Doctoral Thesis

Development and Assessment of a Novel Treatment Process for Municipal Solid Waste Incineration Fly Ashes

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Dissertation

Entwicklung und Bewertung eines neuartigen Behandlungsverfahrens für Flugasche aus der Müllverbrennung

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Abstract

About 3 % of the waste input into a waste incineration plant arise as fly ash, which constitutes a hazardous waste and has to be disposed of accordingly. As the current municipal solid waste incineration (MSWI) fly ash management options are all associated with various disadvantages, this thesis aims at developing a novel fly ash treatment process and comparing its environmental and economic performance with other options known in the state of the art.

The developed process comprises thermal treatment of MSWI fly ash together with combustible waste in existing MSWI plants. In a large-scale experiment, up to 300 kg moistened fly ash per Mg of combustible waste were treated in a rotary kiln hazardous waste incinerator for 102 h (Paper I). The inserted MSWI fly ash as well as bottom ash, fly ash and scrubber water from the rotary kiln were sampled and chemically analysed. The results from this analysis revealed that MSWI fly ash was almost completely transferred to the rotary kiln bottom ash, which, however, still constituted a non-hazardous waste.

The moistening of fly ash effectively prevented dust emissions during transport and storage. However, hydration reactions in the moistened material caused a temperature increase and formation of lumps of hardened fly ash in the waste bunker, which both impair the continuous and safe operation of the incinerator. As a possible solution to this problem, agglomeration was investigated as pretreatment prior to insertion in the waste bunker. About 400 kg of pelletised MSWI fly ash were produced and treated in a pilot-scale electrically heated rotary kiln at different temperatures and angles (Paper II). These experiments showed that MSWI fly ash is well suitable for agglomeration and the mechanical properties of the pellets produced allow their further processing. Thermal treatment at 450 °C for 10 min was already sufficient to generate a non-hazardous material, but treatment at higher temperatures leads to higher volatilisation of heavy metals.

The environmental impact of thermal treatment of MSWI fly ash together with combustible waste was determined by life cycle assessment (LCA) (Paper III). Seven scenarios for the treatment and disposal of MSWI fly ash, some of them comprising recycling processes, were established and modelled within an LCA framework. The LCA results showed that thermal treatment together with combustible waste has a lower environmental impact than stabilisation with cement. The scenario with the lowest impact is the recovery of metals by washing with acidic scrubber water.

As the results from LCA showed that resource recovery from MSWI fly ash has a low environmental impact, resource classification according to the United Nations Framework Classification for Resources (UNFC) was applied on MSWI fly ash (Paper IV). Thereby, an economic assessment of all scenarios comprising resource recovery was conducted. Monetary valuation of the LCA results was applied to subsequently combine the results from economic and environmental assessment and determine the resource potential from a public entity's macro perspective whereas the private investor's micro perspective includes only the results from the economic assessment. Cement production could be classified as commercial project from both perspectives, while metal recovery constitutes a commercial project from the macro perspective only.

Keywords: fly ash, combustion residue, waste incineration, thermal treatment, life cycle assessment, resource classification

Kurzfassung

Bei der Müllverbrennung fallen ca. 3 % des Abfallinputs als Flugasche an. Diese stellt einen gefährlichen Abfall dar und muss dementsprechend entsorgt werden. Da die derzeitigen Behandlungs- bzw. Entsorgungsoptionen für diese Flugaschen alle mit verschiedenen Nachteilen verbunden sind, ist es das Ziel dieser Dissertation ein neues Verfahren zu entwickeln und dessen Umweltauswirkungen und Kosten mit dem Stand der Technik zu vergleichen.

Das entwickelte Verfahren beinhaltet die thermische Behandlung von Müllverbrennungsflugasche zusammen mit brennbaren Abfällen in bereits bestehenden Müllverbrennungsanlagen (MVA). In einem Versuch im Großmaßstab wurden in einem Zeitraum von 102 h bis zu 300 kg befeuchtete MVA-Flugasche pro Mg brennbare Abfälle in einer MVA mit Drehrohrofen behandelt (Paper I). Die Ergebnisse der Analyse zeigten, dass die eingebrachte Flugasche nahezu vollständig in die Drehrohrofenschlacke eingebunden wurde, wobei diese Schlacke weiterhin einen nicht-gefährlichen Abfall darstellte.

Die Befeuchtung der Flugasche verhinderte erfolgreich die Emission von Staub während des Transports und der Lagerung. Allerdings verursachten Hydratationsreaktionen im befeuchteten Material einen Temperaturanstieg und die Bildung von Klumpen aus erhärteter Flugasche im Müllbunker, die beide den kontinuierlichen und sicheren Betrieb der Anlage beeinträchtigen. Als mögliche Lösung für dieses Problem wurde die Aufbauagglomeration als Vorbehandlung untersucht. Ungefähr 400 kg Flugaschepellets wurden hergestellt und in einem elektrisch beheizten Drehrohrofen im Pilotmaßstab bei verschiedenen Temperaturen und Winkel behandelt (Paper II). Diese Versuche zeigten, dass sich MVA-Flugasche sehr gut für die Pelletierung eignet und die mechanischen Eigenschaften der so hergestellten Pellets eine weitere Verarbeitung erlauben. Eine thermische Behandlung bei 450 °C für 10 min war bereits ausreichend um ein nicht-gefährliches Material zu erzeugen.

Die Umweltauswirkungen der thermischen Behandlung von MVA-Flugasche zusammen mit brennbaren Abfällen wurden mittels Ökobilanz bestimmt (Paper III). Insgesamt wurden sieben Szenarien für die Behandlung und Entsorgung von MVA-Flugasche erstellt und modelliert, wobei einige davon auch die Verwertung von Flugasche enthalten. Die Ergebnisse der Ökobilanz zeigten, dass die thermische Behandlung zusammen mit brennbaren Abfällen mit niedrigeren Umweltauswirkungen als die Stabilisierung mit Zement verbunden ist. Das Szenario mit den niedrigsten Umweltauswirkungen ist jedoch für beide Betrachtungszeiträume die Rückgewinnung von Metallen mittels Wäsche mit saurem Wäscherwasser.

Da die Ergebnisse der Ökobilanz zeigten, dass die Rückgewinnung von Ressourcen aus MVA-Flugasche eine Option mit geringen Umweltauswirkungen ist, wurde eine Ressourcenklassifizierung nach der United Nations Framework Classification for Resources (UNFC) für MVA-Flugasche durchgeführt (Paper IV). Dazu wurde eine wirtschaftliche Bewertung aller Szenarien, die eine Rohstoffrückgewinnung enthalten, durchgeführt. Neben Investitions- und Betriebskosten wurden auch Umweltauswirkungen monetarisiert, um eine Beurteilung aus einer Makro- und Mikroperspektive durchzuführen. Die Ergebnisse dieser Analysen zeigten, dass die Verwertung von MVA-Flugasche in der Zementherstellung sowohl aus Mikro- als auch aus Makroperspektive als kommerzielles Projekt aus beiden Perspektiven klassifiziert werden kann, während die Metallrückgewinnung nur aus der Makroperspektive ein kommerzielles Projekt darstellt.

Schlagwörter: Flugasche, Verbrennungsrückstände, Müllverbrennung, thermische Behandlung, Ökobilanz, Ressourcenklassifizierung

List of Appended Papers

I. Huber, F., Blasenbauer, D., Mallow, O., Lederer, J., Winter, F., Fellner, J., 2016. Thermal co-treatment of combustible hazardous waste and waste incineration fly ash in a rotary kiln. Waste Management 58, 181–190. https://doi.org/10.1016/j.wasman.2016.09.013

II. Huber, F., Herzel, H., Adam, C., Mallow, O., Blasenbauer, D., Fellner, J., 2018. Combined disc pelletisation and thermal treatment of MSWI fly ash. Waste Management 73, 381–391. https://doi.org/10.1016/j.wasman.2017.12.020

III. Huber, F., Laner, D., Fellner, J., 2018. Comparative life cycle assessment of MSWI fly ash treatment and disposal. Waste Management 73, 392–403. https://doi.org/10.1016/j.wasman.2017.06.004

IV. Huber, F., Fellner, J., 2018. Integration of life cycle assessment with monetary valuation for resource classification: The case of municipal solid waste incineration fly ash. Resources, Conservation and Recycling 139, 17-26. https://doi.org/10.1016/j.resconrec.2018.08.003

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- Purgar, A., Huber, F., Winter, F. (2016): Alternative Behandlungskonzepte für Flugaschen aus der Abfallverbrennung. Österreichische Ingenieur- und Architekten-Zeitschrift (ÖIAZ), 161 (2016), 75-81

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- Huber, F., Blasenbauer, D., Winter, F., Fellner, J. (2017): Thermische Behandlung von Müllverbrennungsflugasche zusammen mit brennbaren gefährlichen Abfällen in einem Drehrohrofen. In: ProcessNet Jahrestreffen Frankfurt II. 21.-23.03.2017 Frankfurt am Main, Germany.

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Contents

1. Introduction

Waste management has been a necessary activity for centuries, but the global amount of waste to be handled has constantly increased to about 3.5 million Mg (or metric tons) per day in 2010 with an even further rise expected (Hoornweg et al., 2013). This considerable amount can be explained by a pullulating world population and accelerating material consumption rates triggered by economic growth.

The main goal of waste management is the protection of humans and the environment from the hazards potentially caused by waste. Another crucial goal of waste management is conservation of resources. In order to fulfil these two goals of waste management, wastes have to be either recycled or transformed to residues that can be landfilled with little or no aftercare (AWG, 2002; Brunner and Rechberger, 2015). Although recycling is preferable in many cases, it is not always possible, e.g. because materials are used as inseparably composites or are not separated at source. An important waste flow is mixed municipal waste (hereinafter referred to as municipal solid waste), which contains all items of municipal waste that are not separated at source and collected separately.

In most regions of the world, the most common waste disposal options are landfills and open dumps (Hoornweg and Bhada-Tata, 2012; Statista, 2018). However, in high income countries, municipal solid waste incineration (MSWI) has become an essential part of the waste management system (Hoornweg and Bhada-Tata, 2012), as it allows avoiding many negative environmental impacts associated with the landfilling or dumping of untreated waste. MSWI fulfils all of the following objectives and therefore constitutes a key element for sustainable waste management of municipal solid waste:

- hygienisation,
- volume reduction,
- environmental protection,
- mineralisation and immobilisation of hazardous substances,
- resource conservation,
- affordable costs and public acceptance. (Brunner and Rechberger, 2015)

Municipal solid waste is in most MSWI plants treated in grate furnaces with boilers for electricity and/or district heat production. Further possible incinerations technologies comprise fluidised bed combustion and rotary kilns (Stubenvoll et al., 2002).

Solid matter falling down from the grate is generally referred to as bottom ash and accounts for about 20 to 30 % of the waste input (Morf et al., 2000). As this bottom ash usually represents a non-hazardous waste, it can be disposed of on landfills without further treatment or utilised as construction material (Verbinnen et al., 2017).

MSWI plants are equipped with air pollution control (APC) systems in order to prevent emissions of Hg, acidic gases and particulate matter. In Europe, 45 % of the MSWI plants are operated with wet scrubbers, while 55 % are operated with semi-dry and dry APC systems (Fellner et al., 2015b, 2015a). A comparison of different APC systems is given by Beylot et al. (2017). The type of APC system determines the amount and composition of solid residues generated in MSWI (apart from bottom ash).

According to Chandler et al. (1997) fly ash is the "particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material". Depending on the place of generation, fly ash can be classified either as boiler ash or filter ash, making up about 2 % and 1 % of the waste input, respectively (Morf et al., 2000). Some MSWI plants allow separate collection and removal of these different ashes while most of them remove boiler and filter ash together. Accordingly, the term fly ash is used for a mixture of boiler ash and filter ash in this thesis. In addition to fly ash, about 1 % of the input mass is transferred to the wastewater in case a wet APC system is installed. Dissolved solids are precipitated from this wastewater and the solid residue generated thereby is referred to as filter cake. In MSWI plants with semi-dry or dry APC systems, caustic materials (e.g. $Ca(OH)_2$ or $NaHCO_3$) are injected into the flue gas stream upstream of the filter. As a result, a mixture of filter ash, reaction products and excessive neutralising agents is generated and referred to as "semi-dry or dry process residue" (Chandler et al., 1997) and no separate discharge of filter ash is possible in MSWI plants equipped with semi-dry or dry APC system.

The mass of waste treated in MSWI plants in Europe amounted to about 100 million Mg in 2015 (CEWEP, 2017). The application of a transfer coefficient (i.e. the mass flow of a specific output divided by the mass flow of the inputs) of 0.03 yields a MSWI fly ash mass of about 3 million Mg, which has to be managed in some way. Contrary to MSWI bottom ash, fly ash from MSWI is usually classified as hazardous waste due to its high content and leachability of heavy metals and soluble salts (Funari et al., 2017; Jiao et al., 2016; Li et al., 2016; Purgar et al., 2016; Ye et al., 2016; Zhan et al., 2016). The safe disposal of this residue is therefore challenging and costly. Consequently, this thesis focuses on the potential for improved MSWI fly ash treatment, disposal and utilisation processes. This MSWI fly ash does not only contain valuable metals, but also minerals like CaO, SiO_2 and Al_2O_3 and also easily soluble chloride salts (mainly NaCl and KCl).

There is a multitude of different utilisation and disposal options for MSWI fly ash – some of them comprising fly ash treatment. However, most of these studies focus on the experimental, chemical or technological description but do not include any environmental or economic assessment.

MSWI fly ash can be stored in underground deposits (e.g. former salt mines) hundreds or even thousands of meters below the ground surface. However, suitable deposits are not available in all countries and therefore this option is often associated with long transport distances or not feasible at all. Although the disposal in underground deposits is already expensive, further cost increases are expected (Rottlaender, 2013).

Stabilisation and solidification with subsequent disposal at non-hazardous waste landfills above ground is another common practice for MSWI fly ash disposal. Cement and other hydraulic binders are most commonly applied, but Billen et al. (2014) propose a stabilisation process for MSWI fly ash without addition of any material other than water. In addition, stabilisation methods using chemicals like sodium sulphide or phosphoric acid were also investigated (Quina et al., 2010; Sukandar et al., 2009; Youcai et al., 2002). The process of cement stabilisation decreases the mobility of the contaminants in the treated material through encapsulation as a consequence of the reduced surface area and low permeability (Quina et al., 2008). Quina et al. (2008) pointed out that the physical properties of stabilised MSWI fly ashes decreases over time due to leaching of soluble salts. A further disadvantage associated with this practice is the disposal of large amounts of cement on landfills. This means that on the one hand about 1 Mg cement per Mg of MSWI fly ash has to be produced

(Quina, 2016), which is associated with high energy consumption and gaseous emissions (Huntzinger and Eatmon, 2009) and on the other hand the landfill volume required for the disposal of fly ash is increased.

MSWI fly ash from many northern European countries is shipped to Norway and neutralised with waste acid from the local titanium industry prior to disposal (Fruergaard et al., 2010; Stenberg, 2016). This option also implements long distance transport and is not available outside of Norway.

Another group of treatment processes separates soluble MSWI fly ash constituents by extraction. Karlfeldt Fedje et al. (2010) compared the effect of different extracting agents (water, acids, salt solutions). While water is sufficient to extract most of the Na, K, Ca and Cl from the ash, the highest removal rates for heavy metals like Zn and Pb were achieved by using mineral acids, in particular hydrochloric acid, in high concentrations. MSWI plants with wet APC systems already generate hydrochloric acid solution in the first scrubber stage. In the so-called FLUWA process, Hg is removed from this scrubber water by selective ion exchange and the scrubber water is subsequently used for the extraction of MSWI fly ash. Zn can be precipitated from the solution and the metal hydroxide sludge generated thereby can be utilised in the Waelz process to produce secondary Zn (Bühler and Schlumberger, 2010). An alternative to the FLUWA process is the FLUREC process. Thereby, metallic Zn powder is added as reducing agent to the solution after extraction in order to precipitate Pb, Cu and Cd in metallic form. Zn can be separated from the solution by reactive extraction and the application of electrowinning yields high purity metallic Zn (Schlumberger, 2010). Both processes are applied in full scale in Switzerland and until 2021 all MSWI fly ash generated in Switzerland has to be treated and metals have to be recovered (Quina et al., 2018). In the last years, there have been further efforts to improve these metal extraction processes, e.g. by addition of H_2O_2 (Weibel et al., 2017) or addition of NaCl (Weibel et al., 2018). Tang et al. (2018) propose a hydrometallurgical process for the extraction of Cu and Zn from MSWI fly ash using hydrochloric acid and organic compounds in a single step. However, this process has up to date only been conducted in laboratory scale. Further processes for separation of heavy metals from MSWI fly ash in aqueous solutions comprise hydrothermal treatment (Zhang and Itoh, 2006), bioleaching (Funari et al., 2017) and electrodialysis (Jensen et al., 2015). Fellner et al. (2015b) demonstrated that metal recovery is currently only economically viable for MSWI fly ash with very high metal contents (e.g. separately collected filter ashes).

Thermal treatment of MSWI fly ash aims at producing an environmentally stable material mainly by partly separating inorganic compounds with a low boiling point (e.g. Hg, Pb and Cd compounds) and destroying organic compounds (e.g. polychlorinated dibenzodioxins and dibenzofurans). Thermal treatment methods can be divided into the three main groups sintering, melting and vitrification processes (Chandler et al., 1997). Sintering treatment takes place below the melting point of the major MSWI fly ash constituents (about 700-1,200 °C). The decrease in porosity achieved by sintering decreases the leachability of harmful components and volatile metals are partly vaporised (Lindberg et al., 2015). In melting processes the major constituents of MSWI fly ash are transformed to a liquid state, which requires higher temperatures (about 1,100-1,500 °C). Vitrification is carried out at the same temperatures as melting but in vitrification vitrifying additives (e.g. SiO_2, MgO or $NaCO_3$) are used to entrap potentially harmful MSWI fly ash constituents in a glass matrix and to thereby minimise leaching (Lindberg et al., 2015). The volatilisation of heavy metals in thermal treatment processes can be increased by addition of chlorine carriers (Astrup et al., 2011; Jakob et al., 1996;

Nowak et al., 2010, 2013). All thermal treatment processes for MSWI fly ash mentioned above require large amounts of energy (Ecke et al., 2000).

The bulk of the material generated by these thermal treatment processes can be disposed of as non-hazardous waste, while in the APC system of the MSWI fly ash treatment plant a smaller portion arises as secondary fly ash enriched in volatile heavy metals. However, there are also attempts to utilise thermally treated MSWI fly ash. Quina et al. (2014) investigated the production of lightweight aggregates from expanding clay with addition of up to 10 % semi-dry process residue. Their results show that this utilisation option is feasible and that washing of semi-dry process residue prior to utilisation enhances the properties of lightweight aggregates.

The most common utilisation option of MSWI fly ash is replacing raw meal in cement clinker production where the fly ash is exposed to temperatures of about 1,450 °C. However, according to Quina et al. (2018) some influence on cement quality and stack emissions may occur. This is especially relevant, as most cement plants are not equipped with a sophisticated APC system e.g. for the removal of Hg from the flue gas. As the high chlorine content in MSWI fly ash may cause technical problems in the cement kiln, water-washing pretreatment is suggested prior to insertion of the ash into the cement kiln (Quina et al., 2018; Saikia et al., 2007; Tang et al., 2014). Lederer et al. (2017) showed that utilisation of MSWI fly ash in clinker production can severely increase the heavy metal contents of the cement produced, which might impede the further recycling of the concrete made from this cement.

As all of the available MSWI fly ash management options mentioned above are associated with various disadvantages, this thesis is directed at developing innovative processes for the treatment of MSWI fly ash. The application of these processes should comply with the goals of waste management, most importantly protection of humans and the environment and conservation of resources, and also be economically viable. Conservation of resources can be achieved either by providing secondary raw materials (e.g. Zn from MSWI fly ash) or by saving primary raw materials (e.g. cement for the stabilisation of MSWI fly ash). Furthermore, the degree of compliance of novel fly ash treatment processes with these waste management goals is to be assessed within this thesis. Thereto, LCA and resource classification were used to assess the compliance of the newly developed process with the abovementioned goals and to allow the comparison between different MSWI fly ash management options.

2. Objectives and structure of the thesis

A major objective of the Christian Doppler Laboratory for Anthropogenic Resources, within the frame of which this thesis was conducted, was to find novel options for the management, treatment and disposal of MSWI fly ash that comply with the abovementioned goals of waste management.

An approach taken in this thesis is thermal treatment of MSWI fly ash together with combustible waste in already existing MSWI plants. The idea behind this approach is to decrease the large investment and operation costs associated with thermal treatment of MSWI fly ash in a separate plant as described in the literature. Furthermore, this approach seems especially suitable for the City of Vienna, because there are three MSWI plants and one hazardous waste incinerator, which can be used for MSWI fly ash treatment, in Vienna. Paper I aims at answering the following questions:

- How do the inserted MSWI fly ash and its components partition among the hazardous waste incineration residues during the co-treatment with combustible waste in a rotary kiln hazardous waste incinerator?
- Does the co-treatment of MSWI fly ash influence the quality of solid residues in general and bottom ash in particular, generated by the rotary kiln?
- In how far is the impact on the quality of bottom ash observed explainable by simply mixing the inserted MSWI fly ash and rotary kiln bottom ash?

A caveat of the process suggested in Paper I is the insertion of MSWI fly ash into the furnace. Sheer moistening of MSWI fly ash causes hydration and setting reactions in the mixture resulting in a significant temperature increase in the moist ash and the formation of lumps of hardened fly ash. As both effects impair continuous operation of the incinerator, the feasibility of the process is compromised. As a possible solution to the abovementioned problems, a pretreatment process comprising the pelletisation of fly ash was developed within this thesis and is presented in Paper II. The particular research questions addressed in Paper II are:

- Which operation conditions are necessary during pelletisation in order to achieve pellets best suitable for further processing?
- What is the chemical composition of thermally treated MSWI fly ash pellets (total and leachable element contents)?
- How do the MSWI fly ash pellets and their constituents partition between treated MSWI fly ash pellets, dust carried by the flue gas during thermal treatment (secondary fly ash) and flue gas?
- How do the operational conditions of the thermal treatment influence the mass and composition of treated MSWI fly ash pellets and secondary fly ash?

Paper I and Paper II give detailed information about the technical performance of thermal co-treatment of MSWI fly ash with combustible waste with and without pelletisation as pretreatment. However, the economic and ecological performance of this novel process was still unclear. Consequently, life cycle assessment (LCA) was applied to determine the environmental impact of combined thermal treatment of MSWI fly ash and combustible waste and, for comparison, also the environmental impact of other important MSWI fly ash management options (underground deposit, cement stabilisation, FLUREC process, thermal treatment in a separate furnace) and presented in Paper III. This paper aims at answering the following questions:

- What is the environmental impact of thermal co-treatment of MSWI fly ash and combustible waste?

- How does this process compare with other relevant MWSI disposal options with regard to the environmental impact?
- What is the effect of different timeframes considered for the LCA on the total environmental impact and on the ranking of different MSWI fly ash treatment and disposal options?

The experiments conducted within this thesis and described in Paper I and Paper II also showed that the secondary fly ash generated during thermal treatment of MSWI fly ash is enriched in heavy metals like Zn, Pb or Cd. The extraction and subsequent recycling of these heavy metals from MSWI fly ash is technically feasible (Schlumberger, 2010; Tang et al., 2018), but the Zn content is in most cases too low for the recovery to be economic (Fellner et al., 2015b). However, the enrichment of Zn and other metals in secondary fly ash could be a promising aid in establishing facilities for metal recovery from MSWI fly ash. The minerals contained in MSWI fly ash constitute an interesting secondary raw material for cement industry (Guo et al., 2017; Hartmann et al., 2015; Huang et al., 2016; Lederer et al., 2017; Saikia et al., 2007) and chloride salts recovered from fly ash could be utilised as road de-icer (Stena Metall, n.d.; Stenberg, 2016). Consequently, there is a competition between three different MSWI fly ash utilisation options (metal recovery, cement production and salt recovery) and also a competition between secondary and primary raw materials. Paper IV aims at the application of resource classification from a private investor's micro and a public entity's macro perspective to compare the three abovementioned recycling options and to make them comparable with the exploitation of primary raw materials. The particular research questions addressed in Paper IV are:

- What is the environmental impact of the utilisation of MSWI fly ash in cement production, metal recovery and de-icing salt production?
- What is the net present value (NPV) of these recycling options?
- How can environmental impacts in different impact categories be "transformed" to monetary values?
- How can MSWI fly ash be classified in the United Nations Framework Classification for Resources (UNFC) classification scheme from a micro and macro perspective considering the environmental impacts of recycling?

3. Materials and Methods

3.1 Experiments on thermal treatment of MSWI fly ash

3.1.1 MSWI fly ash sampling

The fly ash used for all experiments was collected at a MSWI with a grate furnace. The APC system at this plant comprises an activated coke injector, fabric filters, a two-stage scrubber and a selective catalytic reduction device. Several subsamples were taken and subjected to chemical analysis. The fly ash from this plant generally exceeds the legal limits given in brackets for non-hazardous waste landfills in Austria for the parameters Hg total content (20 mg/kg), total dissolved solids in the leachate (60,000 mg/kg) and Pb in the leachate (10 mg/kg).

3.1.2 Large-scale experiments on thermal co-treatment of MSWI fly ash together with combustible waste

MSWI fly ash was mixed with water in a mass ratio of approximately 3:1, which lead to a water content of 0.23 kg/kg moistened ash. This mixture was inserted into the waste bunker of a hazardous waste incinerator comprising two rotary kilns 12 m in length and 4.5 m in outer diameter and a wet APC system.

During the experimental period of 102 h both rotary kilns were fed with combustible hazardous waste, whereby the amount and composition of the waste feed was as far as possible identical. In addition, moistened fly ash was inserted into rotary kiln 1 at a ratio of 169 kg (130 kg dry matter) per Mg combustible hazardous waste in phase A (54 h) and at a ratio of 300 kg (231 kg dry matter) per Mg combustible hazardous waste in phase B (48 h).

Bottom ash samples were taken every 4 h and samples from rotary kiln fly ash and scrubber water from scrubber stage 1 were taken every 12 h for both kilns. The input (MSWI fly ash and hazardous waste) into both rotary kilns and the sampling times are shown in Figure 1.

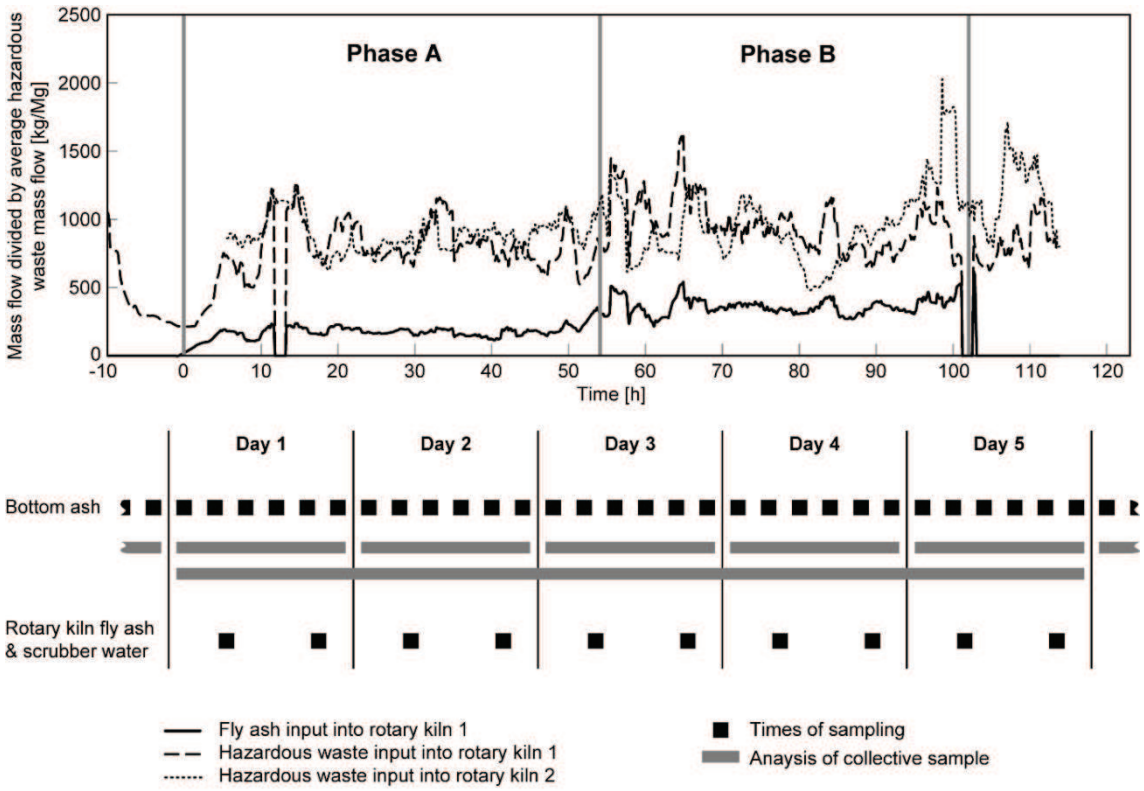


Figure 1. Input into the rotary kilns during the experiment (phase A: 0-54 h, phase B: 54-102 h) and information about the times of sampling and analyses.

3.1.3 Pilot-scale experiments on combined disc pelletisation and thermal treatment of MSWI fly ash

Due to the formation of lumps of hardened MSWI fly ash and temperature increase in the waste bunker in the experiments described in 3.1.2, pelletisation of MSWI fly ash was performed as a pretreatment prior to thermal treatment in a pilot-scale pelletising disc with a diameter of 1.2 m. Upstream of the pelletising disc was a ploughshare mixer where water was added to the ash. At a mass flow of 300 kg/h a batch of about 400 kg of MSWI fly ash pellets with a water content of 0.15 kg/kg and an average diameter of about 8 mm was produced, as these operation conditions yielded the most suitable results. A process scheme of the pelletisation process is shown in Figure 2. The pellets were filled into bulk bags and stored for several months until the experiments on thermal treatment were conducted.

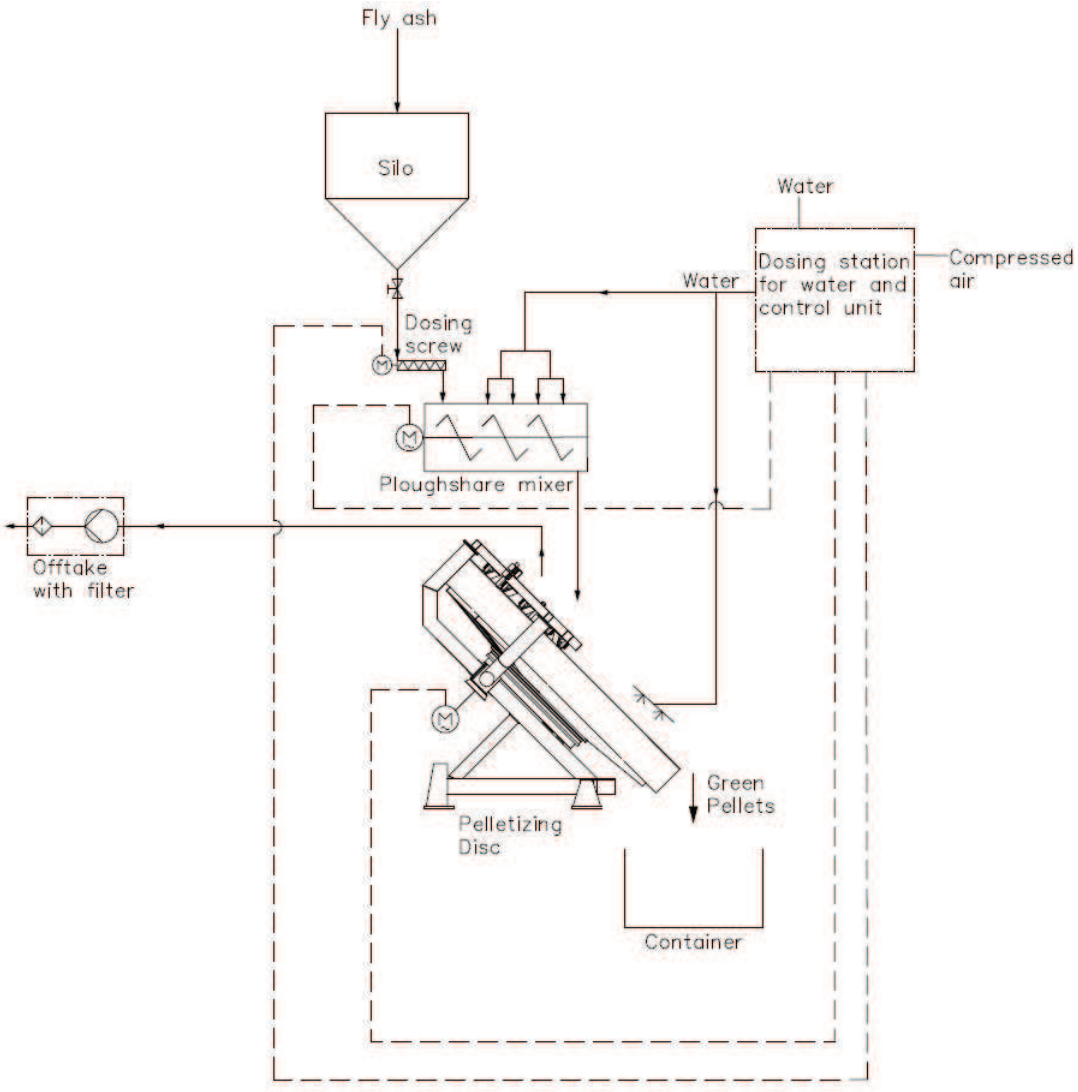


Figure 2. Process scheme of the pelletisation process for MSWI fly ash.

Thermal treatment was conducted in an electrically heated pilot-scale rotary kiln 2 m in length and 0.2 m in inner diameter at different temperatures (450 °C, 550 °C, 650 °C, 750 °C, 850 °C, 950 °C and 1050 °C) and angles (2°, 3°, 6°). The higher the angle of the kiln the lower was the residence time. Contrary to the experiments described in 3.1.2, no other materials than MSWI fly ash were inserted into the kiln. This had the advantage that the effect of thermal treatment on MSWI fly ash pellets without mixing with other materials could be investigated. Pressurised air was injected into the rotary kiln with a relative flow of about 1 m^3 (V_n) per kg of inserted fly ash and the flue gas from the kiln was sucked into a scrubber prior to release in the atmosphere. Pelletised MSWI fly ash was continually transported into the kiln by a conveyer screw and thermally treated pellets were collected in a metal bucket placed on a balance used to determine the mass flow of pellets into the bucket. Thermally treated fly ash pellets were sampled when the outflow of pellets was constant and therefore identical to the inflow of pellets. In this phase of constant outflow, a depth filtration device with glass wool as filter media was mounted between the rotary kiln and the scrubber in order to collect secondary fly ash. This secondary fly ash consists of dust generated by abrasion of the MSWI fly ash pellets and volatile MSWI fly ash compounds that condensed on this dust or on the filter media. The total mass of thermally treated MSWI fly ash pellets and secondary fly ash was determined and samples of the materials were subjected to chemical analysis.

3.1.4 Chemical and physical analysis of MSWI fly ash and products from MSWI fly ash treatment

Total contents as well as leachate contents of MSWI fly ash and products from MSWI fly ash treatment were determined. For the determination of total contents, solid samples were digested in aqua regia according to EN 13657 (2002). Bottom ash samples had to be milled prior to digestion. Different to the other samples analysed, the secondary fly ash samples on glass wool were heated in aqua regia until reaching reflux temperature and kept there for about 2 h prior to filtration (instead of following EN 13657 (2002)). Additionally, two blanks (glass wool without secondary fly ash) were digested.

Leachates were prepared using a liquid-to-solid ratio of 10 L/kg according to EN 12457-4 (2002).

All digested samples, all leachates as well as all liquid samples were analysed by inductively coupled plasma optical emission spectroscopy (ICP-OES) according to EN 11885 (2009).

To determine the total Cl content, samples were milled to a grain size of 250 µm and subsequently analysed by X-ray fluorescence spectroscopy (XRF).

In addition to analysis of inorganic parameters, persistent organic pollutants (POP) were analysed in the original fly ash used for all experiments and in thermally treated fly ash pellets. In particular, polychlorinated dibenzodioxines and furans (PCDD/F), dioxine-like polychlorinated biphenyls (DL-PCB), non-dioxine-like polychlorinated biphenyls (NDL-PCB) and hexachlorobenzene (HCB) were measured. All samples were milled to a grain size of 250 µm and subsequently digested in HCl solution with a concentration of 1 mol/L in an ultrasonic bath for 10 min. PCDD/F, DL-PCB and NDL-PCB were analysed by gas chromatography high resolution mass spectroscopy (GC-HRMS) according to CEN/TS 16190 (2012). HCB was also analysed by GC-HRMS.

The particle size distribution of MSWI fly ash pellets was determined by sieving analysis and the compressive strength of the pellets was measured by crushing 30 pellets (near-mesh size pellets of an 8 mm sieve) on a pressure sensor plate mounted on a hydraulic pellet press. A drop test similar to Gul et al. (2015) was performed by dropping 10 green pellets from a height of 1 m onto a hard surface until they broke.

3.1.5 Determination of transfer coefficients

Based on the recorded mass and volume flows and their respective composition a material flow analysis according to Brunner and Rechberger (2004) aiming at identifying and illustrating the distribution of inserted MSWI fly ash and its elemental components to the different outputs of the rotary kilns (bottom ash/thermally treated fly ash pellets, secondary fly ash and scrubber water) by so-called transfer coefficients (mass transferred to a particular output flow of a process divided by input mass into the process). Transfer coefficients were determined on goods and substances level for the experiments described in 3.1.2 and 3.1.3.

