15.1	56	ifeu - Institut für
scrap				Energie- und
				Umweltforschung
				Heidelberg (2004)

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Glossary

BaP	benzo[a]pyrene
Biodeg.	biodegradability (four days)
BTEX	benzene, toluene, ethylbenzene, and xylenes
C10-C40	hydrocarbons (with 10 to 40 carbon atoms)
CDW	construction and demolition waste
\mathbf{CO}_2	carbon dioxide
\mathbf{CO}_2 -eq.	CO_2 -equivalents (Global Warming Potential: 100 years)
\mathbf{CN}	cyanides
\mathbf{CV}	coefficient of variation
D0	D0-limit value of the German landfill ordinance (DepV, 2009)
D1	D1-limit value of the German landfill ordinance (DepV, 2009)
D2	D2-limit value of the German landfill ordinance (DepV, 2009)
D3	D3-limit value of the German landfill ordinance (DepV, 2009)
D4	D4-limit value of the German landfill ordinance (DepV, 2009)
DOC	dissolved organic carbon
EC	electrical conductivity
ELFM	Enhanced Landfill Mining
EOX	extractable organic halogens
$\mathbf{G}\mathbf{M}$	weighted geometric mean
ISO	International Organization for Standardization
LF ordinance	German landfill ordinance
\mathbf{LFM}	Landfill Mining

LOD	limit of detection
MFA	material flow analysis
MSW	municipal solid waste
MWW	Mann-Whitney U test
PAHs	polycyclic aromatic hydrocarbons
PCB	polychlorinated biphenyl
PEST	method to analyse business environments taking into account political, economic, socio-cultural and technological frameworks
RC guidelines	technical guidelines for recycling soils (StMUV, 2011; LAGA, 2003)
RC1	RC1 limit value of the technical guidelines for recycling soils
RC2	RC2 limit value of the technical guidelines for recycling soils
RDF	refuse-derived fuel
SD	standard deviation
SFA	substance flow analysis
soils	soil-like materials
TOC	total organic carbon
VHH	volatile halogenated hydrocarbons
WPP	waste processing plant
WtE	waste-to-energy



. Kinik, Khiriki, Shiriz (Mirz)