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RESOURCE RECOVERY FROM HAZARDOUS WASTE – THE CASE OF MSW INCINERATION FLY ASHES

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SUMMARY: In the European Union almost 78 million tons of waste are annually combusted in Waste-to-Energy plants. Solid residues generated thereby contain altogether about 72,000 t/a of Zn, of which more than 50 % accumulates in air pollution control residues. Intensive research activities aiming at Zn recovery from such residues recently resulted in a large scale Zn recovery plant at a Swiss waste incinerator. By acidic leaching and subsequent electrolysis this technology (FLUREC) allows to generate metallic Zn of purity >99.9%. In the present paper the economic viability of the FLUREC technology with respect to Zn recovery from different solid residues of waste incineration has been investigated and subsequently been classified according to the mineral resource classification scheme of McKelvey. The results of the analysis demonstrate that recovery costs are generally higher than the current market price of Zn, which implies that none of the identified Zn resources present in incineration residues can be economically extracted and thus cannot be classified as a reserve. Only for about 7,200 t/a of Zn an extraction would be marginally economic, meaning that recovery costs are only slightly (less than 50%) higher than the current market price for Zn. For the remaining Zn resources production costs are between 3 to 5 times (10,800 t/a Zn) and 20 to 80 times (54,000 t/a Zn) higher than the market price.

1. INTRODUCTION

In 2011 about 23% of Municipal Solid Waste (MSW) generated within the European Union has been thermally valorised, which amounts to about 78 million tons of waste. Besides the production of electricity and heat, MSW incineration (MSWI) goes along with the generation of bottom ash and Air-Pollution-Control (APC) residues, namely fly ashes (including boiler ash and filter ash) and filter cake. Filter cake resulting from water purification, of incinerators using a wet flue gas cleaning system, is not considered in this work due to its small mass and low Zn content. While in many countries bottom ash is already processed in order to recover some of the metals contained (mainly iron scrap, but also aluminium and copper), APC residues (which amount in total to about 2 million tons in the European Union) have been hardly considered for resource recovery so far. In all European countries they are classified as hazardous waste, which results from environmental concerns regarding the leaching of easily soluble salts (such as Cl, Na or K) and heavy metals (such as Cd, Pb, Cu or Zn) and their content of dioxins. The latter are hardly leachable, but of major concern because of their toxicity.

Due to these characteristics APC residues are either landfilled at hazardous waste landfills (this includes also the backfilling of former salt mines) or are stabilized with cement or other chemicals in order to comply with regulatory limit values for waste acceptance at non-hazardous landfills. Both practices are associated with significant costs (up to € 200/t fly ash) and the loss of valuable materials.

Only in few European countries attempts are made to recycle APC residues (Astrup, 2008) or at least parts of them. In the Netherlands, for instance, fly ashes partly substitute filler material in asphalt used in road construction. Even though the encapsulation in asphalt may last longer than in cement the dilution and dispersion of pollutants, resulting from this practice has to be criticized from an environmental perspective. In Switzerland several waste incinerators treat their fly ashes by applying acidic washing. Salts/brine (e.g. for the regeneration of ion exchangers or for de-icing roads during winter time) as well as heavy metals are thereby separated from the fly ash, and the processed "almost heavy metal free" fly ash cake is landfilled together with bottom ash. The heavy metals enriched filtrate is neutralized and the hydroxide sludge (rich in Zn) generated thereby can be sent to specific zinc-oxide recycling facility (FLUWA process, Schlumberger, 2010).

Over the last few years this so called FLUWA process has been further developed and extended such that Zinc can be recovered directly at the incineration plant. This new technology (FLUREC) has recently been introduced on large scale at a Waste-to-Energy (WtE) plant in Switzerland (KEBAG., 2013). In general, the recovery of Zn out of MSWI fly ashes has gained increasing interest in recent times, also outside of Switzerland. Numerous research activities in different European countries have been dedicated to a recovery of heavy metals contained in MSWI APC residues (e.g. (Karlfeldt Fedje et al., 2012; Karlfeldt Fedje et al., 2010a; Keppert et al., 2012; Meylan and Spoerri, 2014). However, most studies conducted so far focused mainly on the technical and environmental evaluation of heavy metal recovery (e.g. Boesch et al, 2014). Economic considerations with respect to metal recovery are rather rare and at best limited to residues of certain WtE plants (Fedje el al., 2014).

