INTEGRATING ZN RECOVERY FROM FLY ASHES INTO THE VIENNESE WASTE INCINERATION CLUSTER

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ABSTRACT
In the city of Vienna approximately 1 Million ton of waste is incinerated annually. Therefore 13 lines of waste incineration including fluidized bed incinerators, rotary kilns and grate furnaces are in operation. Beside municipal solid waste, hazardous waste as well as communal sewage sludge is utilized. All incinerator lines use a wet flue gas cleaning system. Based on the determination of the zinc, lead and copper concentrations in the different solid residues and on information about the mass flow of the residues a detailed material flow analysis is carried out, that determines the total flow of these metals through the Viennese waste incineration cluster. These data was subsequently used to evaluate whether a recently developed recycling technology (FLUREC), that allows recovering zinc, lead, copper and cadmium out of fly ash, is evaluated with respect to its application to fly ashes generated at Viennese waste incineration plants. In particular a comparison between the operational costs and the expected revenues from the recovered metals as well reduced disposal costs is discussed within the present work.

ABBREVIATIONS

$m_{i,j}$	mass flow of residue j at site i
$c_{i,j}$	concentration of residue j at site i
s	slag
w	waste
fc	filter cake (dry)
sm	scrap metal
bfa	boiler fly ash
ck	coke
ffa	filter fly ash
fa	fly ash
a	year
t	ton
ls	liquid to solid
rk	rotary kiln
fbc	fluidized bed combustor
MSW	municipal solid waste
fbc	fluidized bed combustor

INTRODUCTION
For a great number of different wastes, such as glass, metal or paper, recycling strategies have been developed. Nevertheless millions of tons of municipal solid waste, various hazardous wastes and communal sewage sludge have to be incinerated each year. In Vienna approximately

one million tons of wastes are incinerated each year at four sites of waste incineration [Purgar 2013]. In Austria and many other countries of the European Union residues of the thermal treatment of waste are considered as hazardous and therefore have to be disposed on landfills for hazardous waste or have to be treated prior to landfilling on a non-hazardous disposal site [Astrup, 2008; DVO, 2008; AVO, 2003]. Whereas slag often can be declared as non-hazardous without any treatment filter cake is disposed of at hazardous waste landfills without considering an alternative due to the low mass flux. Contrary to this, fly ashes with a generation rate of approximately 1,6 to 8,7 % of the incinerated waste, depending on the waste input and combustion technology, represents a serious challenge with respect to a sustainable and legally conform disposal. So far different methods for the treatment of fly ashes have been developed. Some of them are given in the subsequent section.

Wet chemical fly ash treatment methods, like the MR or 3R-process [IAWG, 1997], have been developed and evaluated for decades. The basic concept of these methods is to extract potential hazardous heavy metals and soluble salts from the fly ash with the scrubber water of the first stage of the wet flue gas cleaning system. After this extraction process, typically realized in a stirring reactor, the liquid and the solid phase are separated using a vacuum band filter. The main aim of these processes is typically to fulfill threshold parameter for landfilling. As the product quality after of the washing process is often not sufficient a subsequent thermal treatment is necessary.

Within this work the product quality is neglected and the main focus is given to the filtrate which is meant to contain the extracted potentially valuable metals. In Switzerland recently an industrial - scale facility, based on a wet fly ash decontamination process, which is recovering Zn, Cu, Pb, and Cd from this filtrate went into operation, the so called FLUREC process [Schlumberger, 2013]. It is stated that out of approximately 4 million tons of waste incinerated in Switzerland 4.000 tons of Zinc can be recovered theoretically by implementing the technology used to all Swiss incinerators. The main aim of this work is to investigate Vienna's MSW fly ashes on their zinc content and for their annual zinc load to carry out a preliminary economic evaluation, whether the technology described is applicable for Vienna's fly ashes or not. Moreover, based on a material flow analysis individually over the four sites of waste incineration, regarding to Zn, Cd, Pb and Cu it is investigated whether the incineration technology is the reason for different Zn contents in Vienna's fly ash or simply the metal load into Vienna's waste incineration plants is different to Swiss incinerators.

