71/2015
Lederer, J.; Rechberger, H.; Fellner, J. (2015) “The Utilization of MSWI Fly Ashes in Cement Production and its Impact on Heavy Metal Contents In Cement” In: CD-Proceedings Sardinia 2015, 15th International Waste Management and Landfill Symposium, 5-9 Oct. 2015, S. Margherita di Pula (Cagliari), Sardinia, Ed.: Cossu, R., Pinjing, H.; Kjeldsen, P. Matsufuji, Y.; Reinhart, D.; Stegmann, R., CISA Publisher, Sardina, paper 83, p 1-9, ISSN 2282-0027

THE UTILIZATION OF MSWI FLY ASHES IN CEMENT PRODUCTION AND ITS IMPACT ON HEAVY METAL CONTENTS IN CEMENT

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SUMMARY: The disposal of fly ashes from municipal solid waste incineration (MSWI) is becoming a growing environmental issue in Europe. In order to avoid landfilling and recycle MSWI fly ashes, initiatives aim to recycle them in cement production. While numerous studies have analyzed the impact of MSWI ashes application on the concrete quality and the leaching of potentially hazardous heavy metals from MSWI ash amended concretes, only few studies have determined the consequential heavy metal content in cements if MSWI ashes would be added in large scale in cement production. Therefore, the aim of this study is to determine the inputs of selected heavy metals (Cd, Cr, Hg) into cement production and the heavy metal contents of cements after application of MSWI fly ashes on a national level, taking the case study of cement production in Austria.
In order to do so, data on cement production and corresponding consumption of primary and secondary raw materials and fuels in Austria is retrieved from the environmental reports of the Austrian cement industry. Afterwards, concentrations of heavy metals in the primary and secondary raw materials (and fuels) are retrieved from literature. Both are inserted in a material flow analysis (MFA) model using the STAN freeware in order to calculate the heavy metal concentrations in cements currently produced. Afterwards, a scenario is drafted, assuming that all MSWI fly ashes in Austria are used in the cement industry without prior treatment. The result of the calculation gives the final heavy metal content of cements after application of untreated MSWI fly ashes.
Results show that the application of untreated MSWI fly ashes will significantly increase the Cd content by a factor of five. Hg contents will rise by a factor of three, while the increases of Cr contents are low. Though a pre-treatment of MSWI fly ashes is required due to the salt contents of MSWI fly ashes, it will either only remove some fractions (neutral washing), or be relatively expensive (acidic washing). These results suggest that the secondary raw material value of MSWI in cement production is quite low, particularly if compared to other secondary raw materials currently used.

Proceedings Sardinia 2015, Fifteenth International Waste Management and Landfill Symposium
S. Margherita di Pula, Cagliari, Italy; 5 – 9 October 2015

1. INTRODUCTION

The disposal of fly ashes from municipal solid waste incineration (MSWI) is becoming a growing environmental issue in Europe. Common practices like stabilization and landfilling to hazardous and non-hazardous landfills are challenged by concerns on ecological sustainability (i.e. long-term emissions of salts and heavy metals from disposal sites, utilization of big amounts of cement for stabilization), financial restraints (i.e. high costs for MSWI fly ash treatment and disposal), and low recycling rates of secondary raw materials from MSWI fly ash (i.e. metals, minerals). In order to deal with these concerns, research ambitions aimed to find alternative ways of MSWI fly ash treatment and particularly utilization as secondary raw material. Beside recycling of salts or selected heavy metals in MSWI fly ashes (Ecke, 2003; Karlfeldt Fedje, 2012; Katsuura et al., 1996; Nagib and Inoue, 2000; Schlumberger et al., 2007), one of these ambitions focuses on the utilization of MSWI ashes rich in CaO, SiO2, Al2O3, and Fe2O3 in the building industry, either as filling material, or as a substitute in cement production (Aubert et al., 2007; Aubert et al., 2004; Collivignarelli and Sorlini, 2002; Ferreira et al., 2003; Gao et al., 2008; Katsuura et al., 1996; Kikuchi, 2001; Pan et al., 2008; Rémond et al., 2002; Saikia et al., 2007; Shi and Kan, 2009; Wu et al., 2011).