For the experiments on thermal co-treatment of MSWI fly ash and combustible hazardous waste (Paper I), transfer coefficients for rotary kiln 2 (used as reference) were determined based on the recorded masses of waste input, bottom ash and rotary kiln fly ash assuming that the composition of waste fed into both rotary kilns was identical. These transfer coefficients were applied to the mass of combustible waste input into rotary kiln 1 (used for MSWI fly ash addition) to calculate the mass of

bottom ash generated from combustible waste in rotary kiln 1. The mass of bottom ash generated from inserted MSWI fly ash was finally calculated according to equation 1.

$$m_{FABA} = m_{BA1} - \frac{m_{BA2}}{m_{HW2}} \cdot m_{HW1} \quad (1)$$

m_{FABA} ....mass of inserted fly ash transferred to bottom ash of rotary kiln 1

$m_{BA1,2}$....mass of bottom ash generated in rotary kiln 1 and 2, respectively

$m_{HW1,2}$...mass of hazardous waste inserted into rotary kiln 1 and 2, respectively

To determine the transfer coefficients of elements inserted via MSWI fly ash in kiln 1, in a first step the elemental composition of the hazardous waste combusted was calculated using data of kiln 2 (mass and volume of input and output flows as well as information about their respective elemental composition). Information about the amount and composition of hazardous waste and moistened MSWI fly ash treated in kiln 1 were combined to determine the overall input of different elements and afterwards compared to their overall output flows (via bottom ash, rotary kiln fly ash and scrubber water) observed. This comparison allowed tracing the flows of elements inserted via moistened MSWI fly ash at kiln 1.

Exemplary, equation 2 used to calculate the amount of an element transferred from inserted MSWI fly ash to bottom ash of rotary kiln 1 is given below.

$$m_{E,FABA} = w_{E,BA1} \cdot m_{BA1} - \frac{w_{E,BA2} \cdot m_{BA2}}{m_{HW2}} \cdot m_{HW1} \quad (2)$$

$m_{E,FABA}$ ..mass of element *E* transferred from inserted fly ash to bottom ash of rotary kiln 1

$m_{BA1,2}$....mass of bottom ash generated in rotary kiln 1 and 2, respectively

$m_{HW1,2}$...mass of solid hazardous waste inserted into rotary kiln 1 and 2, respectively

$w_{E,BA1,2}$..mass fraction of element E in bottom ash of rotary kiln 1 and 2, respectively

This calculation was applied to all outputs streams (bottom ash, rotary kiln fly ash, scrubber water).

For the experiments on thermal treatment of pelletised MSWI fly ash (Paper II), no other material was inserted into the kiln. Consequently, all outputs of the kiln were derived from the inserted fly ash pellets. However, the exact amount of pelletised fly ash transported into the rotary kiln by the conveyer screw could hardly be measured. Therefore, the input mass was calculated from the output mass of thermally treated pellets, secondary fly ash and the concentration of Ba and Ca in all input and output flows according to equations 3-5. This calculation is possible because Ba and Ca are not volatised during treatment (as confirmed in preliminary tests) and therefore the mass of Ba and Ca in the thermally treated pellets and in the secondary fly ash is equal the input mass of Ba and Ca.

$$m_I = \frac{c_P \cdot m_P + c_S \cdot m_S}{c_0} \quad (3)$$

$$m_G = m_I - m_P - m_S \quad (4)$$

$$m_{i,G} = m_I \cdot c_{E,0} - m_P \cdot c_{E,P} - m_S \cdot c_{E,S} \quad (5)$$

m_I.....mass of MSWI fly ash pellet input [kg]

m_P.....mass of thermally treated MSWI fly ash pellets [kg]

m_S.....mass of secondary fly ash [kg]

m_G.....mass of MSWI fly ash that is transferred to the flue gas and not captured by the filter [kg]

c_0.....concentration of tracer element in pelletised fly ash before thermal treatment [kg/kg]

c_P.....concentration of tracer element in pelletised fly ash after thermal treatment [kg/kg]

c_S.....concentration of tracer element in secondary fly ash [kg/kg]

$m_{E,G}$.....mass of element *E* that is transferred to the flue gas and not captured in the filter device [kg]

$c_{E,0}$.....concentration of element *E* in pelletised fly ash before thermal treatment [kg/kg]

$c_{E,P}$.....concentration of element *E* in pelletised fly ash after thermal treatment [kg/kg]

$c_{E,S}$.....concentration of element *E* in secondary fly ash [kg/kg]

3.2 Environmental and economic assessment of MSWI fly ash treatment and disposal

3.2.1 Scenarios for MSWI fly ash treatment and disposal and system definition

The goal of the assessment applied in this thesis is the comparison of different scenarios for treatment and disposal of MSWI fly ash based on the environmental impact and economic parameters. In particular, the newly developed process for thermal co-treatment of MSWI fly ash together with combustible waste as described in 3.1 should be compared with other processes.

The functional unit used in Paper III was 1 Mg of MSWI fly ash and the functional unit used in Paper IV was 360,000 Mg of MSWI fly ash, corresponding to the amount of fly ash generated in the city of Vienna during an assumed project time of 20 a (combusted waste mass of about 700,000 Mg/a). However, all LCA results shown in the main body of this thesis refer to the functional unit of 1 Mg MSWI fly ash to facilitate the comparison between all scenarios. In contrast, the results of the economic assessment refer to a project time of 20 a and a total MSWI fly ash mass of 360,000 Mg in Paper IV and the main body of this thesis. The composition of MSWI fly ash was taken from the analysis of the sample described in 3.1.1.

The scenarios modelled in this thesis are described below. Furthermore, Table 1 gives a short overview of all scenarios.

In **scenario 1** (labelled scenario A in Paper III) MSWI fly ash is transported to an underground deposit. As these deposits are only available at certain locations, transport over large distances is necessary.

In **scenario 2** MSWI fly ash is stabilised with cement and subsequently disposed of at a non-hazardous waste landfill. Two alternatives were modelled. The use of blast furnace slag cement CEM III/A in a ratio of 1 Mg cement per Mg of fly ash was assumed in **scenario 2a** (labelled scenario B in Paper III and used as reference system for MSWI fly ash disposal in Paper IV), while 0.3 Mg of cement per Mg of fly ash are used in **scenario 2b** (labelled scenario B* in Paper III).

In **scenario 3** the so-called FLUREC process (acidic fly ash extraction with integrated zinc recovery) is applied as described in detail by Schlumberger (2010). After removal of Hg from the acidic scrubber water, this water is used to extract the metals Zn, Pb, Cu and Cd from the fly ash to be treated. Subsequently, metallic zinc is added to the extract as a reducing agent, whereby a mixture of metallic Cd, Cu and Pb is precipitated. Zn is separated from the liquid by reactive extraction with a selective chelating agent in a liquid-liquid extraction step and metallic Zn with a purity of >99.99 % is produced by electrolysis (average Austrian electricity mix). This Zn can be sold and the mixture of Cd, Cu and Pb can be separated at a non-ferrous metal smelter and subsequently recycled. The solid residue from the acidic washing process complies with the legal limits for non-hazardous waste landfills. In **scenario 3a** (labelled scenario C in Paper III and scenario A in paper IV) the waste water is directly discarded into the sewage system after extraction. In **scenario 3b** (labelled scenario C in Paper IV) chloride salts are recovered from the waste water by evaporation.

Scenario 4 describes the thermal treatment of MSWI fly ash in a furnace exclusively dedicated to this purpose. The main output flow of this process is a glassy slag that can be disposed of at a non-hazardous waste landfill. However, about 10 % of the inserted MSWI fly ash emerge as secondary fly ash, which is enriched in heavy metals and therefore still constitutes hazardous waste (Sakai and Hiraoka, 2000; Yang et al., 2013). It is assumed that this secondary fly ash is disposed of at an underground deposit. In **scenario 4a** (labelled scenario D in Paper III) hard coal is used as a fuel in the furnace necessary for thermal treatment, while in **scenario 4b** (labelled scenario D* in Paper III) natural gas is used as fuel.

In **scenario 5** MSWI fly ash is thermally co-treated together with combustible waste. It was shown by the experiments presented within this thesis that this treatment does not impair bottom ash quality and therefore this residue can be disposed of at a non-hazardous waste landfill. The secondary fly ash was assumed to be disposed of at an underground deposit. Based on the results of the experiments presented within this thesis, it was further assumed that about 5 % of the inserted MSWI fly ash mass is transferred to the secondary fly ash while 95 % are transferred to the bottom ash. In **scenario 5a** (labelled scenario E in Paper III) MSWI fly ash is treated together with combustible hazardous waste in a rotary kiln as described in 3.1.2, while in **scenario 5b** (labelled scenario E* in Paper III) MSWI fly ash is treated together with MSW in a grate furnace.

In **scenario 6** MSWI fly ash is washed with water and subsequently used to substitute primary raw materials in cement clinker production. In accordance with Lederer et al. (2017), it was assumed that about 1.1 % of clinker raw materials are replaced by MSWI fly ash. In **scenario 6a** (labelled scenario B in Paper IV) the waste water from fly ash washing is directly discarded into the sewage system, while in **scenario 6b** (labelled scenario D in Paper IV) chloride salts are recovered from the waste water by evaporation.

Table 1. Overview of all scenarios modelled in this thesis.

Scenario 1	Underground deposit
Scenario 2a	Stabilisation with 1 Mg cement/Mg fly ash
Scenario 2b	Stabilisation with 0.3 Mg cement/Mg fly ash
Scenario 3a	Metal recovery
Scenario 3b	Metal and salt recovery
Scenario 4a	Thermal treatment in coal-fired furnace
Scenario 4b	Thermal treatment in natural gas-fired furnace
Scenario 5a	Thermal co-treatment with combustible waste in a rotary kiln incinerator
Scenario 5b	Thermal co-treatment with combustible waste in a grate furnace incinerator
Scenario 6a	Utilisation of fly ash in cement production
Scenario 6b	Utilisation of fly ash in cement production with salt recovery

The inventory for the scenarios includes all environmental burdens and economic costs, respectively, from the transportation of MSWI fly ash from the MSWI plant to the respective treatment or disposal facilities, the MSWI fly ash treatment and the disposal of residues. The production of secondary metals, the saving of quicklime in the FLUREC process and the decreased demand of primary raw materials in cement production are considered as avoided environmental impacts and avoided costs, respectively. In order to calculate these avoided burdens and costs, reference systems for cement production and scrubber water neutralisation were established and the environmental impacts and costs were subtracted in the respective fly ash disposal scenarios. The inventory does not include upstream burdens associated with MSWI or the production and use of goods prior to their disposal in an MSWI plant (e.g. the extraction and refining of crude oil and the subsequent production of plastic packaging). Hence, the zero burden assumption (Chang and Pires, 2015) was used.

For acidic washing of MSWI fly ash, transfer coefficients for Cd, Cu, Pb and Zn reported by Bühler and Schlumberger (2010) were applied. For all other considered heavy metals in acidic washing as part of the FLUREC process and for all heavy metals in neutral washing as pretreatment for the utilisation in the cement kiln, the transfer coefficients were determined by laboratory experiments using hydrochloric acid solution (c = 1 mol/L) as extracting agent and a liquid-to-solid ratio of 5 as assumed by Fellner et al. (2015b). A detailed description of the experimental setup for the determination of the transfer coefficients can be found in Blasenbauer et al. (2015). The transfer coefficients were used to calculate the amount and composition of washed fly ash and de-icing salt.

3.2.2 Environmental assessment

The environmental impact of all scenarios described in 3.2.1 was determined by LCA. The life cycle inventory data was sourced from ecoinvent database V3.2 (2015). The life cycle impact assessment was conducted using the ReCiPe model (Hierarchist perspective) (Goedkoop et al., 2009). The impact in all midpoint impact categories (agricultural land transformation, climate change, fossil depletion, freshwater ecotoxicity, freshwater eutrophication, human toxicity, ionising radiation, marine ecotoxicity, marine eutrophication, metal depletion, natural land transformation, ozone depletion, particulate matter formation, photochemical oxidant formation, terrestrial acidification, terrestrial ecotoxity, urban land occupation, water depletion) and endpoint impact categories (human health, ecosystem quality, resources, total score) was calculated.

Emissions from landfills caused by the deposition of material in the respective scenarios were considered. For this purpose the mass of each element transferred to the leachate was calculated

using the transfer coefficients given by Doka (2003a). The leachate is transferred to the a wastewater treatment plant and partitioned to sewage sludge and cleaned waste water according to the transfer coefficients given by Doka (2003b). It was assumed that the sewage sludge was incinerated and the resulting ash disposed of at an above-ground landfill. Further assumptions are that the leachate collection system and the liner of the landfill are intact and in operation for 100 years. After this time, leachate is released into the soil (and groundwater) below the landfill.

Stabilisation with cement and thermal treatment of MSWI fly ash decrease the surface available for leaching. Furthermore, thermal treatment as well as the FLUREC process change the chemical composition of the material to be disposed of. Literature data (Blasenbauer et al., 2015; Bühler and Schlumberger, 2010; Fellner et al., 2015b; Guohua et al., 2012; Li et al., 2015; Liu et al., 2010; Mostbauer et al., 1994; TÜV Rheinland, 2012; Wang et al., 2009, 2008, 2015; Yang et al., 2009; Zhao et al., 2010; Zupanič et al., 2012) was used to calculate the composition of the material actually deposited and account for decreased leachability of pollutants compared to the original fly ash. For the modelling of cement production, data from Lederer et al. (2017) were used to calculate transfer coefficients from MSWI fly ash to clinker and flue gas. A detailed description of the models applied is given in Paper III and Paper IV.

Two different time frames were taken into account, which is relevant regarding landfill emissions. The time frame for the assessment of the short-term impact was 100 a. In addition, an infinite timeframe was chosen for the evaluation of the long-term impact of MSWI fly ash disposal.

For the scenarios comprising thermal treatment (4-6) also atmospheric emissions caused by this treatment were accounted for. The APC system necessary for MSWI fly ash treatment in scenarios 4 and 5 was modelled based on literature data (Morf et al., 2000; Stubenvoll et al., 2002; Wang et al., 2008; Wien Energie GmbH, 2016; Wiesenberger and Kircher, 2001). For scenario 6, it was assumed that the cement plant utilising MSWI fly ash is not equipped with any APC system apart from a filtering device. Details on the modelling of atmospheric emissions can be found in Paper III and Paper IV.

3.2.3 Economic assessment

As the results from environmental impact assessment have shown that resource recovery, especially recovery of metals, from MSWI fly ash is preferable over disposal from an environmental view and because this approach also constitutes a step towards a more circular economy, only scenarios comprising at least one utilisation process (3 and 6) were considered in the economic assessment. As the overall objective of the Christian Doppler Laboratory for Anthropogenic Resources, within the framework of which this thesis was conducted, is to develop a methodological framework to identify, characterise and evaluate anthropogenic resources (Christian Doppler Laboratory for Anthropogenic Resources, 2012), resource classification according to UNFC (UNECE, 2010) was applied.

This resource classification framework uses the parameters (axes) socio-economic viability, project feasibility and geological knowledge for the distinction between commercial projects, potentially commercial projects, non-commercial projects, exploration projects and other combinations.

In order to examine the socio-economic viability of generating commodities from MSWI fly ash, a discounted cash flow (DCF) analysis was performed by calculating the NPV before taxes for each scenario based on material and energy flows from the inventory. The economic evaluation of every

scenario was conducted from a private investor's micro view, which is only focussed on revenues and internal costs, and from a public entity's macro view, which includes also external costs.

DCF analysis is a simple tool that is widely applied in the evaluation of mining projects (Baurens, 2010) and was also used by Winterstetter et al. (2015, 2016). It is used to determine whether a certain waste flow can be classified as resource or reserve, whereby a positive NPV indicates a reserve. The NPV was calculated according to equation 6 (Campbell and Brown, 2003).

$$NPV = -c_0 + \frac{c_1}{1+r} + \frac{c_2}{(1+r)^2} + \cdots + \frac{c_T}{(1+r)^T} \quad (6)$$

c_0.....investment cost [EUR]

c_1-c_T.....discounted cash flow for year *1* to *T* [EUR]

T.....time [a]

r.....discount rate [-]

In order to determine the external costs, which are relevant for the macro perspective, the results from the LCA in a 100 years timeframe were used and these environmental impacts were converted into monetary values. Scenario 2a was used as a reference system for the treatment and disposal for MSWI fly ash and, therefore, the environmental impacts and costs of scenario 2a was subtracted from the environmental impact and costs of the scenarios investigated in the economic assessment, respectively.

In order to take into account the economic costs of climate change, the greenhouse gas emissions of every scenario were multiplied by the social cost of carbon (i.e. the welfare loss associated with an additional kg of CO_2 emitted) of 0.17 EUR/kg CO_2 eq. (IPCC, 2014). The damage to human health is expressed in disease adjusted life years (DALY) and the damage to ecosystem quality is expressed in species years in the ReCiPe method. The monetary valuation of the damage to human health and ecosystem quality was conducted using LIME2 according to Itsubo et al. (2012). As the damage to ecosystems is expressed in species instead of species years in LIME2, the respective weighting factor was divided by the inverse of the background extinction rate as estimated by De Vos et al. (2015). The outcome of the endpoint impact category resource consumption is in ReCiPe already displayed in monetary values and expresses the marginal change in efforts to extract future resources (Goedkoop et al., 2009). No other midpoint impact categories apart from climate change were considered, as no data on their monetary valuation was available.

The monetised environmental impact (i.e. the external costs) was added to the sum of DCFs in order to calculate the NPV of the macro view for every scenario. Accordingly, in the following text sum of DCFs refers to the sum of all discounted cash flows without the monetised environmental impact and NPV refers to the sum of DCFs including the monetised environmental impact. This differentiation is only relevant for the macro perspective, as the micro perspective does not account for the environmental impact.

To reflect the high economic risk of an investment into a recycling project, which highly depends on commodity market prices, a high discount rate of 12 % was chosen for the micro perspective, while a lower discount rate of 3 % was used for the macro perspective (Baurens, 2010; Winterstetter et al., 2015). Besides expressing time preference, another justification for discounting is the assumption

that due to economic growth people will be richer in the future (Padilla, 2002). However, unlike the economy, ecosystem quality or resource availability do not grow, but rather decrease. Accordingly, no discounting was applied to monetised environmental impacts in the present study.

3.2.4 Uncertainty analysis

The uncertainty associated with the result of LCA models can be divided into parameter uncertainty, scenario uncertainty and model uncertainty (Huijbregts et al., 2003). The influence of parameter variation on the result and the effect of scenario alterations were investigated, while uncertainties related to the use of alternative modelling concepts (e.g. how to model landfill emissions) are not within the scope of this thesis.

Parameter uncertainty of the output variables was determined by propagating the uncertainty of the input parameters in a Monte Carlo simulation (MCS) with 100,000 runs. A discernibility analysis was conducted for each scenario by calculation the difference between the LCA results and NPV of the single scenarios in all 100,000 iterations as described by Clavreul et al. (2012) in order to determine in how many cases a certain scenario outperforms the other ones.

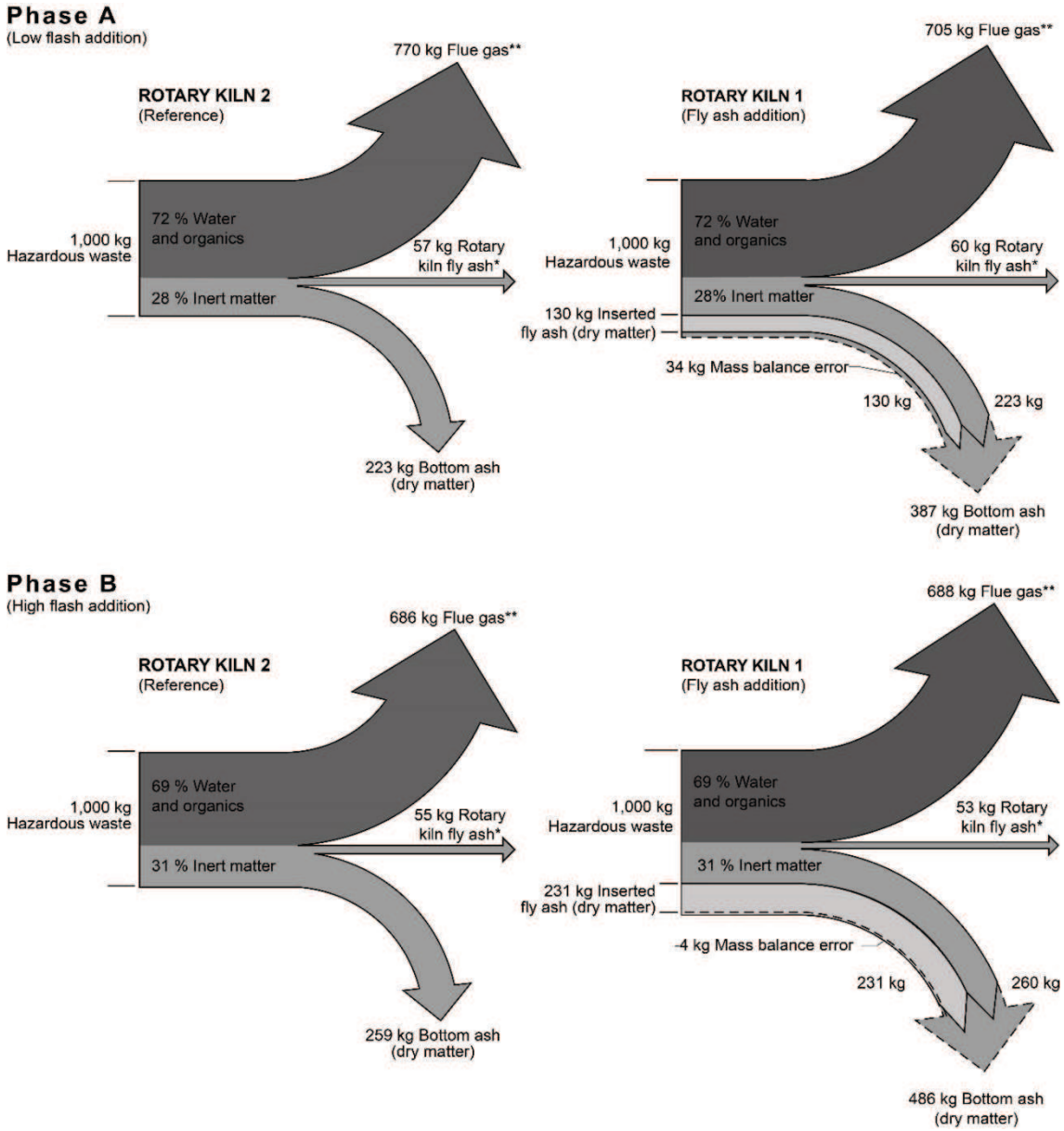
In order to assess scenario uncertainty, alternative scenario choices were modelled and the environmental impact in all midpoint and endpoint impact categories and in case of utilisation scenarios also the NPV was calculated for these alternative scenarios (c.f. 3.2.1).

4. Results and discussion

4.1 Experiments on thermal treatment of MSWI fly ash

4.1.1 Mass flows observed in the experiments

The mass flows observed in the large-scale experiment on thermal treatment of MSWI fly ash together with combustible hazardous waste are shown in Figure 3. This material flows suggest that more than 90 % of the inserted MSWI fly ash are bound into the bottom ash of the rotary kiln, as the amount of bottom ash generated at rotary kiln 1 (fly ash addition) increased accordingly (left side of Figure 3).



*The mass of rotary kiln fly ash could not be determined separately for the two kilns. As discussed above, almost all inserted wet fly ash dry matter was transferred to the bottom ash. Therefore the total amount of rotary kiln fly ash was equally assigned to both rotary kilns.

**The mass of organics and water transferred to the flue gas was calculated as total input mass less bottom ash and rotary kiln fly ash mass generated

Figure 3. Material flows of hazardous waste and inserted fly ash referred on 1 Mg hazardous waste used in kiln 1.

As no other materials apart from pelletised MSWI fly ash were inserted in the pilot-scale experiments, the transfer of solid matter could be studied in more detail. The ratio of secondary fly ash and thermally treated MSWI fly ash pellets is given in Figure 4. The data from these experiments suggests that, depending on the temperature, only between 0.05 % and 1.5 % of the treated fly ash pellets were arising as secondary fly ash.

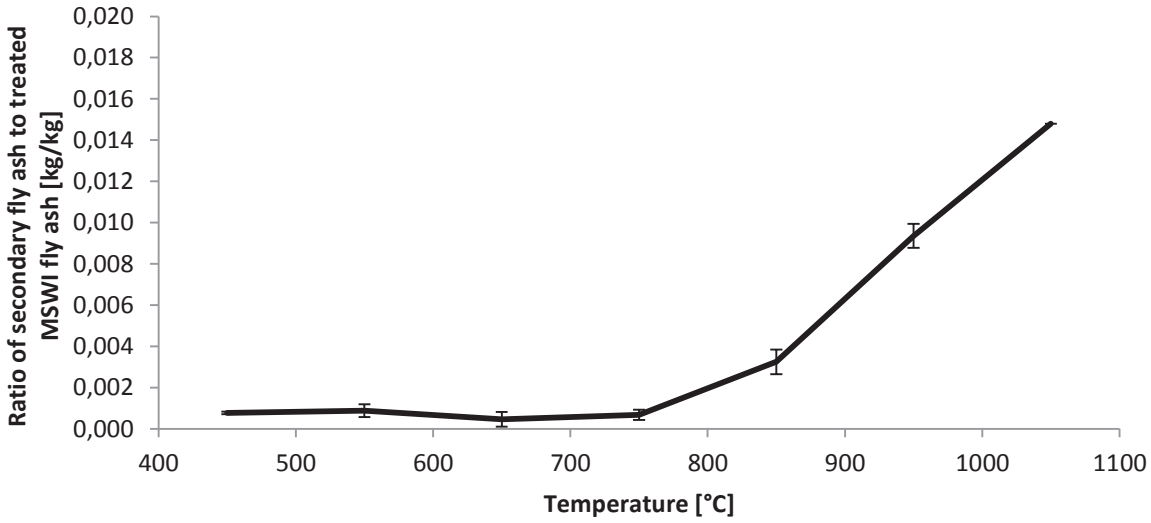


Figure 4. Ratio of secondary fly ash to MSWI fly ash pellets treated at different temperatures. The error bars show the standard deviation.

4.1.2 Quality of materials generated by MSWI fly ash treatment

The chemical analysis of bottom ash and rotary kiln fly ash of both kilns used in the large-scale experiments reveal that the Cl content in bottom ash of kiln 1 increased after the addition of moistened MSWI fly ash by about 75 % (+3,700 mg/kg). Apart from that, no changes in the composition of the bottom ash, significantly deviating from long-time mean values, were observed, still allowing the disposal of bottom ashes from the hazardous waste incinerator at non-hazardous waste landfills with regard to total element contents. The rotary kiln fly ash from kiln 1 showed concentrations of Cd, K and Pb by 81 mg/kg (+54 %), 19,500 mg/kg (+57 %) and 1,660 mg/kg (+22 %), respectively, higher compared to kiln 2. However, this increase is within the long-time fluctuations.

No significant difference in the leachability of the investigated elements in the bottom ash of both kilns could be observed. Also in comparison to the long-term averages no significant impact by co-treatment of moistened MSWI fly ash at kiln 1 on the bottom ash quality in terms of leachability of substances was found. In contrast, the leachate of a mixture of rotary kiln bottom ash and moistened fly ash without thermal treatment does not comply with the limits for non-hazardous waste landfills because of the high content of total dissolved solids. Although the other parameters analysed in this mixture complied with the limit values for disposal as non-hazardous waste, the leachability of Cr, Mo and Zn was significantly higher compared to the bottom ash of kiln 1 (co-treatment of moistened fly ash). Despite the increasing rate of MSWI fly ash treatment during the experimental period of 5 days, no significant changes in the sample composition were observed during the course of the experiment.

The MSWI fly ash pellets produced for the pilot-scale experiments showed an average compressive strength of 131 N (standard deviation: 36 N) and an average drop number of the pellets of 10 (standard deviation 3), which ensures their mechanical suitability for further treatment. More than 60 % of the pellets had a diameter between 7.1 and 9 mm.

The thermally treated MSWI fly ash pellets showed an increased content of Cr, Co and Ni because parts of the kiln material were transferred to the sample as a result of deterioration. As illustrated in Figure 5, the total content of certain elements decreases with higher treatment temperature. As no correlation between residence time and composition could be observed, the mean values of the experiments at each temperature were used. If the measurement result was below the limit of quantification, half of the limit of quantification was used for the calculation of mean values. The total content of Hg was decreased to below 2 mg/kg already by treatment at 450 °C, while temperatures of at least 950 °C are necessary to volatise more than 50 % of the Cd and Pb mass originally contained in pelletised MSWI fly ash. At 1050 °C the total content of Cd and Pb in thermally treated MSWI fly ash pellets is even as low as 23 mg/kg and 44 mg/kg, respectively.

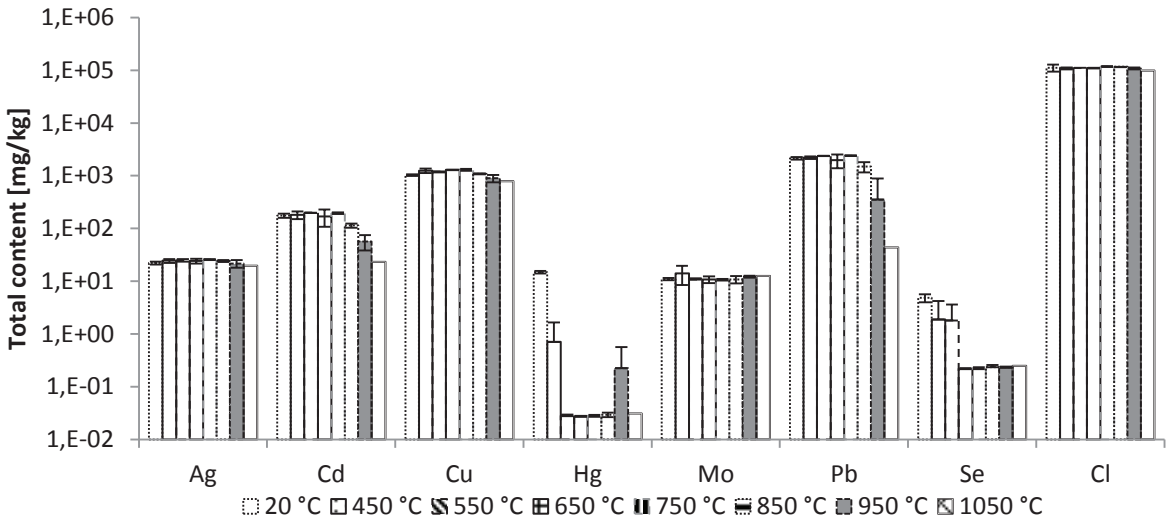


Figure 5. Concentration of volatile constituents in MSWI fly ash pellets after treatment at different temperatures. The error bars show the standard deviation. Treatment temperature of 20 °C means that no thermal treatment was applied.

The concentration of Mo and Se in the leachate increased with higher treatment temperatures. A possible explanation could be an oxidation of Mo and Se to soluble molybdate, selenite and selenate compounds. The Cr content in the leachate was unexpectedly high because of Cr being transferred from the kiln material to the sample. The elements Ag, As, Cd, Co, Cu, Hg, Ni, Pb, Sb and Sn could not be detected in the leachate of most of the thermally treated fly ash pellets, which means that their leachate content was decreased to close to 0. This is especially relevant for Pb, as this parameter of MSWI fly ash usually exceeds the limit values for non-hazardous waste landfills (10 mg/kg) (DVO, 2008) by a factor of more than 10. Interestingly, the Pb content in the leachate of the pellets prior to thermal treatment was already below this limit value, while it was above the limit value for the original fly ash. This observation indicates that already the pelletisation process stabilises heavy metals to some extent.

Analysis of POP in thermally treated MSWI fly ash pellets showed that the toxic equivalency (TEQ) for PCDD/F increased by a factor of almost 3 to a concentration of 1200 ng/kg after treatment at 450 °C

most likely due to de novo synthesis (Huang and Buekens, 1996). However, PCDD/F were destroyed mostly at 550 °C and above. DL-PCB showed a similar pattern like PCDD/F with a TEQ increasing at low treatment temperatures and decreasing at higher treatment temperatures. The sum of NDL-PCB is roughly in the same order of magnitude for all treatment temperatures as well as for untreated MSWI fly ash pellets. No concentration of HCB higher than 10 µg/kg (limit of quantification) could be detected in any of the samples analysed.

As a consequence of the low Hg total content and Pb leachate content, the thermally treated pellets can be classified as non-hazardous waste (AVVO, 2008). On the other hand, the content of total dissolved solids in the leachate still exceeds the legal limit for non-hazardous waste landfills (60,000 mg/kg) (DVO, 2008).

Chemical analysis of the secondary fly ash generated in the pilot-scale experiments revealed that this residue is enriched in Cu (up to 11,000 mg/kg), Pb (up to 91,000 mg/kg) and Zn (up to 21,000 mg/kg), depending on the treatment temperature. Due to this high metal concentration, secondary fly ash could have a considerable potential for resource recovery, e.g. by acidic leaching (Fellner et al., 2015b; Schlumberger, 2010).

4.1.3 Transfer coefficients for selected elements present in MSWI fly ash

The transfer coefficients for selected elements present in MSWI fly ash to the outputs of the hazardous waste incinerator calculated from the data of the large-scale experiment are shown in Figure 6. It can be seen that most elements were mainly transferred to the bottom ash, while the high transfer of Hg into scrubber 1 is desired, as this way Hg can be phased out via the filter cake generated at the on-site wastewater treatment plant.

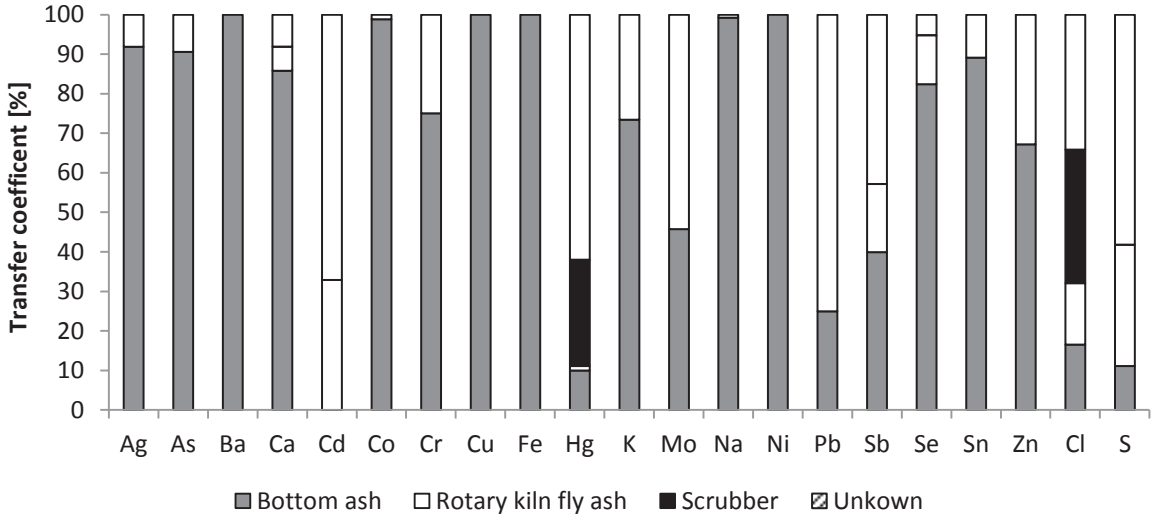


Figure 6. Transfer coefficients of selected elements from MSWI fly ash to the different outputs of kiln 1 in the large-scale experiment (including expanded uncertainty calculated according to Joint Committee for Guides in Metrology (2008)).

The transfer coefficients for selected elements to the outputs of the pilot-scale rotary kiln are shown in Figure 7. The observation from the large-scale experiment that most elements are mainly transferred to the bottom ash could be confirmed in the pilot-scale experiments with pelletised MSWI fly ash. Additionally, Figure 7 shows the correlation between the treatment temperature and the transfer coefficients. The transfer coefficient to flue gas was significantly higher than the transfer coefficient to secondary fly ash for almost all elements and temperatures. As a result, the transfer

coefficients determined in the present study differ significantly from the transfer coefficients determined from the large-scale data. This can be explained by the different experimental setup. The flue gas in the hazardous waste incinerator was cooled in the boiler prior to solids separation in the electrostatic precipitator, allowing volatile heavy metals to condensate on the secondary fly ash, while no cooling of the flue gas was applied in the present study. Furthermore, the results from the pilot-scale experiments indicate that the assumption that the hazardous waste input in both rotary kilns was identical in the experiment described in 3.1.2 is not true. Another difference between co-treatment together with combustible hazardous waste and treatment of MSWI fly ash alone is the different composition of the atmosphere in the kiln. As this can have a significant effect on the volatilisation of heavy metals (Fraissler et al., 2009; Nowak et al., 2010) and because the hazardous waste used for the two different rotary kilns was not identical as assumed, some effects like the volatilisation of Cu that are shown in Figure 7 cannot be seen in Figure 6, while on the other hand Figure 6 e.g. overestimates the transfer of Zn from MSWI fly ash to secondary fly ash.