METHODS

Over a period of four years (2009 to 2012) the solid residues (slag, fly ash, filter cake) of four sites of waste incineration have been analyzed for their Zn, Pb, Cd and Cu content. Almost every day a sample has been taken and merged to a mixed sample which was analyzed every two months. The mixed slag samples have been analyzed every month. The measurements have been accomplished by the plant operator in accordance to mandatory measurements named in [AWG, 2002] due to the basic characterization of wastes. The total contents have been determined by digesting the ash samples in aqua regia (concentrated nitric acid and hydrochloric acid). After digestion the sample has been analyzed using ICP-OES (inductive coupled plasma - optical emission spectrometry). The metal load in the emissions to the hydrosphere and the atmosphere of the investigated plants are given in [BMLFUW, 2009]. It can be observed that these metal fluxes per ton incinerated waste are far beyond 0.1 g and therefore not taken into account, though the metal concentrations in the msw can be slightly higher.

A detailed description of the investigated facilities is given, based on information given by [Böhmer, 2006; Stubenvoll, 2002; WEF, 2012; WEF, 2013] as well as information provided by the plant operators.

Site 1 has two lines of grate furnaces in operation. In the years 2009 to 2012 $m_{1,w}$*=230,000 t/a* of waste have been incinerated whereby the input fuel consists of Viennese MSW and MSW-similar commercial wastes. Besides flue gas thereby $m_{1,s}$=48,000 t of slag, $m_{1,fa}$ =8,100 t of fly ashes, and 268 t of filter cake have been produced annually. The heat transfer system consists of empty flues, followed by boiler flues, and economizer flues in which coarse particles can be collected and subtracted. This coarse particles are defined as boiler fly ash (bfa) within this work ($m_{1,bfa}$*=4,200 t/a)*. After the heat transfer zone an electrostatic precipitator (esp) is used to collect fine particles labeled as filter fly ash – $m_{1,ffa}$=3,800 t/a. After the ESP a two stage wet flue gas cleaning system is installed, which is followed by an activated carbon fixed bed adsorber. The used coke, $m_{1,ck}$=310 t/a, is incinerated at site 4 and is not part of further investigations. Before the flue gas stream is released through the chimney a selective catalytic reduction (scr) system is installed. Grate siftings and slag are collected together and are removed through a water bath. Several tons of scrap metal have been recovered on site but have not been investigated further. The scrubber water is purified in the waste water station of the plant whereby 268 t/a of filter cake with a moisture of approximately 40 percent is produced ($m_{1,fc}$=160 t/a).

At site 2 two lines of grate furnaces with a thermal capacity of 82 MW are in operation. In the years 2009 to 2011 $m_{2,w}$=203,000 tons of mainly Viennese municipal solid waste have been incinerated on average annually. Besides flue gas thereby $m_{2,s}$=44,000 t/a of slag (including grate siftings), and $m_{2,fa}$=3,300 t/a of fly ashes has been produced. Additionally 280 t/a of filter cake with a water content of approximately 43 percent is produced at the water purification station ($m_{2,fc}$=160 t/a). The incineration lines at site 2 are similar to site 1 except for the absence of the fixed bed carbon adsober and boiler ash and filter ash are not collected and subtracted separately and therefore labeled as fly ash (fa) in the following.

At site three 3 lines of grate furnaces with together 57 MW thermal capacity are in operation. In the years 2009 to 2011 $m_{3,w}$=188,000 t/a of mainly Viennese municipal solid waste has been incinerated on average. Besides flue gas thereby $m_{3,s}$=47,500 t/a of slags (including grate siftings), $m_{1,fa}$=3,700 t/a of fly ashes and 95 t/a of filter cake (43% moisture - $m_{1,fc}$=54.15 t/a) has been produced. A mark able difference to site 1 is that between the heat transfer zone and the bag house filter about 24 to 240 t/a crushed coke is injected into the flue gas stream, which adsorbs pollutants and is subtracted in the bag house filter together with the fly ash. Therefore the fixed bed carbon adsorber is waived. At site one Annually 84,000 m^3 of scrubber water with a pH value of 1 is subtracted from the first acidic scrubber stage.