While numerous studies have analyzed the impact of MSWI fly ashes application on the concrete quality and the leaching of potentially hazardous heavy metals from MSWI fly ash amended concretes, only few studies have determined the consequential heavy metal content in cements if MSWI fly ashes would be added in large scale in cement production. Therefore, the aim of this study is to determine the inputs of selected heavy metals (Cd, Cr, Hg) into cement production by introducing MSWI fly ashes and the resulting heavy metal contents of cements after application on a national level, taking the case study of cement production in Austria (Lederer et al., 2015).

2. MATERIALS AND METHODS

MSWI fly ashes contain almost the whole range of heavy metals. Due to their environmental impacts, cadmium (Cd), chromium (Cr), and mercury (Hg) are of particular relevance. For this reason, this study will focus on these particular metals. To answer the research question of heavy metal contents in cements after MSWI fly ashes application, a method that has been successfully applied in prior studies of other authors is material flow analysis (MFA) (Dr. Graf AG, 1998; Fehringer et al., 1999). For this reason, the same method will be applied herein, using the program STAN (Cencic et al., 2012; Cencic and Rechberger, 2008).

The investigated system refers to cement production in Austria, as later shown in Figure 1. In the first step, all heavy metal containing input material flows of primary and secondary raw materials as well as primary and refuse derived energy sources are retrieved from Mauschitz (2013). Afterwards, the concentrations of the selected heavy metals in the input material flows are taken from literature and multiplied with the material flows of inputs, in order to calculate the inputs of the selected heavy metals into the cement production process. The material flows and heavy metal concentrations used in this study are shown in Table 1.

Furthermore, the heavy metal outputs in off-gas are taken from Mauschitz (2013). As the only other output flow from the process is the cement produced, it is then possible to calculate the transfer coefficient of heavy metal inputs to i) the off-gas and ii) the cement produced. Thus, the results of this calculation are i) heavy metal inputs into cement production by primary and secondary raw materials as well as primary and refuse derived fuels, ii) heavy metal outputs through off-gas and cement, and iii) the transfer coefficients of the total heavy metal inputs to off-gas on the one hand and cement on the other hand.

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Table 1. Quantity of cement, clinker, and cement production input materials after Mauschitz (2013) and concentrations of Cd, Cr, and Hg in cement production input materials as well as MSWI fly ashes, based on differend literature sources (see legend).

Input material	Group	Quantity [kt/yr]	Metal concentration [mg/kg] Cd	Cr	Hg	Literature concentrations
Cement	Product	4.446				
Clinker	Clinker	3.227				
Primary raw mix	Primary raw material	4.547	0,15	39	0,05	1)
Slags and sands	Secondary raw material	60	0,50	32	0,20	2), 3)
Debris		423	1,00	76	0,10	2), 3)
Hard coal	Primary fuels	58	0,10	15	0,16	4)
Lignite		65	0,10	39	0,20	4); 5)
Heating oil (light+heavy)		7	1,25	0,63	0,004	6)
Natural gas		3	0	0	0	
Petrol choke & others		25	1,25	0,63	0,004	6)
Waste tyres	Refuse-derived-fuels (RDF)	31	7,50	54	0,001	7); 8); 9)
Plastics		226	4,15	125	0,28	10)
Waste oil		11	3,00	80	0,20	9);11);12)
Waste solvents		14	3,00	80	0,20	9);11);12)
Sewage sludge		27	0,91	38	0,77	13)
Waste fibers		39	2,00	20	0,30	9);14);15)
Slaughter wastes		16	0,22	5	0,10	6);16);17)
Other RDFs		41	2,97	57	0,54	7)-17)
Blast furnace slag	Secondary milling materials	617	0	38	0	2); 18); 21)
Coal fly ash		125	0,50	150	0,10	1); 4)
APC gypsum		48	0,15	27	0,25	1); 4)
Other minerals		98	0,38	71	0,18	1);2);4);18)
Other milling material	Primar milling materials		0,15	39	0,05	1)
MSWI fly ash	*Scenario*	*45*	*291*	*386*	*25*	*19); 20)*