Hence, the aim of the present paper is to evaluate the potential for recovery of Zn from European MSWI residues (including bottom ash and fly ash). Thereto the framework for evaluating anthropogenic resources recently developed by Lederer et al. (2014) has been applied. Their approach foresees a combination of analysing material flows of the resource of interest and a subsequent economic assessment for the recovery of those material flows.

2. MATERIAL AND METHODS

In general, the applied framework for evaluating anthropogenic resources follows the following procedure (see Figure 1). Due to the fact that investigations have a priori been dedicated to the MSWI residues the first step of resource prospection has been left out in the frame of the present investigations.

Evaluation step	Method	Result
1. Prospection	Identification of stocks/flows based on macro-level MFA	Relevant anthropogenic stocks/flow identified and estimated
2. Exploration	Detailed stock/flow characterization based on micro-level studies	Grade, size of stock/flows, incl. uncertainties
3. Evaluation	Selection of technologies and economic analysis of costs and revenues	Costs/revenues ratio
4. Classification	McKelvey cross classification	Reserves, resources, & other occurences of anthropogenic stocks/flows

Figure 1 Procedure for the evaluation of anthropogenic resources (Lederer et al., 2014)

2.1. Exploration of Zn flows in MSWI residues

In order to explore residues from waste incineration as potential secondary resource for Zn a detailed literature analysis focusing on the following issue has been conducted.

- the amounts of waste incinerated in European Waste-to-Energy plants (CEWEP, 2011),
- the technology of incineration applied - distinguishing between grate incineration & rotary kilns on the one hand and fluidized bed incinerators on the other hand (ISWA, 2006, 2013),
- the technology of air pollution control APC systems (wet, dry & semi-dry residue systems) used at European plants and the respective amount of APC residues (ISWA, 2006, 2013),
- the Zn content in different MSWI residues (Abe et al., 2000; Aubert et al., 2007; Aubert et al., 2004a, b; Auer et al., 1995; Boesch et al., 2014; Bontempi et al., 2010; Chiang et al., 2008; De Boom et al., 2011; Fedje et al., 2010; Ferreira et al., 2005; Hallgren and Strömberg, 2004; Hjelmar, 1996; Jakob et al., 1996; Karlfeldt Fedje et al., 2010b; Karlsson et al., 2010; Lam et al., 2010; Mangialardi, 2003; Nagib and Inoue, 2000; Nowak et al., 2013; Quina et al., 2008; Schlumberger, 2010; Van Gerven et al., 2007), and
- transfer coefficients describing the portioning of Zn to the different outputs of incineration plants (Brunner and Mönch, 1986; Morf and Brunner, 1998; Schachermayer et al., 1996)

In most cases numerous data sources (as indicated above) have been utilized, which resulted in particular for the Zn content in MSWI residues as well as for the transfer coefficients of Zn in different figures. The deviations observed between the different sources have been accounted for by using uncertainty ranges for the respective parameters in the frame of the subsequent analyses.

Based on the results of the literature survey a material flow (MFA) model describing the flows of Zn through European waste incinerators has been established.

2.2. Economic Evaluation of Zn flows

The MFA model together with detailed information about the recovery technology and its consumables forms the basis for the economic evaluation of Zn recovery. As to the knowledge of the authors the only technology for recovering Zn from MSWI residues operating at large scale is the FLUREC process. Therefore, this technology has been assumed for the economic evaluation of Zn resources present in MSWI residues.