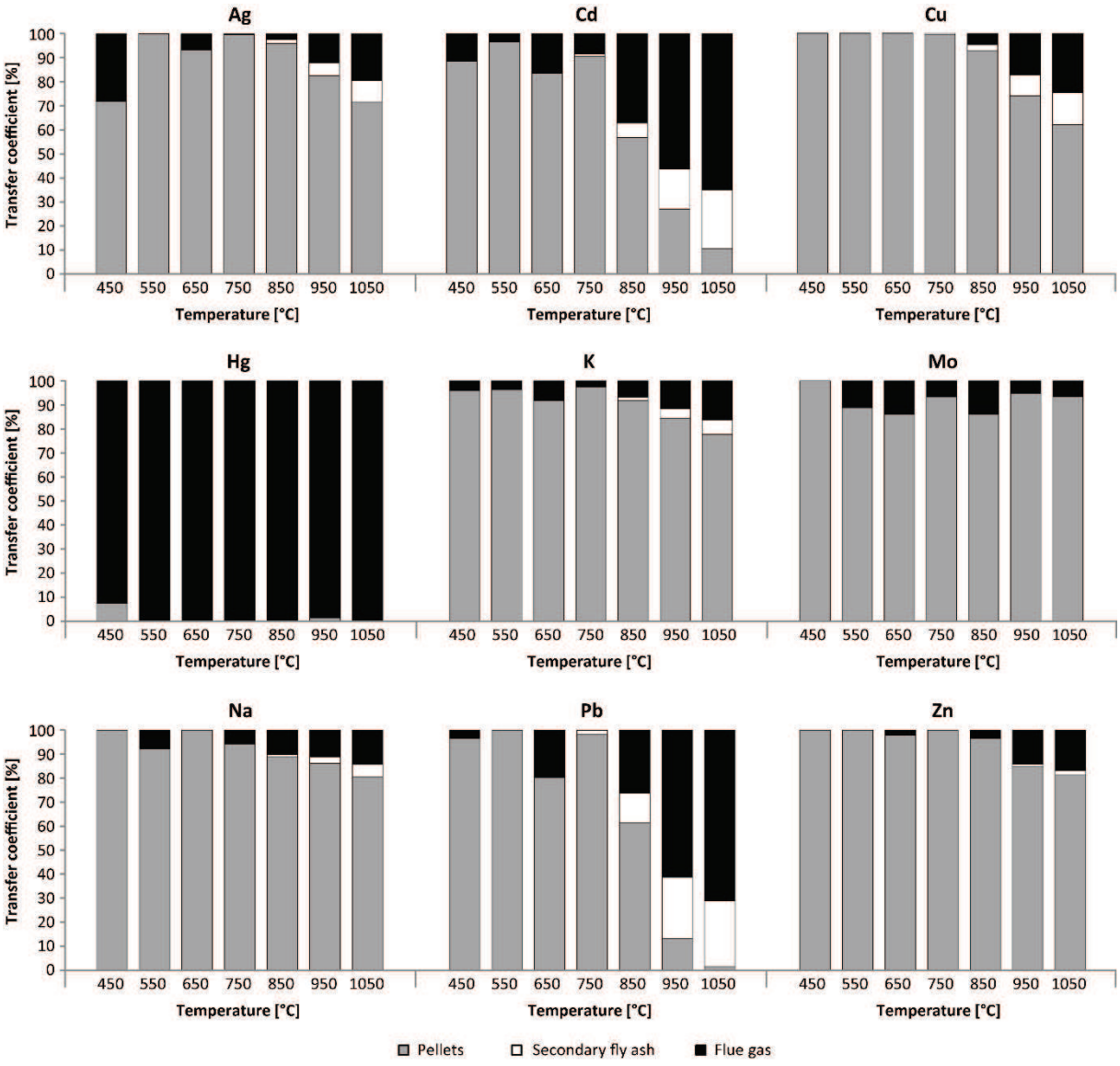


Figure 7. Transfer coefficients of selected elements from pelletised MSWI fly ash to thermally treated pellets, secondary fly ash and flue gas at different treatment temperatures as determined in the pilot-scale experiments.

The behaviour of Cd, Cu, Hg, Mo, Pb and Zn observed in the present study is in line with the results of other studies on thermal treatment of MSWI fly ash (Guohua et al., 2012; Li et al., 2015; Liu et al., 2010; Wang et al., 2008, 2015; Yang et al., 2009; Zupanič et al., 2012). However, in the study by Wang et al. (2015) about 50 % of Co is volatised and in the study by Zupanič et al. (2012) about 40 % of Sb is volatised. These differences are most likely caused by the different composition of the MSWI fly ash used for thermal treatment and the different treatment conditions.

4.2 Environmental and economic assessment of MSWI fly ash treatment and disposal

4.2.1 Environmental assessment

The environmental impacts of all scenarios in all midpoint impact categories in a timeframe of 100 a are shown in Figure 8, while the environmental impact for all endpoint impact categories is shown in Figure 9. The 5- and 95-percentile are indicated by error bars containing 90 % of the MCS results. The environmental impacts for the infinite timeframe as well as the results from the discernibility analysis can be found in Paper III.

The aggregated overall impact is lowest (close to 0) for scenario 3a (metal recovery), mainly due to the benefit in human toxicity and metal depletion caused by production of secondary metals and the low impact in most other midpoint impact categories. The total environmental impact is especially high for the scenarios 4a (thermal treatment in coal-fired furnace) and 6b (utilisation in cement kiln with salt recovery) mainly due to the high consumption of hard coal and natural gas, respectively. This is in agreement with the findings of Fruergaard et al. (2010), which already state that thermal treatment of MSWI fly ash has a very high environmental impact due to the high energy demand. In contrast, this thesis shows that the environmental impact of thermal treatment of MSWI fly ash together with combustible waste (scenarios 5a and 5b) is in many midpoint and all endpoint impact categories significantly lower compared to thermal treatment in a separate furnace (scenarios 4a and 4b). Therefore, it could be shown that the process developed within the frame of this thesis (corresponding to scenarios 5a and 5b) provides a more environmentally friendly option than other thermal treatment processes. Furthermore, the environmental impact of the new process is also lower than many other common disposal options for MSWI fly ash, e.g. stabilisation with cement. The low environmental impact of metal recovery found in this thesis is in line with the results of Bösch et al. (2011).

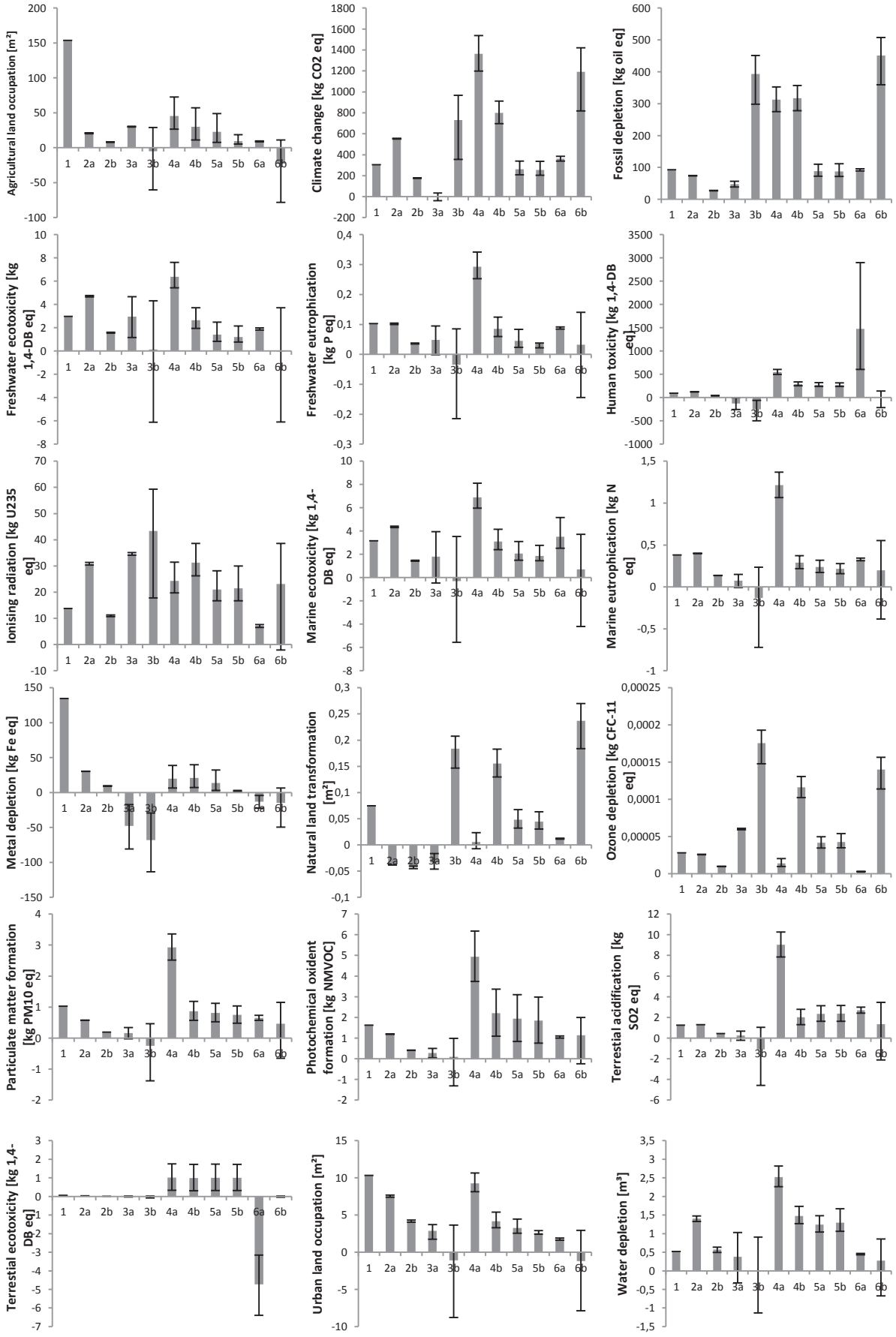


Figure 8. Environmental impact of scenarios 1 to 6 in all midpoint impact categories (5- and 95-percentile as error bars).

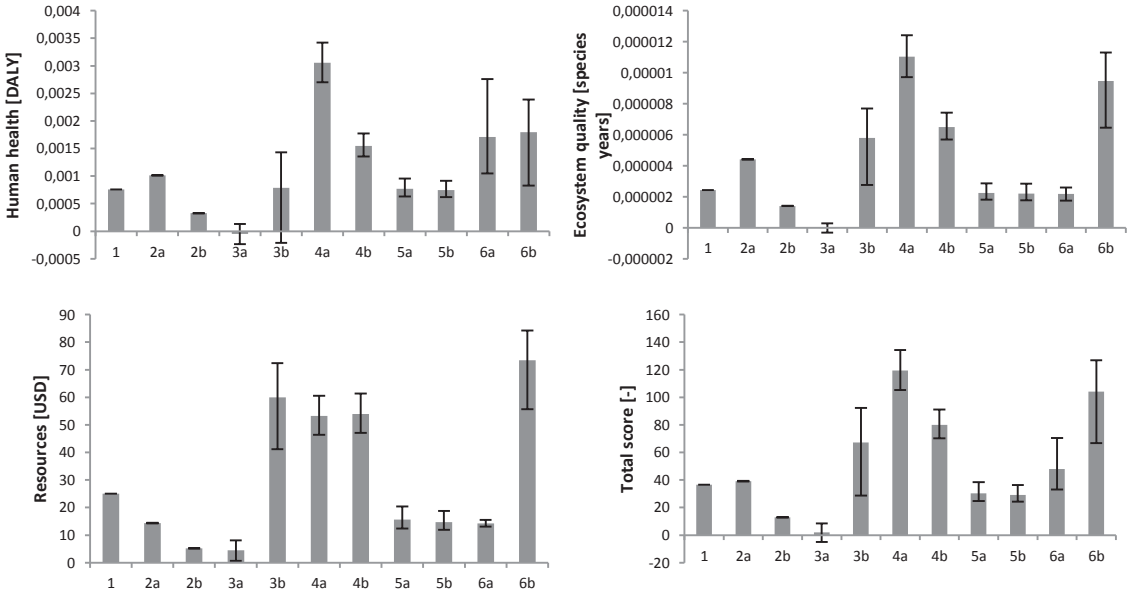
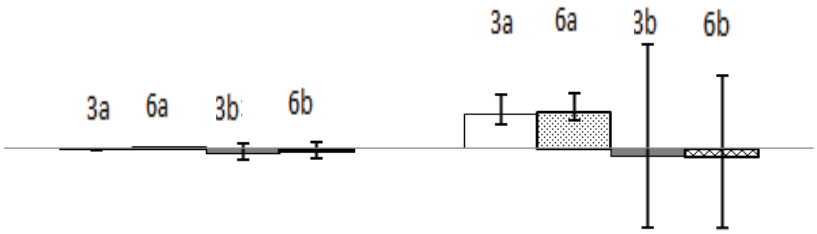


Figure 9. Environmental impact of scenarios 1 to 6 in all endpoint impact categories (5- and 95-percentile as error bars).

4.2.2 Economic assessment

The NPV for all four scenarios is shown in Figure 10 and the percentage of DCF and monetised effect on climate change, human health, ecosystem quality and resources in the total NPV from a macro view is shown in Figure 11. The results from the discernibility analysis can be found in Paper IV.

In scenario 3a (metal recovery), the overall NPV from the macro perspective is dominated by human health and climate change because the absolute value of the DCF is very low compared to the other scenarios. Due to the higher absolute value of the DCF, DCF dominates the NPV from the macro perspective in scenarios 3b (metal and salt recovery), 6a (cement production) and 6b (cement production and salt recovery). In scenario 6a, the damage to human health as a result of Hg emissions also plays an important role. The savings from the current disposal practice make up 88 % of the revenue generated in scenario 6a, while the remaining 12 % are due to the savings of primary raw materials. These savings are also the largest item generating revenue in scenario 3a. However, scenario 3a is associated with significantly higher investment costs than scenario 6a. The economic evaluation results of scenarios 3b and 6b are dominated by the expense for natural gas.



[EUR]	NPV micro	NPV macro
Metal recovery 3a	-28.000.000	1.009.000.000
Cement production 6a	18.000.000	1.088.000.000
Metal and salt recovery 3b	-139.000.000	-217.000.000
Cement production and salt recovery 6b	-93.000.000	-230.000.000

Figure 10. Net present value (given in EUR and including the monetised environmental impacts) of all four recycling scenarios from a private investor's micro and a public entity's macro perspective. The error bars show the range containing 90 % of the MCS results.

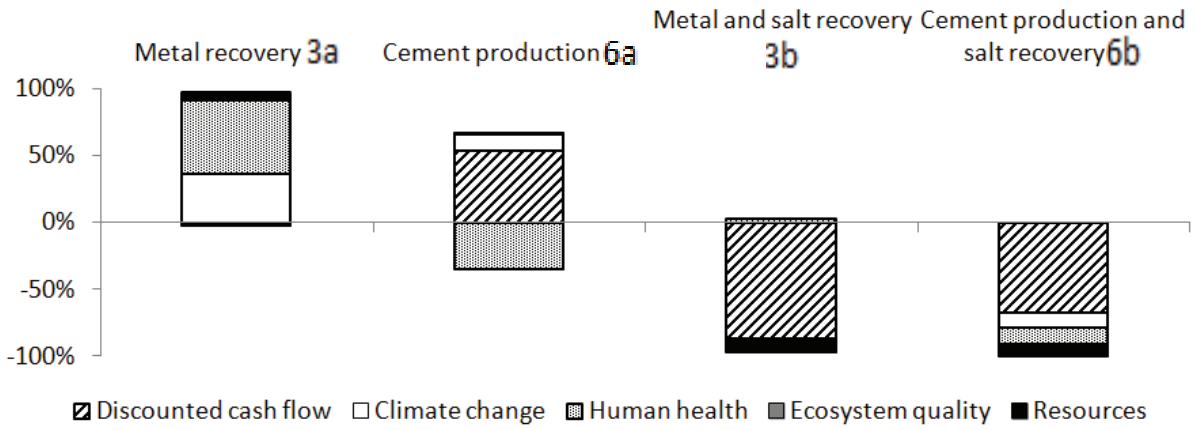


Figure 11. Percentage of DCF and monetised effect on climate change, human health, ecosystem quality and resources (referred to the sum of their absolute values from a macro view). Only median values without uncertainty are displayed for better clarity. Positive values represent revenues and negative values represent costs.

As the NPV for the recycling scenarios including salt recovery is lower compared to the respective scenarios without salt recovery both from a micro and macro perspective, salt recovery cannot be considered as an economically viable option. For a private investor, metal recovery from MSWI fly ash is not an option, because the revenues do not cover the costs for this process. However, for a public entity this option might be more interesting, as on the one hand the discount rate applied is lower (c.f. 3.2.3) and on the other hand the loss of money is overcompensated by the environmental benefit in the impact categories human health, climate change and resources, as can be seen in Figure 11. Production of cement from MSWI fly ash seems to be economically viable from a private investor's micro perspective as well as from a public entity's macro view. The sum of DCFs is positive in both cases. Although the monetised effect on the environment is negative, mainly due to damage to human health, the overall result is still positive and in both cases the median NPV is higher for the cement production scenario compared to the metal recovery scenario. The application of different weighting (valuation) factors could of course yield a different result.

The results from the application of resource classification according to UNFC (UNECE, 2010) to the anthropogenic resource MSWI fly ash are shown in Figure 12. MSWI fly ash is graded with “1” for the utilisation in cement production from a micro as well as from a macro perspective in the category socio-economic viability (E), based on the DCF analysis and LCA applied in the present study. With regard to the recovery of metals, MSWI fly ash is graded as “2” from a micro view because most of the MCS results show a negative NPV and it is graded as “1” from the macro view because the inclusion of external costs yields a positive NPV in most of the MCS results. A positive NPV from the micro perspective could be achieved by Zn prices of about 19 EUR/kg instead of 3 EUR/kg. According to Winterstetter et al. (2015), an increase in metal prices by a factor of 10 is still realistic within the next 20 a. The inclusion of salt recovery into either metal recovery or cement production substantially decreases the NPV from both perspectives. In order to be economically viable either the gas price has to decrease from 0.07 EUR/kWh to 0.0016 EUR/kWh or the salt price has to increase from 0.13 EUR/kg to 5.33 EUR/kg. As such severe price changes (factor 40) are not realistic in the next 20 a, salt recovery from MSWI fly ash is graded as “3”.

With regard to the classification category project feasibility (F), MSWI fly ash can be classified as “1” for the production of cement and the recovery of metals, as metal recovery is currently taking place (e.g. in Switzerland) and the feasibility of cement clinker production has been shown in detailed studies (Guo et al., 2016; Saikia et al., 2007). The recovery of chloride salts from MSWI fly ash is graded as “2” because extraction is subject to further evaluation (Easymining, 2017; Stena Metall, n.d.), but to the authors’ best knowledge no extraction and utilisation of chloride salts is currently taking place in full scale. For this category no difference between micro and macro perspective is made.

MSWI fly ash is sampled and analysed in regular intervals and the amount of this residue generated each year is well known. Consequently, MSWI fly ash can be graded as “1” in the category geological knowledge (G). This grade is applicable to all three recycling options and micro as well as macro perspective.

By combining these three criteria, MSWI fly ash can be classified as “211” for metal recovery from a private investor’s micro perspective, “111” for metal recovery from a public entity’s macro perspective, “111” for cement production and “321” for chloride salt recovery.

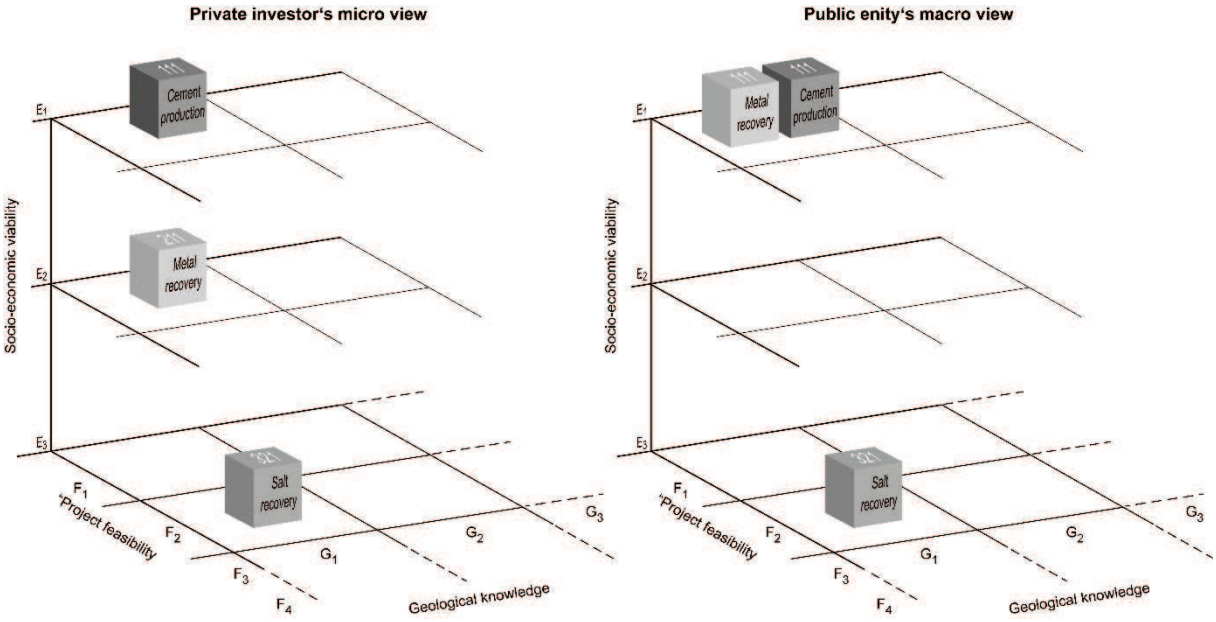


Figure 12. Resource classification of MSWI fly ash for metal recovery, cement production and salt recovery according to UNFC. MSWI fly ash is graded according to socio-economic viability, project feasibility and geological knowledge.

5. Scientific contribution of the thesis

Thermal treatment of waste with integrated energy recovery provides manifold advantages. However, this treatment comes along with the generation of MSWI fly ash, which constitutes a hazardous waste and has to be disposed of accordingly. There are several different possibilities to treat and finally dispose of MSWI fly ash, some of which also include material recovery. The scenarios investigated in this thesis and described in detail in 3.2.1 constitute a non-exhaustive list of such treatment and disposal options.

As all of the above mentioned fly ash management options are associated with disadvantages like high costs, consumption of primary raw materials or geographical constraints leading to dependency from certain countries, a completely novel process was developed within the frame of this thesis. The experiments conducted in the course of this development, their results and connotations are described in detail in Paper I. This paper contributes to the understanding of the behaviour of MSWI fly ash during thermal co-treatment together with combustible waste and gives transfer coefficients on goods and substance level for the transfer from inserted MSWI fly ash to bottom ash, secondary fly ash and scrubber water. It shows that MSWI fly ash is almost completely transferred to the bottom ash, if treated in a rotary kiln waste incinerator, while toxic heavy metals like Cd and Hg are mainly transferred to the secondary fly ash and scrubber water, respectively. As a consequence, the bottom ash still complies with legal limits for non-hazardous waste landfills.

Furthermore, a pelletisation process was developed as a pretreatment prior to thermal treatment to overcome several drawbacks described in Paper I. The combination of pelletisation and thermal treatment of MSWI fly ash, including the chemical and mechanical properties of MSWI fly ash, pelletised MSWI fly ash and thermally treated MSWI fly ash pellets as well as transfer coefficients for the combined treatment process are given in Paper II. This paper provides the basis for a technically feasible process that does not impair continuous operation of the waste incineration plant. It could be demonstrated that MSWI fly ash is well suitable for disc pelletisation with water as the only binder, that the mechanical properties of MSWI fly ash pellets are sufficient and that thermal treatment for 10 min or more at a temperature above 450°C decreases the heavy metal content of MSWI fly ash pellets thereby rendering them non-hazardous. In Paper II the mass and chemical composition of secondary fly ash generated during thermal treatment, a subject hardly discussed in previous literature, are given.

Paper III provides a thorough assessment of the environmental impact of the novel process first described in Paper I and later optimised by investigations mentioned in Paper II and establishes a comparison between this process and other treatment and disposal options known in the state of the art. It showed that the overall environmental impact as determined by LCA of co-treatment together with combustible waste is lower compared to stabilisation with cement and thermal treatment in a separate furnace but still higher compared to acidic extraction with metal recovery (so-called FLUREC process). Additionally, Paper III is an example of how parameter uncertainty can be assessed within LCA and, for the first time, shows the effect that the consideration of different time frames can have on the result of a discernibility analysis of LCA results.

Finally, Paper IV constitutes the first case study for resource classification based on DCF analysis and performed on solid residues from MSWI and therefore contributes to the integration of anthropogenic resources into resource classification and evaluation frameworks. A significant innovation incorporated in Paper IV is the application of LCA with monetary valuation in order to

include external costs into resource classification. While a private investor will only consider internal costs and revenues, a public entity should also take into account the broader implications on human well-being. This aspect is represented by the determination and consideration of external costs in this thesis. The investigation described in Paper IV shows that utilisation of MSWI fly ash in cement production can be considered a commercial project from a private investor's micro view (excluding external costs) and a public entity's macro view (including external costs) and metal recovery can be considered a commercial project from a public entity's macro view, while salt recovery is a non-commercial project from both perspectives.

6. Conclusions and outlook

It could be demonstrated in this thesis that co-treatment of MSWI fly ash together with combustible waste in an existing waste incinerator represent a feasible management option. Pelletisation of MSWI fly ash prior to its thermal treatment is a promising pretreatment process that can facilitate the introduction of fly ash into the incinerator and thereby ensure its continuous operation. Due to the transfer of volatile heavy metals and the destruction of organic compounds contained in MSWI fly ash, effective decontamination of MSWI fly ash takes place and the remaining material (transferred to the bottom ash) complies with legal limits for non-hazardous waste landfills after treatment.

Yet, the combination of pelletisation and thermal treatment of MSWI fly ash was only conducted in an electrically heated pilot-scale kiln without the addition of combustible material. Consequently, further investigations on the co-treatment of MSWI fly ash pellets and combustible waste are advisable in order to confirm the results presented in this thesis. As all experiments within this thesis were performed using rotary kilns, future experiments should also be conducted in grate furnaces, which constitute the most common type of furnace used in MSWI.

The transfer of volatile metals (e.g. Cd, Cu, Pb and Zn) to secondary fly ash is not only a mechanism for decontamination of the bulk of the material but also leads to higher concentrations of these elements in the secondary fly ash. The secondary fly ash can also be reintroduced into the incinerator in order to generate tertiary fly ash with an even higher concentration of metals. As metal recovery is only economically feasible for a few fly ashes with very high Zn concentrations, combined pelletisation and thermal co-treatment together with combustible waste can be used to generate residues with a higher metal concentration and subsequently recover these metals at acceptable costs. This effect could be even increased by selecting combustible wastes high in volatile metals like Zn for the co-treatment together with MSWI fly ash.

The environmental impact assessment performed within this thesis showed that the newly developed MSWI fly ash treatment and disposal process is preferable over stabilisation with cement, regardless of the timeframe taken into account in more than 95 % of the MCS results.

With regard to economic assessment, the research conducted within the frame of this thesis demonstrated that extraction of metals from MSWI fly ash with a composition as typically found in Austrian MSWI plants, cannot be considered a commercial project according to UNFC classification from a private investor's micro view due to the negative NPV in the majority of the MCS results. Nevertheless, the inclusion of external costs and the lower discount rate associated with a public entity's macro view results in a positive NPV in most of the MCS results.

As there is no data available about the repeated reintroduction of MSWI fly ash into the combustion with subsequent metal recovery from fly ash enriched in heavy metals, this MSWI fly ash management option could not be included into the environmental and economic assessment presented in this thesis. Possibly, this option constitutes also a commercial project from a private investor's micro view.

Nevertheless, the work conducted within this thesis paved the way for further case studies on resource classification and evaluation of solid residues from MSWI. Such further case studies in different countries with regard to MSWI fly ashes as well as other combustion residues like MSWI

bottom ashes are necessary to enable private investors and public entities to exploit currently unused resources. This can reduce the amount of wastes disposed of on landfills, ensure the supply with resources and create a more circular economy.

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8. Appendix (Papers I – IV)

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Thermal co-treatment of combustible hazardous waste and waste incineration fly ash in a rotary kiln

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ABSTRACT

As current disposal practices for municipal solid waste incineration (MSWI) fly ash are either associated with significant costs or negative environmental impacts, an alternative treatment was investigated in a field scale experiment. Thereto, two rotary kilns were fed with hazardous waste, and moistened MSWI fly ash (water content of 23%) was added to the fuel of one kiln with a ratio of 169 kg/Mg hazardous waste for 54 h and 300 kg/Mg hazardous waste for 48 h while the other kiln was used as a reference. It was shown that the vast majority (>90%) of the inserted MSWI fly ash was transferred to the bottom ash of the rotary kiln. This bottom ash complied with the legal limits for non-hazardous waste landfills, thereby demonstrating the potential of the investigated method to transfer hazardous waste (MSWI fly ash) into non-hazardous waste (bottom ash). The results of a simple mixing test (MSWI fly ash and rotary kiln bottom ash have been mixed accordingly without thermal treatment) revealed that the observed transformation of hazardous MSWI fly ash into non-hazardous bottom ash during thermal co-treatment cannot be referred to dilution, as the mixture did not comply with legal limits for non-hazardous waste landfills. For the newly generated fly ash of the kiln, an increase in the concentration of Cd, K and Pb by 54%, 57% and 22%, respectively, was observed. In general, the operation of the rotary kiln was not impaired by the MSWI fly ash addition.

1. Introduction

Municipal solid waste incineration (MSWI) reduces the waste volume by 90%, the mass by 60–90%, and the organic matter by nearly 100% (Brunner and Rechberger, 2015; Chandler et al., 1997; Hjelmar, 1996; Kuo et al., 2007), saving valuable landfill space and avoiding gaseous landfill emissions (CH_4) from organic degradation processes. In Europe, almost 90% of MSWI plants are grate furnace combustors equipped with a grate furnace (Fellner et al., 2015). The solid residues produced in the MSWI grate furnace comprise bottom ash, which can be deposited at non-hazardous waste landfills or used as road construction material, and fly ash, which consists of *"particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material"* (Chandler et al., 1997). This MSWI fly ash comprises high contents of easily soluble salts, heavy metals and in some cases also polychlorinated dioxins and furans and is therefore classified as hazardous waste in many countries (Funari et al., 2016; Jiao et al., 2016; Li et al., 2016; Purgar et al., 2016; Ye et al., 2016; Zhan et al., 2016). For this reason, fly ashes from MSWI grate incineration generated in Austria for instance are currently either exported to hazardous waste landfills abroad or stabilised with cement and subsequently disposed of at local non-hazardous waste landfills. Both options involve costs and in the latter case the utilisation of cement requires not only valuable resources but causes significant amounts of CO_2 emissions (up to 500 kg CO_2/Mg fly ash). Hence, in the recent decades numerous studies investigating alternative treatment/disposal options for MSWI fly ash (De Boom and Degrez, 2015; Lindberg et al., 2015; Nowak et al., 2013; Quina et al., 2008; Nowak et al., 2013; Wey et al., 2006; Zacco et al., 2014) have been published.

As the physical, chemical and mineralogical properties of waste incineration fly ashes vary depending on waste composition, type of incinerator and the air pollution control (APC) system installed, it is difficult to compare these results. These facts, as well as the different legislation even in EU member countries, are reflected by various suggestions for alternative treatment and utilisation of fly ashes that can be found in literature.

Several authors propose a wet extraction process (washing) to decrease the content of easily soluble compounds like chloride salts present in MSWI fly ash (Aguiar del Toro et al., 2009;

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Blasenbauer et al., 2015; De Boom and Degrez, 2015; Karlfeldt Fedje et al., 2010; Thomé-Kozmiensky, 2013; Wang et al., 2001; Zhang and Itoh, 2006). These studies show that chloride can effectively be extracted, while the heavy metal content of treated fly ash may still require a stabilisation with cement prior to disposal at a non-hazardous waste landfill. In case that an acidic extraction agent (e.g. acidic scrubber water) is used instead of neutral water (leaching) also some metals (e.g. Zn, Pb) are largely removed.

An extension of the acidic leaching of fly ashes is the so-called FLUREC process that allows extraction and recycling of zinc, cadmium, copper and lead (Boesch et al., 2014; Schlumberger, 2010). However, at current Zn prices, this technique is only economically viable for waste incineration fly ashes showing Zn contents above 50,000 mg/Mg (Fellner et al., 2015).

Another approach is to treat fly ash at temperatures close to or above its melting point with or without additives, thereby transforming it into sintered, melted or vitrified products. These processes decrease the mobility of heavy metals and, as a result, also the heavy metal concentration in the leachate (Li et al., 2015; Sobiecka and Szymanski, 2014; Sobiecka, 2015; Wang et al., 2008; Wunsch et al., 1996; Yang et al., 2009; Zhao et al., 2010; Zupanič et al., 2012) and effectively destroy the polychlorinated dioxins and furans (Miller et al., 1989; Vogg and Stieglitz, 1986; Wu et al., 2011). Sintering trials of MSWI fly ash from grate furnace in a rotary kiln showed that the thermal treatment causes a partial evaporation of lead, thereby rendering the sintered product non-hazardous (Wey et al., 2006). However, in general these high temperature processes require considerable amounts of energy, which calls their cost-effectiveness and sustainability into doubt.

Beside the specific problems mentioned, a common shortcoming of most of these alternative fly ash treatment processes are the rather high investment costs due to the installation of new treatment plants. For this reason, these processes are not widely applied as plant operators fear making such a risky investment. Using already existing treatment facilities in the vicinity of the point of MSWI fly ash generation like existing hazardous waste incineration plants would reduce this risk. However, such co-treatment of MSWI fly ashes has not been investigated so far.

The objectives of the present work are therefore to assess the feasibility of co-treating MSWI fly ash together with hazardous wastes and to evaluate the fate of the inserted fly ash as well as the quality of the solid residues generated. The particular research questions to be addressed are:

- How do the inserted MSWI fly ash and its components partition among the hazardous waste incineration residues during the co-treatment?
- Does the co-treatment of MSWI fly ash influence the quality of solid residues in general and bottom ash in particular, generated by the rotary kiln?
- In how far is the impact on the quality of bottom ash observed explainable by simply mixing the inserted MSWI fly ash and rotary kiln bottom ash?
- Does the co-treatment of fly ash in the rotary kiln impair its safe and continuous operation?

As the study was carried out in Austria, substance limit values for landfills refer to the legal situation in Austria.

2. Materials and methods

In the present study the co-treatment of moistened MSWI fly ash together with hazardous waste in a rotary kiln was investigated. Thereto two rotary kiln lines (1 and 2) of a hazardous waste incinerator were used. At kiln 1 moistened MSWI fly ash was added to the ordinary hazardous waste fuel at different rates for altogether 102 h; whereas kiln 2 served as reference utilising hazardous waste only. During the experimental time the flows of inserted matter (hazardous waste, moistened MSWI fly ash) were recorded and the outputs via bottom ash, rotary kiln fly ash and scrubber water of both kilns were separately recorded, sampled and subsequently analysed. The results served on the one hand to assess the destination of the inserted MSWI fly ash and its components (determination of transfer coefficients) and on the other hand to evaluate the impact of the MSWI fly ash addition on the quality of solid residues generated at the kiln. Finally, moistened MSWI fly ash was mixed with rotary kiln bottom ash without thermal treatment in order to evaluate if observed changes in bottom ash quality after thermal treatment refer to mixing effects only.

2.1. Inserted MSWI fly ash

The fly ash used for the experiment was collected at a MSWI mass-burning combustor with grate furnace. The APC system at this plant comprises an activated coke injector, fabric filters, a two-stage scrubber and a selective catalytic reduction device. In order to avoid dust emissions during the handling of the MSWI fly ash, it was mixed with water in a mass ratio of approximately 3:1, which lead to a water content of 0.23 kg/kg moistened MSWI fly ash. Then it was transported daily by truck to the hazardous waste rotary kiln incineration plant, where it was stored in the waste bunker. Prior to storage the moistened MSWI fly ash was sampled and analysed (see Table 1). The fly ash from this plant generally exceeds the legal limits for non-hazardous waste landfill for the parameters Hg total content, total dissolved solids in the leachate and Pb in the leachate.

2.2. Rotary kilns

The hazardous waste incinerator utilised for the experiment comprises two rotary kilns with a combined capacity of 100,000 Mg/a. The rotary kilns were built in 1980 and each rotary kiln is 12 m long, has an outer diameter of 4.5 m and rotates at approximately 0.5 min^{-1}. The residence time of waste in the rotary kilns is estimated to be about 1 h. The process scheme is illustrated in Fig. 1. The APC system consists of an electrostatic precipitator, a two-stage scrubber, a selective non-catalytic reduction device for injection of NH_3 and an activated coke filter (Stubenvoll et al., 2002). Gypsum is precipitated from the second scrubber stage and deposited on a non-hazardous waste landfill. The scrubber water is treated in an on-site wastewater treatment plant.

For the investigations conducted, the inputs and outputs of the rotary kiln (cf. Fig. 1) were recorded and analysed in order to assess the flows of substances present in MSWI fly ash added to kiln 1.