Literature sources: 1) Zeschmar-Lahl (2003); 2) Nonte (undated); 3) Brameshuber and Vollpracht (2003); 4) Winter et al. (2005); 5) Böhmer et al. (2001); 6) Grech et al. (2001); 7) Fehringer et al. (1999); 8) (2003); 9) Braun (2001); 10) Denner (2009); 11) Möller (2004); 12)VDZ (1999); 13) Kügler et al. (2004); 14) Walter and Tesar (2009); 15) Boubela et al. (2004); 16) Garcia and Rosentrater (2008); 17) Cascarosa et al. (2012); 18) Chernousov and Golubev (2011); 19) Böhmer (2007); 20) Skutan and Rechberger (2007); 21) Proctor et al. (2000)

In the final step, it is assumed that all MSWI fly ash from Austrian MSW grate incinerators are used in cement production as secondary raw material, introduced to the clinker production process. The quantities of MSWI fly ashes and their heavy metal contents are taken from national data sources (see Table 1). Thus, the total input of heavy metals into the cement production process will be increased by the heavy metals introduced by MSWI fly ash. By assuming that the transfer coefficients calculated before remain stable, the increase in i) heavy metal emissions through the off-gas and ii) heavy metal flows and thus concentrations in the cement produced can be calculated.

3. RESULTS

Results show that the Austrian cement industry has a high share of both, refuse derived fuels (RDFs), as well as secondary raw materials and secondary milling materials. The clinker share in the cement is around 70%. Cadmium and mercury are currently mainly introduced into the cement by RDFs from plastics, and to a lesser extends by primary raw materials. Contrary to that, primary raw materials are the main source of chromium input.

For the scenario, it was assumed that all MSWI fly ashes from Austrian grate incinerators will be introduced, which makes in total 45,000 tons per year. As can be seen in Figure 1, this is only 1%

of the total input of raw materials into the clinker production process. Accordingly, the input of Cr into cement production is comparatively small. This is mainly due to the relatively low concentration in combination with the low total material input of MSWI fly ash into the process. The most important input flow for Cr is the mix of primary raw materials (see Figure 2).