Figure 2 gives an overview of the FLUREC technology and summarizes the required operating supplies. Detailed information about the specific quantities of the latter together with data about products and by-products are of major importance for the economic evaluation. Boesch et al. (2014) who performed a LCA on waste incineration enhanced with new technologies for metal recovery, provide detailed information about materials and energy flows associated with the recovery of Zn out of MSWI fly ash. This data about energy and material flows were subsequently linked with market prices for the different operating supplies p_{OP_i} (including electricity) and for the final product, which is Zinc metal of purity > 99.9%, p_{Zn}, as well as specific costs c_{DP_i} for landfilling residues generated and necessary investment costs of the technology C_{INV} (see Equation 1). The overall costs of the FLUREC technology C_{Flurec} were then compared to the costs of the prevailing management practice of MSWI fly ash in Europe C_{CP} (Equation 2), which includes cement stabilization with subsequent disposal at non-hazardous waste landfills or direct landfilling at hazardous waste sites. Merging Equation 1 and 2 by assuming that the overall costs for the FLUREC technology should be not lager than costs for the current practice of fly ash disposal, specific production costs for secondary Zn c_{Zn} [€/kg Zinc] can be determined (see Equation 3). In case that specific production costs c_{Zn} are lower than the market price p_{Zn} for metallic Zn, Zn recycling (using the FLUREC technology) is economically viable and vice versa.

In order to account for the fact that all data required for the economic evaluation (physical mass flows, prices, costs for disposal of residues or investment costs) are uncertain, plausible data ranges were defined and subsequently used to perform Monte Carlo simulations. Thereto the software @risk was utilized. For the definition of the uncertainties ranges temporal (i.e. over the last 5 years) and spatial variations in market prices and costs for disposal have been evaluated. The uncertainty of specific materials flows and energy consumption of the FLUREC technology has been estimated based on information provided by Schlumberger (2006) and Boesch et al. (2014).

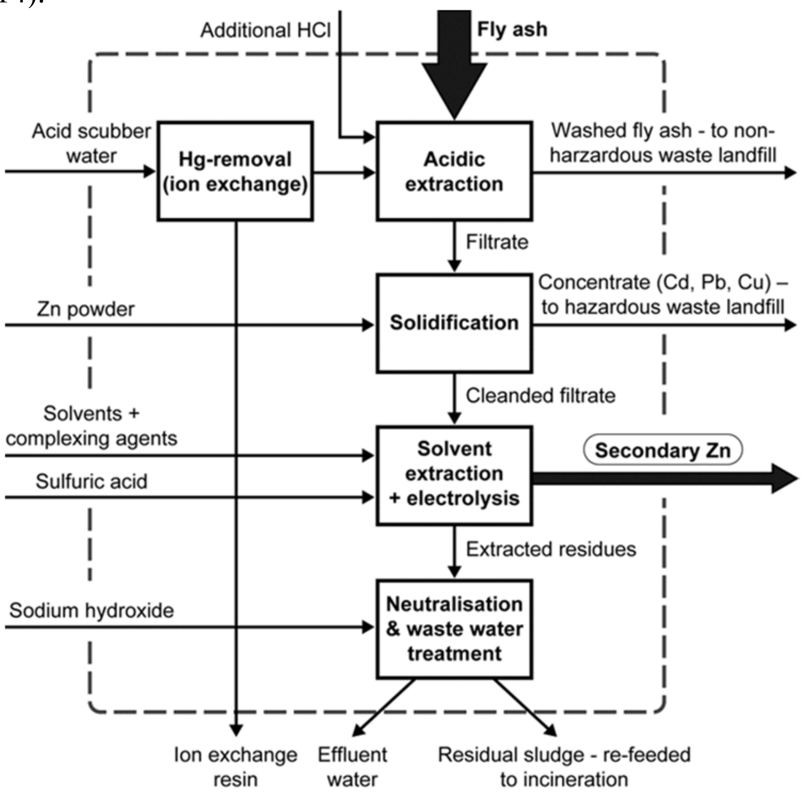

Figure 2 Schematic process diagram of the FLUREC technology (acidic fly ash leaching with integrated zinc recovery)