At site 4 six lines of incineration are in operation, two rotary kilns (rk) and four fluidized bed combustors (fbc). These 6 lines are operated separately, except for sharing a collective scr system and a collective waste water treatment facility. In the years 2009 to 2012 by purifying the scrubber water of these six incineration lines 2,288 t/a of filter cake emerged with a water content of approximately 40 percent. In the investigated period 2009 to 2012 $m_{4,w}$=316,000 tons of wastes, a mixture of hazardous waste, refuse derived fuel (rdf) and communal sewage sludge (css), have been incinerated annually. According to the plant operator 144,000 m^3 of scrubber water with a pH Value of approximately 1 could potentially be used for a fly ash treatment process.

The two lines of rotary kilns (i=4a) at site four are mainly utilizing hazardous waste and have a common capacity of 100,000 t/a and a maximum thermal capacity of 62 MW. In the year 2012 $m_{4a,fa}$=2,500 t of fly ash were produced and about $m_{4a,s}$=16,000 t of slag. Except for the fact that filter ash and boiler ash cannot be subtracted separately the flue gas cleaning system is similar to site 1. At site 4 three fluidized bed combustors (i=4b) with a closed gas distributor bottom are in operation, that are utilizing almost exclusively communal sewage sludge. Together they have a thermal capacity of 60 MW. In the year 2005 and 2006 $m_{4b,w}$=169,000 t/a of sewage sludge has been treated whereby $m_{4b,fa}$=16200 t/a of fly ash emerged. Due to the plant operators these conditions have not changed crucial to the focused time period 2009 to 2012. The flue gas cleaning system is similar to the rotary kilns described above. At site 4 another fluidized bed combustor (i=4c) is in operation that is utilizing almost exclusively refuse derived fuel with a capacity of 110,000 t/a and a thermal capacity of 60 MW. In the years 2011 and 2012 $m_{4c,fa}$=11,200 t/a of fly ash and $m_{4c,s}$=10,100 t/a of slag (bottom ash) have been a result .The flue gas path is similar to the other lines operated at site 4.

With information on the metal concentrations and mass flow of the investigated residue stream the load within the combusted waste is determined and compared to other European incineration plants.

In [Fellner, 2014] an economical evaluation of the FLUREC process is described and benchmarks for the benefits and costs for the operation of the process are given. Additionally to this evaluation parameters have been determined in detail for two scenarios described below and an economical feasibility study was carried out.

Scenario one (SC1): The Fly ash from site 3 is treated at site 3 with the available amount of scrubber water. Different to the calculation of [Fellner, 2014] the parameters, “Zn content of fly ash”, “HCl of wet scrubber”, “HCl – additional” as well as the “savings for quicklime” have been determined in detail for this scenario.

Scenario two (SC2): Filter fly ash of site 1 and fly ash from site 2 and 4a is treated at site 4 with the available amount of scrubber water of site 4. Divergent to the calculation of [Fellner, 2014] the parameters, “Zn content of fly ash”, “HCl of wet scrubber”, “HCl – additional” as well as the “savings for quicklime” have been determined in detail for this scenario.

The parameter “zinc content of fly ash” is determined as described above. The parameter “HCl – additional” is determined with single samples collected at the incineration lines. 10 grams of the fly ash samples were mixed with deionized water in a liquid to solid ratio of 10 and subsequent concentrated hydrochloric acid, in 1 ml steps, was added as long as the mixture had a pH value of 4. The amount of HCl determined for the neutralization minus the available amount of HCl provided by the scrubbers is the amount of additional HCl needed within the process. The amount of HCl of the wet scrubber water available annually (“HCl of wet scrubber”) was determined by the plant operators. Savings for quicklime have been determined under the following assumption: All the quicklime used at the plant is needed for the neutralization of the scrubber water and can be saved by the implementation of the FLUREC process.