2.3. Fly ash treatment

The moistened MSWI fly ash was fed at different rates into rotary kiln 1 of the hazardous waste incinerator for a total period of 102 h. During the first 54 h (phase A) for every Mg of hazardous waste 169 kg (130 kg dry matter) MSWI fly ash were added to rotary kiln 1. Subsequently, the rate of MSWI fly ash addition was increased to 300 kg (231 kg dry matter) fly ash per Mg hazardous waste for a period of 48 h (phase B). During the trial the amount and composition of the hazardous waste inserted into rotary kiln 1 and 2 was, as far as possible, identical so that the only difference between the two kilns was the addition of moistened MSWI fly ash. For all following calculations it was assumed that the composition of hazardous waste inserted into both kilns was

Table 1
Composition of MSWI fly ash co-treated in rotary kiln 1 (TE... toxicity equivalent according to the international toxicity equivalent factor method).

	Unit	Mean	Standard uncertainty (n = 3)
Ag	mg/kg	29	5
As	mg/kg	47	3
Ba	mg/kg	1,220	138
Ca	mg/kg	205,000	7510
Cd	mg/kg	140	35
Co	mg/kg	39	4
Cr	mg/kg	250	9
Cu	mg/kg	702	109
Fe	mg/kg	12,100	1210
Hg	mg/kg	12	3
K	mg/kg	32,300	6990
Mo	mg/kg	18	2
Na	mg/kg	35,800	6800
Ni	mg/kg	54	4
Pb	mg/kg	1730	340
Sb	mg/kg	552	89
Se	mg/kg	6	1
Sn	mg/kg	557	76
Zn	mg/kg	9750	1660
C	mg/kg	22,500	2970
Polychlorinated dioxins and furans	ng TE/kg	298 (Wien Energie, 2015)	–

identical. The mass flows of moistened MSWI fly ash and hazardous waste into both rotary kilns are illustrated in Fig. 2.

2.4. Sampling and analysis of different output flows of the hazardous waste incinerator

The sampling times are shown in Fig. 2. During the experiment bottom ash of both kilns was sampled every 4 h. Equal amounts of these samples were mixed to daily composite samples and the daily composite samples were subsequently combined to a total composite sample for analysis. This procedure of sample aggregation resembles the weekly composite samples usually analysed by the plant operator and required for the verification as non-hazardous waste prior disposal at a non-hazardous waste landfill. Further samples separately taken (every 12 h) for both kilns include rotary kiln fly ash and scrubber water of scrubber 1.

The concentration of CO, NO_x and SO_2 in the cleaned flue gas was monitored continuously, as well as the volume of flue gas.

For the chemical analysis, total contents as well as the leachability were determined. For the total contents, samples were digested in aqua regia according to EN 13657 (2002) and subsequently analysed by ICP-OES (inductively coupled plasma optical emission spectroscopy) according to EN 11885 (2009). Leachates were prepared using a liquid to solid ratio of 10 L/kg according to EN 12457-4 (2002) and analysed as above. To determine the total Cl content, the samples were milled to a grain size of 250 µm and subsequently analysed by X-ray fluorescence spectroscopy. The total S and C contents were measured with a total CHNS analyser. Chloride and sulphate concentrations in liquids were determined by ion chromatography. The analysis parameter for the different solid and liquid residues are summarised in Table 2. As the mass of carbon inserted via fly ash is far below the mass of carbon inserted via ordinary hazardous waste, no mass balance for this element can be established. As a consequence, carbon was not measured in the incineration residues of the rotary kilns. Vogg and Stieglitz (1986) demonstrated that polychlorinated dioxins and furans are nearly quantitatively decomposed at temperatures of 600 °C. As the temperature in the rotary kilns was even higher, it can be assumed that no significant amount of dioxins and furans contained in the inserted fly ash is transferred to the rotary kiln incineration residues or flue gas. Even if dioxins and furans from inserted fly ash are transferred to the flue gas, they will be adsorbed by the fixed-bed activated coke filter (cf. Section 2.2).

2.5. Determination of transfer coefficients

Based on the recorded mass and volume flows and their respective composition a material flow analysis of the experiment was conducted according to Brunner and Rechberger (2004) aiming at identifying and illustrating the distribution of inserted MSWI fly ash and its elemental components to the different outputs of the rotary kilns (bottom ash, rotary kiln fly ash and scrubber water) by so-called transfer coefficients.

The transfer coefficients on a goods level (e.g. total mass flows of inserted MSWI fly ash and hazardous waste) were first calculated for kiln 2 by dividing each output flow by the inserted

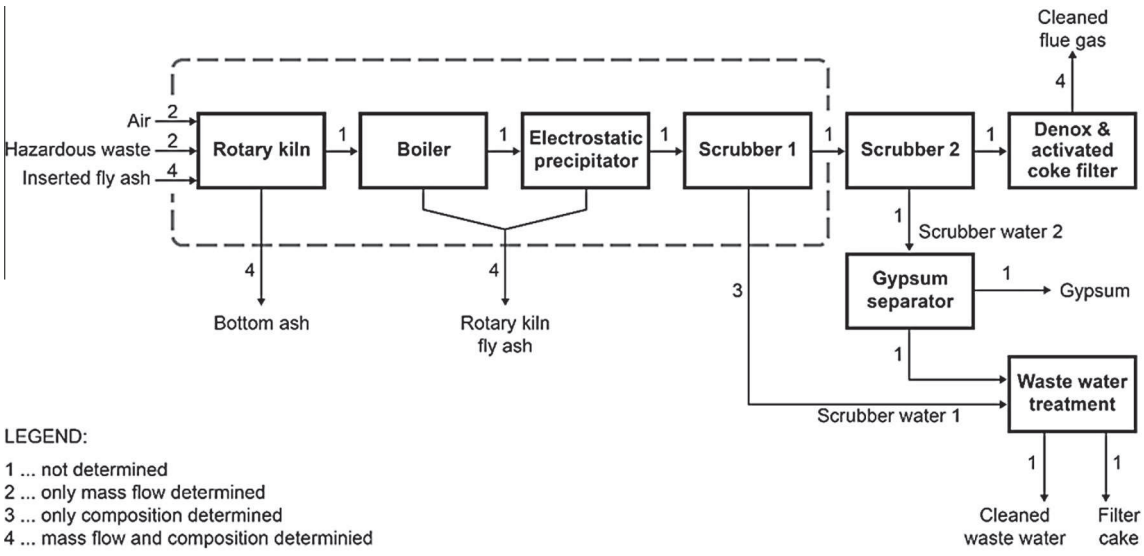


Fig. 1. Process scheme of the rotary kiln for hazardous waste incineration (including input and output flows measured, as well as the system boundary used for the investigations).

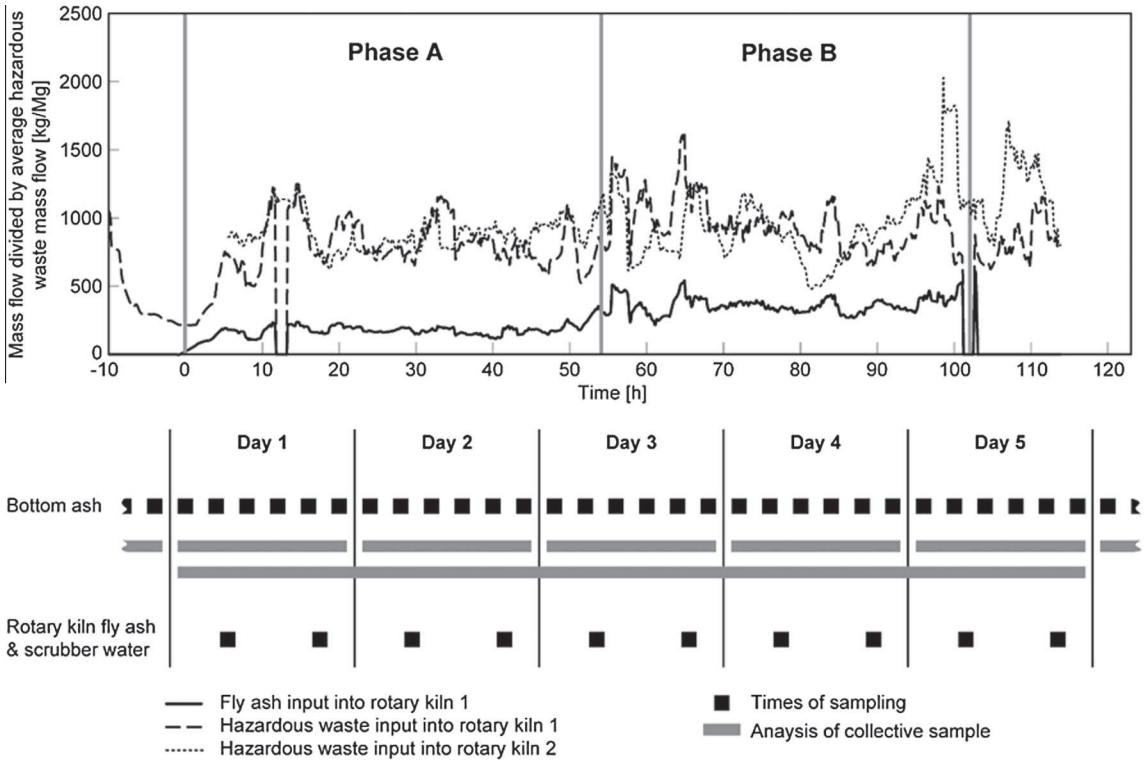


Fig. 2. Input into the rotary kilns during the experiment (phase A: 0–54 h, phase B: 54–102 h) and information about the times of sampling and analyses.

Table 2
Summary of analysis parameters. The measured parameters for bottom ash, rotary kiln fly ash and water of scrubber stage 1 are indicated. For bottom ash the total content as well as the concentration in the leachate were determined. For the rotary kiln fly ash only total contents were analysed. In scrubber water to concentration of Cl^-, SO_4^{2-}, Na, K and selected volatile heavy metals were determined.

	Bottom ash	Rotary kiln fly ash	Scrubber stage 1
Total dissolved solids	x		
pH	x		
Ag	x	x	
As	x	x	
Ba	x	x	
Ca	x	x	
Cd	x	x	x
Co	x	x	
Cr	x	x	x
Cu	x	x	
Fe	x	x	
Hg	x	x	x
K	x	x	x
Mo	x	x	x
Na	x	x	x
Ni	x	x	
Pb	x	x	x
Sb	x	x	x
Se	x	x	
Zn	x	x	x
Cl	x	x	
S	x	x	
Cl^-			x
SO_4^{2-}			x

amount of hazardous waste. Based on the calculated transfer coefficient for bottom ash of kiln 2 and the hazardous waste input of kiln 1, the part of bottom ash produced in kiln 1 solely from hazardous waste incineration was estimated. The difference between this calculated mass of bottom ash from hazardous waste combustion in kiln 1 and the mass of bottom ash observed at kiln 1 was used to determine the amount of inserted MSWI fly ash transferred into bottom ash as shown in formula 1.

$$m_{FABA} = m_{BA1} - \frac{m_{BA2}}{m_{HW2}} \cdot m_{HW1} \quad (1)$$

m_{FABA} mass of inserted fly ash transferred to bottom ash of rotary kiln 1
$m_{BA1,2}$ mass of bottom ash generated in rotary kiln 1 and 2, respectively
$m_{HW1,2}$ mass of hazardous waste inserted into rotary kiln 1 and 2, respectively

To determine the transfer coefficients of elements inserted via MSWI fly ash in kiln 1 (Ag, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mo, Na, Ni, Pb, Sb, Se, Sn, Zn, Cl and S), in a first step the elemental composition of the hazardous waste combusted was calculated using data of kiln 2 (mass and volume of input and output flows as well as information about their respective elemental composition). Information about the amount and composition of hazardous waste and moistened MSWI fly ash treated in kiln 1 were combined to determine overall input of different elements and afterwards compared to their overall output flows (via bottom ash, rotary kiln fly ash and scrubber water) observed. This comparison allowed tracing the flows of elements inserted via moistened MSWI fly ash at kiln 1.

Exemplary, formula 2 used to calculate the amount of an element transferred from inserted MSWI fly ash to bottom ash of rotary kiln 1 is given below.

$$m_{E,FABA} = w_{E,BA1} \cdot m_{BA1} - \frac{w_{E,BA2} \cdot m_{BA2}}{m_{HW2}} \cdot m_{HW1} \quad (2)$$

$m_{E,FABA}$ mass of element E transferred from inserted fly ash to bottom ash of rotary kiln 1
$m_{BA1,2}$ mass of bottom ash generated in rotary kiln 1 and 2, respectively

$m_{HW1,2}$ mass of solid hazardous waste inserted into rotary kiln 1 and 2, respectively
$w_{E,BA1,2}$ mass fraction of element E in bottom ash of rotary kiln 1 and 2, respectively

This calculation was applied to all outputs streams and elements mentioned above. However, for several elements the so calculated mass in the output was lower than the mass in the input, resulting in an "unknown" fraction. Consequently, the respective expanded uncertainties (combination of the variation in analysis results for one sample and the standard deviation of different samples multiplied by an extension factor of 2, calculated according to the Guide to the Expression of Uncertainty in Measurement (2008)) were added to the transfer coefficients to reduce this unknown fraction.

2.6. Mixing of moistened MSWI fly ash and rotary kiln bottom ash without thermal treatment

Bottom ash from rotary kiln 2 was mixed with moistened MSWI fly ash in a ratio of 5:4 corresponding to the ratio of inert matter from hazardous waste and inserted MSWI fly ash in rotary kiln 1 (average of phase A and phase B). This mixture was analysed the same way as bottom ash samples and compared to bottom ash from rotary kiln 1 in order to assess the effect of thermal treatment in relation to mere mixing.

3. Results and discussion

3.1. Mass flows of hazardous waste and inserted fly ash

During phase A for every Mg of hazardous waste inserted, in rotary kiln 1 (with MSWI fly ash addition) a total of 460 kg (387 kg dry matter) of bottom ash was produced, contrary to only 249 kg (223 kg dry matter) rotary kiln 2 (without MSWI fly ash addition). If the transfer coefficient from kiln 2 (0.223) is applied to the hazardous waste treated in kiln 1, the calculated amount of inserted MSWI fly ash that should have been transferred to the bottom ash (164 kg dry matter/Mg hazardous waste) was larger than the overall dry matter of inserted MSWI fly ash (130 kg dry matter/Mg hazardous waste). This mismatch of 34 kg dry matter/Mg hazardous waste may be attributed to the fact that most probably hazardous waste treated at both kilns was not perfectly identical and showed slight difference in their ash content. Furthermore, uncertainties with respect to the determination of mass flows of inserted MSWI fly ash and generated bottom ash, and water contents may explain the mismatch between calculated and observed mass flows of inert matter.

During phase B, for every Mg of hazardous waste used in rotary kiln 1 a total of 571 kg (486 kg dry matter) bottom ash was produced. At kiln 2 the bottom ash generation amounted to 294 kg (259 kg dry matter) per Mg hazardous waste. Applying the transfer coefficient of kiln 2 (0.259) to kiln 1 and comparing the calculated hazardous waste related amount of bottom ash with the observed quantity at kiln 1, the residual calculated mass of inserted fly ash transferred to the bottom ash was 226 kg dry matter/Mg hazardous waste, which accounts for 98% of the inserted MSWI fly ash (231 kg dry matter/Mg hazardous waste).

Based on these results it may be assumed that the above mentioned mismatch in phase A is apparently due to the relatively low share of inserted MSWI fly ash dry matter in the total inserted inert matter (25%) compared to phase B (47%).

As no separate recording of the fly ash generation at each kiln was possible, only a common quantity for both kilns was determined. Comparing this figure to the annual average generation of rotary kiln fly ash reveals that during the experiment the generation rate was only slightly higher (less than 10%).

All in all the results of the material flow analysis suggest that >90% of the moistened fly ash inserted into rotary kiln 1 were bound into the bottom ash of the kiln.

Furthermore, the transfer coefficient from hazardous waste to wet bottom ash in kiln 2 (no fly ash addition) was 0.27 during the whole experimental period compared to an annual average of 0.29, which allows concluding that the hazardous waste incinerated during the experiment is representative regarding its bottom ash content.

All material flows of hazardous waste, inserted fly ash and residues based on 1 Mg of inserted hazardous waste are illustrated in Fig. 3.

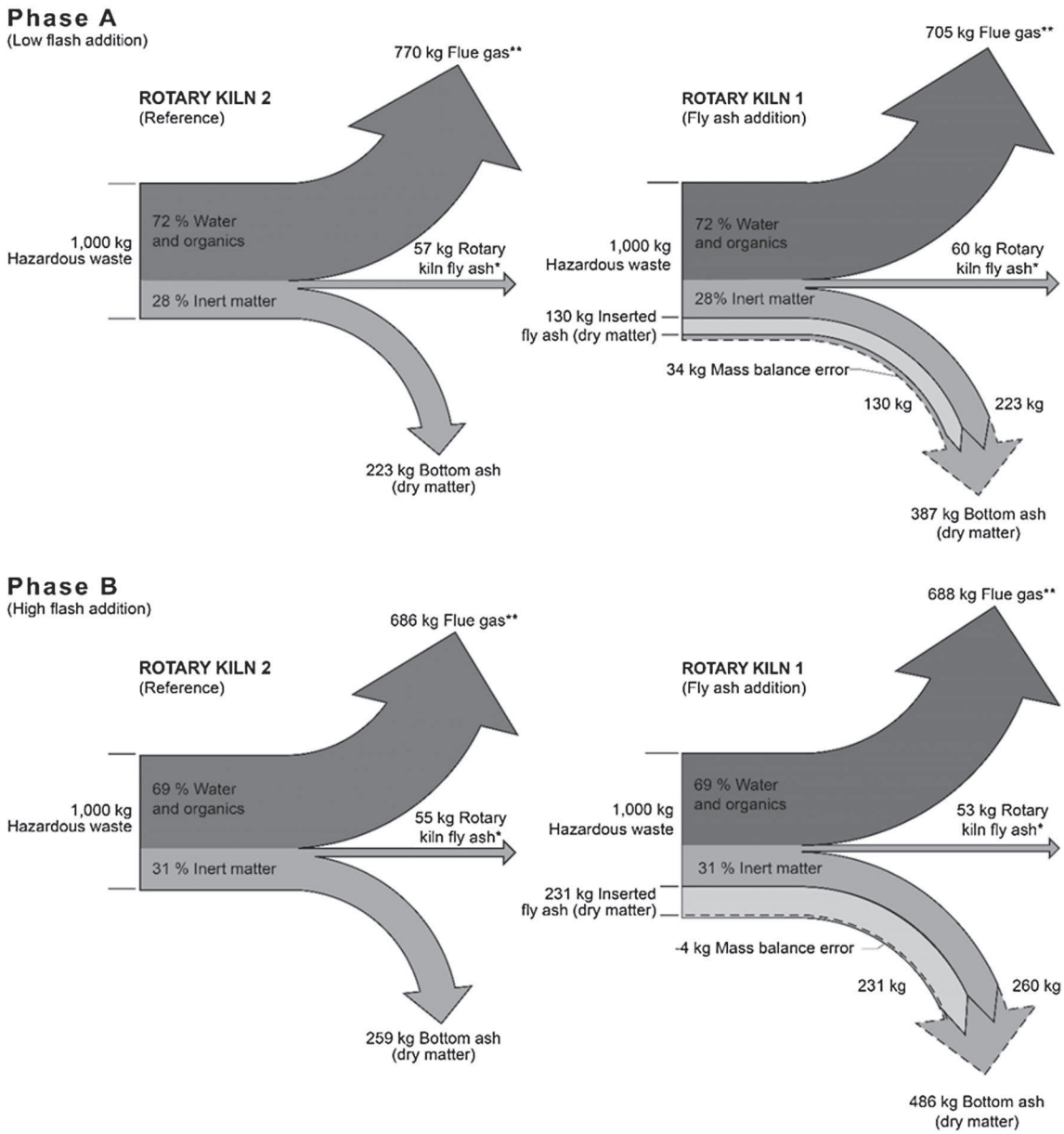
3.2. Quality of solid residues

The chemical analysis (total contents of elements) for composite samples (representative for the entire experimental period) of bottom ash and rotary kiln fly ash of both kilns reveal that the Cl content in bottom ash of kiln 1 increased after the addition of moistened MSWI fly ash by about 75% (+3700 mg/kg) (see Table 3). Apart from that, no changes in the composition of the bottom ash, significantly deviating from long time mean values (see Supplementary information 1), were observed, still allowing the disposal of bottom ashes from the hazardous waste incinerator at non-hazardous waste landfills with regard to total element contents. The rotary kiln fly ash from kiln 1 showed concentrations of Cd, K and Pb by 81 mg/kg (+54%), 19,500 mg/kg (+57%) and 1660 mg/kg (+22%), respectively, higher compared to kiln 2. However, this increase is within the long time fluctuations.

The chemical composition of the leachates for composite samples of the bottom ash and the bottom ash fly ash mixture (without thermal treatment, cf. Section 2.6) are summarised in Table 4. No significant difference in the leachability of the investigated elements in the bottom ash of both kilns could be observed. Also in comparison to the long term averages (see Supplementary information 1) no significant impact by co-treatment of moistened MSWI fly ash at kiln 1 on the bottom ash quality in terms of leachability of substances was found. This was surprising as the vast majority of the inserted MSWI fly ash was transferred to the kiln's bottom ash. In contrary, the leachate of the mixture of rotary kiln bottom ash and moistened fly ash without thermal treatment, made as described in Section 2.6, does not comply with the limits for non-hazardous waste landfills because of the high content of total dissolved solids. Although the other parameters analysed comply with the limit values for disposal as non-hazardous waste, the leachability of Cr, Mo and Zn is significantly higher compared to the bottom ash of kiln 1 (co-treatment of moistened fly ash).

Besides composite samples representative for the entire experimental period, also daily composite samples were made and analysed for their elemental composition. The results of these analyses for selected elements are illustrated in Figs. 4 and 5. Data for elements other than the ones shown in Figs. 4 and 5 are given in Supplementary information 2.

The increasing rate of MSWI fly ash treatment during the experimental period of 5 days is not reflected in the composition of the bottom ash of kiln 1. Heavy metal contents of the bottom ash during the days 4 and 5 (higher rate of fly ash addition) are almost identical to ones observed for the first two days of the experiment, when lower quantities of fly ash have been treated. In general, results for the daily composite samples reveal a slightly varying composition of the bottom ash regardless fly ash has been treated (kiln 1 – Fig. 4) or not (kiln 2 – Fig. 5).



*The mass of rotary kiln fly ash could not be determined separately for the two kilns. As discussed above, almost all inserted wet fly ash dry matter was transferred to the bottom ash. Therefore the total amount of rotary kiln fly ash was equally assigned to both rotary kilns.

**The mass of organics and water transferred to the flue gas was calculated as total input mass less bottom ash and rotary kiln fly ash mass generated

Fig. 3. Material flows of hazardous waste and inserted fly ash referred on 1 Mg hazardous waste used in kiln 1.

3.3. Transfer coefficients for elements present in the MSWI fly ash treated

The ratio of the input of different elements into rotary kiln 1 via hazardous waste and via inserted moistened MSWI fly ash is illustrated in Fig. 6. The amount inserted via MSWI fly ash compared to the amount inserted via hazardous waste is especially high (>60%) for Ca, Cd, K, Sb, and Se. On the other hand, >80% of the overall input of Ba, Cr, Cu, Fe, Hg, Mo, and Ni into kiln 1 originate from hazardous waste.

The mass flows and transfer coefficients of Ag, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mo, Na, Ni, Pb, Sb, Se, Sn, Zn, Cl and S in the rotary kiln fly and bottom ashes were calculated as described in Section 2.5. The resulting transfer coefficients are shown in Fig. 7. The transfer coefficients plus their respective expanded uncertainties (cf. Section 2.5) are presented in Fig. 8.

For most heavy metals considered the total mass found in the different outputs of kiln 1 was below the mass inserted via hazardous waste and moistened MSWI fly ash. This observation might most likely be explained by the limited representativeness of the bottom ash samples with respect to heavy metal contents.

When attempting to trace the substance flows inserted via MSWI fly ash, it was shown that for major elements present in inserted MSWI fly ash, like Ca and Na, about 80% of the mass could be detected in the output streams. For S, only about 20% of mass inserted via fly ash was found in the different outputs of kiln 1.

Table 3
Total contents of elements of rotary kiln fly ash and bottom ash generated during the experiment (in mg/kg dry matter).

	Rotary kiln fly ash		Bottom ash		Limit for acceptance at non-hazardous waste landfills ("Reststoffdeponie") in Austria
	Kiln 1	Kiln 2	Kiln 1	Kiln 2	
Ag	78.3	67.6	17.2	13.5	
As	68.9	98.8	31.5	29.7	5000
Ba	330	419	7920	10,300	
Ca	64,900	40,300	114,000	84,300	
Cd	233	152	<0.36	<0.36	5000
Co	68.1	62,8	68.8	77.2	
Cr	445	449	688	1140	
Cu	1620	1820	1730	2360	
Fe	18,900	19,500	84,300	85,000	
Hg	0.199	1.65	0.344	<0.06	20
K	53,700	34,200	9960	5960	
Mo	155	191	65.4	118	
Na	129,000	158,000	20,700	15,000	
Ni	191	181	232	224	
Pb	9380	7720	201	137	
Sb	795	701	110	88,8	
Se	3.13	5.04	1.55	0.579	
Sn	1380	1140	416	430	
Zn	31,200	37,400	3450	2720	
Cl	61,100	101,000	8750	5030	
S	210,000	173,000	4870	4160	

Table 4
Leachable content of elements in leachate from bottom ash of kiln 1 and 2 (in mg/kg dry bottom ash).

	Bottom ash		Bottom ash - fly ash mixture (without thermal treatment)	Limit for acceptance at non-hazardous waste landfills in Austria
	Kiln 1	Kiln 2		
pH	12.11	12.34	11.95	6–13
Total dissolved solids	7950	7050	96,400	60,000
Cd	<0.018	<0.018	<0.018	1
Cr	<0.06	<0.06	3.66	10
Hg	0.02	0.02	0.02	0.1
Mo	3.19	2.27	4.6	10
Ni	<0.033	<0.033	<0.033	10
Pb	0.74	0.77	<0.36	10
Sb	<0.021	<0.021	0.05	0.7
Zn	1.10	0.98	3.30	50

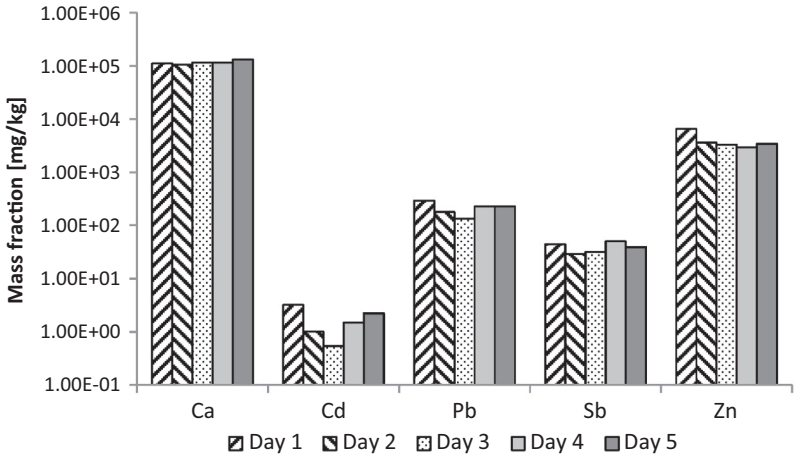


Fig. 4. Temporal variation of bottom ash composition in rotary kiln 1 (with fly ash addition) based on daily composite samples (total contents given in mg/kg).

However, as the dominating sulphur compound in inserted fly ash is $CaSO_4$ (Mahieux et al., 2010), which decomposes at temperatures prevailing in the kiln (>850 °C) (Holleman et al., 2007), most of sulphur will be transferred to the raw flue gas and removed in the second scrubber stage, which has not been analysed in the frame of this study. This might explain the mismatch of sulphur in the input and output flows.

If major amounts of heavy metals present in the inserted fly ash were transferred to the rotary kiln fly ash, this would lead to an enrichment of heavy metals, which was not observed. The vast

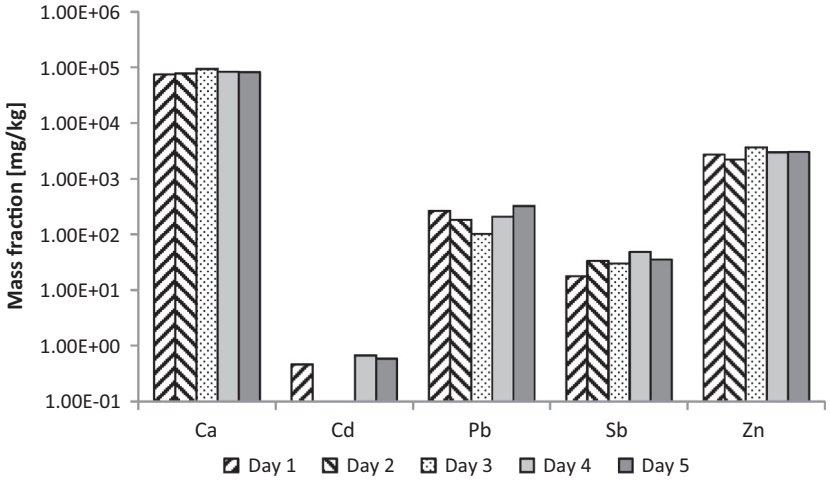


Fig. 5. Temporal variation of bottom ash composition in rotary kiln 2 (without fly ash addition) based on daily composite samples (total contents given in mg/kg).

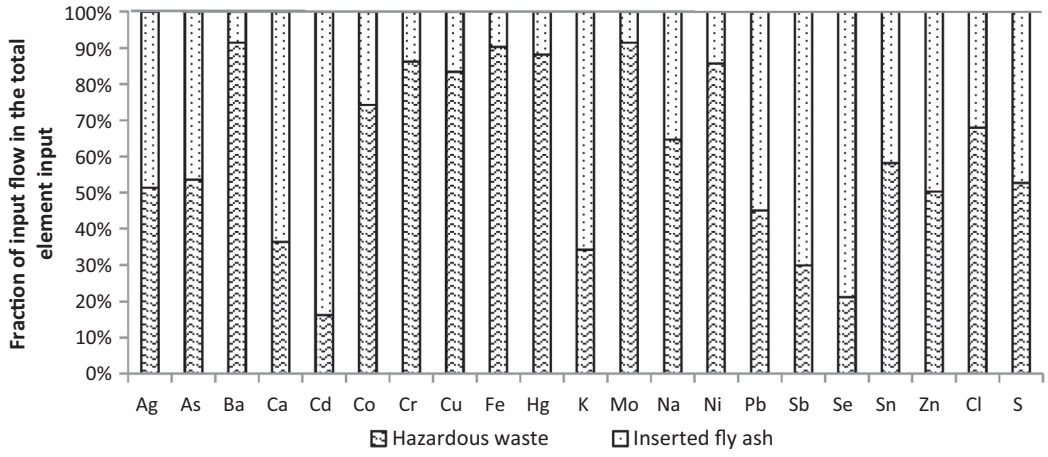


Fig. 6. Ratio of input into rotary kiln 1 via hazardous waste and wet MSWI fly ash.

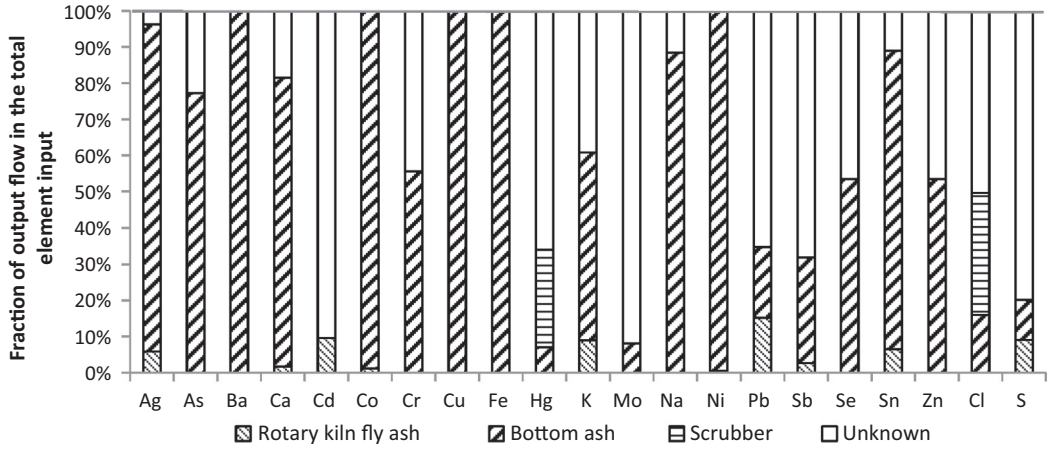


Fig. 7. Partitioning of the elements contained in the inserted MSWI fly ash treated to the different outputs of kiln 1.

majority of the heavy metals inserted via fly ash were transferred into the bottom ash (as indicated in Figs. 7 and 8). Hence, co-treatment of moistened fly ash together with hazardous waste in a rotary kiln has only minor effects on the composition of rotary kiln fly ash. The observed transfer of Hg into scrubber 1 is desired, as this way Hg can be phased out via the filter cake generated at the on-site wastewater treatment plant.

3.4. Impacts on operation conditions

It was possible to maintain continuous operation of the kiln during the time of the experiment without any major negative impacts arising from the MSWI fly ash addition. However, hydration reactions taking place in the moistened MSWI fly ash during storage in the waste bunker caused a large lump of several Mg

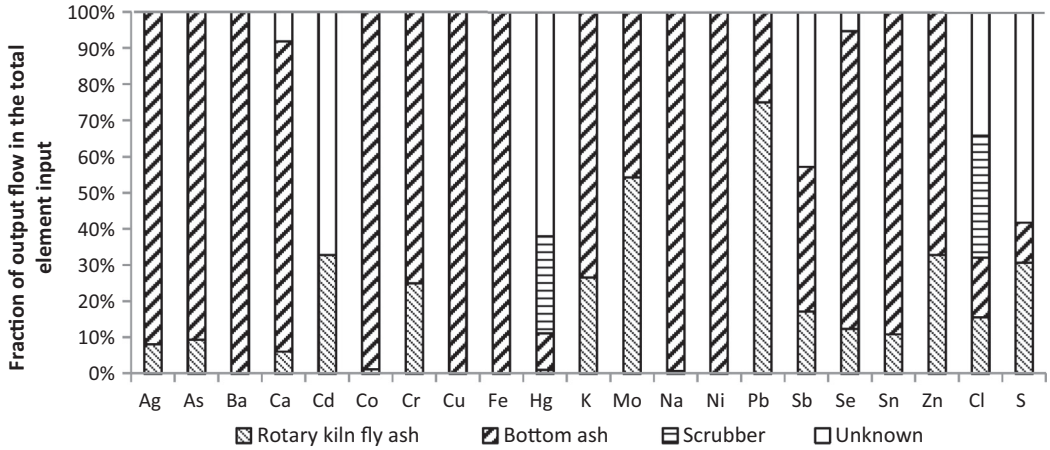


Fig. 8. Partitioning of the elements contained in the inserted MSWI fly ash treated to the different outputs of kiln 1 (including expanded uncertainty calculated according to Joint Committee for Guides in Metrology (2008)).

which had to be destroyed mechanically and removed. This confirms the finding of Billen et al. (2014) that MSWI fly ash shows significant cementitious properties. An additional challenge for the safe operation of the hazardous waste incinerator is the temperature increase in the bunker due to exothermal reaction of MSWI fly ash hydration. This temperature increase also leads to partial evaporation of the water used to moisten the inserted fly ash. Although the evaporation rate could not be measured, it is estimated that only a very limited amount of water evaporates during the residence time in the waste bunker (about 24 h) because of the small area available for evaporation and the high relative humidity in the waste bunker. This was confirmed by an approximate calculation (see Supplementary information 3). Apart from that, a slight increase in corrosive species like Cl in the raw flue gas of kiln 1 was observed (+23%) in comparison to kiln 2, which might cause a higher risk of corrosion for all parts in contact with the raw gas. However, the concentration increase was far below the temporal variations in raw gas composition observed during ordinary operation of the hazardous waste incinerator. The concentration of CO, NO_x and SO_2 in the cleaned flue gas was not affected by the addition of moistened MSWI fly ash.

4. Conclusions

The results of the study clearly demonstrate the general feasibility of a co-treatment of moistened MSWI fly ash and hazardous waste in a rotary kiln hazardous waste incinerator. More than 90% of the inserted MSWI fly ash was transferred to the bottom ash of the rotary kiln, whose quality was not impaired with regard to non-hazardous waste landfills disposal criteria.

As the heavy metal contents of the leachate of bottom ash generated from the co-treatment of moistened MSWI fly ash is considerably lower compared to the leachate of a mixture of rotary kiln bottom ash and MSWI fly ash without thermal treatment, it can be concluded that the thermal treatment goes along with an immobilisation of harmful heavy metals.

To ensure that this immobilisation takes place also under different operation conditions of the rotary kilns, additional studies are necessary to investigate the co-treatment of hazardous waste and MSWI fly ash at different temperatures and speeds.

The enrichment of heavy metals in the rotary kiln fly ash, observed during the experiment, was much lower than expected. Only the concentration of Cd, K and Pb in the rotary kiln fly ash generated increased after co-treatment of moistened fly ash by 54%, 57% and 22%, respectively. To which extent this unexpected observation results from varying and unequal composition of the hazardous waste fed into both kilns cannot be quantified. Thus, further investigations into the fate of heavy metals in a long-term trial are suggested.

Because of the hydration reactions taking place in moistened fly ash, it is suggested to keep the residence time of the inserted fly ash in the waste bunker as low as possible and to mix the fly ash with the hazardous waste present in the bunker and not to separately store the moistened fly ash as was done during the experiment. The addition of setting retarders known from concrete technology to moistened fly ash might also reduce or at least retard the setting of the fly ash and the therewith associated heat generation.

