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COMPARATIVE LIFE CYCLE ASSESSMENT OF THE UTILISATION OF MSWI FLY ASH IN CEMENT PRODUCTION AND METAL RECOVERY

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SUMMARY: Municipal solid waste incineration (MSWI) fly ash constitutes a hazardous waste that is nowadays disposed of either at underground deposits or after cement stabilisation on an above-ground landfill. In the present study, a life cycle assessment model for two recycling options (acidic washing with integrated metal recovery and utilisation as clinker raw material) was developed with regard to two different time horizons (100 years and infinite). The uncertainties of the input parameters were propagated by Monte Carlo simulations (MCS). The results show that the metal recovery has a lower environmental impact than utilisation in cement production in more than 99% of the MCS, regardless of the time horizon considered. Consequently, a clear decision support can be given in this case study although there is still no standardised method to account for long-term emissions.

1. INTRODUCTION

Municipal solid waste incineration (MSWI) fly ash constitutes a hazardous waste that is nowadays disposed of either at underground deposits or after cement stabilisation on an above-ground landfill (Huber et al., 2016).

However, in recent years a process for metal recovery (Zn, Pb, Cd, Cu) from MSWI fly ash, namely the FLUREC process, was developed (Schlumberger, 2010). In this process acidic scrubber water from the waste incineration plant is mixed with MSWI fly ash and used as an extraction agent. After a 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 recovery Zn from the solution.

Alternatively, MSWI fly ash can be utilised as a raw material for cement production, making use of its cementitious properties and high content of Ca, Si and Al. This utilisation option leads to a decreased primary raw material consumption as MSWI fly ash is used as secondary raw materials 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., 2016). In order to prevent corrosion in the cement kiln and to obtain cement of sufficient quality, Cl has to be
removed from MSWI fly ash. 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).

2. METHODOLOGY

2.1 Goal and scope of the life cycle assessment

The goal of the present study is the evaluation and comparison of two different scenarios for the utilisation of MSWI fly ash. The functional unit is the processing of 1 Mg of MSWI fly ash from a grate incinerator with wet flue gas cleaning. A representative sample of such fly ash was collected at an incinerator in Vienna equipped with activated coke injection into the flue gas stream, a filtering separator and a multistage scrubber. The composition of the fly ash was determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) according to EN 11885 (2009). 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

<table>
<thead>
<tr>
<th>Element</th>
<th>Mass fraction [mg/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>39,000</td>
</tr>
<tr>
<td>Sb</td>
<td>520</td>
</tr>
<tr>
<td>As</td>
<td>21</td>
</tr>
<tr>
<td>Ba</td>
<td>900</td>
</tr>
<tr>
<td>Pb</td>
<td>2,300</td>
</tr>
<tr>
<td>Cd</td>
<td>180</td>
</tr>
<tr>
<td>Cr</td>
<td>190</td>
</tr>
<tr>
<td>Co</td>
<td>24</td>
</tr>
<tr>
<td>Fe</td>
<td>12,000</td>
</tr>
<tr>
<td>Cu</td>
<td>780</td>
</tr>
<tr>
<td>Mn</td>
<td>710</td>
</tr>
<tr>
<td>Mo</td>
<td>15</td>
</tr>
<tr>
<td>Ni</td>
<td>51</td>
</tr>
<tr>
<td>Hg</td>
<td>13</td>
</tr>
<tr>
<td>Se</td>
<td>0.05</td>
</tr>
<tr>
<td>Ag</td>
<td>27</td>
</tr>
<tr>
<td>Zn</td>
<td>13,000</td>
</tr>
<tr>
<td>Sn</td>
<td>480</td>
</tr>
</tbody>
</table>

The life cycle inventory includes all burdens from the transport of MSWI fly ash from the MSWI plant to the respective treatment facilities, the MSWI fly ash treatment and the disposal of residues. The benefits for decreased primary raw material consumption were included into the
LCA system. 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.

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 fuel 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 ecotoxicity, urban land occupation, water depletion) and endpoint impact categories (human health, ecosystem quality, resources, total score) was calculated.

2.2 Modelling of landfill emissions

The solid residue of MSWI fly ash extraction in the FLUREC process is disposed of at above-ground landfills. In case of utilisation of MSWI fly ash in cement production, it is assumed that no heavy metals are released during the use phase of the released concrete, as several studies investigating the leachability of toxic substances from concrete mortar with MSWI fly ash addition show very low leachability (Colangelo et al., 2015; Guo et al., 2016; Tan et al., 2016) and the area of buildings that is in contact with water is usually very low. However, eventually buildings are deconstructed and the concrete is disposed of at a landfill for inert material.