$$\sum_{i=1}^{n} m_{OP_i} \cdot p_{OP_i} + \sum_{j=1}^{m} m_{DP_j} \cdot c_{DP_j} - m_{Zn} \cdot p_{Zn} + C_{INV} = C_{FLUREC} \qquad \textbf{Equation 1}$$

$$\sum_{j=1}^{m} m_{DP_j} \cdot c_{DP_j} = C_{CP} \qquad \textbf{Equation 2}$$

$$c_{Zn} = \frac{\sum_{i=1}^{n} m_{OP_i} \cdot p_{OP_i} + \sum_{j=1}^{m} m_{DP_j} \cdot c_{DP_j} + C_{FLUREC} - C_{CP}}{m_{Zn}} \qquad \textbf{Equation 3}$$

$m_{OP_i} \cdot$	specific mass of operating supply i [kg/t fly ash] and specific energy demand [kWh/t fly ash]
m_{DP_i}	specific mass of residue j to be disposed of [kg/t fly ash]
m_{Zn}	specific mass of metallic Zn recovered [kg Zn/t fly ash]
p_{OP_i}	market price for operating supply i and energy demand [€/kg] or [€/kWh]
p_{Zn}	market price for metallic Zn [€/kg]
c_{Zn}	production costs for metallic Zn [€/kg]
c_{DP_j}	specific costs for the disposal of residue j [€/kg]
c_{INV}	specific investment costs for the FLUREC technology [€/t fly ash]
c_{FLUREC}	specific overall costs for the FLUREC technology [€/t fly ash]
c_{CP}	specific overall costs for the current disposal management of fly ashes [€/t fly ash]

2.3. Classification of Zn flows

The classification of Zn present in MSWI residues has been accomplished in accordance with Lederer et al. (2014) who based their evaluation framework for anthropogenic resource on McKelvey (1972). The approach considers both, the economic viability of extracting a secondary raw material from a resource and producing a tradable good, and the knowledge of the existence of the resource. For the economic classification, McKelvey suggests the following terms. Resources are *economic* or *recoverable* if they can be extracted with a profit. Therefore, the production costs must be below the market price of the product achievable, which means in our case $c_{Zn} < p_{Zn}$. Resources for which the production costs are higher than the price, but not by more than a factor of 1.5, are *marginally economic*. Resources above this value are termed as *submarginal* or *subeconomic,* whereby according to Lederer et al. (2014) a threshold factor of 10 times the market price is assumed ($p_{Zn} < c_{Zn} < 10 \times p_{Zn}$). Resource flows whose production costs are above the threshold (of 10 times the market price) are counted as *other occurrences*.
The classification according to the certainty of the existence of a resource flow is structured as *identified – demonstrated*, *identified – inferred*, and *potentially undiscovered*. To perform this classification, the uncertainties determined for each Zn flow in the residues of MSWI are used. *Identified – demonstrated* resources are of proven existence and knowledge is highly certain (confidence that the actual flow of Zn is at least this size is 90%). *Identified – inferred resources* are defined here as the amount of Zn flows between the lower uncertainty bound (confidence 90%) and the mean value of the flow). The same amount of the material (due to symmetric uncertainty ranges) is designated as *potentially undiscovered* resources, which may exist but are highly uncertain. Finally, a cross-classification is accomplished considering both, economic viability and knowledge. Therein, reserves are resources that are both identified – demonstrated and economically extractable. The reserve base further includes the part that is identified – demonstrated and not profitably extractable with current technology and market conditions (classified as marginally economic).