As a consequence of the lower energy demand and the lower volume of solid residues to be finally disposed of at landfills compared to stabilisation with cement, the thermal co-treatment of moistened MSWI fly ash and combustible hazardous waste in a rotary kiln appears to be an ecologically advantageous alternative to current fly ash management practices (Huber et al., 2016). As the use of already existing facilities (hazardous waste incinerator and conditioning drum to moisten the MSWI fly ash) does not cause investment costs, this treatment option could also be economically promising.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2016.09.013.

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Combined disc pelletisation and thermal treatment of MSWI fly ash

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ABSTRACT

An environmentally friendly and cost efficient way for the management of municipal solid waste incineration (MSWI) fly ash represents its thermal co-treatment together with combustible waste. However, the safe introduction and storage of MSWI fly ash in the waste bunker is challenging and associated with severe problems (e.g. dust emissions, generation of undefined lumps and heat in case of moistened MSWI fly ash). Therefore, the aim of this study is to investigate the suitability of pelletisation as a pretreatment of MSWI fly ash. In particular, MSWI fly ash was characterised after sampling, pelletisation and thermal treatment and the transfer of constituents to secondary fly ash and flue gas was investigated. For this purpose, MSWI fly ash pellets with a water content of about 0.15 kg/kg and a diameter of about 8 mm have been produced by disc pelletiser and treated in an electrically heated pilot-scale rotary kiln at different temperatures, ranging from 450 °C to 1050 °C. The total contents of selected elements in the MSWI fly ash before and after thermal treatment and in the generated secondary fly ash have been analysed in order to understand the fate of each element. Furthermore, leachable contents of selected elements and total content of persistent organic pollutants of the thermally treated MSWI fly ash were determined. Due to the low total content of Hg (0.7 mg/kg) and the low leachate content of Pb (<0.36 mg/kg), even at the lowest treatment temperature of 450 °C, thermally treated MSWI fly ash pellets can be classified as non-hazardous waste. However, temperatures of at least 650 °C are necessary to decrease the toxic equivalency of PCDD/F and DL-PCB. The removal of toxic heavy metals like Cd and Pb is significantly improved at temperatures of 850 °C, 950 °C or even 1050 °C. The observed metal removal led to relatively high contents of e.g. Cu (up to 11,000 mg/kg), Pb (up to 91,000 mg/kg) and Zn (up to 21,000 mg/kg) in the secondary fly ash. This metal enriched secondary fly ash might represent a potential raw material for metal recovery (e.g. via acidic leaching). Due to the high content of total dissolved solids observed in the leachate of thermally treated MSWI fly ash pellets, a wet extraction procedure is suggested to enable its safe disposal at non-hazardous waste landfills.

1. Introduction

Incineration is an effective option for the management of MSW (municipal solid waste), as it decreases the mass and volume of material that has to be disposed of at a landfill and enables the utilisation of energy from MSW for the generation of electricity or district heating. Apart from bottom ash, which accounts for about 25% of the MSW mass inserted into a grate furnace, about 3% of the inserted mass emerges as fly ash (Morf et al., 2000). Fly ash can be defined as "*particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material*" (Chandler et al., 1997) and constitutes a hazardous waste that has to be handled appropriately.

State of the art management options for MSWI (municipal solid waste incineration) fly ash comprise disposal at underground deposits (Doka, 2003; Quina et al., 2008) or, after stabilisation with cement, disposal at non-hazardous waste landfills (Polettini et al., 2001; Quina et al., 2008). Both options are costly and require landfill space (either underground or above ground). In case of stabilisation, a considerable amount of cement is necessary, resulting in emissions from cement production (Salas et al., 2016). Consequently, in recent years several studies investigating the thermal treatment of MSWI fly ash as an alternative management option have been conducted (Guohua et al., 2012; Li et al., 2015; Sobiecka, 2015; Sobiecka and Szymanski, 2014; Wang et al., 2008, 2015; Zupanič et al., 2012).

However, due to their high energy demand the above-mentioned thermal treatment processes fail to represent a cost-effective

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treatment and disposal option (Huber et al., 2017), which has so far impaired the wide-scale implementation of thermal MSWI fly ash treatment. Lately, a thermal co-treatment of MSWI fly ash together with combustible hazardous waste in a rotary kiln incinerator was suggested (Huber et al., 2016). In this previous study, MSWI fly ash from a grate furnace was moistened with water to avoid dust emissions during transport. Subsequently, the ash was unloaded into the bunker of a rotary kiln incineration plant and treated in the kiln at temperatures of 850 °C. Almost the entire MSWI fly ash mass inserted into the rotary kiln was transferred to the bottom ash of the rotary kiln incinerator. Volatile heavy metals like Hg and Cd were mainly transferred to the scrubber water and rotary kiln fly ash, respectively, and thereby phased out. Despite the transfer of considerable amounts of non-volatile heavy metal compounds from MSWI fly ash to rotary kiln bottom ash, the bottom ash still complies with the legal limits for non-hazardous waste landfills in Austria (Huber et al., 2016) and the environmental impact of this co-treatment is lower than that of MSWI fly ash stabilisation with cement and thermal treatment in a separate furnace (Huber et al., 2018).

Nevertheless, the previous studies mentioned above do not reveal, if thermal treatment of MSWI fly ash is sufficient to create a material that fulfils the criteria for disposal at non-hazardous waste landfills. So far only the mixture of thermally treated MSWI fly ash and rotary kiln bottom ash (generated from hazardous waste incineration) was analysed, as both residues arise together. The rotary kiln fly ash constitutes a hazardous waste anyway independent of the thermal co-processing of MSWI fly ash. Another shortcoming of the previous work is that large lumps of hardened MSWI fly ash were generated in the waste bunker due to setting reaction taking place in moistened fly ash (similar to cement paste) and the temperature in the waste bunker locally increased due to these exothermic reactions, which might pose a safety risk. Neither the hardening of MSWI fly ash nor the temperature increase is acceptable in routine operation. One possible solution for these two problems could be the agglomeration of MSWI fly ash prior to its thermal co-treatment together with combustible waste.

Agglomeration of MSWI fly ash in order to obtain particles of a larger size is already known in the literature. Colangelo et al. (2015) applied a double step cold bonding pelletisation with cement addition to MSWI fly ash in order to produce lightweight aggregates for construction purposes. Nowak et al. (2010) already combined the agglomeration of MSWI fly ash with thermal treatment, but they used a flat die press for agglomeration instead of a pelletising disc and added $CaCl_2$ to the ash.

The objective of the present work is, therefore, the assessment of combined disc pelletisation and thermal treatment of MSWI fly ash without addition of further substances in pilot-scale experiments. The particular research questions to be addressed are:

- Which operation conditions are necessary during pelletisation in order to achieve pellets best suitable for further processing?
- What is the chemical composition of thermally treated MSWI fly ash pellets (total and leachable element contents)?
- How do the MSWI fly ash pellets and their constituents partition between treated MSWI fly ash pellets, dust carried by the flue gas during thermal treatment (secondary fly ash) and flue gas?
- How do the operational conditions of the thermal treatment influence the mass and composition of treated MSWI fly ash pellets and secondary fly ash?

2. Materials and methods

2.1. MSWI fly ash

The fly ash used for the investigation was collected at a MSW incinerator with grate furnace. The air pollution control system at this plant comprises an activated coke injector, fabric filters, a two-stage scrubber and a selective catalytic reduction device. About 1000 kg of MSWI fly ash were collected and stored in big bags prior to use. Six random samples were taken from this MSWI fly ash batch and analysed for total content and leachate content. The fly ash from this plant generally exceeds the legal limits for non-hazardous waste landfills in Austria concerning the parameters Hg total content, total dissolved solids as well as and Pb in the leachate.

2.2. Pelletisation

The pelletisation process is illustrated in Fig. 1. MSWI fly ash was mixed and moistened with water in a ploughshare mixer and subsequently pelletised using a pilot scale pelletising disc with a diameter of 1.2 m. Additional water was sprayed onto the MSWI fly ash as needed and no other additives were used.

The mass flows of MSWI fly ash and water were varied in order to find suitable operation conditions (no dust emissions and acceptable consistency of the pellets). At a mass flow of 300 kg/h a batch of about 400 kg of pellets with a water content of about 0.15 kg/kg and an average diameter of about 8 mm was produced. The pellets were filled into big bags and aged until the experiments on thermal treatment were conducted. Six random samples were taken from this pelletised MSWI fly ash batch and analysed for total and leachate contents. Additionally, the mechanical properties of the MSWI fly ash pellets were measured.

2.3. Thermal treatment

2.3.1. Thermal treatment in a muffle furnace

Pelletised fly ash was filled into corundum crucibles and weighted on an analytical balance. Filled crucibles were heated in a muffle furnace at 450 °C, 750 °C and 1050 °C for 10 min, 30 min and 60 min. After thermal treatment the mass of the pelletised fly ash was measured and the chemical composition of the treated fly ash pellets was determined as described in 2.4. A concentration factor was calculated for each sample (corresponding to a specific temperature and residence time) by dividing the mass of the pellets prior to thermal treatment by the mass of the pellets after thermal treatment. The total content of selected elements in the fly ash pellets before and after thermal treatment was used to find suitable tracer elements that are not transferred to the gas phase. This is the case when the ratio of the concentration of the element after thermal treatment to the concentration before thermal treatment is identical to the concentration factor determined for the sample mass.

2.3.2. Thermal treatment in a rotary kiln

Thermal treatment was conducted in an electrically heated pilot-scale rotary kiln at different temperatures (450 °C, 550 °C, 650 °C, 750 °C, 850 °C, 950 °C and 1050 °C) and angles (2°, 3° and 6°). The higher the angle of the rotary kiln the lower was the residence time of the pelletised MSWI fly ash. The heated part of the rotary tube was 1.5 m long (total length 2.0 m). The rotary tube had an inner diameter of 0.2 m, consisted of a high temperature nickel/chromium alloy (Nicrofer 6025 HT) and was rotating at 2 min^{-1}. Pressurised air was injected into the kiln with a relative flow of about 1 Nm^3/kg MSWI fly ash pellets and the flue gas from the kiln was sucked into a scrubber prior to release into the atmosphere. Pelletised MSWI fly ash was transported into the kiln by a conveyer screw rotating at constant speed. At 1050 °C it was only possible to conduct an experiment at an angle of 6° because of deformation of the conveyer screw that occurred at high temperatures and long treatment times associated with angles of 2° and 3°.

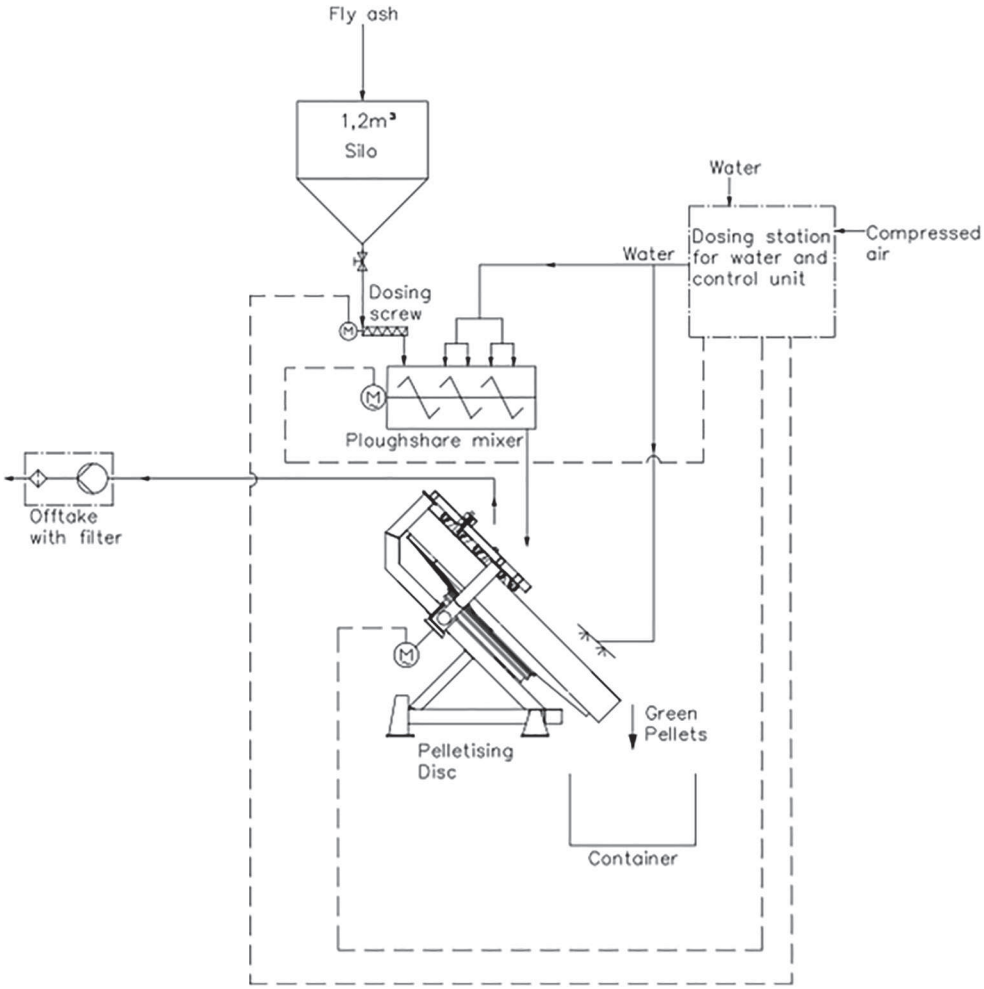

Fig. 1. Process scheme of the pelletisation process for MSWI fly ash.

The course of the experiments is illustrated in Fig. 2. Treated MSWI fly ash pellets were collected in a metal bucket placed on a balance in order to determine the mass flow into the bucket. The mass flow of treated MSWI fly ash pellets was determined every minute until a constant value was obtained (phase A). At this time an empty bucket was placed at the outlet of the rotary kiln and the conveying of MSWI fly ash pellets was continued until this second bucket was filled with about 1 kg of thermally treated pellets (phase B). Subsequently, a third bucket was placed at the outlet of the rotary kiln and, simultaneously, the rotation of the conveyer screw was stopped. Due to the continued rotation of the rotary kiln, the rotary kiln was slowly emptied and the MSWI fly ash pellets still in the kiln at the end of the phase B were collected in the third bucket (phase C). The residence time was calculated by dividing the mass of thermally treated MSWI fly ash pellets in the third bucket by the mass flow of fly ash pellets out of the kiln in phase B. The thermally treated MSWI fly ash pellets from phase B were analysed as described in 2.4.

During phase B, when the mass of MSWI fly ash pellets in the rotary kiln was constant, a depth filtration device with glass wool as filter media was mounted in the flue gas flow. In this manner, the mass of the dust that was carried out of the rotary kiln (secondary fly ash) could be determined. Furthermore, the chemical composition (total content) of the secondary fly ash was analysed.

The mass of thermally treated MSWI fly ash pellets and of secondary fly ash could be determined directly by weighing. However,

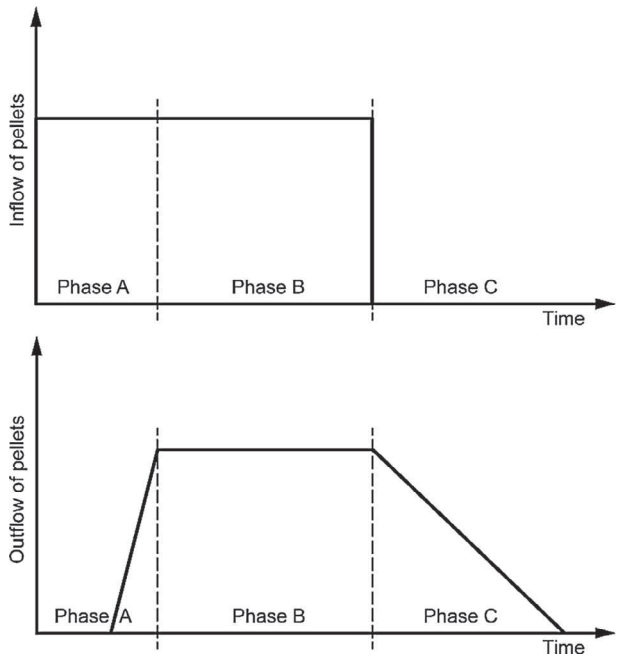

Fig. 2. Input and output flows of solid matter for the rotary kiln in the course of one experiment.

this was not possible for the input mass that left the rotary kiln as part of the gas phase. Therefore, in a first step, the total dry matter input mass of MSWI fly ash was calculated on the basis of a mass balance of a tracer element (i.e. an element that is not transferred to the gas phase as determined in 2.3.1) in the thermally treated MSWI fly ash and the secondary fly ash according to formula (1). The mass that left the rotary kiln as part of the flue gas was calculated according to formula (2). The mass of a selected element *i* that was transferred to the flue gas was calculated according to formula (3). Therefore, chemical analysis of the flue gas was not necessary.

$$m_I = \frac{c_P \cdot m_P + c_S \cdot m_S}{c_0} \quad (1)$$

$$m_G = m_I - m_P - m_S \quad (2)$$

$$m_{i,G} = m_I \cdot c_{i,0} - m_P \cdot c_{i,P} - m_S \cdot c_{i,S} \quad (3)$$

m_I – mass of MSWI fly ash pellet input [kg],
m_P – mass of thermally treated MSWI fly ash pellets [kg],
m_S – mass of secondary fly ash [kg],
m_G – mass of MSWI fly ash that is transferred to the flue gas and not captured by the filter [kg],
c_0 – concentration of tracer element in pelletised fly ash before thermal treatment [kg/kg],
c_P – concentration of tracer element in pelletised fly ash after thermal treatment [kg/kg],
c_S – concentration of tracer element in secondary fly ash [kg/kg],
$m_{i,G}$ – mass of element i that is transferred to the flue gas and not captured in the filter device [kg],
$c_{i,0}$ – concentration of element i in pelletised fly ash before thermal treatment [kg/kg],
$c_{i,P}$ – concentration of element i in pelletised fly ash after thermal treatment [kg/kg],
$c_{i,S}$ – concentration of element i in secondary fly ash [kg/kg].

2.4. Chemical and physical analysis

For the chemical analysis, total contents as well as the leachability of all MSWI fly ash samples (untreated powder and pellets as well as thermally treated pellets) were determined. For the total metals contents, samples were digested in aqua regia according to EN 13657 (2002) and subsequently analysed according to EN 11885 (2009) by a PerkinElmer Optima 8300 ICP-OES (inductively coupled plasma optical emission spectroscopy) spectrometer equipped with a SC-2 DX FAST sample preparation system. In general, non matrix-elements are determined via axial view and triple determination followed by an arithmetic averaging. A customised single-element (Merck, Roth) standard was used for the calibration.

Leachates were prepared using a liquid-to-solid ratio of 10 L/kg according to EN 12457-4 (2002) and analysed as above. To determine the total Cl content, the samples were milled to a grain size of 250 μm and subsequently analysed by XRF (X-ray fluorescence spectroscopy). All XRF analyses have been done with a NITON XL3t Air handgun, conducting the measurement in a lead-chamber and using the analysation-mode "TestAll Geo".

Different to the other samples analysed, the secondary fly ash samples on glass wool were heated in aqua regia until reaching reflux temperature and kept there for about 2 h (instead of following EN 13657 (2002)). The digestion was followed by filtration and the solution was subsequently analysed by ICP-OES according to EN 11885 (2009). Additionally, two blanks (glass wool without secondary fly ash) were analysed and considered in the calculation of the results.

For the analysis of the persistent organic pollutants PCDD/F (polychlorinated dibenzodioxines and furans), DL-PCB (dioxine like polychlorinated biphenyls), NDL-PCB (non-dioxine like polychlorinated biphenyls) and HCB (hexachlorobenzene), samples were digested in HCl solution (concentration 1 mol/L) in an ultrasonic bath for 10 min. PCDD/F, DL-PCB and NDL-PCB were analysed by GC-HRMS (gas chromatography-high resolution mass spectrometry) according to CEN/TS 16190 (2012). HCB was analysed by GCMS. The TEQ (toxic equivalency) was calculated for PCDD/F and DL-PCB according to WHO (2005). Three samples of untreated MSWI fly ash pellets and one sample of treated MSWI fly ash pellets for every temperature at an angle of 6° were used for organic pollutant analysis.

The particle size distribution of the MSWI fly ash pellets was determined by sieving of about 3 kg pellets with 7.1, 8, 9 and 10 mm sieves.

The compressive strength of the pellets was assessed by crushing a total of 30 pellets (near-mesh size pellets of an 8 mm sieve). Individual pellets were placed on a pressure sensor plate (Portable Precision USB Sensor Interface 9205) which was mounted on a hydraulic pellet press. The pressure sensor measured the required pressure to fracture the pellets. Compressive strength data were collected by the software DigiView (version 2012.2.1). Additionally, a drop test was conducted immediately after the production similar to Gul et al. (2015). Ten pellets were dropped from a height of 1 m onto a hard surface until they broke and the drop number was counted.

Four big bags filled with fresh pellets were stacked on top of each other (height about 1.5 m) and stored for several months to determine, if the MSWI fly ash pellets tend to stick together or if the spherical shape of the pellets is conserved even under considerable pressure.

3. Results and discussion

3.1. Properties of MSWI fly ash prior to thermal treatment

The chemical composition (total and leachate contents) of the original MSWI fly ash collected at the MSWI plant and the pelletised MSWI fly ash is summarised in Table 1.

Obviously, the total content of the unpelletised and the pelletised MSWI fly ash are substantially identical. Nevertheless, the pelletisation process has a significant influence on the leaching behaviour of fly ash. The leachate content of total dissolved solids, Ba, Hg, Mo, Pb and Zn were decreased while the leachate content of Cr was slightly increased by the pelletisation process. Especially the decrease of Pb in the leachate from 41.6 mg/kg to 0.78 mg/kg, which is even below the limit value for non-hazardous waste landfills in Austria (10 mg/kg) (DVO, 2008), is remarkable. However, the comparison of the MSWI fly ash sample used for the pelletisation and the average MSWI fly ash composition reveals that the sample used for the present study is not representative regarding the parameter Pb in the leachate, as the average value of the last four years was 10 times higher (see Table 1). Pelletised fly ash still exceeds the legal limits for total dissolved solids in the leachate (100,000 mg/kg) and the total content of Hg is still critical although slightly below the limit value of 20 mg/kg.

The particle size distribution of MSWI fly ash is given in Table 2. Almost half of the pellets had a diameter between 8 and 9 mm.

The average compressive strength was 131 N (standard deviation: 36 N) and the average drop number of the pellets was 10 (standard deviation 3) which is very high compared to other pellets made on pelletising discs (Gul et al., 2015). These values ensure

Table 1
Total and leachate contents of MSWI fly ash used for the experiments and average values for MSWI fly ash from the same MSWI plant for 4 consecutive years. SD...standard deviation.

	MSWI fly ash used for the pelletisation		Pelletised MSWI fly ash		Average (4 years) MSWI fly ash	
	Mean	SD	Mean	SD	Mean	SD
Total content [mg/kg dry matter]						
As	<3.6	–	<3.6	–	37	11
Ag	22.7	1.4	22.2	1.4	49.1	7.9
Ba	1212	27	1118	28	927	82
Cd	190.2	2.7	176	16	347	85
Co	30.5	5.8	30.3	5.8	24.5	9.7
Cr	296	21	271	22	245	52
Cu	1083	59	1027	31	1010	200
Fe	13,360	430	12,790	630	11,000	4000
Hg	14.02	0.81	14.81	0.81	12.5	5.1
Mo	11.6	1.0	10.96	0.56	24	15
Ni	55	23	44.6	9.2	59	15
Pb	2227	45	2140	110	4900	1300
Sb	682	29	677	17	740	180
Sn	396	18	400	14	950	170
Zn	13,930	190	13,260	860	20,700	6400
Leachate content [mg/kg dry matter, except for pH]						
pH	10.52	0.37	10.93	0.052	12.05	0.27
Total dissolved solids	232,000	32,000	174,200	5800	296,000	36,000
As	<0.18	–	<0.18	–	<0.1	–
Ag	<0.03	–	<0.03	–	<0.2	–
Ba	2.392	0.050	1.703	0.081	5.18	0.95
Cd	<0.018	–	<0.018	–	<0.2	–
Co	<0.09	–	<0.09	–	<0.2	–
Cr	1.33	0.14	4.97	0.41	2.7	1.3
Cu	<0.03	–	<0.03	–	0.20	0.19
Hg	0.0917	0.0041	0.0750	0.0055	<0.01	–
Mo	3.35	0.10	2.80	0.17	7.0	3.7
Ni	<0.033	–	<0.033	–	<0.2	–
Pb	41.6	6.3	0.78	0.31	520	260
Sb	<0.18	–	<0.18	–	<0.1	–
Se	0.49	0.13	0.49	0.11	<0.1	–
Sn	<0.123	–	<0.123	–	<0.2	–
Zn	18.4	1.3	8.9	3.0	25.8	9.7

Table 2
Particle size distribution of MSWI fly ash pellets.

Diameter [mm]	<7.1	7.1–8	8–9	9–10	>10
Mass fraction [%]	17.2	21.9	42.4	14.0	4.5

that the MSWI fly ash pellets are mechanically stable for further processing. A low mechanical strength could lead to breakage of the pellets and thereby increase the amount of secondary fly ash that is generated during thermal treatment. Furthermore, the break of pellets prior to their thermal treatment might results in emissions of hazardous dust during the handling of the pellets. The result of the stacking test revealed spherical pellets even at the bottom. Although some sticking was observed individual MSWI fly ash pellets could easily be separated from each other by hand.

3.2. Identification of suitable tracer elements

The mass ratio of MSWI fly ash pellets before and after treatment at 450 °C, 750 °C and 1050 °C was determined in the muffle furnace as 1.14, 1.16 and 1.21, respectively. The ratio of the concentration after and before thermal treatment in the muffle furnace was 1.05, 1.08 and 1.14, respectively, for Ba and 1.08, 1.12 and 1.14, respectively, for Ca. As the concentration ratios for these two elements are very close to the mass ratio, the input mass was calculated as described in 2.3.2 based on the Ba and Ca concentration. For all further calculations the mean values of these input mass values was used.

3.3. Chemical composition of thermally treated MSWI fly ash pellets and secondary fly ash

Analysis of the samples generated during the experiments with the rotary kiln revealed that significant amounts of Ag, Cd, Cu, Hg, Mo, Pb, Se and Cl were volatised by thermal treatment of pelletised MSWI fly ash, while Ba, Ca, Fe, K, Na, Sb, Sn and Zn remained to a large extent in the pellets. The contents of Cr, Co and Ni are increased in thermally treated fly ash pellets compared to untreated MSWI fly ash. This observation can be explained by parts of the kiln material that are transferred to the samples as a result of deterioration.

Fig. 3 shows the concentration of volatile elements in MSWI fly ash pellets treated at different temperatures. As no correlation between residence time and composition could be observed the mean values of the experiments at each temperature were used. If the measurement result was below the limit of quantification, half of the limit of quantification was used for the calculation of mean values. The total content of Hg was decreased to below 2 mg/kg already by treatment at 450 °C, while temperatures of at least 950 °C are necessary to volatise more than 50% of the Cd and Pb mass originally contained in pelletised MSWI fly ash. At 1050 °C the total content of Cd and Pb in thermally treated MSWI

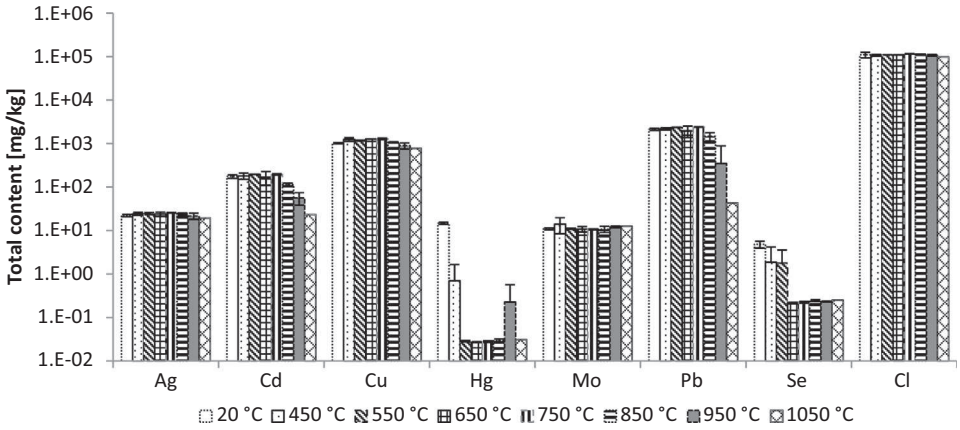

Fig. 3. Concentration of volatile constituents in MSWI fly ash pellets after treatment at different temperatures. The error bars show the standard deviation. Treatment temperature of 20 °C means that no thermal treatment was applied.

fly ash pellets is even as low as 23 mg/kg and 44 mg/kg, respectively.

The leachate content of Ba, Mo, Se and Zn is shown in Fig. 4. The concentration of Mo and Se in the leachate increases with higher treatment temperatures. A possible explanation could be an oxidation of Mo and Se to soluble molybdate, selenite and selenate compounds. The Cr content in the leachate is unexpectedly high because of Cr being transferred from the kiln material to the sample. Therefore it is not shown in Fig. 4. The elements Ag, As, Cd, Co, Cu, Hg, Ni, Pb, Sb and Sn could not be detected in the leachate of most of the thermally treated fly ash pellets, which means that their leachate content was decreased to close to 0. This is especially relevant for Pb, as this parameter of MSWI fly ash usually exceeds the limit values for non-hazardous waste landfills (10 mg/kg) (DVO, 2008) by a factor of more than 10.

As a consequence of the low Hg total content and Pb leachate content, the thermally treated pellets can be classified as non-hazardous waste (AVVO, 2008). On the other hand, the content of total dissolved solids in the leachate still exceeds the legal limit for non-hazardous waste landfills (100,000 mg/kg) (DVO, 2008).

The composition of secondary fly ash from thermal treatment at different temperatures is shown in Table 3. The higher the treatment temperature, the higher is the mass fraction of Ag, Cd, Cu, K and Na in the secondary fly ash. Several elements like Ca or Ba show a decreasing concentration in secondary fly ash with increasing treatment temperature, which means that they are not volatised. For yet another group of elements like Hg, Pb, Se or Zn the total content in secondary fly ash increases with rising temperature but decreases again at a certain point, e.g. Pb contents in secondary fly ash increase from 442 mg/kg at treatment temperatures of 450 °C to 91,000 mg/kg at 850 °C and decrease to 48,000 mg/kg in treatments at 1050 °C. A likely explanation for this behaviour is that at relatively low treatment temperatures these elements are volatised in the rotary kiln and subsequently condense on the filter media while at higher temperatures they condense to a lower extent on the filter media and are partly transferred to the scrubber. Secondary fly ash contains total contents of Cu up to 11,000 mg/kg (1050 °C), Pb up to 91,000 mg/kg (850 °C) and Zn up to 21,000 mg/kg (1050 °C), which might enable economically viable metal recovery, e.g. by applying the FLUWA or FLUREC process (Bühler and Schlumberger, 2010; Schlumberger, 2010).

The content of organic pollutants is shown in Table 4. OCDD has the highest concentrations of all organic compounds analysed but due to its relatively low toxicity has only a negligible contribution to the TEQ. The TEQ for PCDD/F was increased at 450 °C by a factor of almost 3 from 440 to 1200 ng/kg. This increase in toxicity is mainly caused by the formation of 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF and 2,3,4,6,7,8-HxCDF. The reason for this PCDD/F

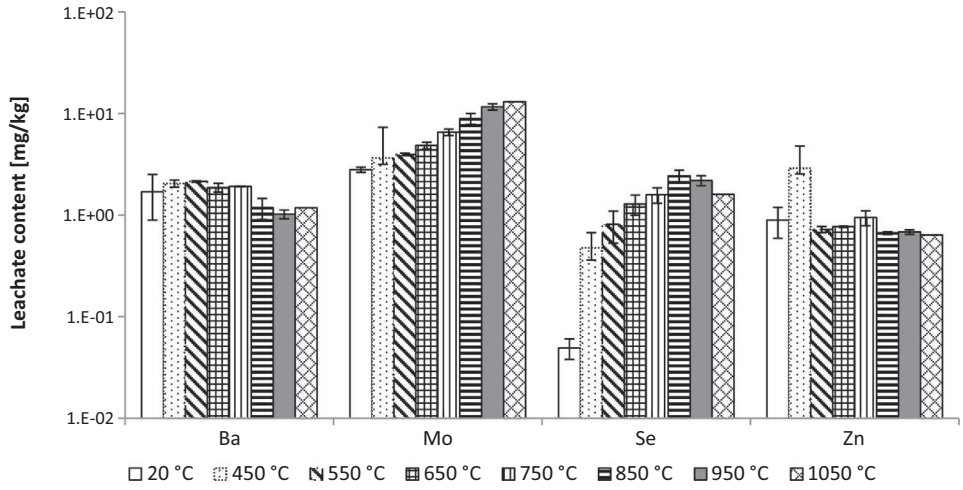

Fig. 4. Leachate contents of Ba, Mo, Se, Zn in MSWI fly ash pellets treated at different temperatures. The error bars show the standard deviation. Treatment temperature of 20 °C means that no thermal treatment was applied.

Table 3
Total contents in secondary fly ash generated at different treatment temperatures. As for 1050 °C no standard deviation could be given, two digits are shown as significant for treatment at this temperature. The concentration of Mo and Se in several samples (glass wool with secondary fly ash) was below the limit of quantification. SD...standard deviation.

T [°C]	450		550		650		750		850		950		1050
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean
Ag [mg/kg]	12.3	2.9	11.9	6.0	30	31	99	42	126	16	149	22	160
Ba [mg/kg]	910	150	650	370	970	90	181	60	73	61	36	24	23
Ca [mg/kg]	200,000	31,000	191,000	69,000	208,000	38,000	62,000	19,000	25,000	21,000	12,700	8400	7200
Cd [mg/kg]	247	30	267	87	529	25	2350	560	3820	550	3640	990	3500
Co [mg/kg]	15.8	1.6	19.8	8.0	25	17	5.5	2.6	<1.8	–	<1.8	–	<1.8
Cr [mg/kg]	990	650	470	330	2400	1400	390	150	234	72	550	310	280
Cu [mg/kg]	1160	120	980	360	1550	160	6100	2900	9000	1300	10,920	540	11,000
Fe [mg/kg]	8600	4600	5400	2000	11,300	7800	990	890	1330	330	700	370	1100
Hg [mg/kg]	160	110	140	51	24	19	23	23	12.3	8.1	2.04	0.70	<1.14
K [mg/kg]	47,500	6700	42,000	14,000	61,580	140	206,000	77,000	192,800	7500	193,000	20,000	190,000
Mo [mg/kg]	26	32	5.3	8.2	210	160	<0.66	–	<0.66	–	<0.66	–	<0.66
Na [mg/kg]	22,000	23,000	29,000	20,000	40,000	34,000	156,000	79,000	86,000	13,000	113,100	7600	140,000
Ni [mg/kg]	2900	1600	1350	990	9000	15,000	340	260	82	96	160	240	37
Pb [mg/kg]	2990	300	3600	1300	7800	6400	58,000	21,000	91,000	16,000	68,000	9500	48,000
Sb [mg/kg]	1110	120	1020	420	1080	350	186	82	16	28	<3.6	–	<3.6
Se [mg/kg]	<3.6	–	<3.6	–	<3.6	–	<3.6	–	<3.6	–	15.3	4.3	11
Sn [mg/kg]	622	76	580	250	803	26	126	29	56	42	26	20	13
Zn [mg/kg]	19,200	1000	16,600	5900	19,800	2200	8400	2500	10,800	1500	13,300	1400	21,000

Table 4
Content of persistent organic pollutants in MSWI fly ash pellets treated at different temperatures. Treatment temperature of 20 °C means that no thermal treatment was applied. SD...standard deviation.