The leachate collected from landfills is commonly fed into the municipal sewer (Calabrò, et al., 2016; Tosti et al., 2016). One part of the heavy metals 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.

The emissions from landfills were calculated based on the composition of the deposited material and transfer coefficients given by Doka (2003b) for both time frames.

The composition of FLUREC residue was calculated based on transfer coefficients provided by Bühler and Schlumberger (2010) (for Cd, Cu, Pb and Zn). Transfer coefficients for other heavy metals 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 from MSWI fly ash to clinker and flue gas given by Lederer et al. (2016) were used.
2.3 Uncertainty analysis

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 the total score for each scenario by calculation of 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).

3. RESULTS

3.1 Material flow analysis

The material flows of the two recycling options are shown in Figure 1 and Figure 2. The functional unit of each system is the utilisation of 1 Mg of MSWI fly ash. The system shown in Figure 1 additionally fulfils the function of neutralising 4,660 kg of acidic scrubber water and the system shown in Figure 2 additionally fulfils the function of providing 83,410 kg cement. The reference process for the function scrubber water neutralisation is illustrated in Figure 3 and the reference process for the function cement production is illustrated in Figure 4. The environmental impact of these reference processes was substracted in the respective scenario to determine the impact that is attributed to MSWI fly ash utilisation.
Figure 1. Material flows of the FLUREC process [kg]. The mass of landfill emissions depends on the time frame considered (100 years or infinite).
Figure 2. Material flows of MSWI fly ash utilisation in cement production [kg]

Figure 3. Material flows for default scrubber water neutralisation [kg]
3.2 Environmental impact for 100 years

The environmental impact of both utilisation options for a period of 100 years in all midpoint impact categories is shown in Figure 5.
The largest difference between the two investigated recycling options with regard to the midpoint impact categories was found for human toxicity. The application of the FLUREC process results in an environmental benefit (-127 kg 1,4-DCB equivalents) because of the substitution of primary metal production by secondary metal from MSWI fly ash, while utilisation of MSWI fly ash in the cement industry causes considerable damage in the category human toxicity due to high Hg emissions to air (1,473 kg 1,4-DCB equivalents). On the other hand, utilisation of MSWI fly ash in cement production results in a benefit in terrestrial ecotoxicity (-3.2 kg 1,4-DCB equivalents) due to substitution of primary gypsum, while the FLUREC process is associated with a burden (0.24 kg 1,4-DCB equivalents). The benefit in natural land transformation caused by the FLUREC process (-0.35 m²) is a result of depositing material on an above-ground landfill, while the impact of the cement industry scenario is caused by the increased demand of coal as a fuel in the cement kiln. This fuel demand is also responsible for the higher impact in the categories particulate matter formation, photochemical oxidant formation, and terrestrial acidification.
formation and terrestrial acidification. The benefit in metal depletion that is caused by utilising MSWI fly ash in cement production (-13.1 kg Fe equivalents) is a result of a decreased demand for iron ore in clinker production as MSWI fly ash contains a considerable amount of iron oxide. However, it is still lower than the benefit of the FLUREC process (-47.9 kg Fe equivalents).

The LCA results for the endpoint impact categories are shown in Figure 6. This figure shows that the environmental impact in all endpoint impact categories is significantly lower for the metal recovery scenario.

![Figure 6](image.png)

**Figure 6.** Environmental impact for 100 years in all endpoint impact categories. The value on the primary vertical axis is the impact of the utilisation 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.

### 3.3 Environmental impact for an infinite time period

Figure 7 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.
Figure 7. Environmental impact for an infinite timeframe in all toxicity midpoint impact categories. The value on the vertical axis is the impact of the utilisation 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.

Contrary to the short-term perspective, no environmental benefit is caused by recycling of MSWI fly ash, if long-term emissions are included in the assessment. The impact in all toxicity impact categories is higher for the utilisation of MSWI fly ash in cement industry compared to zinc recycling because heavy metals are extracted from the MSWI fly ash. Therefore the mass of heavy metals finally disposed of at an above-ground non-hazardous waste landfill is lower, causing lower damage after leaching. The LCA results for the endpoint impact categories are shown in Figure 8. This figure shows that the environmental impact of utilising MSWI fly ash in cement production is higher for all endpoint impact categories, if long-term emissions from landfills are considered. The total score (aggregated impact) is increased by a factor of approximately 2 compared to the total score excluding long-term emissions.