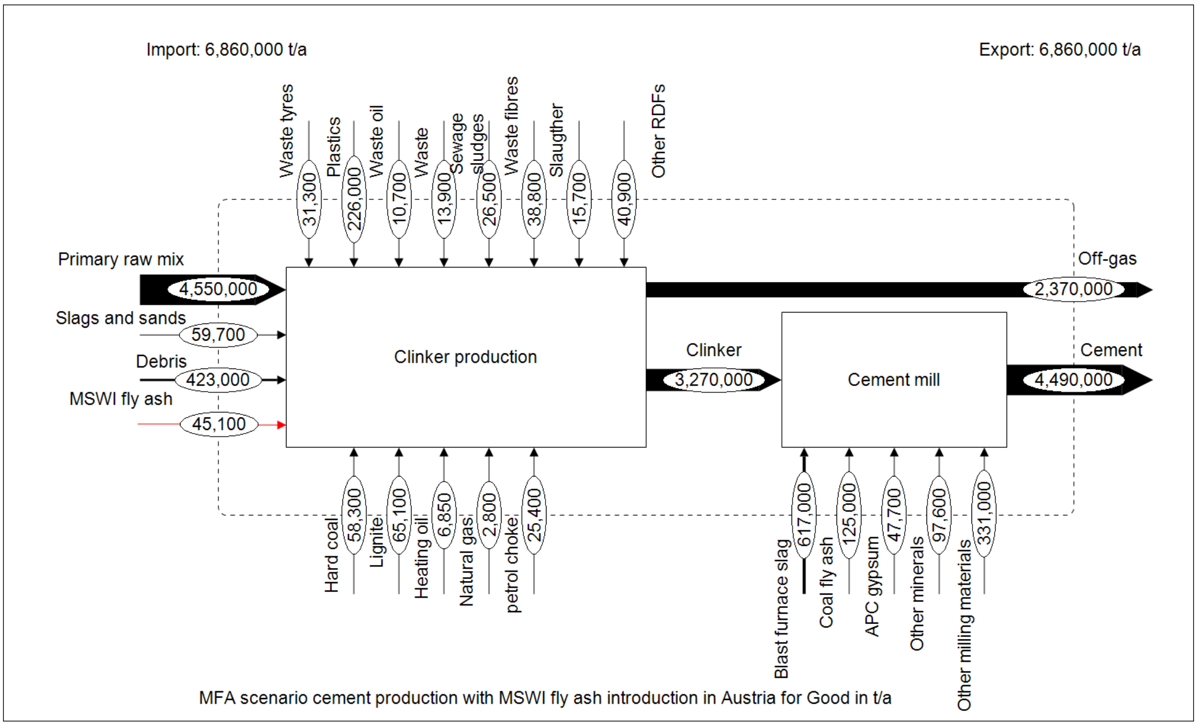

Figure 1. MFA scenario for introducing MSWI fly ash into Austrian cement production – calculation for total material flows.

However, contrary to the total input material flows and the input of Cr, the input of Cd by introducing untreated MSWI fly ash is relatively high (see Figure 3). The same counts for Hg (see Figure 4). For both substances, the reason is that the concentrations in MSWI fly ashes are significantly higher than for all other primary and secondary raw materials and fuels which are currently applied in the Austrian cement industry (see Table 1).

By assuming in the calculation that the transfer coefficients of heavy metals in input materials distributing to the two output flows (off-gas and cement) are stable, meaning not depending on the heavy metal input, the consequences for the concentrations of Cd, Cr, and Hg in the MSWI fly ash containing cement produced are clear: a negligible increase in Cr concentrations is faced by a large increase of the concentrations of Hg by a factor of two, while the increase of Cd concentrations in cements are up to a factor of five (see Table 2).

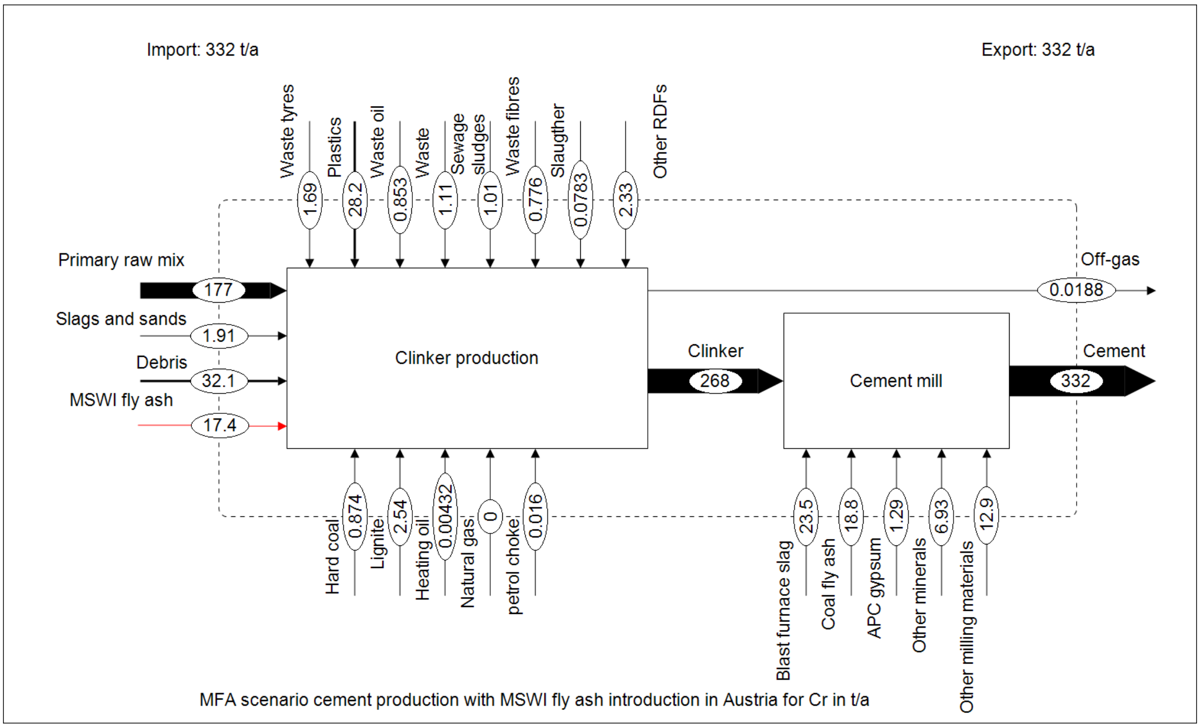

Figure 2. MFA scenario for introducing MSWI fly ash into Austrian cement production – calculation for Cr flows.