Temperature [°C]		20 (mean)	20 (SD)	450	550	650	750	850	950	1050
Polychlorinated dibenzodioxins and dibenzofurans [ng/kg]	TEQ PCDD/F	440.00	36.06	1200	5.9	3	1.9	1.3	1.2	1.4
	2,3,7,8-TCDD	38.67	2.52	9.5	<0.54	<0.54	<0.54	<0.54	<0.54	<0.54
	1,2,3,7,8-PeCDD	121.67	23.63	140	<0.56	<0.56	<0.56	<0.56	<0.56	<0.56
	1,2,3,4,7,8-HxCDD	120.00	10.00	320	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
	1,2,3,6,7,8-HxCDD	183.33	5.77	130	<0.60	0.67	0.64	<0.60	<0.60	0.79
	1,2,3,7,8,9-HxCDD	170.00	20.00	170	1.3	2.1	1.3	<0.61	<0.61	<0.61
	1,2,3,4,6,7,8-HpCDD	1800.00	100.00	1500	<11	<11	<11	<11	<11	<11
	OCDD	6700.00	655.74	3300	<3.6	35	<3.6	<3.6	<3.6	<3.6
	2,3,7,8-TCDF	243.33	5.77	290	6.2	<0.19	<0.19	<0.19	<0.19	<0.19
	1,2,3,7,8-PeCDF	286.67	15.28	520	9.7	2.9	<0.23	0.36	<0.23	<0.23
	2,3,4,7,8-PeCDF	230.00	10.00	760	7.3	2.2	1.5	0.8	<0.34	<0.34
	1,2,3,4,7,8-HxCDF	323.33	15.28	2000	5.8	2.9	1.5	1	<0.28	0.64
	1,2,3,6,7,8-HxCDF	313.33	20.82	1300	4.8	3.6	1.2	<0.35	<0.35	0.9
	2,3,4,6,7,8-HxCDF	276.67	23.09	1800	0.49	1.7	0.7	<0.26	<0.26	0.78
	1,2,3,7,8,9-HxCDF	40.67	2.08	180	<0.85	<0.85	<0.85	<0.85	<0.85	<0.85
	1,2,3,4,6,7,8-HpCDF	1060.00	69.28	13,000	12	7.6	9.4	2.9	<0.31	9.2
	1,2,3,4,7,8,9-HpCDF	143.33	15.28	750	1.3	1.4	0.83	<0.17	<0.17	0.19
	OCDF	1010.00	81.85	5500	<3.5	18	16	<3.5	<3.5	<3.5
Dioxin like polychlorinate biphenyls [ng/kg]	TEQ DL-PCB	5.70	1.51	26	9.4	0.12	1.1	0.049	0.057	0.049
	PCB 77	44.23	33.67	280	280	<0.13	82	68	91	75
	PCB 81	1.73	3.00	210	330	<0.15	<0.15	<0.15	<0.15	<0.15
	PCB 126	47.67	9.07	240	90	<0.13	9.7	<0.13	<0.13	<0.13
	PCB 169	28.67	20.74	45	7.8	0.86	5	<0.31	<0.31	<0.31
	PCB 105	283.67	227.53	120	<24	240	280	190	270	200
	PCB 114	16.67	14.47	27	<11	19	<11	<11	23	<11
	PCB 118	1973.33	1431.83	130	260	1800	340	350	470	360
	PCB 123	90.33	52.35	67	89	<3.3	38	36	<3.3	<3.3
	PCB 156	358.00	246.76	87	55	310	40	43	51	43
	PCB 157	27.67	18.45	35	<3.3	16	8.7	6.7	13	<3.3
	PCB 167	237.67	159.93	43	36	230	18	16	14	9.4
	PCB 189	20.00	7.81	41	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Non-dioxin like polychlorinate biphenyls [ng/kg]	Sum NDL-PCB	6246.67	1506.40	1747	5430	0	2583	3077	4600	2010
	PCB 28	<25	–	36	<25	<25	450	460	710	420
	PCB 52	106.67	184.75	81	310	<25	260	490	640	320
	PCB 101	1166.67	152.75	350	1300	<25	590	710	1100	430
	PCB 138	1833.33	577.35	480	1500	<25	660	710	830	420
	PCB 153	2633.33	838.65	600	2000	<25	540	630	1100	300
	PCB 180	506.67	166.23	200	320	<25	83	77	220	120
[μg/kg]	Hexachlorobenzene	<10	–	10	<10	<10	<10	<10	<10	<10

formation is most likely de novo synthesis, as 450 °C is still in the temperature window for this process (Huang and Buekens, 1996). However, PCDD/F are destroyed mostly at 550 °C and above. DL-PCB show a similar pattern like PCDD/F. Here the TEQ is increased at 450 °C and to a lesser extent also at 550 °C mainly due to the formation of PCB 126. The toxicity of DL-PCB decreases at

temperatures of 650 °C and above. The sum of NDL-PCB is roughly in the same order of magnitude for all samples, although the NDL-PCB pattern changes as a result of thermal treatment. The concentration of PCB 28 and PCB 52 increases at higher temperatures while the concentration of PCB 101, PCB 138, PCB 153 and PCB 180 decreases. No concentration of HCB higher than 10 μg/kg (limit of quantification) could by detected in any of the samples analysed. No data about the concentration of organic pollutants in the flue gas is given. However, as PCDD/F are destroyed mostly at 550 °C and above, it can be presumed that no organic pollutants are present at these treatment temperatures. Even if minor amounts of organic pollutants were present in the flue gas, they could be removed by an activated coke adsorber in the air pollution control system. Such adsorbers are used in state of the art waste incineration plants.

3.4. Mass flows of thermally treated MSWI fly ash pellets and secondary fly ash

The ratio of secondary fly ash to treated MSWI fly ash pellets at different temperatures is shown in Fig. 5. The higher this ratio the more mass is volatised during thermal treatment of pelletised MSWI fly ash. Up to a temperature of about 800 °C that ratio of secondary fly ash to treated fly ash is essentially constant and at temperatures higher than about 800 °C a linear increase could be observed. This increase is caused mainly by volatilisation of K, Na, Pb and Zn as shown in Fig. 6. No correlation between mean residence time and secondary fly ash generation could be found.

3.5. Transfer of selected elements from pelletised MSWI fly ash to thermally treated pellets, secondary fly ash and flue gas

The transfer coefficients from pelletised MSWI fly ash to thermally treated pellets, secondary fly ash and flue gas at different treatment temperatures for the elements Ag, Cd, Cu, Hg, K, Mo, Na, Pb and Zn are shown in Fig. 7. The transfer coefficients for Ba and Ca are not shown as these two elements were used as tracers (c.f. 2.3). Cr, Co, Ni, Sb and Sn are not shown as they are not transferred to secondary fly ash or flue gas. As and Se are not shown as their concentration in secondary fly ash is too low to establish reliable mass balances. For some elements, the transfer coefficient to flue gas fluctuates at different treatment temperatures. This can most likely be explained by the standard deviation of these elements at certain temperatures.

Fig. 7 shows that the transfer coefficient to flue gas is significantly higher than the transfer coefficient to secondary fly ash for almost all elements and temperatures. As a result, the transfer coefficients determined in the present study differ significantly from the transfer coefficients determined by Huber et al. (2016). This can be explained by the different experimental setup. The flue gas in the experiments of Huber et al. (2016) was cooled in the boiler prior to solids separation in the electrostatic precipitator,

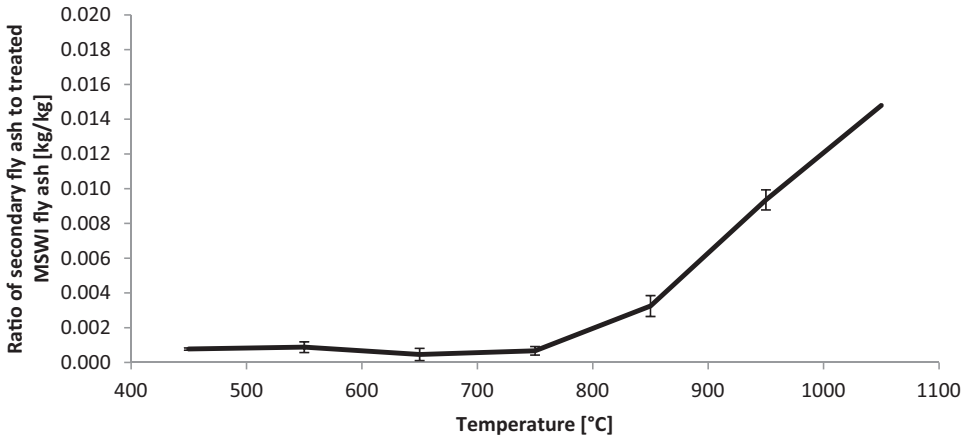

Fig. 5. Ratio of secondary fly ash to MSWI fly ash pellets treated at different temperatures. The error bars show the standard deviation.

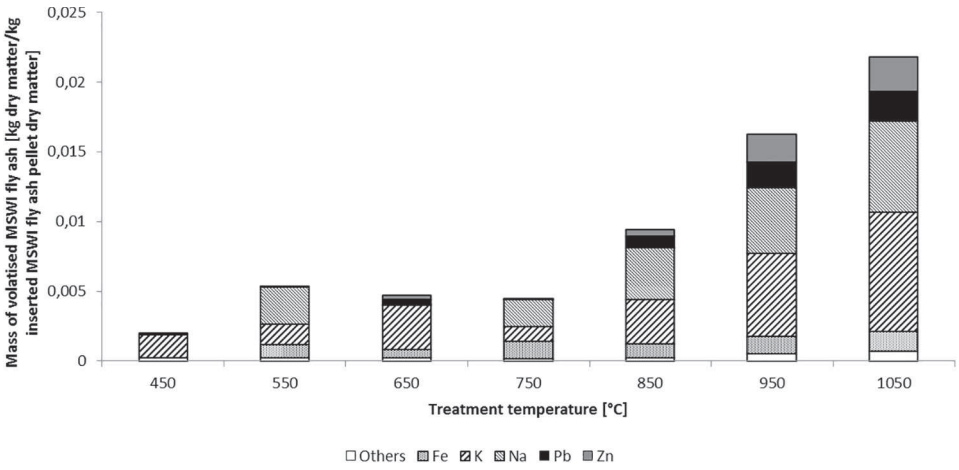

Fig. 6. Mass and composition of volatised MSWI fly ash determined from the transfer coefficients calculated and the composition of MSWI fly ash pellets.

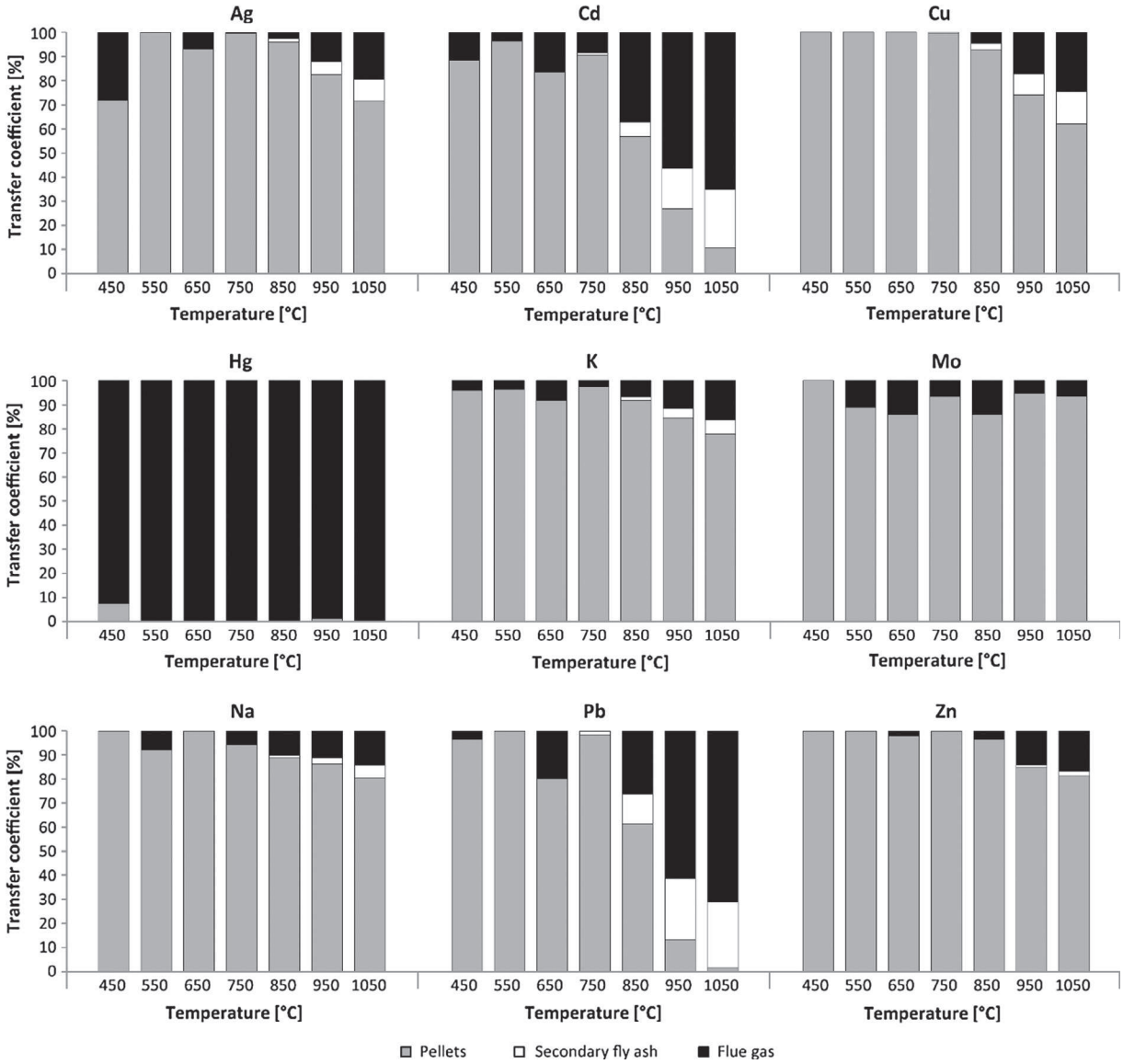

Fig. 7. Transfer coefficients of single elements from pelletised MSWI fly ash to thermally treated pellets, secondary fly ash and flue gas at different treatment temperatures.

allowing volatile heavy metals to condensate on the secondary fly ash, while no cooling of the flue gas was applied in the present study. Due to this condensation almost no metals (except Hg) were present in the flue gas after the electrostatic precipitator in Huber et al. (2016).

Furthermore, in Huber et al. (2016) MSWI fly ash was treated together with combustible hazardous waste with the assumption that the composition of hazardous waste in the rotary kiln used for the experiment and the reference rotary kiln is identical. However, this assumption is not true. Another difference between co-treatment together with combustible hazardous waste and treatment of MSWI fly ash alone is the different composition of the atmosphere in the kiln. As this can have a significant effect on the volatilisation of heavy metals (Fraissler et al., 2009; Nowak et al., 2010) and because the hazardous waste used for the two different rotary kilns in Huber et al. (2016) is not identical as assumed, some effects like the volatilisation of Cu that are shown in Fig. 7 were not observed by Huber et al. (2016), while on the other hand they e.g. overestimate the transfer of Zn from MSWI fly ash to secondary fly ash.

The behaviour of Cd, Cu, Hg, Mo, Pb and Zn observed in the present study is in line with the results of other studies on thermal treatment of MSWI fly ash (Guohua et al., 2012; Li et al., 2015; Liu et al., 2010; Wang et al., 2008, 2015; Yang et al., 2009; Zupanič et al., 2012). However, in the study by Wang et al. (2015) about 50% of Co is volatised and in the study by Zupanič et al. (2012) about 40% of Sb is volatised. These differences are most likely caused by the different composition of the MSWI fly ash used for thermal treatment and the different treatment conditions. As described above, no mass balance for Se could be established. However, as the Se concentration is very low in pelletised MSWI fly ash and higher in secondary fly ash from treatments at 950 °C and 1050 °C, it can be presumed that a major part of Se from MSWI fly ash is volatised. This presumption corresponds also to the results of Zupanič et al. (2012).

4. Conclusion

The results of the present study demonstrate the general feasibility of combined disc pelletisation and thermal treatment of

MSWI fly ash. Pellets with a water content of about 0.15 kg/kg show high mechanical strength, and thus enabling further processing and safe handling of the material.

As no correlation between residence time and composition of the thermally treated pellets could be observed, it can be concluded that a residence time of 10 min is sufficient for thermal treatment. Thermal treatment at 450 °C was sufficient to generate a non-hazardous material, although higher temperatures improve the separation of toxic heavy metals like Cd and Pb. However, it has to be noted that the MSWI fly ash used for the experiments is not representative with regard to the parameter Pb in the leachate and further investigations are necessary to confirm that combined pelletisation and thermal treatment of fly ashes characterised by a higher leachate content of Pb also generates non-hazardous waste. Furthermore, treatment temperatures of at least 650 °C are necessary to decrease the TEQ of PCDD/F and DL-PCB.

The high concentration of Cu, Pb and Zn observed in secondary fly ash makes this residue a potential raw material for metal recovery, e.g. by applying the FLUREC process.

In the present study, the transfer of selected elements from pelletised MSWI fly ash to the output streams of the kiln (thermally treated MSWI fly ash pellets, secondary fly ash and flue gas) could be observed. However, in an industrial plant for thermal fly ash treatment, the transfer coefficient to the flue gas would most likely be lower and transfer coefficients to the secondary fly ash would be higher due to cooling of the flue gas (with integrated heat recovery) prior to filtration, causing condensation of volatised substances.

With respect to the quality of the thermally treated MSWI fly ash pellets, it can be concluded that the proposed treatment steps allowed generating a non-hazardous waste out of a hazardous waste. Nonetheless, the high content of total dissolved solids in the leachate of thermally treated MSWI fly ash pellets prevents a disposal at non-hazardous waste landfills in Austria. For the later an additional solid-liquid extraction procedure before or after thermal treatment would be necessary.

Hence, further research is still necessary to investigate the thermal co-treatment of pelletised MSWI fly ash together with combustible waste and to assess the resource potential of the secondary fly ash thereby generated.

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Comparative life cycle assessment of MSWI fly ash treatment and disposal

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ABSTRACT

Municipal solid waste incineration (MSWI) fly ash constitutes a hazardous waste. The possibilities for managing this waste comprise disposal at underground deposits or at above-ground landfills after cement stabilisation, application of the FLUREC process, thermal treatment in a dedicated furnace or thermal co-treatment together with combustible hazardous waste. A comparative life cycle assessment (LCA) study was conducted in order to assess the environmental impact of these five MSWI fly ash disposal options with regard to two different time horizons (100 years, indefinite). The uncertainties of the input parameters were propagated by Monte Carlo simulations (MCS). As could be shown by the discernibility analysis, the FLUREC process has the lowest impact in more than 90% of the MCS results. In case long-term emissions (beyond 100 years) are neglected, the second lowest impact is caused by thermal co-treatment in more than 90% of the MCS results. Consideration of long-term emissions indicates the disposal at underground deposits as second best option. Furthermore, it is shown that stabilisation with cement has the second highest and thermal treatment in a dedicated furnace has the highest environmental impact, mostly due to high CO_2 emissions. Therefore these two treatment options should be avoided in the future. Besides the comparative evaluation of the different options, it could be shown that uncertainty analysis is useful to determine the relevance of long-term emissions for the ranking of different systems.

1. Introduction

Municipal solid waste incineration (MSWI) reduces the waste volume by 90%, the mass by 60–90%, and the organic matter by nearly 100% (Brunner and Rechberger, 2015; Chandler et al., 1997; Hjelmar, 1996; Kuo et al., 2007), thereby saving valuable landfill space and avoiding gaseous landfill emissions (CH_4) from organic degradation processes. In Europe, almost 90% of MSWI plants in operation are grate furnace combustors (Fellner et al., 2015). The solid residues produced at these plants comprise bottom ash, which can be deposited at non-hazardous waste landfills or used as road construction material, and fly ash, which consists of "particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material" (Chandler et al., 1997). This MSWI fly ash comprises high contents of easily soluble salts, heavy metals and in some cases also polychlorinated dioxins and furans and is therefore classified as hazardous waste in most countries (Funari et al., 2017; Jiao et al., 2016; Li et al., 2016; Purgar et al., 2016; Ye et al., 2016; Zhan et al., 2016).

The safe disposal of this residue is challenging and costly. MSWI fly ash can be stored in underground deposits (e.g., former salt mines) hundreds or even thousands of meters below the ground surface. However, suitable deposits are not available in all countries and therefore this option is often associated with long transport distances or not feasible at all. Stabilisation with cement or other hydraulic binders and subsequent disposal at non-hazardous waste landfills above ground is another common practice for MSWI fly ash disposal, although the volume of material to be disposed of is significantly increased due to cement addition. Less used disposal options are thermal treatment of MSWI fly ash in a furnace heated to a temperature close to or above the melting point of most MSWI fly ash constituents (700–1200 °C (Ecke et al., 2000; Mangialardi, 2001; Wey et al., 2006)) and extraction of heavy metals with acidic scrubber water as part of the FLUWA or FLUREC process (Bühler and Schlumberger, 2010; Schlumberger, 2010), currently applied in Swiss waste incinerators. Another process for decontamination of fly ash currently under development is thermal co-treatment together with combustible hazardous waste (Huber et al., 2016). This seems prima facie to be a promising

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approach as existing infrastructure necessary for the disposal of unavoidable combustible hazardous waste can be used and the resulting bottom ash complies with legal limits of non-hazardous waste landfills despite the fact that significant amounts of heavy metals from inserted MSWI fly ash are transferred to the rotary kiln bottom ash. This can be explained by vaporisation of volatile elements (e.g. Hg and Cd) and sintering of the remaining residue, which decreases the leaching concentration of the contained pollutants. Detailed information about this treatment option can be found in the paper cited above. However, to date the economic performance and environmental impact of this process are still widely unknown. Huang et al. (2016) compared in their study the environmental impact of different utilisation options of MSWI fly ash in the building material industry. However, the utilisation of MSWI fly ash in buildings materials significantly increases the heavy metal content of building materials and might impair recycling of these materials after demolition (Lederer et al., 2017).

The objectives of this study are to address the environmental performance of fly ash disposal based on a case study from the city of Vienna and to provide decision support by doing a comparative life cycle assessment (LCA) of different MSWI fly ash treatment and disposal options, which can be useful for Vienna as well as for other places where MSWI is incinerated and therefore MSWI fly ash is generated.

2. Methodology

2.1. Goal and scope of the life cycle assessment

The goal of the present study is the evaluation and comparison of five different scenarios for treatment and disposal of MSWI fly ash. The functional unit is the treatment and disposal of 1 Mg of MSWI fly ash from a grate incinerator in Vienna. A representative sample of this fly ash was collected at the incinerator equipped with activated coke injection into the flue gas stream, a filtering separator and a multistage scrubber. The activated coke is injected upstream of the filtering separator. The composition of the fly ash was determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) according to EN 11885 (2009). The total C content was measured with a total CHNS analyser. The results of this analysis are summarised in Table 1.

The model developed for the life cycle assessment can easily be adapted to any other fly ash composition and, additionally, all assumptions, which are part of the model, can be altered to reflect a specific fly ash generated at a specific location. This way a plant operator or waste owner can find the disposal option most beneficial in terms of environmental impacts.

Table 1
Composition of fly ash from Viennese MSW incinerator.

Element	Mass fraction [mg/kg]
Al	39,000
Sb	520
As	21
Ba	900
Pb	2300
Cd	180
Cr	190
Co	24
Fe	12,000
Cu	780
Mn	710
Mo	15
Ni	51
Hg	13
Se	0.05
Ag	27
Zn	13,000
Sn	480
C	23,000

2.2. System definition and scenarios for MSWI fly ash treatment and disposal

The life cycle inventory includes all burdens from the transportation of MSWI fly ash from the MSWI plant to the respective treatment facilities, the MSWI fly ash treatment and its disposal on an above-ground landfill or underground deposit. The production of secondary metals and the saving of quicklime in the FLUREC process are considered as avoided burdens. The inventory does not include upstream burdens associated with MSWI or the production and use of goods prior to their disposal in an MSWI plant (e.g. the extraction and refining of crude oil and the subsequent production of plastic packaging). Hence, the zero burden assumption (Chang and Pires, 2015) was used.

Two different time frames were taken into account, which is relevant regarding landfill emissions (cf. 2.3). The time frame for the assessment of the short-term impact was 100 years. In addition, an indefinite timeframe was chosen for the evaluation of the long-term impact of MSWI fly ash disposal.

A material flow analysis according to Brunner and Rechberger (2004) was conducted for all scenarios in order to determine the import and export flows of the system. The life cycle inventory data was sourced from ecoinvent database V3.2 (2015). As not all necessary data was available in this database the life cycle inventory was complemented with additional data from scientific literature. The life cycle impact assessment was conducted using the ReCiPe model (Hierarchist perspective) (Goedkoop et al., 2009). In order to enable the waste holder or plant operator to apply its own weighting of impact categories, the environmental impact in all midpoint impact categories (agricultural land transformation, climate change, fossil depletion, freshwater ecotoxicity, freshwater eutrophication, human toxicity, ionising radiation, marine ecotoxicity, marine eutrophication, metal depletion, natural land transformation, ozone depletion, particulate matter formation, photochemical oxidant formation, terrestrial acidification, terrestrial ecotoxity, urban land occupation, water depletion) and endpoint impact categories (human health, ecosystem quality, resources, total score) was calculated.

Five different scenarios for treatment and disposal of MSWI fly ash were comparatively assessed.

In **scenario A** MSWI fly ash is transported to an underground deposit. As these deposits are only available at certain locations, transport over large distances is necessary. In LCA it is generally assumed that the hazardous substances deposited in such underground deposits are not transferred to the environment (Doka and Hischier, 2004).

In **scenario B** MSWI fly ash is stabilised with cement and subsequently disposed of at a non-hazardous waste landfill. The use of blast furnace slag cement CEM III/A in a ratio of 1 Mg cement per Mg of fly ash is assumed.

In **scenario C** the so-called FLUREC process (acidic fly ash extraction with integrated zinc recovery) is applied. This process is already utilised in a waste incineration plant in Switzerland (KEBAG, 2013) and was described in detail by Schlumberger (2010). After removal of Hg from the acidic scrubber water, this water is used to extract the metals Zn, Pb, Cu and Cd from the fly ash to be treated. Subsequently, metallic zinc is added to the extract as a reducing agent, whereby a mixture of metallic Cd, Cu and Pb is precipitated. Zn is separated from the liquid by reactive extraction with a selective chelating agent in a liquid-liquid extraction step and metallic Zn with a purity of >99.99% is produced by

electrolysis (average Austrian electricity mix). This Zn can be sold and the mixture of Cd, Cu and Pb can be separated at a non-ferrous metal smelter and subsequently recycled. The solid residue from the acidic washing process complies with the legal limits for non-hazardous waste landfills.

Scenario D describes the thermal treatment of MSWI fly ash in a furnace exclusively dedicated to this purpose. It is assumed that this furnace is heated by coal. The main output flow of this process is a glassy slag that can be disposed of at a non-hazardous waste landfill. However, about 10% of the inserted MSWI fly ash emerge as secondary fly ash, which is enriched in heavy metals and therefore still constitutes hazardous waste (Sakai and Hiraoka, 2000; Yang et al., 2013). It is assumed that this secondary fly ash is disposed of at an underground deposit.

In **scenario E** MSWI fly ash is thermally treated together with combustible hazardous waste in a rotary kiln as described in a previous study (Huber et al., 2016). It was shown that this treatment does not impair bottom ash quality and therefore this residue can be disposed of at a non-hazardous waste landfill. The rotary kiln fly ash is assumed to be disposed of at an underground deposit. Based on investigations of Huber et al. (2016), it is further assumed that about 5% of the inserted MSWI fly ash mass is transferred to the rotary kiln fly ash while 95% are transferred to the bottom ash.

2.3. Modelling of landfill emissions

The substances deposited on landfills do not stay there forever, simply due to the concentration gradient between landfill and surrounding environment. Toxic substances like heavy metals can be mobilised by leachate generated as a result of rainfall over a very long time period. Additionally, erosion processes may transfer the landfilled material to the surrounding environment, which were, however, not considered in the present study. In the European Union, collection and treatment of landfill leachate is mandatory (Ramke, 2008). The collected leachate is commonly fed into the municipal sewer (Calabrò, et al., 2016; Tosti et al., 2016). One part of the heavy metal contained in landfill leachate is incorporated into the sewage sludge generated at the wastewater treatment plant while the other part is emitted to the receiving water. Transfer coefficients for heavy metals in wastewater treatment are given by Doka (2003a). In the present study it is assumed that the sewage sludge is incinerated and the resulting ash is disposed of at a non-hazardous waste landfill. The subsequent landfill emissions from the disposal of sewage sludge ash were considered. Further assumptions are that the leachate collection system and the liner of the landfill are intact and in operation for 100 years. After this time, leachate is released into the soil (and groundwater) below the landfill.

As the landfill emissions largely depend on the composition of the deposited material, they can be assessed using the transfer coefficients for a period of 100 years provided by Doka (2003b). For the indefinite timeframe, the total heavy metal content of fly ash was assumed to be emitted to the environment with the exception of Cr. It is assumed that 75% of this element is present as chromite and therefore not available even under most aggressive conditions (Doka, 2003b). However, in scenarios B-E the MSWI fly ash is treated prior to disposal on a landfill, which changes the chemical and physical properties of fly ash.

In the case of stabilisation treatment, the chemical composition of MSWI fly ash is not altered, but physical stabilisation prevents the contact between rainwater and fly ash to a large extend. In central Europe, about 15% of precipitation on landfills are found as leachate (TÜVRheinland, 2012). According to Mostbauer et al. (1994) this value is reduced to 2% for cement stabilised waste. Conclusively, the transfer coefficients from Doka (2003b) were multiplied by a factor of 0.133333, thereby assuming that the cement stabilisation reduces the pore space of the fly ash accessible for water flow by factor 7.5.

For the solid residue from acidic washing of MSWI fly ash, transfer coefficients for Cd, Cu, Pb and Zn are given by Bühler and Schlumberger (2010). For all other considered heavy metals, the transfer coefficients were determined by laboratory experiments using hydrochloric acid solution (c = 1 mol/L) as extracting agent and a liquid to solid ratio of 5 as assumed by Fellner et al. (2015) and are given in the supplementary information. A detailed description of the experimental setup can be found in Blasenbauer et al. (2015). The transfer coefficients were used to calculate the composition of washed fly ash. The emissions from washed fly ash at a landfill to the environment was calculated using the transfer coefficients from Doka (2003b).

Thermal treatment (scenario D) changes the chemical composition of MSWI fly ash as volatile elements are vaporised at high temperatures. Furthermore, the surface area of the resulting slag is much lower compared to the original fly ash (Wang et al., 2008) and therefore the mass of heavy metals available for short-term leaching is decreased. Several studies (Guohua et al., 2012; Li et al., 2015; Liu et al., 2010; Wang et al., 2009, 2008, 2015; Yang et al., 2009; Zhao et al., 2010; Zupanič et al., 2012) provided data for the chemical composition of fly ash before and after thermal treatment, which enabled the determination of average transfer coefficients. The multiplication of the heavy metal concentration in the original fly ash by these transfer coefficients yielded the concentration in thermally treated fly ash. To account for the decreased mobility of the contained heavy metals, correction factors were introduced. These average correction factors were calculated by dividing the leachable heavy metal content after thermal treatment by the leachable heavy metal content before thermal treatment. The necessary data was sourced from the abovementioned studies. The short-term heavy metal emissions via landfill leachate were calculated by multiplying the heavy metal concentration in original MSWI fly ash by these correction factors (depletion of heavy metals by thermal treatment and decreased mobility) and the transfer coefficients from Doka (2003b).

The thermal co-treatment of MSWI fly ash together with combustible hazardous waste in a rotary kiln (scenario E) is described in detail by Huber et al. (2016), who provided also transfer coefficients for the transfer of heavy metals from inserted fly ash to the output streams of the hazardous waste incinerator. These transfer coefficients were applied to the heavy metal concentrations given in 2.1 to determine the composition of MSWI fly ash after co-treatment, which is disposed of at a non-hazardous waste landfill as part of the rotary kiln bottom ash. As the temperatures in the rotary kiln are significantly lower than those typically used for thermal ash treatment in a separate furnace, no correction factor accounting for the decrease in surface area and thus reduced mobility of heavy metals was used for the evaluation of thermal co-treatment contrary to scenario D.

2.4. Modelling of thermal treatment emissions

If MSWI fly ash is treated thermally (scenarios D and E), an air pollution control (APC) system will be necessary. It is assumed that this APC system consists of an electrostatic precipitator and a two-stage scrubber. Transfer coefficients from MSWI fly ash to secondary fly ash of 0.1 and 0.05 in scenario D and E, respectively, are assumed. As in MSWI the transfer coefficient from MSW to fly ash is about 0.033 (Morf et al., 2000), the dedusting unit for the treatment of flue gas from thermal treatment of 1 t of MSWI fly ash corresponds to the dedusting unit for the treatment of flue gas from incineration of 3.0 t MSW and 1.5 t MSW, respectively. According to Wang et al. (2008), thermal treatment of MSWI fly

ash causes SO_2 emissions of 2.86 kg per Mg fly ash treated. However, the incineration of 1 Mg MSW causes SO_2 emissions of 0.87 kg per Mg MSW (Wien Energie, 2016). Therefore, it was assumed that the scrubber necessary to treat the flue gas from the treatment of 1 Mg/h MSWI fly ash equals a scrubber for the treatment of flue gas from thermal treatment of 3.27 Mg/h MSW. As the NO_X emissions from thermal treatment of MSWI fly ash are below the legal limits (Wang et al., 2008), it was concluded that no DeNOx unit is necessary.

The APC system for scenarios D and E was modelled based on the information given above and the data about energy and consumables necessary for flue gas treatment from Stubenvoll et al. (2002) and Wiesenberger and Kircher (2001). The emissions into air were sourced from Wang et al. (2008). It was assumed that the emissions into air from thermal treatment in scenario D and E are equal. However, there is some uncertainty related to this assumption, because on the one hand chlorinated solvents or polymers in the waste input can increase the Cl^- concentration in the scrubber and therefore improve Hg removal from the raw gas (Abanades et al., 2002; Vogg et al., 1986) and on the other hand NaCl and KCl contained in MSWI fly ash might be able to volatise heavy metals contained in combustible hazardous waste as heavy metal chlorides or organic Cl compounds present in combustible hazardous waste might volatise heavy metals contained in MSWI fly ash (Morf et al., 2000; Nowak et al., 2010).

2.5. Uncertainty analysis

The uncertainty associated with the result of LCA models can be divided into parameter uncertainty, scenario uncertainty and model uncertainty (Huijbregts et al., 2003). The influence of parameter variation on the result and the effect of scenario alterations were investigated, while uncertainties related to the use of alternative modelling concepts (e.g. how to model landfill emissions) were not within the scope of the present study.

Parameter uncertainty of the output variables was determined by propagating the uncertainty of the input parameters in a Monte Carlo Simulation (MCS) with 100,000 runs. The distributions of all input parameters can be found in the supplementary information. A discernibility analysis was conducted for the total score for each scenario by calculation the difference between the LCA results of the single scenarios in all 100,000 iterations as described by Clavreul et al. (2012) in order to determine in how many cases a certain scenario outperforms the other ones. Additionally an uncertainty contribution analysis was conducted for the total score in order to determine which parameters contribute most to the overall uncertainty of the LCA result (Clavreul et al., 2012). Values for the total score above 1 person equivalent were excluded for the uncertainty contribution analysis.

In order to assess scenario uncertainty, alternative scenario choices were modelled and the environmental impact in all midpoint and endpoint impact categories was calculated for these alternative scenarios. In scenario B∗ 0.3 Mg of cement were used instead of 1 Mg in scenario B as Billen et al. (2014) conclude that the environmental impact of cement production might exceed any savings from decreased emissions due to stabilisation. The heat necessary for thermal treatment in scenario D∗ was provided by combustion of natural gas instead of coal in scenario D. In scenario E the thermal treatment of MSWI fly ash in a rotary kiln hazardous waste incinerator is assumed. In comparison, in scenario E∗ the re-introduction of MSWI fly ash into the grate furnace together with MSW instead of co-treatment in a rotary kiln hazardous waste incinerator is evaluated. In this case The partial transfer of the volatile elements Hg and Cl to the scrubber water and subsequently to the filter cake was modelled for the treatment of primary, secondary and tertiary fly ash using the transfer coefficients given by Huber et al. (2016). Furthermore two different time frames (100 years and indefinite time) where considered for leachate emissions from landfills.

3. Results

3.1. Material flow analysis

The material flows of scenario A, B, C, D and E are shown in Fig. 1 and Fig. 2, respectively. The function of each system is the treatment and disposal of 1 Mg of MSWI fly ash. The system of scenario C additionally fulfils the function of neutralising 4660 kg of acidic scrubber water and the system of scenario E additionally fulfils the function of disposing 3333 kg of combustible hazardous

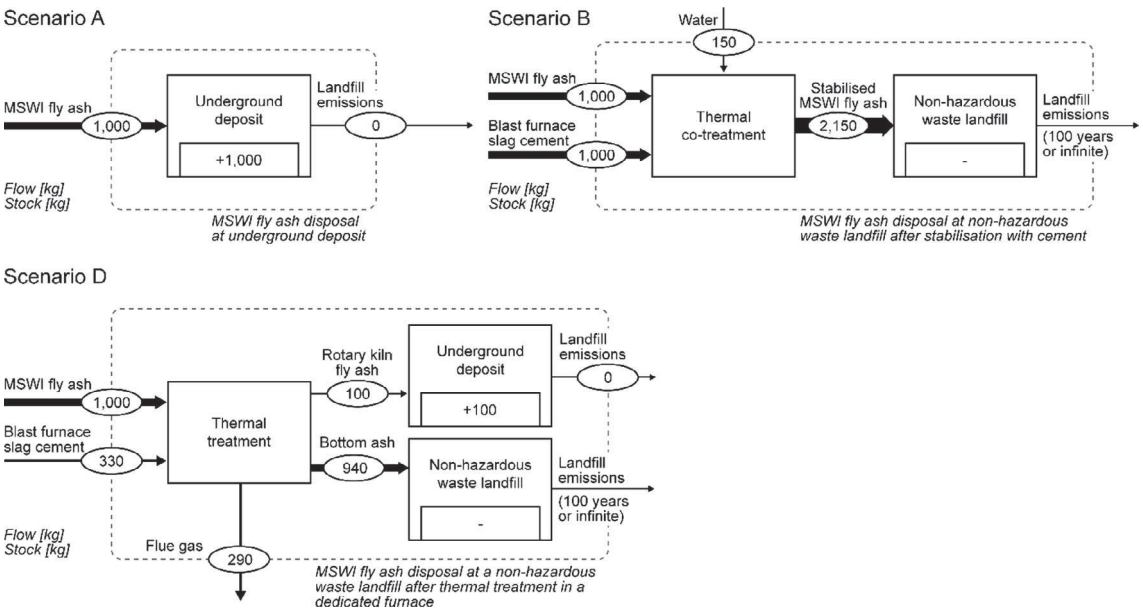

Fig. 1. Material flows of scenarios A, B and D. The mass of landfill emissions from non-hazardous waste landfills depends on the timeframe considered.