Figure 8. Environmental impact for an infinite timeframe in all endpoint impact categories. The value on the primary vertical axis is the impact of the utilisation 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 Uncertainty analysis

As described in section 2.3 parameter uncertainty was 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 Figure 5 to Figure 8. The parameter uncertainty ranges between about 1 and 33% of the median value.

The discernibility analysis showed that the environmental impact of the FLUREC process is in 99.96% (excl. long-term emissions) or 99.95% (incl. long-term emissions), respectively, of the MCS results lower compared to the utilisation of MSWI fly ash in cement production.

For both scenarios, the three input parameters contributing most to the uncertainty of the aggregated overall environmental impact were identified and are shown in Table 2.

Table 2. Parameters with the highest contribution to uncertainty of aggregated overall environmental impact (excl. LTE…excluding long-term emissions, incl. LTE…including long-term emissions).

<table>
<thead>
<tr>
<th>Recycling option</th>
<th>Parameters with highest contribution to uncertainty</th>
<th>Highest</th>
<th>Second highest</th>
<th>Third highest</th>
</tr>
</thead>
<tbody>
<tr>
<td>FLUREC process (excl. LTE)</td>
<td>Zn content of MSWI fly ash (28.9%)</td>
<td>Transfer coefficient from landfill to environment in 100 a for Sb (10.5%)</td>
<td>Transfer coefficient from landfill to environment in 100 a for Mn (7.6%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Hg content of MSWI fly ash (57.0%)</td>
<td>Hg content of ordinary clinker raw material (21.8%)</td>
<td>Hg content in cement kiln off gas without MSWI fly ash utilisation (7.7%)</td>
<td></td>
</tr>
<tr>
<td>FLUREC process (incl. LTE)</td>
<td>Ag content of MSWI fly ash (14.9%)</td>
<td>Transfer coefficient from landfill to environment in 100 a for Sb (10.4%)</td>
<td>Transfer coefficient of MSWI fly ash to washed fly ash (8.2%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Zn content of MSWI fly ash (45.9%)</td>
<td>Hg content of MSWI fly ash (31.1%)</td>
<td>Hg content of ordinary clinker raw material (9.1%)</td>
<td></td>
</tr>
</tbody>
</table>

The data indicates that the content of Hg in MSWI fly ash and its transfer coefficient to the cement kiln off gas (calculated from Hg content of clinker raw material and Hg content in cement kiln of gas) are responsible for the highest share of the uncertainty of the environmental impact of MSWI fly ash recycling in cement production, while the uncertainty of the environmental impact of the FLUREC process is determined mainly by the Zn content of MSWI fly ash and the transfer coefficients from landfills to the environment.

### 4. DISCUSSION

The presented model allows comparing the environmental impact of different recycling...
options for MSWI fly ash. Both recycling options counter metal depletion, as primary Zn, Cd, Cu and Pb or iron ore, respectively, can be substituted by MSWI fly ash constituents and the mass of waste that has to be disposed of at a landfill is decreased in both cases. Consequently, both options can contribute to a shift towards a circular economy.

Nevertheless, the recycling of MSWI fly ash has a considerable environmental impact on human health, ecosystem quality and fossil fuel depletion. Compared to utilisation of MSWI fly ash in cement production, the FLUREC process has a rather low environmental impact, 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 discernibility analysis shows the same result for both timeframes considered (100 years and infinite). This means that the way how long-term emissions from landfills are treated in LCA has no effect on the ranking in this case study. Consequently, a clear decision support can be given although there is no standardised methodology to account for long-term metal emissions.

5. CONCLUSION

A LCA model for the determination of the environmental impact of two different recycling options for MSWI fly ash was developed. The presented results show that application of the FLUREC process is preferable to utilisation of MSWI fly ash in cement production.

Furthermore, it could be demonstrated that the consideration of long-term landfill emissions has no effect on the ranking of the two options.

Additional research is still needed to determine the environmental impact of recycling options not considered within the scope of this study, like salt recovery, as well as the environmental impact of combined recycling process. It still has to be determined, if the positive effect of the FLUREC process (heavy metal extraction and recycling) can be combined with the positive effects of utilisation in cement production (mineral recycling, saving of landfill volume). For this future research experimental approaches are equally important as environmental impact assessment.

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