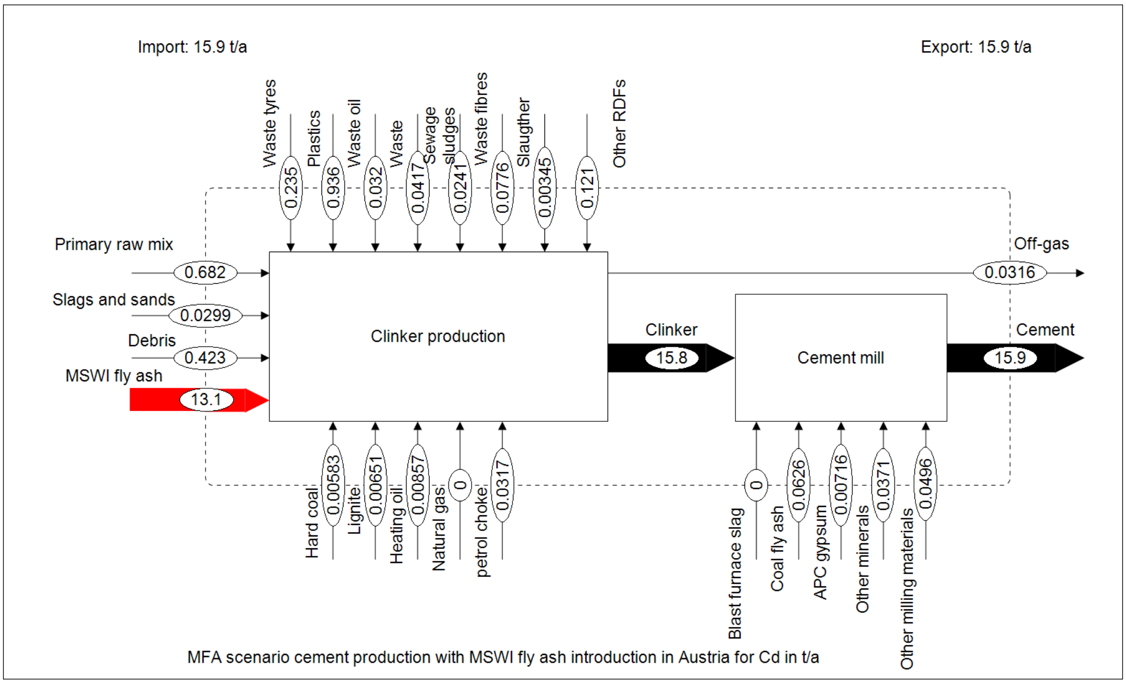

Figure 3. MFA scenario for introducing MSWI fly ash into Austrian cement production – calculation for cadmium flows.