RESULTS

Table 1 shows the total content of the metals Zn, Pb, Cu and Cd in the investigated residue streams. With equation 1 the concentrations $c_{w,i}$ in the waste input of the investigated sites is determined and can be seen in Table 2. Also in Table 2 concentrations of the investigated metals of Austrian and Swiss wastes determined by [Morf, 2005 and Morf, 2006] are summarized.

$$c_{w,i} \cdot \mathrm{m_{w,i}} = \sum_{j=1}^{n} \mathrm{c_{j,i}} \cdot \mathrm{m_{j,i}} \quad (1)$$

Tab. 1: Zinc (Zn), Lead (Pb), Copper (Cu) and Cadmium (Cd) concentrations ($c_{j,i}$) in the investigated residue streams ($m_{j,i}$) in kg/kg; mn, mean value; sd, standard deviation; n number of analyzed samples

		Zn		Pb		Cu		Cd		
site(i)	residue(j)	m	sd	m	sd	m	sd	m	sd	n
3	fly ash	1,84e-02	2,64e-03	4,39e-03	8,63e-04	1,00e-03	1,56e-04	3,21e-04	5,53e-05	24
3	slag	2,31e-03	6,27e-04	9,53e-04	4,85e-04	2,35e-03	1,83e-03	5,95e-06	3,14e-06	48
3	filter cake	2,90e-04	1,51e-04	9,85e-05	4,04e-05	1,32e-04	1,30e-04	9,77e-06	2,16e-06	24
1	filter ash	2,16e-02	4,97e-03	7,02e-03	2,14e-03	1,90e-03	8,39e-04	3,75e-04	7,21e-05	24
1	boiler ash	5,68e-03	1,41e-03	1,10e-03	5,28e-04	1,01e-03	3,77e-04	3,80e-05	1,27e-05	24
1	slag	2,62e-03	6,51e-04	1,21e-03	8,66e-04	3,61e-03	2,10e-03	6,12e-06	7,40e-06	48
1	filter cake	7,54e-04	4,48e-04	3,63e-04	3,15e-04	1,13e-04	1,07e-04	1,87e-05	1,22e-05	24
2	filter cake	3,07e-03	2,26e-03	9,49e-04	6,82e-04	1,98e-04	1,40e-04	6,66e-05	5,01e-05	23
2	fly ash	2,07e-02	6,42e-03	4,88e-03	1,33e-03	1,01e-03	1,95e-04	3,47e-04	8,48e-05	23
2	slag	2,28e-03	9,30e-04	8,23e-04	4,78e-04	1,96e-03	1,01e-03	6,52e-06	6,56e-06	46
4	filter cake	1,63e-03	2,02e-03	2,22e-04	1,22e-04	2,20e-04	9,45e-05	5,60e-05	1,53e-04	24
4a	slag	2,94e-03	9,80e-04	6,97e-04	3,89e-04	3,24e-03	7,59e-04	3,68e-06	2,81e-06	48
4a	fly ash	3,81e-02	8,11e-03	1,29e-02	4,33e-03	3,32e-03	8,53e-04	3,21e-04	2,32e-04	24
4b	fly ash	3,75e-03	6,28e-04	6,77e-04	2,44e-04	2,93e-03	9,64e-04	1,49e-05	4,21e-06	48
4c	fly ash	3,87e-03	7,41e-04	1,25e-03	4,29e-04	3,61e-03	8,36e-04	1,38e-05	5,37e-06	26
4c	bottom a.	4,68e-04	4,01e-04	5,10e-04	6,90e-04	7,90e-04	3,91e-04	1,15e-06	3,29e-07	12