3. RESULTS

3.1. Exploration of Zn flows in MSWI residues

According to CEWEP (2011) about 78 million tons of waste have been utilized in European (EU-28 + Norway and Switzerland) WtE plants in 2011. This equals to about 90% of the total waste incineration capacity (about 86 million tons) installed. Out of the 78 million tons 4.2 million tons are combusted in fluidized bed incinerators (FBI), the remaining part is utilized in rotary furnaces or grate incinerators (GI). According to information (data of 350 plants out of 470 plants in total have been available) provided by ISWA (2006, 2013), about 45% of the incineration plants are equipped with wet flue gas cleaning systems, 29% with semidry and 26% with dry systems. The discrimination of the incineration technology (grate vs. fluidized bed) and APC system (wet-semidry-dry) is of significant importance for the exploration of Zn flows, since the technologies do not only strongly influence the size and grade (with respect to Zn content) of fly ashes (see Table 1), but also the need of operating supplies (e.g. consumption of HCl) as well as costs for the current disposal practice. Fly ashes from FBI, for instance, are likely to be disposed of at non-hazardous waste landfills (due to their comparatively low contents of heavy metals and salts), whereas fly ashes from grate incinerators (GI) are definitely classified as hazardous and have therefore to be landfilled at hazardous waste sites. In Table 1 main outcomes of exploring MSWI residues as potential resource for Zn recovery are summarized. Based on the analysis of data from more than 50 WtE plants, it becomes obvious that the APC control but also the type of fly ash (filter ash vs. boiler ash) strongly influences the Zn content of the fly ash. Whereas for dry or semidry APC systems average Zn contents of fly ashes amount to about 11,000 mg Zn/kg, wet systems may generate residues with Zn contents of about 20,000 mg Zn/kg fly ash (in case boiler and filter ash are collected together) or even above 40,000 mg Zn/kg fly ash (in case that filter ash is withdrawn separately). In comparison to the contents given in Table 1 Zn contents of bottom ashes form waste incineration are almost 1 order or magnitude smaller (1,800 to 6,000 mg Zn/kg bottom ash - Hjelmar, 1996).

Table 1 Statistical analysis of fly ash amounts (kg/t waste) generated at Waste-to-Energy (WtE) plants with different flue gas cleaning systems and their respective Zn contents (mg Zn/kg fly ash)

	Amount of fly ash [kg/t waste input]			**Zn-content in flue gas cleaning residues** [mg Zn/kg fly ash]			
	Flue gas cleaning system			Boiler and filter ash of			filter ash of wet systems*
	Wet	semidry	dry	wet systems	semidry systems	dry systems	
no. of WtE plants	*27*	*14*	*9*	*22*	*15*	*9*	*14*
no. of countries	*10*	*8*	*5*	*9*	*7*	*5*	*5*
mean	24	40	38	**21,600**	**11,300**	**12,000**	**41,300**
median	25	39	39	17,300	10,300	10,800	42,900
10% quantile	16	33	27	12,400	7,300	7,600	20,200
90% quantile	30	49	48	35,800	15,500	18,500	60,300

* plants with separate collection of filter ash and boiler ash

Based on transfer coefficients describing the portioning of Zn during waste incineration (between 50% and 60% of Zn is transferred to the fly ash) and the data given in Table 1 the average content of Zn in the waste feed of European WtE plants have been determined to about 930±150 mg Zn/kg waste. Considering this content and the overall mass of waste combusted the following material flow analysis diagram has been derived (see Figure 3). In total about 72±12 kt of Zn are annually fed into European waste incineration plants. Almost half of it (34.5±3.5 kt) accumulates in MSWI residues (bottom ashes and fly ashes of fluidized bed incinerators) at average concentrations below 5,000 mg Zn/kg ash. About 18±3.5 kt of Zn are present in boiler and filter ash of grate incinerators equipped with wet APC systems (average Zn content about 21,000 mg Zn/kg ash) and almost the same amount (18.8±2.4 kt) can be found in fly ashes from dry and semidry APC systems (average Zn content of 11,000 mg Zn/kg ash).