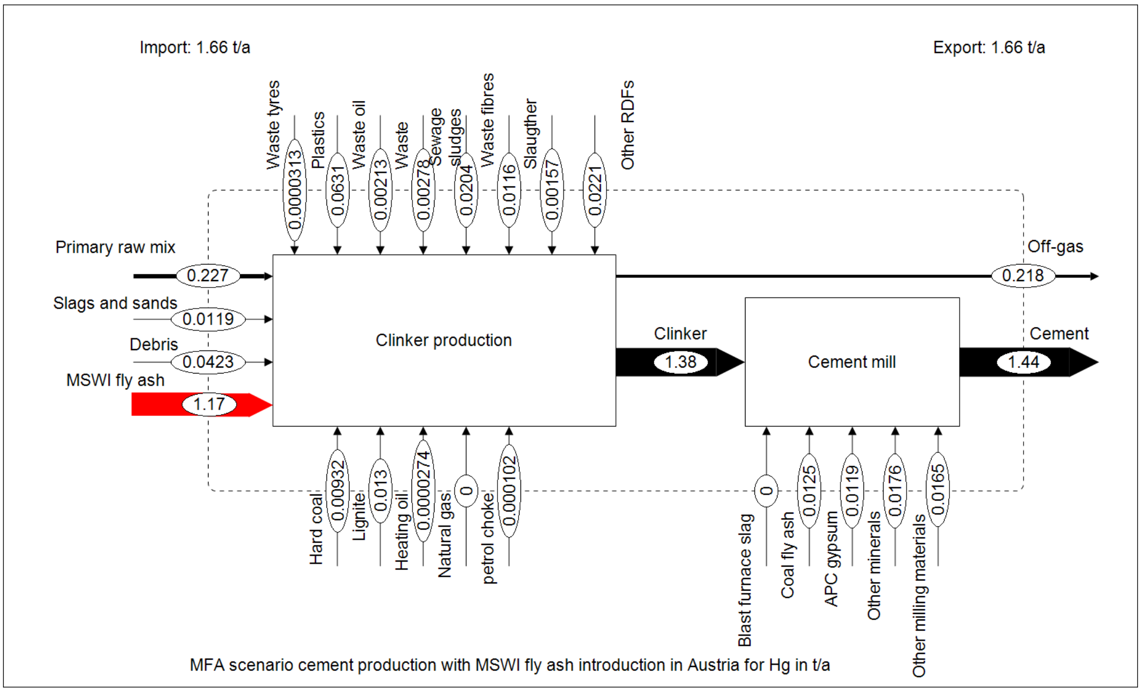

Figure 4. MFA scenario for introducing MSWI fly ash into Austrian cement production – calculation for mercury flows.

Table 1. Resulting concentrations of selected heavy metals (Cd, Cr, and Hg) in cement by introducing MSWI fly ash.

		Unit	Cd	Cr	Hg
Calculated concentrations in cement with current raw material mix		mg/kg	0.63	71	0.10
Calculated concentrations in cement with introduction of MSWI fly ashes		mg/kg	3.54	74	0.32
Increase of calculated concentrations in cement through MSWI fly ashes		%	+460%	+4%	+220%
Averatge concentrations in selected Austrian cements [1]		mg/kg	0.80	42	nd
Average concentrations in European cements according to literature [2),3),4),5)]	min	mg/kg	0.03	41	0.01
	mean	mg/kg	0.40	53	0.15
	max	mg/kg	0.70	70	0.37

Literature: 1) Murphy (2013); 2) VDZ (2001); 3) Anger et al. (2011); 4) Dr. Graf AG (1998)5) Zeschmar-Lahl (2003); nd…not data available.

4. DISCUSSION AND CONCLUSION

The results show that the ambitions of introducing untreated MSWI fly ashes from great incinerators into cement production on a national scale will lead for some substances to a significant increase of their concentrations in cement, particularly Cd and Hg. Though the leachability of Cd and Hg from the concrete produced by using this cement has been estimated as small to negligible by other authors, other problems might appear. For instance, the new Austrian ordinance on recycling of construction and demolition waste contains a criterion on total heavy metal contents for secondary raw materials produced from crushed concrete (Starke, 2015). It is not clear yet if a concrete produced from MSWI fly ash containing concrete will meet this criterion.

Due to the high salt content of MSWI fly ashes, most authors that have investigated the utilization of MSWI fly ashes in cement production suggest some treatment before utilization. This is usually some wet treatment (fly ash washing). If this treatment is on a neutral basis (water, ph 7), it is likely that some heavy metals will not be dissolved to a sufficient extend, and thus remain in the washed MSWI fly ash. This counts for instance for Hg. Therefore, acidic leaching, as currently sometimes practiced with the acidic washing water of MSWI treatment plant before landfilling the MSWI fly ashes, will be a more successful strategy to desalinate and de-metalize the MSWI fly ashes before utilization in cement production. However, it must be beard in mind that for other secondary raw materials usually used in cement production (e.g. brick debris, blast furnace slags, coal fly ashes), no such intensive and expensive pre-treatment before utilization is required.

AKNOWLEDGEMENTS

The presented work is part of a large-scale research initiative on anthropogenic resources (Christian Doppler Laboratory for Anthropogenic Resources). The financial support of this research initiative by the Federal Ministry of Economy, Family and Youth and the National Foundation for Research, Technology and Development is gratefully acknowledged.

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