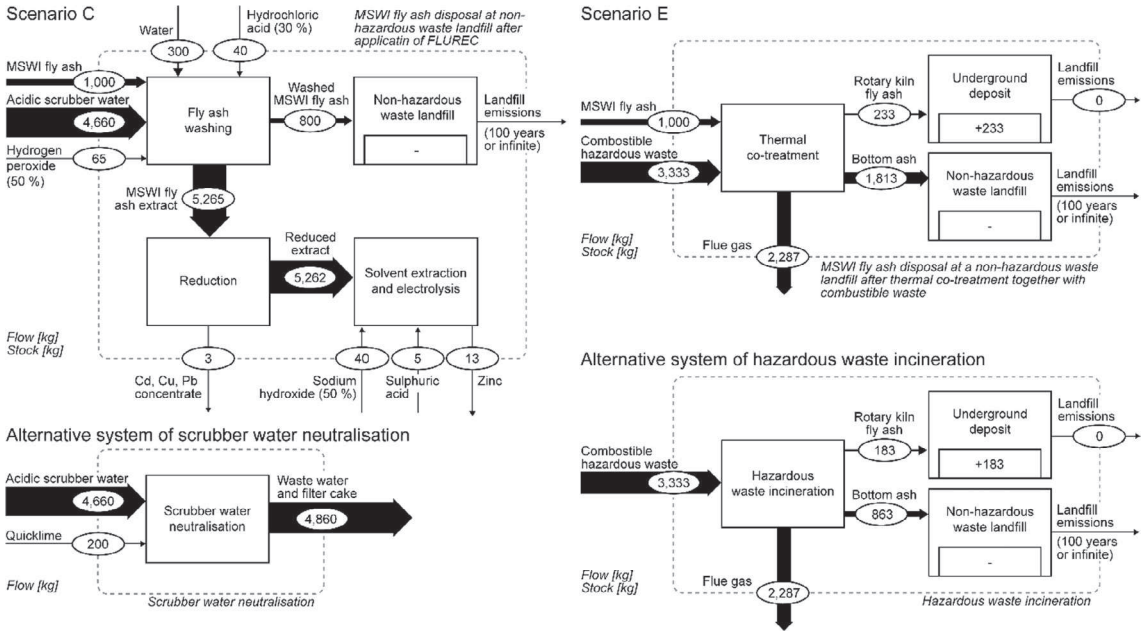

Fig. 2. Material flows of scenarios C and E. The material flows of the reference systems are shown below the material flows for MSWI fly ash treatment. The mass of landfill emissions from non-hazardous waste landfills depends on the timeframe considered.

waste. The reference processes for the function scrubber water neutralisation and the function combustible hazardous waste disposal are illustrated in Fig. 2 below the systems for MSWI fly ash disposal. The environmental impact of these reference processes was subtracted in scenario C and E, respectively to determine the impact that is attributed to MSWI fly ash treatment and disposal.

3.2. Environmental impact for 100 years

The environmental impact of scenarios A–E for a period of 100 years in all midpoint impact categories is shown in Fig. 3. This diagram shows that for most scenarios human toxicity, ecotoxicity, eutrophication and natural land transformation are the midpoint impact categories with the highest impact with regard to the total impact caused by an average European (Sleeswijk et al., 2008).

The LCA results for the endpoint impact categories are shown in Fig. 4. Scenario C has the lowest and scenario D has the highest environmental impact in all endpoint impact categories. The total environmental impact of scenarios A, B and E for a period of 100 years is in the same order of magnitude. The aggregated impact (total score) is dominated by the midpoint impact categories climate change, human toxicity, terrestrial ecotoxicity and fossil depletion.

3.3. Environmental impact for an indefinite time period

Fig. 5 shows the environmental impact including long-term landfill emissions in midpoint impact categories. As these emissions only affect the toxicity impact categories, only these are shown here. The impact in all other midpoint categories is equal to the one shown in part 3.2. As in LCA it is generally assumed that material disposed of in underground deposits is not transferred to the environment, the impact of scenario A is equal to the one shown in part 3.2 and therefore this disposal option is associated with the lowest long-term toxicity. Scenarios B–E show a significantly higher toxicity, if long-term landfill emissions are considered. This is especially true for terrestrial ecotoxicity, as it is assumed that the leachate migrates into the soil below the landfill. Scenario C shows a lower long-term toxicity than scenarios B, D and E because of the benefit from secondary metal production and the extraction of heavy metals from fly ash. Because in scenario B all heavy metals contained in MSWI fly ash are disposed of at a non-hazardous waste landfill, this scenario shows the highest long term toxicity.

The environmental impact including long-term landfill emissions in endpoint impact categories is shown in Fig. 6. The impact on resource scarcity is the same as shown in part 3.2. As in LCA it is assumed that there are no long-term emissions from underground deposits, the impact of scenario A in all categories is equal to the one shown in part 3.2. If long-term emissions are considered, the impact of MSWI fly ash treatment and disposal on human health and ecosystem quality is significantly higher than in part 3.2. This is especially true for scenario B, because here the highest mass of heavy metals is disposed of at an above-ground landfill. Regarding the total score, the lowest impact is caused by scenario C, followed by scenario A. The long-term impact of scenario E is higher than the short-term impact and therefore, if long-term emissions are considered, scenario E has only the third lowest environmental impact.

3.4. Uncertainty analysis

3.4.1. Parameter uncertainty

As described in part 2.5, parameter uncertainty and scenario uncertainty were assessed in the present study. The parameter uncertainty is shown as error bars representing the range between the 5 percentile and the 95 percentile, which contains 90% of the Monte Carlo Simulation (MCS) results, in Figs. 3–9. Detailed data can be found in the supplementary information.

As the result for scenario A is only depending on the mass of MSWI fly ash (functional unit) and the transportation distance, no parameter uncertainty is displayed. The parameter uncertainty for the scenarios B, D and E ranges between about 1% and 25% of the median value. For scenario C the relative uncertainty is higher (up to 8600%) as the environmental impact is close to 0 in many impact categories.

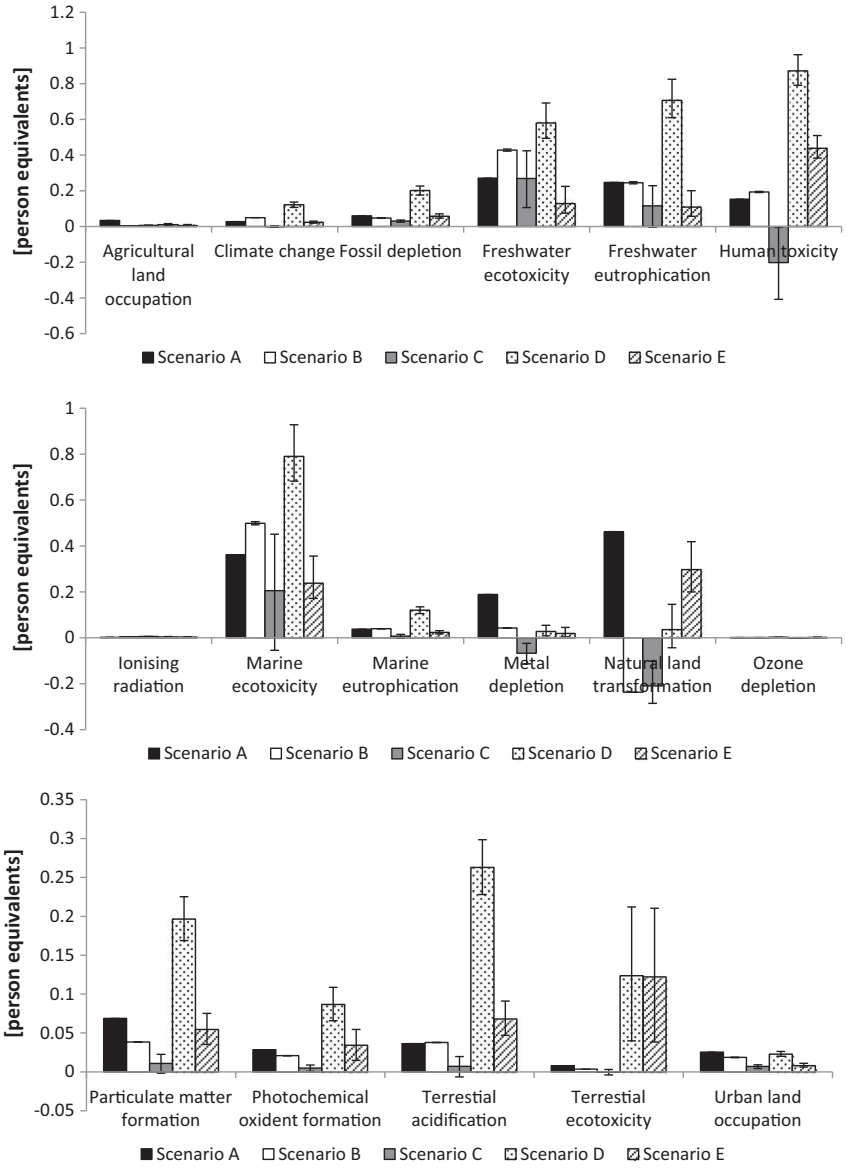

Fig. 3. Environmental impact for 100 years in all midpoint impact categories. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range between the 5-percentile and the 95-percentile (containing 90% of the MCS results).

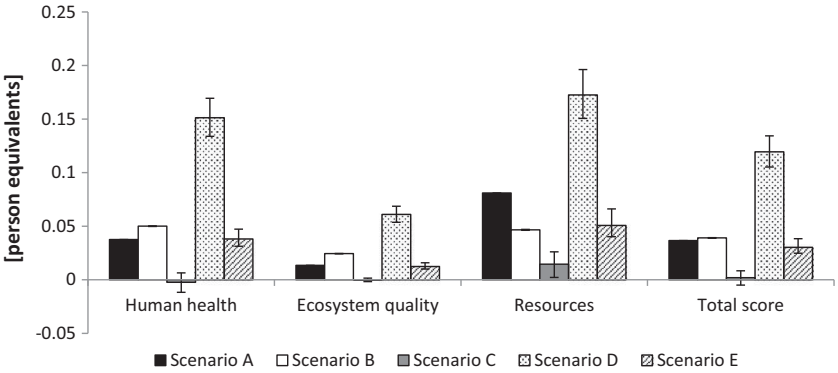

Fig. 4. Environmental impact for 100 years in all endpoint impact categories. The value on the primary vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

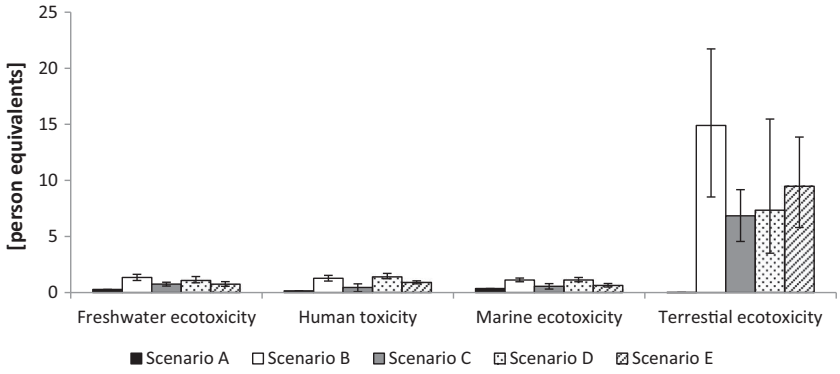

Fig. 5. Environmental impact for an indefinite timeframe in all toxicity midpoint impact categories. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

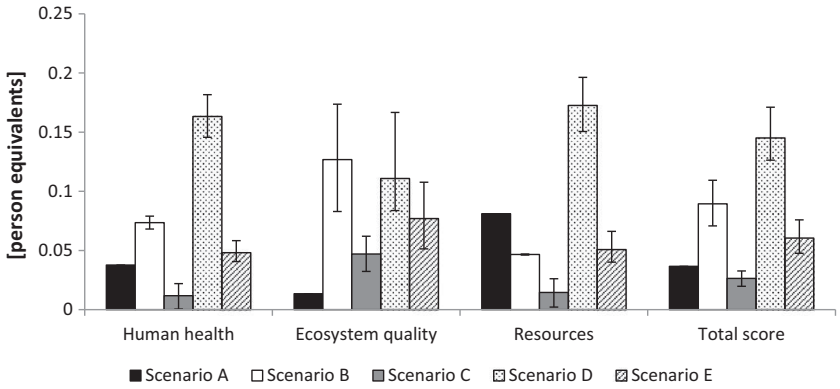

Fig. 6. Environmental impact for an indefinite timeframe in all endpoint impact categories. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

3.4.2. Scenario uncertainty

3.4.2.1. Amount of cement used for stabilisation. The short-term and long-term environmental impact of scenario B with high amount of cement (1000 kg) and low amount of cement (300 kg) is shown in Fig. 7. As production of the cement required for stabilisation of MSWI fly ash causes significant emissions to the environment, the impact in all endpoint categories can be decreased significantly, if the cement to MSWI fly ash ratio is optimised in scenario B. The use of 300 kg instead of 1000 kg cement lowers the short-term environmental impact by almost 70% and the long-term environmental impact by almost 30% (total score).

3.4.2.2. Fuel used for thermal treatment. The short-term and long-term environmental impact of scenario D with hard coal and natural gas as fuel for thermal treatment are shown in Fig. 8. The impact on human health can be decreased by about 50% (short-term and long-term). This decrease as well as the lower impact on ecosystem quality can be mainly attributed to the lower CO_2 emissions from the combustion of natural gas compared to hard coal.

3.4.2.3. Reintroduction of MSWI fly ash into the grate furnace. The short-term and long-term environmental impact of scenario E with

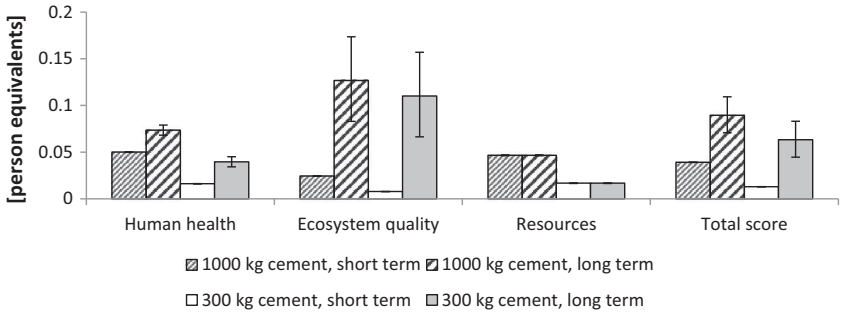

Fig. 7. Short-term and long-term environmental impact of scenario B with different amounts of cement used. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

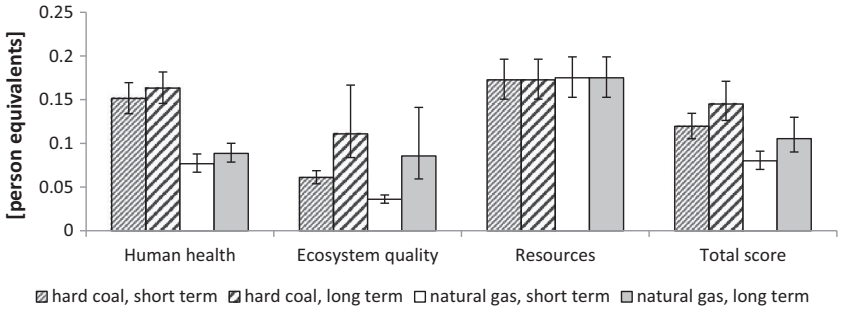

Fig. 8. Short-term and long-term environmental impact of scenario D with different fuels for thermal treatment of MSWI fly ash. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

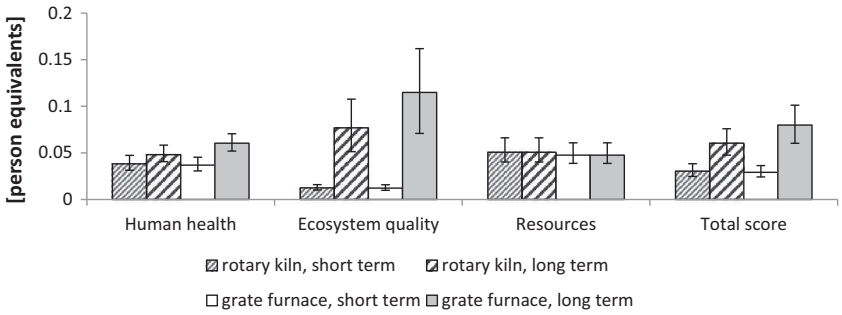

Fig. 9. Short-term and long-term environmental impact of scenario E with different furnaces for thermal treatment of MSWI fly ash. The value on the vertical axis is the impact of the treatment and disposal of 1 Mg MSWI fly ash divided by the impact of an average European in each impact category (Sleeswijk et al., 2008). The error bars show the range containing 90% of the MCS results.

thermal co-treatment in a rotary kiln and thermal co-treatment at a grate furnace are shown in Fig. 9. If MSWI fly ash is reintroduced into the grate furnace where it is generated, the impact on human health and ecosystem quality is increased by 15% and 30%, respectively, compared to thermal co-treatment together with combustible hazardous waste in a rotary kiln. This difference is only visible, if long-term emissions are considered. The lower environmental impact of co-treatment in the rotary kiln is explained by the partial transfer of toxic heavy metals (mainly Pb and Cd) to the rotary kiln fly ash (Huber et al., 2016), which is disposed of at an underground deposit. If, however, MSWI fly ash is co-treated in a grate furnace, most heavy metals contained in this residue are finally disposed of at a non-hazardous waste landfill. Only Hg is mainly transferred to the filter cake generated at the MSWI plant and subsequently disposed of at an underground deposit.

3.4.3. Discernibility analysis

The results of the discernibility analysis for the total score are shown in Table 2 (without long-term emissions) and Table 3 (with long-term emissions). These results show that scenario C has the lowest environmental impact in almost all cases. If only short-term emissions are considered, the total score of scenario E is in more than 90% of the cases lower compared to scenarios A, B and D. If also long-term emissions are considered, scenario A has in almost 100% of the cases a lower total score than the scenarios B, D and E. Scenario D has the highest environmental impact in almost all cases.

3.4.4. Uncertainty contribution analysis

For every scenario, the three input parameters contributing most to the aggregated overall environmental impact were identified and are shown in Table 4. In scenario A no uncertainty is displayed as the environmental impact of this scenario is only influenced by the mass of MSWI fly ash disposed of at an underground deposit and the transport distance. Both parameters are not associated with an uncertainty. The uncertainty of the other scenarios is dominated by the uncertainty of the heavy metal concentration in MSWI fly ash, various transfer coefficients used in the LCA model, and the energy required for the respective fly ash treatment processes.

4. Discussion

Every year 65 million Mg of MSW are incinerated in the European Union (EUROSTAT, 2016). Applying a transfer coefficient from MSW to MSWI fly ash of 0.033 (Morf et al., 2000), results in a MSWI fly ash mass flow of 2.1 million Mg/a or 4.2 kg/a per capita, which equals to about 0.08% of total waste generation in the EU. Based on the results of the LCA conducted, the contribution of MSWI fly ash disposal to the total environmental impact of an average EU citizen is between 0.008 and 0.47% in case only short-term emissions are considered and between 0.12 and 0.61% if long-term emissions are accounted for as well.

The presented model allows comparing the environmental impact of different treatment and disposal options for MSWI fly ash. The lowest environmental impact causes the application of the FLUREC process (scenario C), which corresponds to the results of Bösch et al. (2011). However, based on findings of Fellner et al. (2015) this process is at current commodity prices only economically feasible for a few MSWI filter ashes from MSWI plants with wet flue gas treatment that contain high amounts of Zn (>40,000 mg Zn/kg fly ash).

The second best results with respect to environmental impacts are determined for the thermal co-treatment together with combustible hazardous waste (scenario E) or disposal in underground deposits (scenario A), depending on whether long-term emissions

Table 2
Results of the discernibility analysis for the total score without consideration of long-term emissions. The number display the percentage of MCS results that give a lower environmental impact for the scenario in the first column compared to the scenario of the first line.

	A	B	C	D	E
A	–	100	0.1	100	8.8
B	0.0	–	0.1	100	3.8
C	99.9	99.9	–	99.9	100
D	0.0	0.0	0.1	–	0.0
E	91.2	96.2	0.0	100	–

Table 3
Results of the discernibility analysis for the total score with consideration of long-term emissions. The number display the percentage of MCS results that give a lower environmental impact for the scenario in the first column compared to the scenario of the first line.

	A	B	C	D	E
A	–	100	0.4	100	100
B	0.0	–	0.1	100	0.3
C	99.6	99.9	–	99.9	100
D	0.0	0.0	0.1	–	0.0
E	0.0	99.7	0.0	100	–

Table 4
Parameters with the highest contribution to uncertainty of aggregated overall environmental impact (excl. LTE. . .excluding long-term emissions), incl. LTE. . .including long-term emissions).

Scenario	Parameters with highest contribution to uncertainty		
	Highest	Second highest	Third highest
A (excl. LTE)	–	–	–
B (excl. LTE)	Transfer coefficient from landfill to environment in 100 a for Sb (3.2%)	Transfer coefficient from landfill to environment in 100 a for Mn (0.9%)	Electricity demand for mixing (0.5%)
C (excl. LTE)	Zn content of MSWI fly ash (28.9%)	Transfer coefficient from landfill to environment in 100 a for Sb (10.5%)	Transfer coefficient from landfill to environment in 100 a for Mn (7.6%)
D (excl. LTE)	Amount of coal used (48.4%)	Transfer coefficient from MSWI fly ash to secondary fly ash (9.7%)	SO_2 emissions from thermal treatment (3.2%)
E (excl. LTE)	Transfer coefficient from MSWI fly ash to bottom ash (11.9%)	SO_2 emissions from thermal treatment (4.9%)	Amount of fuel oil used (3.6%)
A (incl. LTE)	–	–	–
B (incl. LTE)	Zn content of MSWI fly ash (87.4%)	Ag content of MSWI fly ash (5.6%)	Hg content of MSWI fly ash (1.0%)
C (incl. LTE)	Ag content of MSWI fly ash (14.9%)	Transfer coefficient from landfill to environment in 100 a for Sb (10.4%)	Transfer coefficient of MSWI fly ash to washed fly ash (8.2%)
D (incl. LTE)	Amount of coal used (25.0%)	Immobilisation factor for Zn (23.3%)	Zn content of MSWI fly ash (16.6%)
E (incl. LTE)	Zn content of MSWI fly ash (25.0%)	Transfer coefficient from MSWI fly ash to bottom ash (11.0%)	Ag content of MSWI fly ash (4.4%)

(beyond 100 years) from landfills are considered or not. However, the good result for the disposal in underground deposits is based on the assumption, that the deposited material and therein contained substances are not transferred to the environment, even in the long run. However, in practice there is a certain risk that heavy metals might migrate from the deposit into the surrounding environment, e.g. as a result of flooding in the long term (Doka, 2003b; Doka and Hischier, 2004; Morf et al., 2016). Furthermore, underground deposits are only available in a few countries like Germany or France. If a country does not want to export its hazardous waste or the countries with underground deposits do not want to accept MSWI fly ash at some time, this disposal option will not be feasible any more.

The good LCA result of thermal co-treatment of MSWI fly ash together with combustible hazardous waste in a rotary kiln can be explained by the fact that already existing infrastructure can be used. However, the knowledge about this process has still to be solidified prior to its large scale application in routine operation (Huber et al., 2016). The short-term environmental impact is below the one of scenarios A and B, which are the prevailing disposal options for MSWI fly ash in Europe. With regard to the long-term impact, the factor determining the relative environmental impact of the three abovementioned scenarios is the mass of heavy metals deposited on an above-ground landfill. This mass is lowest in scenario A (underground deposit) and highest in scenario B (stabilisation with cement). For the same reason the long-term environmental impact will be higher, if MSWI fly ash is treated in a grate furnace MSW incinerator instead of a rotary kiln hazardous waste incinerator, as for the latter the secondary fly ash generated will be disposed of at underground deposits.

Stabilisation with cement (scenario B) represents the probably most common practice of treating MSWI fly ash in Europe, because it is simple and the required cement is abundantly available at an acceptable price. However, all heavy metals contained in MSWI fly ash are disposed of at an above-ground landfill and emissions from mostly fossil fuel-fired cement kilns have a significant environmental impact. A lower amount of cement used for stabilisation of MSWI can lead to a better LCA result as was shown in the uncertainty analysis of this study, which is in accordance with the findings of Billen et al. (2014). Therefore, the optimisation of the cement to MSWI fly ash ratio is imperative with regard to environmental burden as well as economic costs, if this treatment and disposal option is used.

In comparison to thermal co-treatment in an existing facility, thermal treatment in a furnace dedicated to this purpose (scenario D) has a remarkable environmental impact. Even a fuel switch from hard coal to natural gas, does not improve the performance of this treatment and disposal option sufficiently. As a result of

the high energy requirement and the associated greenhouse gas emissions this treatment process should be avoided.

In order to assess the composition of treated fly ash (c.f. 2.3) and the emissions from landfills to the environment transfer coefficients from literature were used, which are associated with a rather high uncertainty and therefore contribute considerably to the overall uncertainty of the LCA result which was shown by the uncertainty contribution analysis. An LCA study should advantageously be based on the chemical analysis results from fly ash before and after treatment. This would require large scale experiments, in which one representative MSWI fly ash sample of several Mg is divided into three parts and treated as in scenarios C, D and E. However, even if this considerable effort was made, it would not be certain that the operation conditions chosen for the treatment are the ones best suitable for the MSWI fly ash from a particular plant. Another important source of uncertainty is the variation of heavy metal contents in fly ash.

The abovementioned points are especially relevant for the present study as emissions from landfills account for about 50% (scenario D) to up to almost 100% (scenario E) of the impact in the midpoint categories freshwater ecotoxicty, human toxicity, marine ecotoxicty and terrestrial ecotoxicity.

The second relevant processes with regard to the impact on human health and ecosystem quality are the ones responsible for the provision of energy (combustion of fossil fuels for thermal treatment of MSWI fly ash, in the cement kiln or power plant). Additionally in scenario C the production of ancillaries has a significant environmental impact while the production of secondary metals is associated with significant environmental benefits. In scenario A about 75% of the total environmental impact are caused by the underground deposit while the remainder is caused by the transport. As there is still no standardised methodology to account for long-term metal emissions, two extreme approaches (cut off after 100 years and indefinite time horizon) were applied to assess the environmental impact of MSWI fly ash treatment and disposal. In cases where both approaches yield the same ranking based on the discernibility analysis (c.f. part 3.4.3), a decision whether long-term emissions should be considered or not is not necessary. However, if the two approaches yield different results (like the ranking of scenario A and scenario E in the present study), no clear decision support can be given.

This could be done by calculating the share of heavy metals contained in the residue deposited on a landfill that has to be released in order to get equal results for the two options (17.2% for the aggregated total impact in scenario A and E). The timeframe corresponding to these 17.2% can be calculated by using one of the landfill models, which can be found in the literature (Astrup et al., 2006; Belevi and Baccini, 1989; Hellweg et al., 2005; Hyks et al., 2009). After such modelling, which is beyond the scope of the present study, it is possible to decide whether a timeframe shorter or longer than this calculated period is considered. The different possibilities for handling long-term emissions in LCA are discussed in detail by Bakas et al. (2015), Hellweg (2000) and Laner (2009). For example, discounting or time-dependent characterisation factors could be applied in order to rank the two scenarios in doubt. However, it might also be possible that this additional effort is not necessary as the decision on which scenario to choose can also be taken based on the present LCA result and other decision factors like economic costs, risks that cannot be assessed in an LCA or aims like autarchy of disposal.

5. Conclusion

A LCA model for the determination of the environmental impact of five different scenarios for treatment and disposal of MSWI fly ash was developed. The presented results show that stabilisation with cement and thermal treatment in a separate furnace have a considerable environmental impact and should therefore be avoided in the future. The lowest environmental impact is achieved by the FLUREC process. However, this option might not be economically feasible. Therefore, the thermal co-treatment of MSWI fly ash together with combustible waste seems to be a promising scenario that results in a comparably low environmental impact. Nevertheless, further experiments on thermal co-treatment of MSWI fly ash are necessary to confirm the data used in this study.

Furthermore, our results show that the way how long-term emissions from landfills are handled in an LCA framework can have a significant effect on the ranking of different waste management scenarios. In this case study, it determines whether disposal at an underground deposit or thermal co-treatment together with hazardous waste has the lower environmental impact. Therefore decision makers have to decide which time frame they want to consider or alternatively base their decision on other factors complementary to the LCA result.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2017.06.004.

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Full length article

Integration of life cycle assessment with monetary valuation for resource classification: The case of municipal solid waste incineration fly ash

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ABSTRACT

Recently, recycling of municipal solid waste incineration (MSWI) fly ash has gained increasing interest, although it is still unclear if recycling is preferable over disposal from an economic and environmental point of view. The objective of the present study is to assess the resource potential of MSWI fly ash for the production of three different commodities (cement, metals, de-icing salts) from a private investor's micro and a public entity's macro perspective. Therefore, the environmental impacts and economics were determined by life cycle assessment (LCA) and discounted cash flow (DCF) analysis, respectively. Monetary valuation was applied to the LCA results in order to determine the external costs, which were subsequently added to the DCF analysis results from the macro perspective (i.e. internal costs) to determine the socio-economic viability (i.e. total costs) of MSWI fly ash recycling. The resource classification scheme UNFC was used to classify MSWI fly ash for utilisation in cement production, metal and salt recovery based on the combined result from LCA and DCF analysis. The consideration of project feasibility and geological knowledge leads to the classification as commercial project (111) for cement production (micro and macro view) and for metal recovery (macro view). Metal recovery is classified as other combination (211) from a micro view due to a negative median net present value and salt recovery is classified as non-commercial project (321) from a micro and macro view. It was demonstrated that monetary valuation of LCA results is a useful tool for including external costs into resource classification.

1. Introduction

Thermal treatment of municipal solid waste (MSW) is considered a key element for sustainable waste management (Brunner and Rechberger, 2015). The solid residues from this treatment comprise bottom ash and air pollution control residues such as fly ash (*"particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material"*), wet scrubber sludge or dry and semi-dry system residues (Chandler et al., 1997). While bottom ash is already utilised in many countries mainly as road construction material (Verbinnen et al., 2017), fly ash and other air pollution control residues are usually disposed of either in underground landfills for hazardous waste or after stabilisation (e.g. with cement) on above-ground landfills for non-hazardous waste (Fellner et al., 2015b; Huber et al., 2016). There are increasing efforts to recycle also MSWI fly ash by different processes but it is still not clear if this recycling is preferable over safe disposal.

Fly ash from municipal solid waste incineration (MSWI) consists mainly of mineral phases with a high content of Ca, Si and Al (Le Forestier and Libourel, 1998). Therefore MSWI fly ash can be utilised as a raw material for cement production. This utilisation option leads to a decreased primary raw material consumption as MSWI fly ash is used as secondary raw material and as the entire MSWI fly ash mass can be used, no disposal on a landfill is necessary. Several studies investigated the feasibility of the utilisation of MSWI fly ash in cement production (Guo et al., 2017; Hartmann et al., 2015; Huang et al., 2017; Saikia et al., 2007). If fly ash is supposed to replace raw meal in clinker production, Cl has to be removed from MSWI fly ash in order to prevent operational problems and corrosion in the cement kiln and to obtain cement of sufficient quality (Reeves et al., 2006; Tang et al., 2014). This can be achieved by simply extracting easily soluble salts with water as an extracting agent (De Boom and Degrez, 2015; Karlfeldt Fedje et al., 2010; Wang et al., 2001; Zhang and Itoh, 2006). Depending on the ratio of MSWI fly ash and other primary and secondary raw materials, this utilisation option however may have a significant effect on the total content of heavy metals in cement and concrete (Lederer et al., 2017).

MSWI fly ash has a total Zn content of up to about 60,000 mg/kg (Fellner et al., 2015b) and contains other potentially valuable metals. In recent years, a process for recovery of Zn, Pb, Cd and Cu from MSWI fly ash, namely the FLUREC process, was developed (Schlumberger, 2010).

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In this process acidic scrubber water from the waste incineration plant is mixed with MSWI fly ash and used as an extraction agent. After sufficient contact time, solid liquid separation is applied and the remaining solid material is disposed of at a non-hazardous waste landfill. Metallic Zn powder is added as a reducing agent in order to precipitate Cd, Cu and Pb in metallic form. Subsequently, reactive extraction and electrowinning are applied to recover Zn from the solution.

Fellner et al. (2015b) evaluated the resource potential of Zn in fly ash, semi-dry and dry system residue from MSWI in Europe by conducting an economic analysis of the application of the FLUREC process compared to stabilisation and disposal at underground landfills. They conclude that Zn recovery is only economically viable for some filter ashes that are collected separately from the boiler ashes in MSWI plants with wet air pollution control systems. The environmental impact of the FLUREC process compared to other MSWI fly ash management practices was assessed by Bösch et al. (2011) and Huber et al. (2018, 2017) using life cycle assessment (LCA) and several midpoint and endpoint impact categories. Bösch et al. (2011) show that the application of the FLUWA and especially FLUREC process lead to greenhouse gas savings, while disposal in underground landfills and especially stabilisation with cement are associated with greenhouse gas emissions. Huber et al. (2018) compared five different MSWI management scenarios (underground landfill, stabilisation with cement, FLUREC process, thermal treatment in a dedicated facility, thermal treatment together with combustible waste). This study shows that the FLUREC process has the lowest overall environmental impact, while the ranking of disposal in underground landfills and thermal treatment together with combustible waste depends on the timeframe considered. Thermal treatment in a dedicated facility and stabilisation with cement both have relatively high environmental impacts. Huber et al. (2017) demonstrated that the environmental impact of metal recovery of MSWI fly ash is significantly lower compared to the environmental impact of utilising MSWI fly ash in cement industry.

Currently, there are also first attempts to recover chlorides as de-icing salt from MSWI fly ash by liquid extraction. The heavy metals contained in the resulting solution are precipitated by addition of TMT15 and pH adjustment and the water is evaporated to generate a salt mixture that can be used as a road de-icer (Easymining, 2017; Stena Metall, n.d.; Tang et al., 2014).

As mentioned above, it is not clear which MSWI fly ash utilisation option is preferable from an economic and environmental point of view. A multitude of different ways to perform economic and environmental assessment has been developed so far. The environmental impact can be assessed inter alia by life cycle assessment (Curran, 2012), cost-benefit analysis (Pearce et al., 2006) or statistical entropy analysis (Laner et al., 2017). However, these methods often do not take into account the economic circumstances and especially they do not allow the comparison of waste recycling projects with primary raw materials exploitation projects.

One possible way to fill this omission is by using a resource classification method that is usually applied to natural resources. The application of the resource classification scheme UNFC (UNECE, 2010) for anthropogenic resources, specifically for a landfill mining project in Belgium, was already demonstrated by Winterstetter et al. (2015). They calculated the net present value (NPV) by discounted cash flow (DCF) analysis for the economic evaluation of the project from a private investor's micro perspective as well as from a public entity's macro perspective. For the macro perspective an arbitrary carbon tax of 10 €/t CO_2 was assumed to include the benefit of avoided greenhouse gas emissions compared to a "do nothing" scenario in the evaluation. However, other impact categories apart from global warming were not considered.

The objective of the present study is therefore to assess the resource potential of MSWI fly ash for the production of three different commodities (cement, metals and de-icing salt) from a private investor's micro and a public entity's macro perspective considering a wide range of environmental impact categories. The particular research questions to be addressed are:

- What is the environmental impact of the utilisation of MSWI fly ash in cement production, metal recovery and de-icing salt production?
- What is the NPV of these recycling options?
- How can environmental impacts in different impact categories be "transformed" to monetary values?
- How can MSWI fly ash be classified in the UNFC classification scheme from a micro and macro perspective considering the environmental impacts of recycling?

2. Methodology

2.1. System definition and recycling scenarios

The goal of the present study is the evaluation of MSWI fly ash as a resource for different commodities, namely cement, metals and salt, from a micro and macro perspective, which includes the external costs based on an environmental assessment using several impact categories. The functional unit of the environmental and economic analysis is the treatment of 18,000 Mg/a MSWI fly ash from MSWI plants equipped with wet air pollution control systems. The MSWI fly ash is therefore not mixed with any other materials. This amount corresponds to the annual MSWI fly ash mass arising in Vienna (combusted waste mass of about 700,000 Mg/a). The project lifetime was defined as 20 a. Hence, it was assumed that the investment necessary for the different scenarios is depreciated after 20 a and after this time new equipment is purchased, which requires further investment. Accordingly, all environmental burdens and economic costs refer to a total amount of 360,000 Mg of MSWI fly ash processed in the time of 20 a.

The composition of MSWI fly ash is given in Table 1. The socio-economic circumstances (e.g. the legal situation in a specific country might rule out one recycling option) and technology applied for MSWI (e.g. metal contents are generally higher in fly ash compared to dry and semi-dry process residue) can have a significant effect on the environmental and economic evaluation as well as on the resource classification. Therefore, fly ash from every MSWI plant has to be assessed on a case-by-case basis. This approach was also used by Winterstetter et al. (2015) contrary to Fellner et al. (2015b).

As the recovery of chloride salts from fly ash washing liquid can be combined with the FLUREC process and the production of cement

Table 1
Composition of MSWI fly ash used for the assessment (Huber et al., 2018; Wien Energie GmbH, 2016).