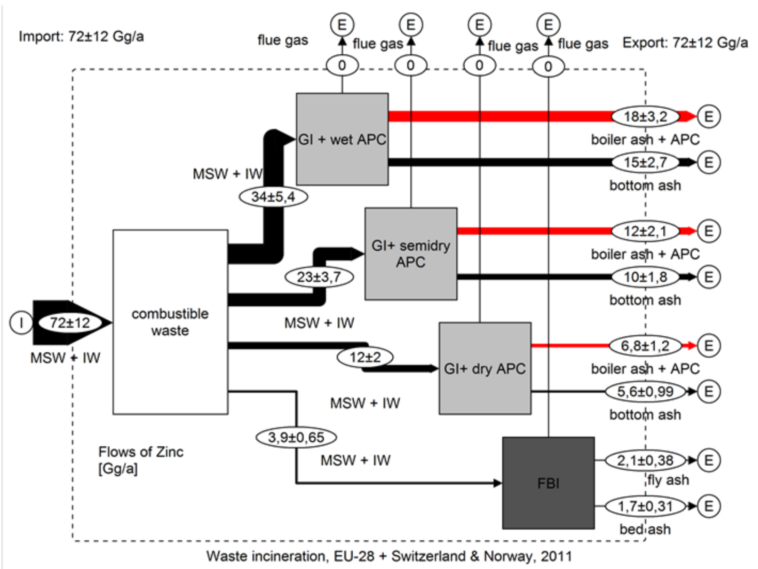

Figure 3 Annual Zn flows (in 1,000 t) through European WtE plants (uncertainties represent the 10% and 90% confidence interval, respectively) – red flows indicate flows of MSWI residues with Zn contents above 8,000 mg Zn/kg

3.2. Economic evaluation and classification of Zn flows

Based on the material and energy demand of the FLUREC technology and the potential recovery rates for Zn (provided by Boesch et al., 2014) the different MSWI residues have been evaluated regarding their specific costs for Zn recovery. In Table 2 all assumptions made for the economic evaluation of Zn recovery from filter ashes of wet APC systems are summarized. The results indicate that despite the comparatively high Zn contents (around 41,000 mg Zn/kg ash) of those ashes, the specific production costs for Zn are about 2.4±1.2 €/kg Zn and thus, above the current market price of 1.6 €/kg Zn. For the other fly and bottom ashes of European waste incinerators, specific production or recovery costs of Zn are even much higher (see Figure 4).

Table 2 Economic evaluation of Zn recovery from MSWI residues (using the example of filter ash from wet APC systems) applying the FLUREC technology

Means of production + outputs		Materials and energy (per 1 t of fly ash)				Specific costs (positive) & spec. benefits-savings (negative)					Total costs/savings
		unit	mean	σ		*unit*	mean	σ	source		mean
"inputs"	fly ash disposal (current practice)	*kg*	1000			€/kg	-0.2	0.02	*1*		-€ 200.0
	zinc content of fly ash	***kg***	**41**	1							
	HCl (30%) of wet scrubber	*kg*	550	100		€/kg	0				-
	HCl (30%) - additional	*kg*	40	15		€/kg	0.11	0.015	*2*		€ 4.4
	H2SO4	*kg*	15	1.5		€/kg	0.16	0.02	*2*		€ 2.2
	NaOH (50%)	*kg*	125	12.5		€/kg	0.11	0.015	*3*		€ 13.9
	solvents & complexing agents	*kg*	0.4	0.08		€/kg	0.4	0.1	*3*		€ 0.2
	quicklime	*kg*	-200	20		€/kg	0.08	0.01	*2*		-€ 16.0
	electricity	*kWh*	351	18		€/kWh	0.094	0.005	*4*		€ 33.0
	Total investment costs (per 1000 kg fly ash)					€	180	20	*8*		€ 180.0
"outputs"	leached fly ash (non-hazardous waste landfill)	*kg*	800	30		€/kg	0.045	0.005	*5*		€ 36.0
	concentrate (hazardous waste landfill)	*kg*	12	2		€/kg	0.2	0.02	*1*		€ 2.4
	depleted resin material	*kg*	1	0.1		€/kg	18.4	2.8	*6*		€ 18.4
	residual sludge (re-fed to incinerator)	*kg*	24	5		€/kg	0	0			-
	Recovery rate of Zn	-	*0.75*	*0.025*							
					necessary revenues from Zn production [€/t fly ash]						€ 74.7
	secondary Zinc production	*kg*	**30.8**	**3.0**	**specific production costs for Zn [€/kg Zn]**						**€ 2.4**