Tab. 2: Mean concentrations of metals Zinc, Lead, Copper and Cadmium ($c_{w,i}$) in the input stream ($m_{w,i}$) of the investigated sites in kg/kg; msw, municipal solid waste; iw, industrial and commercial wastes.

source:	Zn	Pb	Cu	Cd
Site 1	1,00e-3	3,87e-4	11,6e-4	8,15e-6
Site 2	0,83e-3	2,57e-4	4,4e-4	7,06e-6
Site 3	0,94e-3	3,26e-4	6,1e-4	7,76e-6
Site 4	0,78e-3	2,26e-4	4,8e-4	4,15e-6
[morf 2006] msw	1,1 e-3 ±1 e-4	4,00 e-4 ±4,3 e-5	9,1e-4 ±1 e-4	7,8 e-6 ±6,3 e-7
[morf 2006] iw	3,10 e-3 ±2,8 e-4	8,80 e-4 ±8,3 e-5	9,00e-4 ±1,8 e-4	19 e-6 ±17 e-7
[morf 2005] msw	0,56 e-3 ± 0,25 e-4	2,9 e-4 ± 2,4 e-5	3,0 e-4 ± 0,024 e-3	5,8 e-6 ± 3,5 e-7

In Table3 benchmarks for the material and energy demand of the FLUREC Process per ton of fly ash are given. Also in table 3 specific costs and benefits are given. Scenario 0 (SC0) equals the economical evaluation of [Fellner, 2014]. SC1 and SC2 are adapted calculations due to the scenarios 1 and 2 described above.

Discussion

Table 1 shows that the concentrations of zinc in the different fly ashes. Neglecting fly ashes from fluidized bed combustors it is seen that filter fly ashes as well as fly ashes in total show a zinc concentration of 1.5 to 4.6 percent which is not unusual for fly ashes of this type [Karlfeldt, 2010; Fellner, 2014]. Together with the mass flow and the zinc concentrations of the other solid residues the zinc concentration in the input of the incinerators is determined. Zinc concentrations determined range from 0.5 to 1.3 g per kg waste incinerated which is not unusual as long as no industrial or commercial waste is incinerated, [Morf, 2005 and Morf, 2006].

Tab. 3: Material and energy demands and specific costs for the implementation of the FLUREC process according to [Fellner, 2014] (SC0), scenario 1 (SC1) and scenario 2 (SC2)

		Materials and Energy			Specific costs (positive) & spec. benefits-savings (negative)		Costs and Revenues		
	unit	SC0	SC1	SC2	unit		SC 0	SC 1	SC 2
fly ash disposal (current practice)	kg	1000	3.7 e6	13.3 e6	€/kg	-0.2	-200	-740 e3	-2658 e3
zinc content of fly ash	kg	41	6.8 e4	3.1 e5	€/kg	-1.6	-65.6	-109 e3	-501 e3
HCl (30%) of wet scrubber	kg	550	1.0 e6	1.75 e6	€/kg	0	0	0	0
HCl (30%) - additional	kg	40	1.3 e6	5.93 e6	€/kg	0.11	4.4	104 e3	652 e3
H2SO4	kg	15	5.6 e4	2.0 e5	€/kg	0.16	2.4	8.9 e3	32 e3
NaOH (50%)	kg	125	4.6 e5	1.66 e6	€/kg	0.11	13.8	501 e3	183 e3
solvents & complexing	kg	0.4	1.5 e3	5.3 e3	€/kg	0.4	0.16	0.6 e3	2.1 e3
quicklime	kg	-200	-2.4 e5	-4.0 e5	€/kg	0.08	-16	-19 e3	-32 e3
electricity	kWh	351	1.3 e6	4.67 e6	€/kWh	0.094	33	122 e3	439 e3
Total investment costs (per 1000 kg fly ash)		1	3.7 e3	1.3 e4	€	180	180	666 e3	2393 e3
leached fly ash (non-hazardous waste landfill)	kg	800	3.0 e6	10.6 e6	€/kg	0.045	36	133 e3	479 e3
concentrate (hazardous waste landfill)	kg	12	4.4 e4	1.6 e5	€/kg	0.2	2.4	9 e3	32 e3
depleted resin material	kg	1	3.7 e3	1.3 e4	€/kg	18.4	18.4	68 e3	245 e3
residual sludge (re-fed to incinerator)	kg	24	8.8 e4	3.2 e5	€/kg	0	0	0	0
Total annual operation costs	€						8.9	331 e3	1263 e3