Element	Mass fraction [mg/kg]
Al	36,000
Sb	520
As	21
Ba	900
Pb	2,300
Cd	180
Ca	190,000
Cr	190
Co	24
Fe	12,000
Cu	780
Mn	710
Mo	15
Ni	51
Hg	13
S	34,000
Se	0.05
Ag	27
Si	58,000
Zn	13,000
Sn	480

clinker, the following scenarios were assessed

- FLUREC process without salt recovery (A),
- cement clinker production without salt recovery (B),
- FLUREC process with salt recovery (C) and
- cement clinker production without salt recovery (D).

The inventory of the systems includes all environmental burdens and economic costs, respectively, from the transportation of MSWI fly ash from the MSWI plant to the respective treatment facilities, the MSWI fly ash treatment and the disposal of residues from fly ash recycling. The complete inventory is given in the supplementary information (spreadsheet B).

The production of secondary metals and the saving of quicklime in the FLUREC process, decreased demand for primary raw materials in cement production as well as the impact of ordinary MSWI fly ash disposal (without resource recovery) are considered as avoided environmental burdens and avoided economic costs, respectively. In order to calculate these avoided burdens and costs, reference systems for cement production, scrubber water neutralisation and stabilisation and subsequent above-ground disposal of MSWI fly ash were established and the environmental impacts and costs were subtracted in the respective fly ash recycling scenarios. An overview about the functions (MSWI fly ash disposal, scrubber water neutralisation, cement production) provided by different systems and the mass flows associated with these systems is illustrated in Fig. 1 in 3.1. The amount of primary raw materials (gypsum, limestone, clay, quartz, iron ore) replaced by MSWI fly ash was determined based on the mineralogy of the fly ash (c.f. Table 1).The inventory does not include upstream burdens associated with MSWI or the production and use of goods prior to their disposal in an MSWI plant (e.g. the extraction and refining of crude oil and the subsequent production of plastic packaging). Hence, the zero burden assumption (Chang and Pires, 2015) was used in accordance with Huber et al. (2018).

All burdens in a timeframe of 100 a were considered for the environmental assessment, as this is the most common timeframe used for LCA of MSWI in the literature (Hellweg et al., 2005). According to Huber et al. (2017), the ranking of different MSWI fly ash utilisation scenarios based on their environmental impact is the same for a timeframe of 100 a and an infinite timeframe.

For acidic washing of MSWI fly ash, transfer coefficients for Cd, Cu, Pb and Zn are given by Bühler and Schlumberger (2010). For all other considered heavy metals in acidic washing as part of the FLUREC process and for all heavy metals in neutral washing as pretreatment for the utilisation in the cement kiln, the transfer coefficients were determined by laboratory experiments using hydrochloric acid solution (c = 1 mol/L) as extracting agent and a liquid to solid ratio of 5 as assumed by Fellner et al. (2015b) and are given in the supplementary information (spreadsheet A). In addition, the water content of the washed MSWI fly ash sent to clinker production was determined by these experiments as 0.37 kg/kg. A detailed description of the experimental setup for the determination of the transfer coefficients can be found in Blasenbauer et al. (2015). The transfer coefficients were used to calculate the composition of washed fly ash and de-icing salt. The transfer coefficients used in this study are the same as already used by Huber et al. (2018).

For the modelling of cement production, data from Lederer et al. (2017) were used to calculate transfer coefficients from MSWI fly ash to clinker and flue gas.

A material flow analysis according to Brunner and Rechberger (2004) was conducted for all recycling systems and all reference systems based on the above mentioned transfer coefficients in order to determine the import and export flows of all systems.

2.2. Evaluation of environmental impacts

A life cycle assessment (LCA) was conducted in order to determine the environmental impacts of every scenario. The life cycle inventory data was sourced from ecoinvent database V3.2 (2015). The leachate emissions from materials disposed of on landfills and gaseous emissions additionally generated due to MSWI fly ash utilisation in the cement kiln were determined by models described in detail in Huber et al. (2017, 2018). The life cycle impact assessment was conducted using the ReCiPe model (Hierarchist perspective) (Goedkoop et al., 2009). The impact in the all endpoint and midpoint impact categories was calculated. However, only the three endpoint impact categories human health, ecosystem quality and resources as well as the midpoint impact category climate change were used for further calculations (c.f. 2.3).

It was assumed that the cement from MSWI fly ash is used in construction of buildings and that these buildings are not demolished within the timeframe of 100 a. This assumptions seems plausible as, according to Kohler and Yang (2007), more than 90% of residential buildings are not demolished within the first 100 a. As the leaching of heavy metals from concrete is very low (Colangelo et al., 2015; Guo et al., 2016; Tan et al., 2016) and the surface available for leaching is very small in buildings, heavy metal emissions from buildings were not considered in the present study. The modelling of landfill emissions was described in detail by Huber et al. (2018).

2.3. Economic evaluation

In order to examine the socio-economic viability of generating commodities from MSWI fly ash, a discounted cash flow (DCF) analysis was performed by calculating the net present value (NPV) before taxes for each scenario based on material and energy flows from the life cycle inventory. The economic evaluation of every scenario was conducted from a private investor's micro view, which is only focussed on revenues and internal costs, and from a public entity's macro view, which includes also external costs.

DCF analysis is a simple tool that is widely applied in the evaluation of mining projects (Baurens, 2010) and was also used by Winterstetter et al. (2016, 2015). It is used to determine whether a certain waste flow can be classified as resource or reserve, whereby a positive NPV indicates a reserve. The NPV is calculated according to Eq. (1) (Campbell and Brown, 2003).

$$NPV = -c_o + \frac{c_1}{1+r} + \frac{c_2}{(1+r)^2} + \bullet\bullet\bullet + \frac{c_T}{(1+r)^T} \quad (1)$$

c_0- investment cost [EUR]

c_1-c_T- discounted cash flow for year 1 to T [EUR]

T- time [a]

r- discount rate [-]

For the macro perspective, the external costs are relevant. In order to determine these costs, the results from the LCA are used and in the present study these environmental impacts were converted into monetary values.

In order to take into account the economic costs of climate change, the greenhouse gas emissions of every scenario were multiplied by the social cost of carbon (SCC) of 0.17 EUR/kg CO_2 eq. (IPCC, 2014). This corresponds to a global SCC (referring to the damage occurring in all countries) contrary to the domestic SCC (referring to the damage occurring only in the country where the emissions take place). The global SCC was used here, as Kotchen (2016) mathematically showed that it is individually rational for a single country to apply the global SCC if the discount rate is sufficiently low. The SCC given by IPCC (2014) is based on integrated assessment models. Pindyck (2016) used expert judgement of the probability of catastrophic outcomes of climate change and thereby determined a SCC in the range of 0.07 and 0.17 EUR/kg CO_2 eq., which is in line with the number used in this study. The damage to human health is expressed in disease adjusted life years (DALY) and the

Scenario	System		
	MSWI fly ash disposal	Scrubber water neutralisation	Cement production
Reference case (subtracted from all scenarios)	Stabilisation and disposal of MSWI fly ash **(1)**	Neutralisation with quicklime **(2)**	Cement production **(3)**
A	FLUREC process **(4)**	FLUREC process **(4)**	Cement production **(3)**
B	Cement production with MSWI fly ash utilisation **(5)**	Neutralisation with quicklime **(2)**	Cement production with MSWI fly ash utilisation **(5)**
C	FLUREC process with salt recovery **(6)**	FLUREC process with salt recovery **(6)**	Cement production **(3)**
D	Cement production with MSWI fly ash utilisation and salt recovery **(7)**	Neutralisation with quicklime **(2)**	Cement production with MSWI fly ash utilisation and salt recovery **(7)**

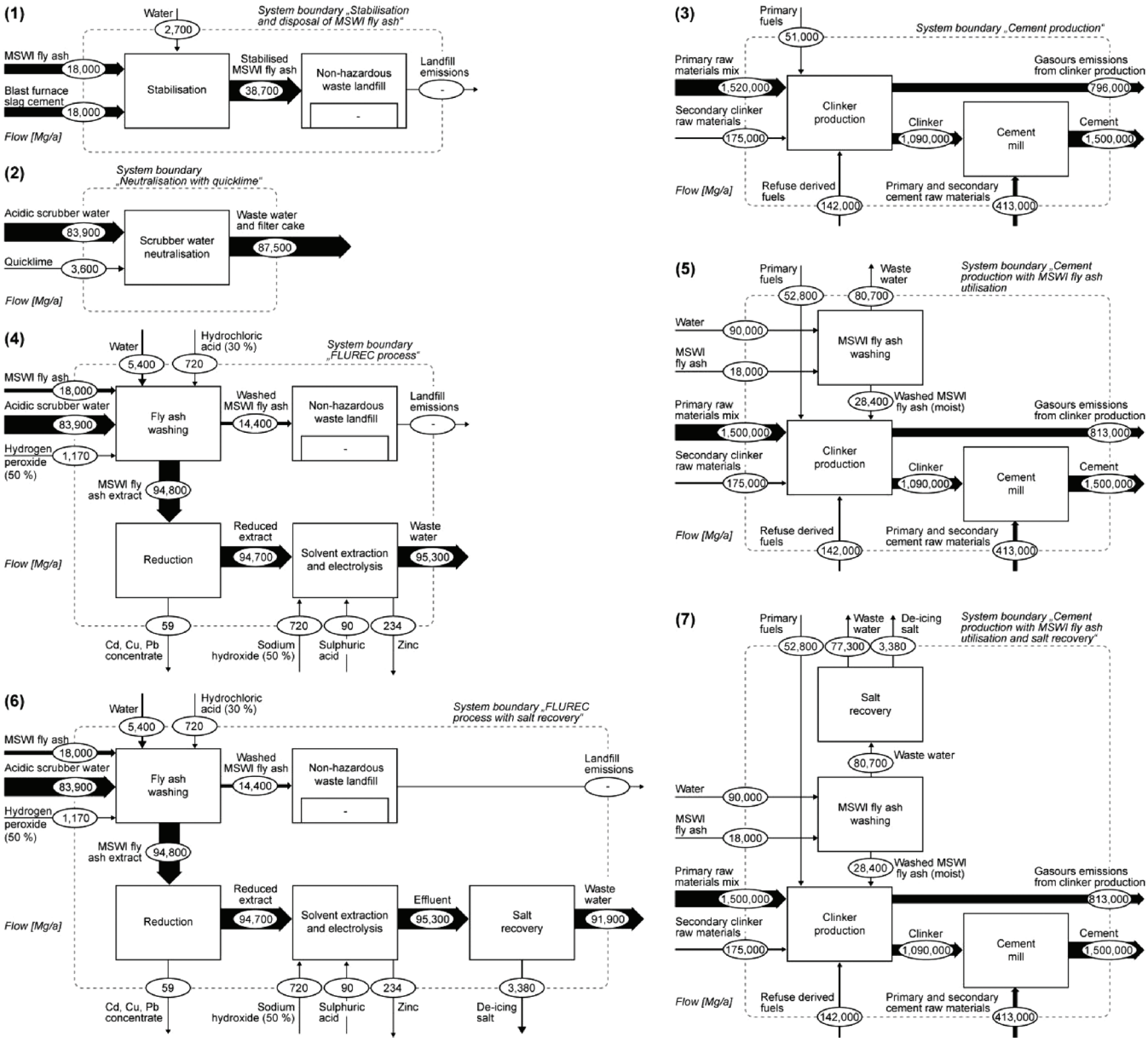


Fig. 1. Material flows of all MSWI fly ash recycling scenarios. Each scenario consists of 2–3 systems. The table gives the name and number of the system for each function (MSWI fly ash disposal, scrubber water neutralisation and cement production) in all four scenarios and the reference case. The material flows for these systems are shown in the diagrams below the table.

damage to ecosystem quality is expressed in species years in the ReCiPe method. The outcome of the endpoint impact category resource consumption is in ReCiPe already displayed in monetary values and expresses the marginal change in efforts to extract future resources (Goedkoop et al., 2009).

The monetary valuation of the damage to human health and

ecosystem quality was conducted using LIME2 according to Itsubo et al. (2012). LIME2 is a monetary valuation method based on conjoint analysis, a stated preference valuation method where the marginal value of a good is identified on the basis of stated choices between two or more alternative goods where at least one attribute of one good is systematically varied across respondents and at least one good has a market price or market price equivalent (Weidema et al., 2013). Specifically, a sample of 1000 persons was selected and a questionnaire that contained different policies was designed. Each policy comprised different levels of loss of human health, social assets, plant productivity and biodiversity as well as different tax levels. Based on the results of the survey, weighting factors were determined by application of a random parameter logit model (Itsubo et al., 2012). The weighting factors of 117,000 EUR/DALY and 113,000,000,000 EUR/species as estimated by LIME2 were used in the present study. As the damage to ecosystems is expressed in species years instead of species in ReCiPe, the respective weighting factor was divided by the inverse of the background extinction rate as estimated by De Vos et al. (2015), resulting in 11,300 EUR/species year. As the impact to resource scarcity is already expressed in monetary values, no further conversion of the LCA result is necessary. LIME2 was chosen for the monetary valuation as it applies conjoint analysis, which is considered the most appropriate approach for monetary valuation in LCA for most impact categories (Weidema et al., 2013). As the social cost of carbon only comprises the economic costs of climate change but not its effect on non-market goods like human health or ecosystem quality, no double counting takes place.

An exchange rate of 1 USD = 1 CHF = 0.86 EUR = 116 JPY was used and all monetary values were transformed to EUR in the year 2017 using the consumer price index (Statistik Austria, 2017). The monetised environmental impact (i.e. the external costs) was added to the sum of DCFs in order to calculate the NPV of the macro view for every scenario. Accordingly, in the following text sum of DCFs refers to the sum of all discounted cash flows without the monetised environmental impact and NPV refers to the sum of DCFs including the monetised environmental impact. This differentiation is only relevant for the macro perspective, as the micro perspective does not take into account the environmental impact.

To reflect the high economic risk of an investment into a recycling project, which highly depends on commodity market prices, a high discount rate of 12% was chosen for the micro perspective, while a lower discount rate of 3% was used for the macro perspective (Baurens, 2010; Winterstetter et al., 2015). Besides expressing time preference, another justification for discounting is the assumption that due to economic growth people will be richer in the future (Padilla, 2002). However, unlike the economy, ecosystem quality or resource availability do not grow, but rather decrease. Accordingly, no discounting was applied to monetised environmental impacts in the present study.

2.4. Uncertainty analysis

Parameter uncertainty of the output variables was determined by propagating the uncertainty of all input parameters in a Monte Carlo Simulation (MCS) with 100,000 runs. The parameters and the distributions used for this MCS are given in the supplementary information (spreadsheets B and E). A discernibility analyses was conducted for the NPV from a micro and macro perspective for all scenarios by calculating the difference between the MCS results of the single scenarios in all 100,000 iterations as described by Clavreul et al. (2012) and Huber et al. (2018).

2.5. Assumptions

The assumptions made in the models applied in this study are summarised in Table 2.

Table 2
Assumptions made in the models.

	Assumptions
1.	The composition of MSWI fly ash during the project lifetime is identical to the average composition of the year 2015.
2.	The share of primary clinker raw materials replaced by MSWI fly ash is 1.8 % in scenario A and C.
3.	Emission occurring after 100 a are not relevant.
4.	The investment is depreciated after 20 a.
5.	A liquid to solid ratio of 5 is used for MSWI fly ash washing/extraction.
6.	No further treatment except evaporation (fuelled by natural gas) is necessary for producing road de-icing salt from brine. The content of pollutants in this road de-icing salt produce is the same as for primary salt.
7.	The cement produced with MSWI fly ash is used for construction of buildings, which are not demolished within the timeframe of 100 a.
8.	The weighting factors determined by Itsubo et al. (2012) are applicable to Austria although they were determined for Japan.
9.	The price for the treatment of MSWI fly ash in the cement kiln is only based on the treatment costs and does not contain any potential profit of the cement producer.

3. Results and discussion

3.1. Material flow analysis

The material flows for scenario A, B, C, D and the reference case are shown in Fig. 1. The functions of each scenario comprise the treatment of 18,000 Mg/a of MSWI fly ash, the neutralisation of 83,900 Mg/a acidic scrubber water from MSWI and the production of 1,500,000 Mg/a cement. Thus, each scenario consists of 2 to 3 of the systems shown in Fig. 1 together providing all 3 functions.

The environmental impacts and economic costs of the systems of the reference case were subtracted in all scenarios to determine the environmental impacts and economic costs that are attributed to MSWI fly ash utilisation.

The annual amounts of primary raw materials that can be replaced by MSWI fly ash utilisation, as shown by the material flow analysis with the systems considered in the this study, are given in Table 3 in 3.3.

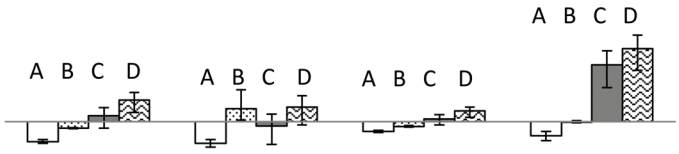
In Europe, 868,000 Mg of MSWI fly ash (not mixed with any other materials like dry system residue) were generated in the year 2011. This accounts for about 30% of air pollution control residues from MSWI (Fellner et al., 2015a).

3.2. Evaluation of environmental impacts

The environmental impact in the midpoint impact category climate change and the endpoint impact categories human health, ecosystem quality and resources for a timeframe of 100 a is shown in Fig. 2. The environmental impact in all midpoint and endpoint impact categories is given in the supplementary information (spreadsheet C). In all four

Table 3
Mass of primary raw materials replaced by secondary raw materials in all four scenarios.

[Mg/a]	Metal recovery (A)	Cement production (B)	Metal and salt recovery (C)	Cement production with salt recovery (D)
Zn	234	0	234	0
Pb	41	0	41	0
Cu	14	0	14	0
Cd	3	0	3	0
Gypsum	0	3,213	0	3,213
Limestone	0	1,868	0	1,868
Clay	0	6,190	0	6,190
Quartz	0	1,612	0	1,612
Iron ore	0	295	0	295
De-icing salt	0	0	3,382	3,382



[person equivalents/a]	Climate change	Human health	Ecosystem quality	Resources
☐ Metal recovery (A)	-945.790	-1022.356	-466.914	-672.332
⚄ Cement production (B)	-307.928	620.430	-222.336	-1.132
■ Metal and salt recovery (C)	286.092	-198.648	137.418	2676.488
☒ Cement production and salt recovery (D)	1022.055	697.712	501.373	3458.466

Fig. 2. Environmental impact for 100 a in the impact categories climate change, human health, ecosystem quality and resources. The value in the data table is the impact of the utilisation of 18,000 Mg/a MSWI fly ash divided by the annual impact of an average European in each impact category (Goedkoop et al., 2015; Sleeswijk et al., 2008). 1 person equivalent amounts to 11,200 kg CO_2 eq, 0.0202 DALY, 0.000181 species years and 308 USD, respectively. The error bars show the range containing 90% of the MCS results. Positive values indicate damages and negative values indicate benefits.

scenarios shown in Fig. 2, the impacts of the reference system landfilling of fly ash are already subtracted, as described in 2.1. Contrary to this, the environmental impact of the reference system for MSWI fly ash disposal (i.e. stabilisation with cement and subsequent disposal at a non-hazardous waste landfill) is not subtracted from the data in the supplementary information (spreadsheet C) to allow the comparison between the four recycling scenarios investigated and the reference disposal system.

The damage in all impact categories shown is larger, if salt recovery is added to either metal recovery or cement production. The reason for this is the large amount of natural gas consumed for the vaporisation of water in order to obtain a marketable product (i.e. road de-icing salt). Consequently, the environmental impact and thereby also the external costs of salt recovery could be reduced by using renewable energy (e.g. solar heat) for water vaporisation or by using brine derived from MSWI fly ash washing water instead of solid salt for de-icing of roads. In case of using brine instead of solid salts, less water has to be evaporated. The use of solar heat for evaporation was not considered in the present study, as all-season solar heat is mainly available in countries with a warmer climate, which then do not need road de-icing salt. The results also show that from an environmental point of view, disposal of wastewater from MSWI fly ash might be preferable over utilisation of chloride salts.

The environmental impact of metal recovery and cement production from MSWI fly ash is lower compared to the disposal after stabilisation with cement. The only exception is the impact category human health for cement production due to the high emissions of gaseous Hg from the cement kiln, if MSWI fly ash is utilised. As a result, both recycling options show an environmental benefit (i.e. negative values in Fig. 2). This environmental benefit is larger for the metal recovery scenario in all impact categories. The reason for this comparably large benefit is the substitution of primary metals (Zn, Pb, Cu, Cd) by secondary metals derived from MSWI fly ash.

3.3. Economic evaluation

The mass of primary raw materials that are replaced by secondary raw materials in each of the investigated scenarios is given in Table 3. The NPV for all four scenarios is shown in Fig. 3 and the percentage of DCF and monetised effect on climate change, human health, ecosystem quality and resources in the total NPV from a macro view is shown in Fig. 4. The amount of all costs and revenues for every scenario is given in the supplementary information (spreadsheet D). In scenario A, the overall NPV from the macro perspective is dominated by human health and climate change because the absolute value of the DCF is very low compared to the other scenarios. Due to the higher absolute value of the DCF, DCF dominates the NPV from the macro perspective in scenarios B–D. In scenario B, the damage to human health as a result of Hg emissions also plays an important role. The savings from the current disposal practice make up 88% of the revenue generated in scenario B, while the remaining 12% are due to the savings of primary raw materials. These savings are also the largest item generating revenue in scenario A. However, scenario A is associated with significantly higher investment costs than scenario B. The economic evaluation results of scenarios C and D are dominated by the expense for natural gas.

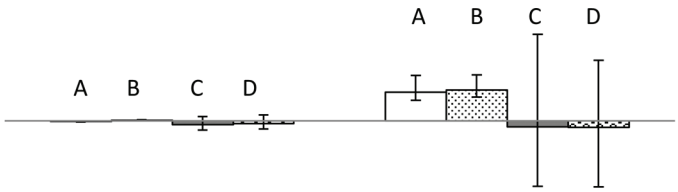
As the NPV for the recycling scenarios including salt recovery is lower compared to the respective scenarios without salt recovery both from a micro and macro perspective, salt recovery cannot be considered as an economically viable option.

For a private investor, metal recovery from MSWI fly ash is not an option, because the revenues do not cover the costs for this process. However, for a public entity this option might be more interesting, as on the one hand the discount rate applied is lower (c.f. 2.3) and on the other hand the loss of money is overcompensated by the environmental benefit in the impact categories human health, climate change and resources, as can be seen in Fig. 4.

Production of cement from MSWI fly ash seems to be economically viable from a private investor's micro perspective as well as from a public entity's macro view. The sum of DCFs is positive in both cases. Although the monetised effect on the environment is negative, mainly due to damage to human health, the overall result is still positive and in both cases the median NPV is higher for the cement production scenario compared to the metal recovery scenario. The application of different weighting (valuation) factors could of course yield a different result. Furthermore, it is assumed that the treatment of MSWI fly ash in the cement kiln is associated with no additional cost apart from the expenditure for fly ash washing and the increased demand for hard coal in the cement kiln. However, cement producers usually charge fees for the treatment of waste that are higher than their costs associated with this treatment. If cement producers charged 100 EUR/Mg of treated MSWI fly ash, the NPV would decrease by 13.4 million EUR for the micro view and by 26.8 million EUR for the macro view, due to the different discount rates applied. Nevertheless, the median NPV would still be positive for both perspectives.

3.4. Uncertainty analysis

The parameter uncertainty is shown as error bars representing the range between the 5 percentile and the 95 percentile, which contain 90% of the MCS results, in Figs. 2 and 3. For the micro perspective, the parameter uncertainty for the NPV in scenarios A and B (no salt recovery) is about 10–30 million EUR, while it amounts to about 200–300 million EUR in scenario C and D (with salt recovery). The reason for this high uncertainty in salt recovery is the high consumption of natural gas and the significant price fluctuations associated with this energy carrier. Contrary to this, the parameter uncertainty is about 200–600 million EUR in scenarios A and B and about 2–3 billion EUR in scenarios C and C if the macro perspective is considered. The high uncertainty of the results for the macro perspective reflects the high uncertainty associated with monetary valuation of LCA results. In the supplementary information (spreadsheets B, C and D), the median value, 5 percentile and 95 percentile are given for every parameter.



[EUR]	NPV micro	NPV macro
Metal recovery (A)	-2,80,00,000	1,00,90,00,000
Cement production (B)	1,80,00,000	1,08,80,00,000
Metal and salt recovery (C)	-13,90,00,000	-21,70,00,000
Cement production and salt recovery (D)	-9,30,00,000	-23,00,00,000

Fig. 3. Net present value (including the monetised environmental impacts) of all four scenarios from a private investor's micro and a public entity's macro perspective. The error bars show the range containing 90% of the MCS results.

The results of the discernibility analysis for the NPV from a micro and macro view are shown in Table 4. The results show that the NPV of the two recycling options metal recovery (scenario A and C) and cement production (scenario B and D) is higher without salt recovery in at least 78.1% of the MCS results from a micro and at least 99.4% from a macro perspective. Cement production is more profitable than metal recovery in 100% of the MCS results from a micro perspective due to lower cost of this recycling option (if treatment in the cement kiln is done at no charge). However, metal recovery is more profitable than cement production in 57.4% of the MCS results from a macro perspective due to its large environmental benefit.

3.5. Resource classification

The application of resource classification according to UNFC (UNECE, 2010) to the anthropogenic resource MSWI fly ash is shown in Fig. 5. In this classification framework occurrences are graded according to socio-economic viability, project feasibility and geological knowledge, resulting in a three-digit code. Low numbers indicate preferable conditions, while higher numbers indicate low feasibility or low knowledge, respectively. Every three-digit code is associated with one of the classes commercial projects, potentially commercial projects, non-commercial projects, additional quantities in place associated with known deposits, exploration projects, additional quantities in place with potential deposits and other combinations.

MSWI fly ash is graded with "1" for the utilisation in cement production from a micro as well as from a macro perspective in the category socio-economic viability (E), based on the DCF analysis and LCA applied in the present study. It has to be noted that this option is feasible from a legal point of view despite the Hg emissions to the atmosphere (BMLFUW, 2016). With regard to the recovery of metals, MSWI fly ash is graded as "2" from a micro view because most of the MCS results show a negative NPV and it is graded as "1" from the macro view because the inclusion of external costs yields a positive NPV in most of the MCS results. A positive NPV from the micro perspective could be achieved by Zn prices of about 19 EUR/kg instead of 3 EUR/kg. According to Winterstetter et al. (2015), an increase in metal prices by a factor of 10 is still realistic within the next 20 a. The inclusion of salt recovery into either metal recovery or cement production substantially decreases the NPV from both perspectives. In order to be economically viable either the gas price has to decrease from 0.07 EUR/kWh to 0.0016 EUR/kWh or the salt price has to increase from 0.13 EUR/kg to 5.33 EUR/kg. As such severe price changes (factor 40) are not realistic in the next 20 a, salt recovery from MSWI fly ash is graded as "3". In case of use of brine instead of solid salts, less water has to be evaporated. This could improve the grade of salt recovery. The use of solar heat for evaporation was not considered in the present study, as all-season solar heat is mainly available in countries with a warmer

Table 4
Results of the discernibility analysis for the NPV from a private investor's micro and a public entity's macro perspective. The number displays the percentage of MCS results that give a higher NPV for the scenario in the first column compared to the scenario in the first line (e.g. the table shows that the NPV from the micro view of scenario C is in 1.9% of the MCS results higher than that of scenario A). A…metal recovery, B…cement production, C…metal and salt recovery, D…cement production and salt recovery.

Private investor's micro perspective				
	A	B	C	D
A	–	0.0	98.1	78.1
B	100.0	–	100.0	98.1
C	1.9	0.0	–	0.0
D	21.9	1.9	100.0	–
Public entity's macro perspective				
	A	B	C	D
A	–	57.4	99.4	99.5
B	42.6	–	97.8	99.4
C	0.6	2.2	–	24.1
D	0.5	0.4	75.9	–

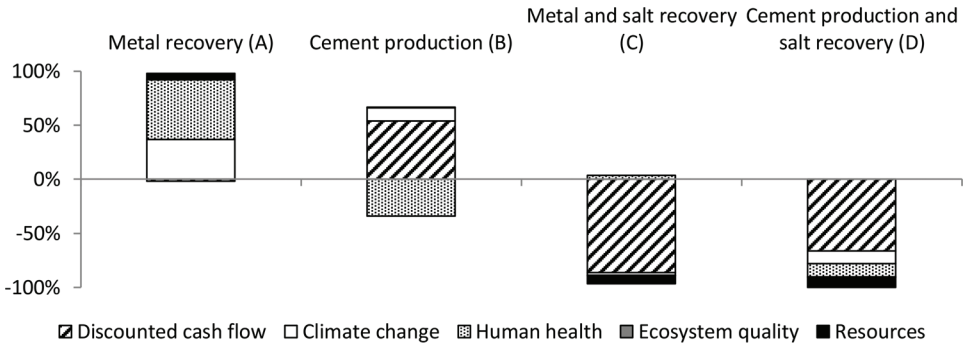


Fig. 4. Percentage of DCF and monetised effect on climate change, human health, ecosystem quality and resources (referred to the sum of their absolute values from a macro view). Only median values without uncertainty are displayed for better clarity. Positive values represent revenues and negative values represent costs. The amount of all costs and revenues is given in the supplementary information (spreadsheet D).

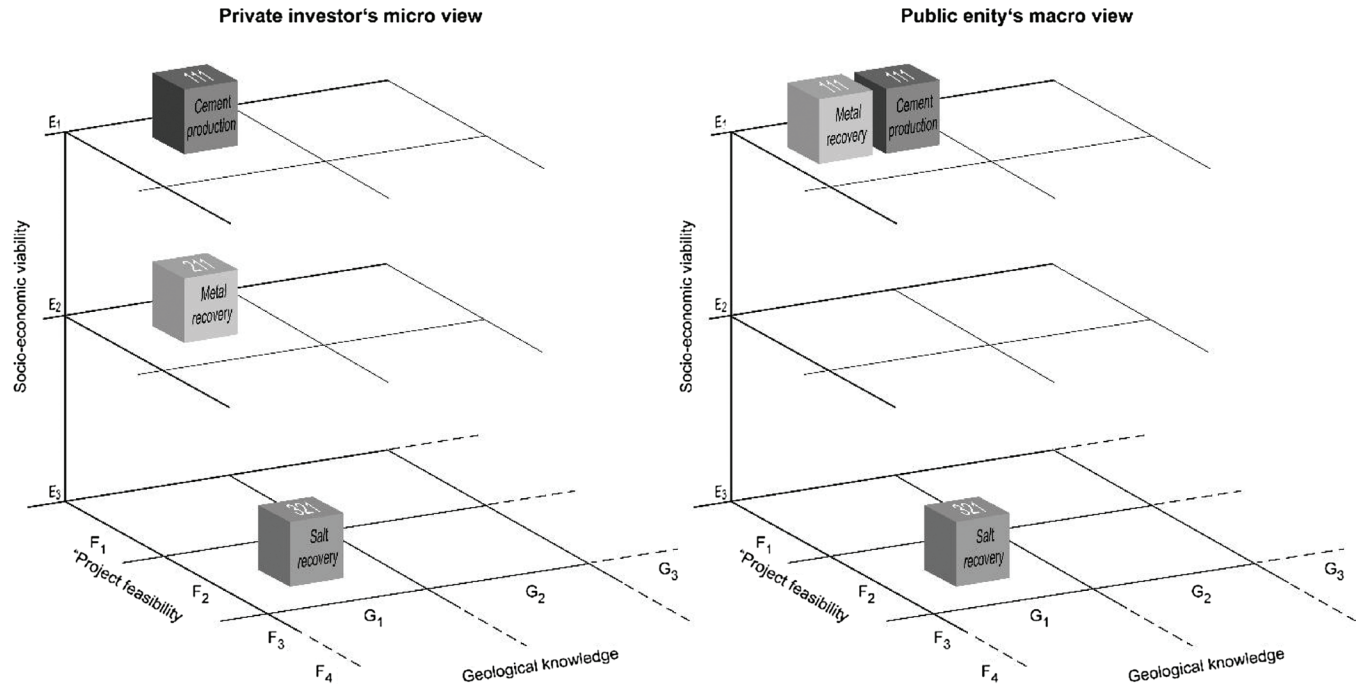


Fig. 5. Resource classification of MSWI fly ash for metal recovery, cement production and salt recovery according to UNFC. MSWI fly ash is graded according to socio-economic viability, project feasibility and geological knowledge.

climate, which then do not need road de-icing salt.

With regard to the classification category project feasibility (F), MSWI fly ash can be classified as "1" for the production of cement and the recovery of metals, as metal recovery is currently taking place (e.g. in Switzerland) and the feasibility of cement clinker production has been shown in detailed studies (Guo et al., 2016; Saikia et al., 2007). The recovery of chloride salts from MSWI fly ash is graded as "2" because extraction is subject to further evaluation (Easymining, 2017; Stena Metall, n.d.), but to the authors' best knowledge no extraction and utilisation of chloride salts is currently taking place in full scale. For this category no difference between micro and macro perspective is made.

MSWI fly ash is sampled and analysed in regular intervals and the amount of this residue generated each year is well known. Consequently, MSWI fly ash can be graded as "1" in the category geological knowledge (G). This grade is applicable to all three recycling options and micro as well as macro perspective.

By combining these three criteria, MSWI fly ash can be classified as "211" for metal recovery from a private investor's micro perspective, "111" for metal recovery from a public entity's macro perspective, "111" for cement production and "321" for chloride salt recovery.

3.6. Sources of error, limitations and caveats

Due to limited availability of data, only four impact categories of the LCA were used for the monetary valuation. However, the economic effect of damages or benefits in other midpoint impact categories (acidification, eutrophication, etc.) should also be accounted for, if the total external costs are to be quantified and included into the DCF analysis.

Another caveat is the application of LIME2 for monetary valuation, which aims at representing the environmental attitude of the Japanese public (Itsubo et al., 2012). Further investigations on conjoint analysis have to be conducted in order to verify that the weighting factors of LIME2 are also valid for other high income countries. Furthermore, it has to be considered that LIME2 is based on the stated preference of individuals and these individuals might be ill-informed and not adequately incorporate social fairness, environmental sustainability and other important goals (Costanza et al., 1997).

For the monetary valuation of greenhouse gas emissions, the SCC as estimated by IPCC (2014) was used. This value corresponds to a more commonly used utilitarian SCC contrary to a prioritarian SCC as proposed by Adler et al. (2017). According to Adler et al. (2017) a prioritarian SCC could be higher or lower than a utilitarian SCC and therefore the monetary value of greenhouse gas emission cuts could be increased or decreased.

According to Costanza et al. (1997) the supply and demand curves for essential ecosystem services do not match curves for normal goods. This means that the price of these services approaches infinity as quantity goes to zero. This issue is not accounted for in LCA with monetary valuation, as linear damage functions and linear valuation are assumed.

For the LCA conducted in the present study, only emissions taking place during the first 100 a were accounted for. However, in the metal recovery scenarios and the reference scenario (stabilisation of MSWI fly ash with cement) solid residues are disposed of at above-ground landfills. From these landfills pollutants like heavy metals are emitted to the environment over time periods considerably longer than 100 a (Doka, 2003; Hellweg et al., 2005; Hyks et al., 2009). The utilisation of MSWI fly ash in cement production also increases the heavy metal content of cement (Lederer et al., 2017). However, additional emissions from buildings during and after their lifetime due to a higher heavy metal content of cement are not considered in presented calculations and their results. The inclusion of these long-term emissions from landfills would slightly increase the NPV from a public entity's macro perspective, as the long-term emissions are lower in all recovery scenarios compared to the reference MSWI fly ash disposal option (i.e. stabilisation with cement and subsequent above ground disposal).

A general limitation of resource classification of waste streams compared to geogenic or anthropogenic deposits is that only the composition of the material as it arose in the past can be determined and therefore form the basis of the economic and environmental assessment. The utilisation of these waste streams, on the contrary, can only take place in the future, which is why any potential changes in composition of the waste streams investigated that might be relevant cannot be accounted for.

Because of the limitations noted above, it can be expected that the current estimate of the monetary value of environmental damages and benefits represents a minimum value. As a consequence, it cannot be precluded that metal recovery from MSWI fly ash is preferable over cement production from a public entity's macro perspective, contrary to the results shown in 3.3 and 3.4. This could also negatively affect the resource classification in the category socio-economic viability from a public entity's macro view, but will not change the classification from a private investor's micro view.

4. Conclusion

In this study, the application of the resource classification framework UNFC on the anthropogenic resource MSWI fly ash was demonstrated. Cement production, metal recovery and chloride salt recovery from MSWI fly ash were investigated. In order to account for the environmental impacts, an LCA was performed and the results in the impact categories climate change, human health, ecosystem quality and resources were transformed to monetary values and added to the NPV.

It can be concluded that the integration of LCA into resource classification is possible after monetary valuation. The approach used in the present study enables the comparison of waste utilisation projects with primary raw materials exploitation projects. This comparison represents valuable information for those who are taking decision about investments in such projects, but also for waste managers in general. The methodology applied can also be especially useful in view of circular economy, as e.g. recycling projects can be evaluated based on their environmental and economic performance, which enable the assessment of recycling rates (i.e. up to which extent is recycling preferable over primary raw material use).

The study shows that from a micro view cement production is classified as commercial project (111) and from a macro view cement production and metal recovery are classified as commercial projects, while recovery of chloride salt is graded as non-commercial project (321) from both perspectives.

Due to the limitations of the monetary valuation applied in the present study, a potentially higher value of environmental damages and benefits and, hence, also a worse classification in the category socio-economic viability cannot be precluded.

As a consequence, further efforts to establish valid weighting factors for monetary valuation of environmental impacts are still necessary.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.resconrec.2018.08.003.

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