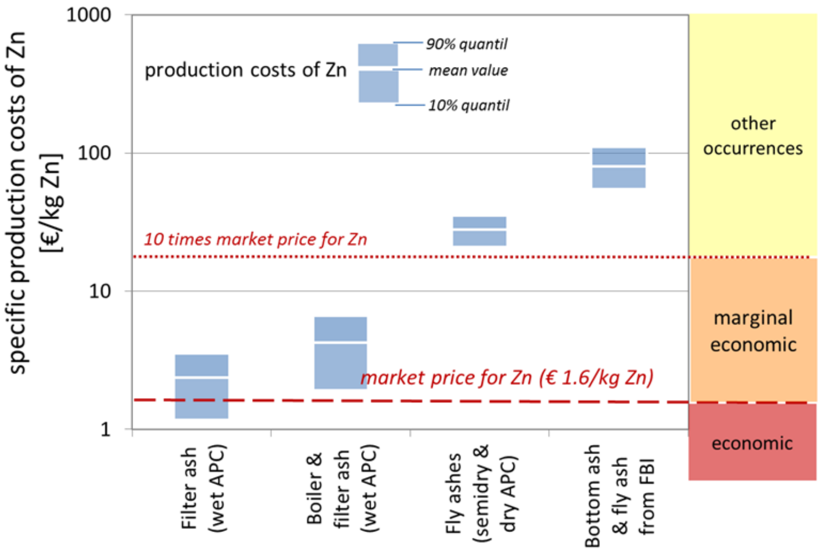

Figure 4 Specific production costs for Zinc (given in €/kg Zn) for different MSWI residues

Combining information about the size of Zn flows (incl. their uncertainties) and the specific recovery costs allows classifying Zn flows in accordance to the commonly applied classification scheme for mineral resource (Mc Kelvey, 1972). The results of this classification (see Table 3) demonstrate that total size of the identified Zn flows in European MSWI residues is 72,000 t/a. Based on the current market prices for Zn (1.6 €/kg), none of the identified resources can be economically extracted and thus cannot be classified as a reserve. The reserve base containing marginally economic and identified – demonstrated resources amounts to 5,900 t (separately collected filter ash from wet APC systems). A total of 8,900 t of Zn is classified as subeconomic, with production costs approximately 3-5 times above the current market price of Zn (boiler ash and jointly collected boiler and filter ash from wet APC systems). The residual bulk of Zn is either low-grade fly ash from dry or semidry APC systems (18,800 t of Zn), from bottom ash (22,300 t) or from fly ash generated at fluidized bed incinerators (2,100 t). Regarding certainty, 83% of the identified resources are demonstrated, and 17% are inferred stocks.

Table 3 McKelvey diagram for annual Zn flows (in t/a) in European MSWI residues (the uncertainty ranges of the estimates form the basis for the distinction between *demonstrated*, *inferred*, and *potentially undiscovered resources*).

	identified resources		potentially undiscovered resources
	demonstrated	inferred	
economic	0#	0#	0#
marginally economic	5,900*	1,300*	1,300*
subeconomic	8,900*	1,900*	1,900*
other occurrences (low grade)	45,200	8,800	8,800
total	72,000		12,000

\# *an economically viable recovery of Zn from fly ashes would (at current market prices) only be possible at Zn contents above 70,000 mg/kg ash*

* *assuming that at 50% of all WtE plants with wet APC systems filter ashes can be separately collected from the boiler ash*

In comparison to total European Zn imports, which amount to about 1.3×10^6 t (Spatari et al., 2003), Zn recovery from *marginally economic* and *subeconomic* MSWI residues could at maximum substitute 1.1% of European imports.

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