In total 800 (± 200) tons of Zinc, 300 (±100) tons of Lead, and 550 (±250) tons of Copper pass through Vienna's waste incineration cluster annually. Thereof 450 (± 100) tons of Zinc 120 (±40) tons of Lead and 115 (±33) tons of Copper are found in fly ashes. For Zn an overall transfer coefficient, from waste incinerated into the fly ash, of 0.56 is determined which fits to results given in [Brunner, 1986].

Neglecting fly ashes from fluidized bed combustors and fly ashes subtracted in the heat transfer zone (boiler ash), because of their low zinc concentrations, 300 (± 70) tons of Zink 90 (±30) tons of lead and 22 (±6) tons of copper can be recovered implementing the FLUREC process according to scenario 2 assuming a recovery rate of 100 percent which can theoretically be achieved according to [Karlfeldt, 2010] as long as the pH value in the extraction vessel is very low. By implementing the FLUREC process to a single incineration site with a capacity of 200 kilotons of waste 68 tons of zinc can be recovered annually.

Lead and copper are recovered as mixture polluted with cadmium and therefore it is assumed that no revenues are generated out of this fraction. With a current market price of 1.6 €/kg for zinc at best 0.6 million € can be gained by selling zinc at scenario 2. In contrast 0.6 million Euros are indispensable for additional hydrochloric acid to reach a sufficient extraction rate for zinc and another 0.4 Million € are necessary to cover the costs for electricity. Neglecting the investment costs which is the most uncertain parameter in this evaluation 1.1 million € can be gained in total annually. Comparing the savings due to the alternative disposal on a non hazardous waste landfill and revenues due to the sale of zinc it is seen that 80 % of the revenues result from the alternative disposal. Savings due to a reduced or absent demand for quicklime in the waste water treatment facility is less than 2 % of the total revenues.

At scenario 1 only 109,000 Euros can be gained by selling zinc. Almost the same amount has to be spent for additional hydrochloric acid and electricity. Comparing the revenues resulting from an alternative disposal (85 %), savings because of reduced quick lime demands (2%) and the sale of zinc (13%) shows a similar situation to scenario 2.

CONCLUSION

Due to the material flow analysis carried out the Zinc flows through Vienna's waste incineration cluster has been investigated precisely. The determined Zinc concentration in the waste input of the investigated incinerator lines is not crucial different to others of its type in Europe. The transfer coefficient into the fly ash for zinc is determined to be approximately 50 % which is also not uncommon.

By implementing the FLUREC process to the waste incineration cluster of Vienna 450 (± 100) tons of Zinc 120 (±40) tons of Lead and 115 (±33) of Copper can potentially be recovered, whereby only from the selling of Zinc revenues can be generated. By carrying out two case studies (scenario 1 and 2) it is seen that the major intention of implementing the FLUREC process is neither the expected revenue due to the selling of zinc, nor savings of quicklime. The major driver for the implementing of the FLUREC process to the waste incineration cluster of Vienna is the possibility of an alternative disposal of the washed fly ash alternatively to the current cement stabilization process. The technical feasibility whether the washed fly ash is valid for a disposal on a non hazardous waste landfill regarding Austrian landfill regulations or not is of major interest for a similar fly ash treatment process. It has to be considered to exclude the Zn recovery parts of the process and focus exclusively on the decontamination part of the evaluated process.

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